



Commission of the European Communities

# radiation protection - 53

Proceedings of  
**Seminar on**  
**Comparative Assessment of the Environmental**  
**Impact of Radionuclides Released during**  
**Three Major Nuclear Accidents:**  
**Kyshtym, Windscale, Chernobyl**

Luxembourg, 1-5 October 1990

**Volume II**



**Report**

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## Volume II

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# **Radioecological Monitoring of the Black Sea Basin Following the Chernobyl NPP Accident**

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## ABSTRACT

A monitoring programme was drawn up to study the radioecological situation of the Black Sea basin following the Chernobyl NPP accident, with studies being carried out from May 1986 onwards to determine the levels of radioactive contamination in various parts of the Black Sea, the Sea of Azov and the Aegean Sea, including the estuaries of major rivers (Dnieper, Danube, Dniester and Don) and shelf areas of the Black Sea and the Sea of Azov.

The work focused on long-lived radionuclides ( $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ ), with the migration dynamics of these radionuclides in the aquatic environment, bed sediments and aquatic biota (including plants, molluscs, crustacea and fish) being studied. We compared the behaviour of radionuclides in the aquatic environment of the Dnieper reservoirs following the Chernobyl accident (our data) with the behaviour of radionuclides in lakes in the Urals following the Kyshtym accident (published data). As in the case of the lakes in the Urals, the Dnieper waters contain substantial concentrations of  $^{90}\text{Sr}$  as a result of the Chernobyl accident, and  $^{90}\text{Sr}$  therefore enters the Black Sea with the Dnieper waters. The paper compares the contribution of the Chernobyl accident to radioactive contamination of the Black Sea with that of global fallout.

Radioecological research has been carried out in the Black Sea Basin by staff of the Southern Seas Institute of Biology since the beginning of the 1960s (Marine radioecology, 1970<sup>10</sup>; Polikarpov et al., 1970<sup>15</sup>; Sokolova, 1971<sup>19</sup>; Kulebakina, Zesenko, 1984<sup>8</sup>).

Thus, when the news of the Chernobyl accident first broke, we started to take samples in the Sevastopol rayon (district) and later, as part of a more extensive programme of radioecological monitoring, in the Black Sea, the Sea of Azov and the Aegean Sea, the estuaries of the Danube, Dniester, Dnieper and Don Rivers and in the small inland basins and rivers of the Crimea. The main components of the ecosystems (water, bed sediments, aquatic plants, molluscs, crustacea and fish) were analysed. This paper presents the results of observations made between 1986 and 1989 of the main long-lived, biologically significant radionuclides (<sup>90</sup>Sr and <sup>137</sup>Cs).

Gamma-spectrometry measurements were carried out using Al-1024 gamma analyzers with NaI(Tl) crystals (150x100 mm) and, from 1988 onwards, Ge(Li) detectors with a volume of 98 cm<sup>3</sup>. A low-background UMF-1500 unit with SBT-13 counters was used for beta-radiometry. The radiochemical methods used and the radiometrical measurements taken were subjected to international calibration.

### <sup>137</sup>Cs

In the aftermath of the Chernobyl accident the maximum <sup>137</sup>Cs concentrations in the surface waters of the Black Sea reached 815-840 mBq·l<sup>-1</sup> in the summer of 1986. Prior to the Chernobyl accident the <sup>137</sup>Cs concentration in the Black Sea water amounted to 19.61±1.11 mBq·l<sup>-1</sup> (Vakulovsky et al., 1980). In the course of 1986 considerable heterogeneity was observed in the distribution of the <sup>137</sup>Cs concentrations in surface waters of the Black Sea: the values ranged from 18.5 to 815-840 mBq·l<sup>-1</sup> (Polikarpov, Kulebakina, 1989). Between 1986 and 1989 hydrophysical and biological processes led to redistribution and migration of <sup>137</sup>Cs into the depths of the water which led, in turn, to a reduction in the maximum <sup>137</sup>Cs concentration and an increase in its minimum level in the surface waters of the Black Sea (Fig. 1).

Other scientists who carried out radioecological studies in the Black Sea in 1986 produced similar data on the concentration of <sup>137</sup>Cs in surface waters: 518 Bq·m<sup>-3</sup> in the eastern area of the Black Sea and 250-340 Bq·m<sup>-3</sup> north of the Bosphorus (Nikitin et al., 1988; Livingston et al., 1986; Buesseler, 1987).

The irregular, "patchy" distribution of <sup>137</sup>Cs in the surface waters of the Black Sea, which was observed in 1986, tended to obscure the latitudinal reduction in <sup>137</sup>Cs fallout levels from north to south which was clearly seen in 1987 (Fig. 2) and 1988.

However, as early as 1989 the concentration of <sup>137</sup>Cs in the surface waters of the shallow north-west area of the Black Sea was lower than in the southern area around the Bosphorus (Fig. 2). This can be explained by the flow of the Danube and Dnieper Rivers which had lower <sup>137</sup>Cs concentrations than those of the water in the Black Sea and therefore diluted the <sup>137</sup>Cs contamination of the Black Sea.

In the Lower Danube (town of Vil'kovo) the concentrations of <sup>137</sup>Cs between 1986 and 1989 ranged from 7.4 to 25.9 mBq·l<sup>-1</sup>, the maximum

concentration being  $55.5 \text{ mBq}\cdot\text{l}^{-1}$  (Kulebakina, Polikarpov, 1990). In the Lower Dnieper (town of Kherson) the  $^{137}\text{Cs}$  concentrations in the water ranged from  $3.7$  to  $18.5 \text{ mBq}\cdot\text{l}^{-1}$ . The maximum concentrations ( $92.5$  and  $181.3 \text{ mBq}\cdot\text{l}^{-1}$ ) were observed during the spring flood of 1987. The  $^{137}\text{Cs}$  concentration in the estuaries of these rivers therefore fell between 1986 and 1989 (Fig. 3). The influence of river waters is also confirmed by the direct correlation between salinity and the  $^{137}\text{Cs}$  concentration (Fig. 4).

Throughout this period the maximum concentrations in the surface waters of the Black Sea decreased and the minimum concentrations increased. The histograms (Fig. 5) show the percentage distribution of  $^{137}\text{Cs}$  concentrations in the years concerned. They indicate that, in the autumn of 1987, 34% of the sample ( $n = 0.54$ ) had a concentration of between  $77.7$  and  $92.5$ , in the autumn of 1988 36% ( $n = 0.60$ ) of the sample had a concentration of between  $59.2$  and  $74.0$  and in the spring of 1989 51% ( $n = 0.33$ ) of the water had a concentration of between  $40.7$  and  $55.5 \text{ mBq}\cdot\text{l}^{-1}$ . Irregular "patchy" contamination of the area of water as a result of atmospheric fallout in 1986 was also observed in the Dnieper-Bug Estuary (Ilman) where  $^{137}\text{Cs}$  concentrations ranged from  $3.7$  to  $470 \text{ mBq}\cdot\text{l}^{-1}$ . Prior to the Chernobyl accident the  $^{137}\text{Cs}$  concentration in this area ranged from  $3.7$  to  $7.4 \text{ mBq}\cdot\text{l}^{-1}$  (Vintsukevich, Tomilin, 1987).

The maximum  $^{137}\text{Cs}$  concentrations in the water of the Lower Dnieper were observed in the 1987 spring flood (March-May) as a result of the removal of soil particles with adsorbed radionuclides from the upper areas of the Dnieper River basin situated in the most contaminated area. In the autumn of 1988 we mapped the radioactive contamination level of the Dnieper Waters (Fig. 7), from the cooling pond of the Chernobyl NPP (10-km zone), down the Dnieper and through all the Dnieper reservoirs to the Dnieper-Bug Estuary.

The results of this research showed that the  $^{137}\text{Cs}$  concentration levels remained high in the upper areas of the Dnieper (Fig. 7).

In 1988 the concentrations of  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  in the Chernobyl cooling pond were  $21.7 \text{ Bq}\cdot\text{l}^{-1}$  and  $2.8 \text{ Bq}\cdot\text{l}^{-1}$  respectively, the figures for the Pripyat River (settlement of Kopachi) were  $3.1$  and  $0.44 \text{ Bq}\cdot\text{l}^{-1}$  respectively, while in the Kiev and Kremenchug reservoirs the figures were  $0.188$  and  $0.033 \text{ Bq}\cdot\text{l}^{-1}$  respectively. Further towards the Black Sea the Dnieper reservoir system facilitated the settling of suspended particles with adsorbed radiocaesium, thereby affecting the level not only of  $^{90}\text{Sr}$  (Polikarpov, Timoshchuk, Kulebakina, 1987), but also in particular of  $^{137}\text{Cs}$ , since  $^{137}\text{Cs}$  is typically found in the form of compounds of low solubility (Kuznetsov, Generalova, 1989) and the majority of the radiocaesium in the Chernobyl NPP zone in the pluvial runoff period migrates in solid form (Perepeyatnikova et al., 1989).

Between 1987 and 1989 the  $^{137}\text{Cs}$  concentration ranged from  $3.7$  to  $18.5 \text{ mBq}\cdot\text{l}^{-1}$ , higher levels being observed only during the spring floods. An increase in the  $^{137}\text{Cs}$  concentration in rivers as a result of the spring flood was also observed for global radionuclide fallout prior to the Chernobyl accident (Bochkov et al., 1983).

Atmospheric fallout contaminated bed sediments with radionuclides fairly quickly. Radioactive contamination of bed sediments in the shelf area of the Black Sea and the lower part of the Dnieper River was also of a "patchy" nature: the concentrations of  $^{137}\text{Cs}$  in 1986 ranged from

3 to 122 Bq·kg<sup>-1</sup> wet mass. The <sup>137</sup>Cs concentration level in Black Sea bed sediments increased in certain places by two orders of magnitude in comparison with the level observed prior to the Chernobyl accident (Kulebakina, Zesenko, 1984).

Radiocaesium passed rapidly through the waters onto the bed because the Chernobyl accident produced a high level of fuel particles and aggregated forms of radionuclides (Informatsiya, 1986). This was recorded by direct observations using traps in the Black Sea and the Mediterranean Sea (Buesseler, 1987; Fowler et al., 1987). Thus, although, in the case of global fallout, an increase was observed in the specific activity of <sup>137</sup>Cs in bed sediments with depth (Yegorov et al., 1986), our observations revealed the opposite, namely a decrease in <sup>137</sup>Cs concentration in bed sediments with depth (Fig. 8). "Patchy" contamination of bed sediments was also observed in the lower part of the Dnieper (from Zaporozhe to the Dnieper-Bug Estuary). <sup>137</sup>Cs concentrations of between 2.44 and 38.18 Bq·kg<sup>-1</sup> were observed in the Kherson area from June to September 1986.

Subsequently we observed horizontal and vertical migration of radiocaesium in bed sediments (Fig. 9).

Of all the aquatic biota in the Black Sea, green and red algae are among the bioindicators of radiocaesium. Observations of the change in the <sup>137</sup>Cs concentration in sea lettuce (Ulva rigida) showed that the maximum <sup>137</sup>Cs concentrations observed in macrophytes in the summer of 1986 were between 12.76 and 22.2 Bq·kg<sup>-1</sup> of wet mass and quickly decreased after a fall in the <sup>137</sup>Cs concentration in water. In 1988-1989 the <sup>137</sup>Cs concentrations in Black Sea algae ranged from 0.67 to 6.74 Bq·kg<sup>-1</sup>, the levels in Cystoseira crinita brown algae being higher than in Ulva rigida green algae. This can be explained by the fact that Cystoseira is a perennial alga, whereas sea lettuce is an annual.

Some 140 varieties of plant grow in the Dnieper reservoirs and the Lower Dnieper, 69 of which are higher aquatic plants (Korelyakova, Gorbik, 1989) which differ in their ecological and biological features. We studied various representatives of the ecological and biological groups: submerged plants (parrot feather, hornwort, pondweed), plants with vegetative organs which float on the surface of the water (cow lily, naiad, wild celery) and amphiphytes (common reed, great bulrush).

The highest <sup>137</sup>Cs accumulation coefficients were observed in thorrowort pondweed (Potamogeton perfoliatus) (750 - 1 164 - 2 005), Canada parrot feather (Myriophyllum verticillatum) (983 - 1 006 - 1 143 - 2 508) and fennel-leaved pondweed (Potamogeton pectinatum) (1 053 - 1 000).

Prior to the Chernobyl accident the <sup>137</sup>Cs concentrations in aquatic vegetation (cladophora, pondweed, cat's-tail, duck's-meat) of the Yuzhny Bug River ranged from 0.12 to 7.94 Bq·kg<sup>-1</sup> (Vintsukevich, Tomilin, 1984), with higher <sup>137</sup>Cs accumulation being recorded in the case of thorrowort pondweed.

The dynamics of the <sup>137</sup>Cs concentration in Dnieper plants between 1986 and 1988 (Figs. 10, 11) reflected the dynamics of the <sup>137</sup>Cs concentration in the water of the Dnieper River. We have no data on the <sup>137</sup>Cs concentrations in Dnieper plants prior to the Chernobyl accident.

After the Chernobyl accident an increase was observed in the maximum concentrations of  $^{137}\text{Cs}$  in plants of the Yuzhny Bug Estuary: 60 times in the case of amphiphytes (bulrush, cat's-tail, reed), seven times in the case of pondweed and 10 times in the case of cladophora.

It was essential to record not only the visible differences but also the ecological and biological differences in radiocaesium accumulation in aquatic plants. In 1986, the highest  $^{137}\text{Cs}$  concentrations were recorded in amphiphytes (up to  $138 \text{ Bq}\cdot\text{kg}^{-1}$  for great bulrush (Scirpus lacustris) or  $24.67 \text{ Bq}\cdot\text{kg}^{-1}$  for the common reed (Phragmites australis)), this direct plant contamination being caused by atmospheric fallout.

The same factors may account for the high concentrations of  $^{137}\text{Cs}$  in 1986 in common duckweed (Lemna minor) ( $77 \text{ Bq}\cdot\text{kg}^{-1}$ ), which is a submerged plant but has leaves floating on the surface.

Subsequently, when aquatic plants accumulated radionuclides primarily from water, submerged plants displayed the greatest capacity for radiocaesium concentration.

$^{137}\text{Cs}$  accumulation in molluscs and fish of the Black Sea and the Dnieper River was of a more complex nature than that in plants. In the case of Black Sea mussels, the  $^{137}\text{Cs}$  concentration in 1986 reached  $55.5 \text{ Bq}\cdot\text{kg}^{-1}$  in soft tissue and  $14.8 \text{ Bq}\cdot\text{kg}^{-1}$  in whole molluscs. Subsequently, after the  $^{137}\text{Cs}$  concentration in mussels fell in 1987, it began to rise again in 1988 (Fig. 12).

The same dynamics of  $^{137}\text{Cs}$  accumulation were noted in the case of Black Sea fish (Fig. 13), and in Dnieper molluscs (Fig. 14) and Dnieper fish (Fig. 15). A wide range of values was recorded, ranging from minimum values (of the order of  $0.5 \text{ Bq}\cdot\text{kg}^{-1}$ ) corresponding to the  $^{137}\text{Cs}$  concentration level in Black Sea fish prior to the Chernobyl accident (Kulebakina, Zesenko, 1984), to maximum values ( $30 \text{ Bq}\cdot\text{kg}^{-1}$ ), which are 60 times higher than the pre-Chernobyl values. An analysis of the dynamics of the  $^{137}\text{Cs}$  concentration for particular species of fish (Fig. 16) shows how this process is dependent on the trophic level of fish in the food chains: an increase in the trophic level increases the  $^{137}\text{Cs}$  accumulation: in the case of first-degree consumers (annular bream (Diplodus annularis), sprats (Sprattus sprattus sprattus), Mediterranean scad (Trachurus mediterraneus euxinus) etc.) the ratio between the  $^{137}\text{Cs}$  concentrations and  $^{40}\text{K}$  ranged from 0.024 to 0.135; in the case of second-degree consumers (whiting (Qdontogadus merlangus), goby (Gobius sp.), peacock wrasse (Crenilabrus tinca), stingray (Dasyatis pastinaca) etc.), the ratio ranged from 0.154 to 0.203; in the case of third-degree consumers (scorpion fish (Scorpaena porcus), shark (Squalus acanthius) and Blacksea turbot (Scophthalmus maeoticus maeoticus)), the ratio ranged from 0.272 to 0.834.

### $^{90}\text{Sr}$

In addition to  $^{137}\text{Cs}$ , we also studied  $^{90}\text{Sr}$ , another long-lived radionuclide. As a result of nuclear explosions in the atmosphere, the quantity of  $^{90}\text{Sr}$  released in 1980 ( $6.04\cdot 10^{17} \text{ Bq}$ ) was almost the same as that of  $^{137}\text{Cs}$  ( $9.6\cdot 10^{17} \text{ Bq}$ ) (Sources, 1988).

The Chernobyl accident released  $8.1\cdot 10^{15} \text{ Bq}$  of  $^{90}\text{Sr}$  into the atmosphere (Informatsiya, 1986), which is one order of magnitude less than the quantity of  $^{137}\text{Cs}$  released. The biogeochemistry of

radiostrontium in aquatic ecosystems and its biological action differ from the behaviour of  $^{137}\text{Cs}$ .

In 1986 the  $^{90}\text{Sr}$  concentrations in surface waters of the Black Sea following the Chernobyl accident ranged from 16.6 to 157  $\text{mBq}\cdot\text{l}^{-1}$ . The maximum concentrations of  $^{90}\text{Sr}$  we recorded in June were five times lower than those of  $^{137}\text{Cs}$ . In the Dnieper-Bug Estuary and the Lower Dnieper in September 1986 the concentrations of  $^{90}\text{Sr}$  ranged from 21.1 to 20.4  $\text{mBq}\cdot\text{l}^{-1}$  (translators note: sic) and were usually higher than the concentrations of  $^{137}\text{Cs}$ .

Following the Chernobyl accident the concentrations of  $^{90}\text{Sr}$  in the surface waters of the Black Sea decreased between 1986 and 1989, but the dynamics of this process for  $^{90}\text{Sr}$  differed from those for  $^{137}\text{Cs}$  (Fig. 17). As a result, there was a change in the ratio between the concentrations of  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ , as shown in Fig. 18 for the area of water near the coast of the Crimean peninsula. This can be explained by differences in the biogeochemical behaviour of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  in the aquatic environment.

In this period a gradual increase in the concentration of  $^{90}\text{Sr}$  in the water was observed in the lower part of the Dnieper River as a result of the arrival of more highly contaminated water from the upper part of the Dnieper (Fig. 19). The maximum concentration of  $^{90}\text{Sr}$  in the water of the Lower Dnieper occurred during the spring flood of 1987 (Fig. 20) when the maximum concentrations of  $^{90}\text{Sr}$  in the Kakhovka reservoir amounted to 910-992  $\text{mBq}\cdot\text{l}^{-1}$ . The gradient of  $^{90}\text{Sr}$  concentration from Zaporozhe (the uppermost point of the Kakhovka reservoir) to the Dnieper-Bug Estuary remained the same. In 1988-1989 we recorded a reduction in the concentration of  $^{90}\text{Sr}$  in the lower part of the Dnieper River and its virtually uniform distribution between Zaporozhe and Kherson. In 1988 the  $^{90}\text{Sr}$  concentration in the water of the Lower Dnieper ranged from 281 to 397  $\text{mBq}\cdot\text{l}^{-1}$ , while at the end of 1988 it ranged from 192 to 289  $\text{mBq}\cdot\text{l}^{-1}$ ; in 1989 it ranged from 211 to 282  $\text{mBq}\cdot\text{l}^{-1}$  in the Kakhovka reservoir, and from 290 to 341  $\text{mBq}\cdot\text{l}^{-1}$  near the town of Kherson. However, it is essential to take account of the fact that the upper section of the Dnieper River, which formed part of the area of maximum  $^{90}\text{Sr}$  contamination following the Chernobyl accident, will continue to be a source of  $^{90}\text{Sr}$  uptake further down the Dnieper to the Black Sea (Fig. 21). Although in the area of the Black Sea adjacent to the Dnieper Estuary the  $^{90}\text{Sr}$  concentrations in 1986-1987 were comparable to those at other points in the north-western part of the Black Sea, there was an increase in the  $^{90}\text{Sr}$  concentration in this area as early as 1988 as a result of removal with Dnieper waters (Fig. 21).

According to our calculations (Table 1, Fig. 22) based on data on the mean monthly flow of the Dnieper and the dynamics of the  $^{90}\text{Sr}$  concentration in the water of the lower part of the Dnieper, in all  $39.7\cdot 10^{12}$  Bq  $^{90}\text{Sr}$  entered the Black Sea with Dnieper waters between 1986 and 1989, which amounts to approximately 0.5% of all the  $^{90}\text{Sr}$  released into the atmosphere during the Chernobyl accident. Maximum  $^{90}\text{Sr}$  removal took place during the spring flood of 1987 (Fig. 23) due to  $^{90}\text{Sr}$  concentrations in the water and water flow being at their highest.

In the lower part of the Dnieper we observed movement of  $^{90}\text{Sr}$  downstream to the Black Sea with bed sediments as well (Fig. 24).

The maximum  $^{90}\text{Sr}$  concentrations recorded by us in the spring of 1987

In the water of the Lower Dnieper were accompanied by the highest levels of  $^{90}\text{Sr}$  in bed sediments (Fig. 25).

As for accumulation of  $^{90}\text{Sr}$  by Black Sea aquatic biota, we observed, above all, an increase of several times in its concentration in algae in comparison with pre-Chernobyl levels (Kulebakina, 1970<sup>7</sup>; Parchevsky, Kulebakina, Sokolova, 1971<sup>12</sup>; Sokolova, 1971<sup>19</sup>), and its dynamics in the brown alga Cystoseira corresponded to the dynamics of the  $^{90}\text{Sr}$  concentration in water. Of all the plants in the Lower Dnieper which we studied, the ones with the highest  $^{90}\text{Sr}$  accumulation levels were members of the pondweed family (up to  $52.5 \text{ Bq}\cdot\text{kg}^{-1}$ ), hornwort and common cat's-tail. The  $^{90}\text{Sr}$  concentration in the plants of the Lower Dnieper also reflected the dynamics of the  $^{90}\text{Sr}$  concentration in the water, maximum values being observed in 1987 (Fig. 26).

The  $^{90}\text{Sr}$  concentrations in Black Sea mussels reached their highest levels in 1986, after which they fell in 1987 and increased again slightly in 1988 (Fig. 27).  $^{90}\text{Sr}$  accumulation was observed in Dnieper molluscs between 1986 and 1988 (Fig. 28). The maximum  $^{90}\text{Sr}$  levels in Dreissenacea were observed in 1988 ( $432.4 \text{ Bq}\cdot\text{kg}^{-1}$  of wet mass). The pre-Chernobyl  $^{90}\text{Sr}$  concentration levels in Black Sea mussels and fish increased several-fold in 1986 (Fig. 29), this increase varying for the different species (Fig. 30). The maximum  $^{90}\text{Sr}$  levels for Black Sea animals were recorded in 1986, while  $^{90}\text{Sr}$  accumulation in Dnieper fish and molluscs rose from 1986 to 1988 (Fig. 31).

It can be assumed that  $^{90}\text{Sr}$  accumulation in molluscs and fish takes place not only from water but also via food although, in contrast to  $^{137}\text{Cs}$ , no clear relationship was observed between  $^{90}\text{Sr}$  accumulation and the trophic level of the aquatic biota.

The following general conclusions can therefore be drawn from the four-year radioecological research carried out in the Black Sea Basin:

1. The main contamination of the Black Sea ecosystems following the Chernobyl accident resulted from atmospheric fallout, and  $^{137}\text{Cs}$  is mainly to blame (long-lived radionuclides).
2.  $^{90}\text{Sr}$  enters the Black Sea with water from the Dnieper River.  $^{137}\text{Cs}$  movement is restricted by its non-soluble compounds in soils and bed sediments.
3. The  $^{90}\text{Sr}$  accumulation process is continuing in the molluscs and fish of the Dnieper River.

Given the high levels of contamination of the upper part of the Dnieper Basin with long-lived radionuclides, there is still a danger that these radionuclides will move into the Black Sea via river flow.

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Fig. 1: Dynamics of the concentration of Cs-137 in surface waters of the Black sea (1986-1990)

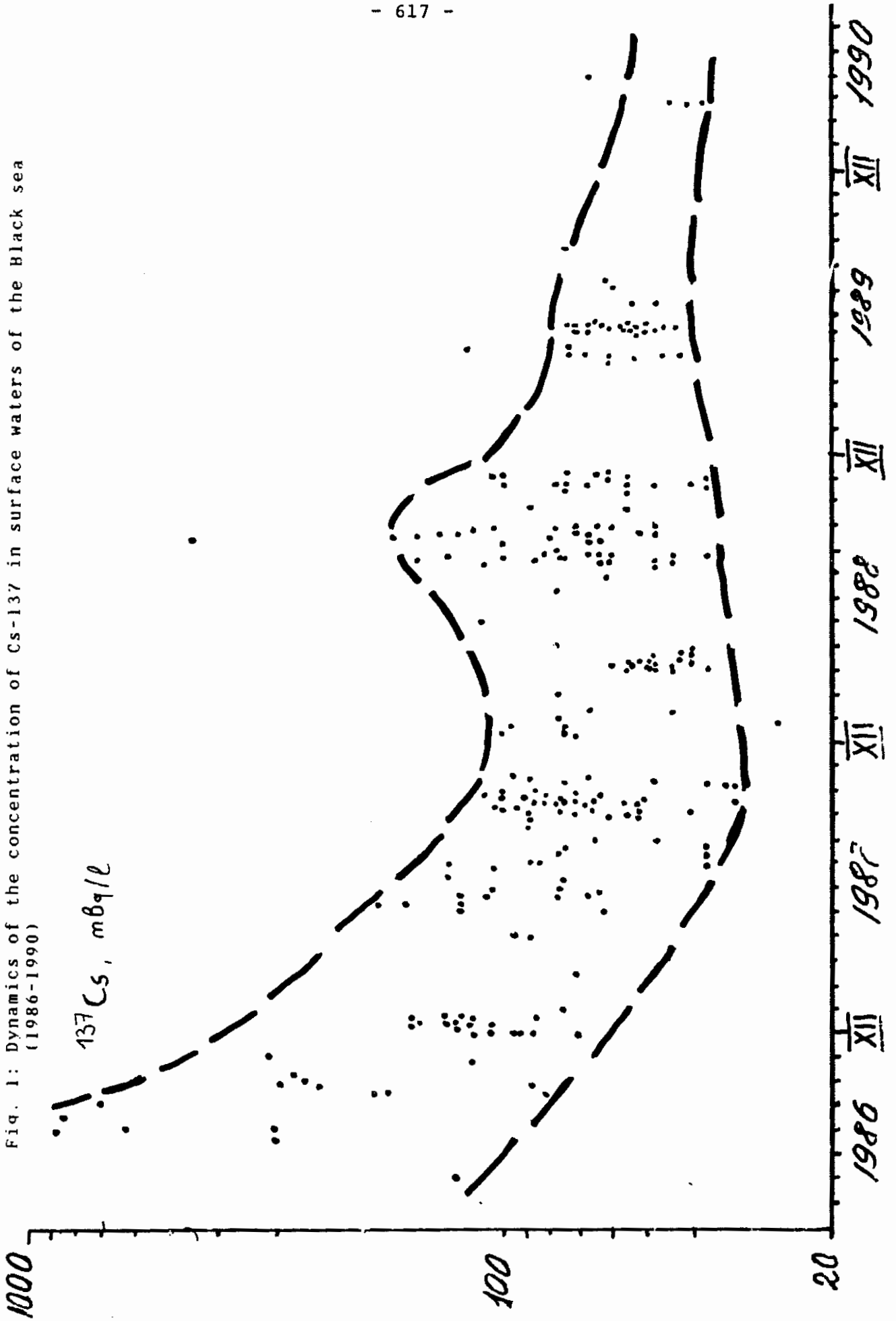


Fig. 2: Latitudinal distribution of Cs-137 in the western area of the Black sea from the Dnieper estuary to the Bosphorus (1986-1989)

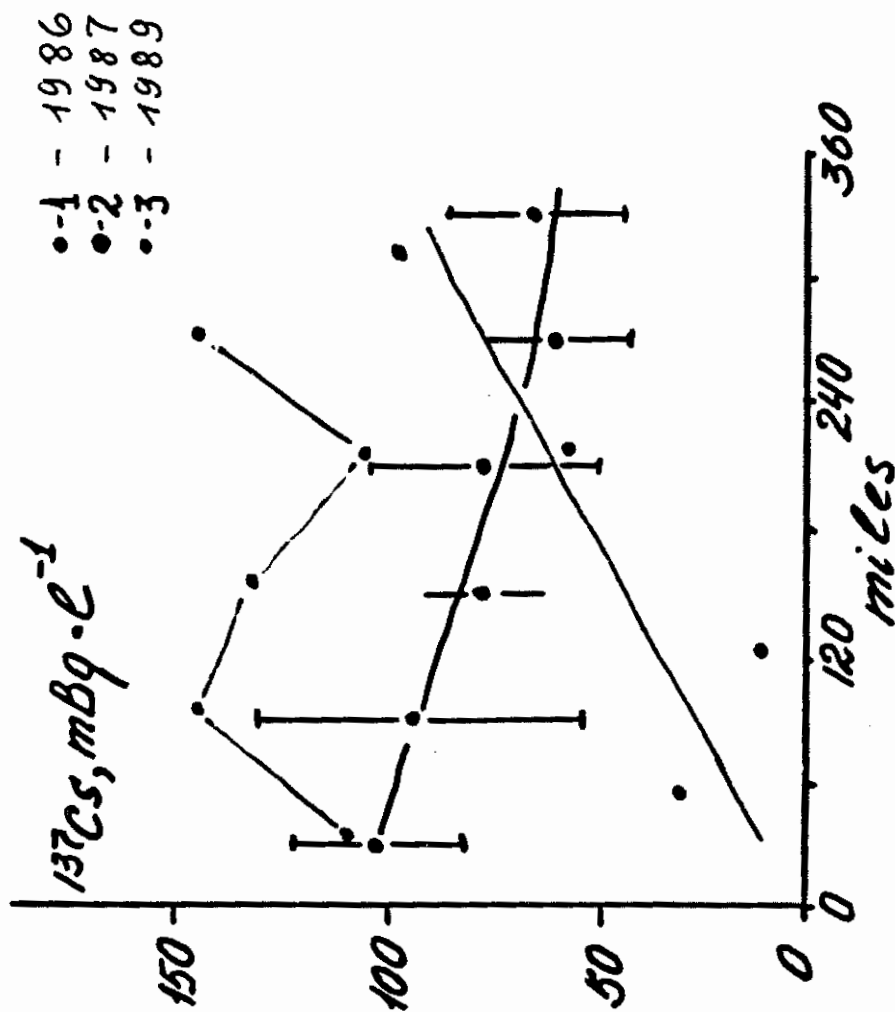


Fig. 3: Dynamics of the concentration of Cs-137 in the areas around the estuaries of the Dnieper, Dniester and Danube rivers and the Tarkhankut promontory (Crimea) in 1986-1989

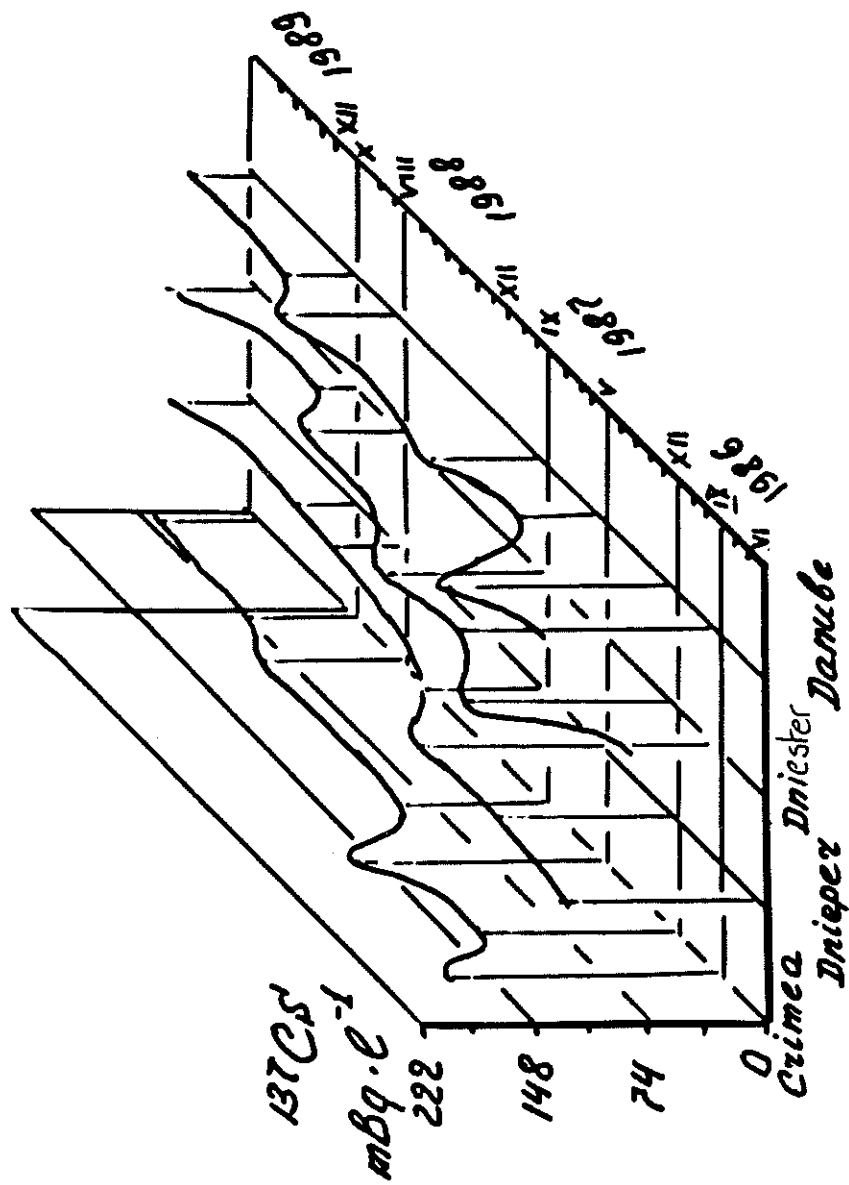


Fig. 4: Relationship between the Cs-137 concentration and the water salinity (1989)

*Black Sea*

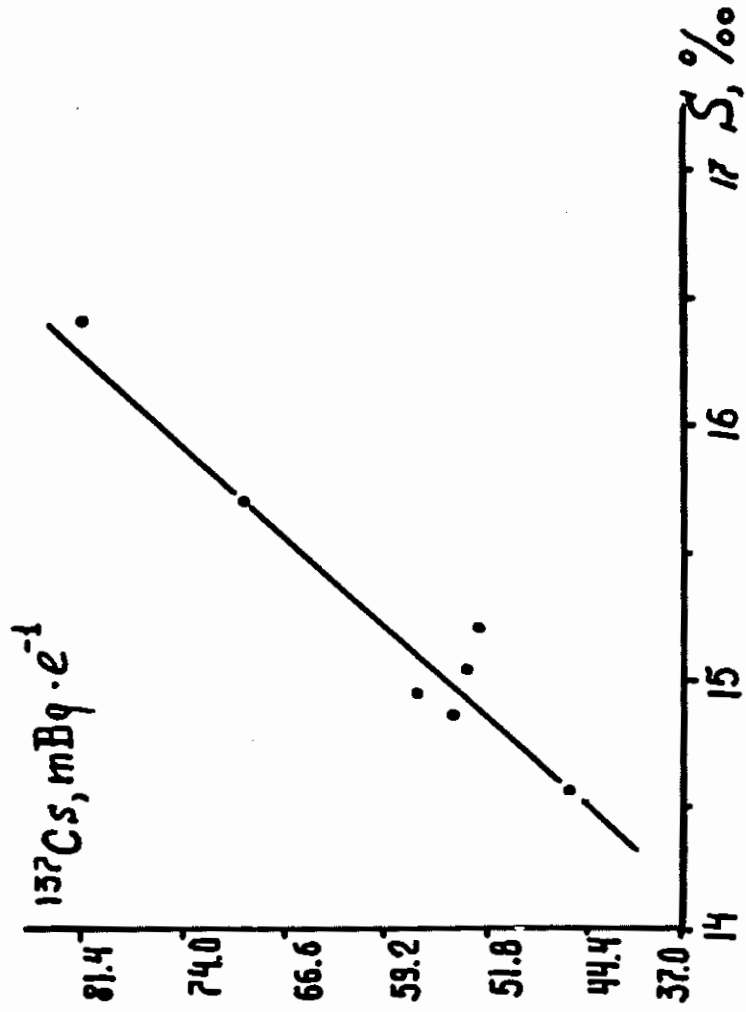


Fig. 5: Histograms of the distribution of Cs-137 concentrations in surface waters of the Black sea in 1987, 1988 and 1989

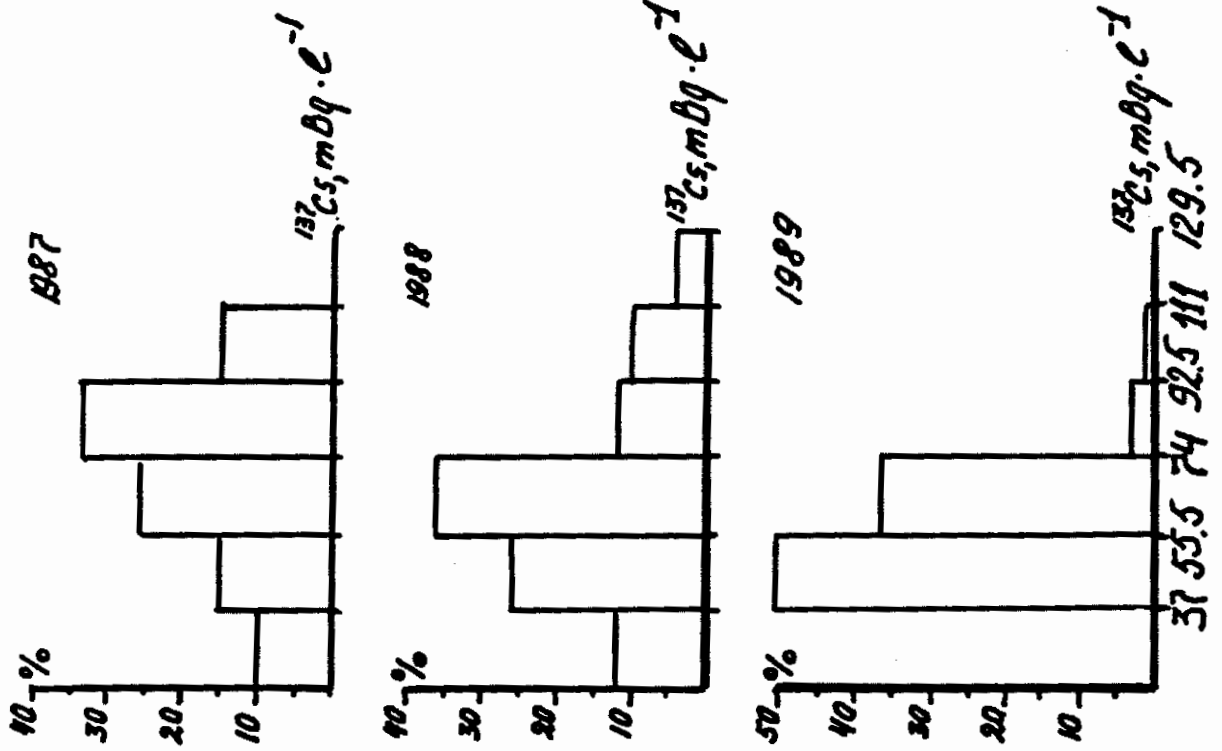


Fig. 6: The Dnieper reservoir system <sup>622</sup>

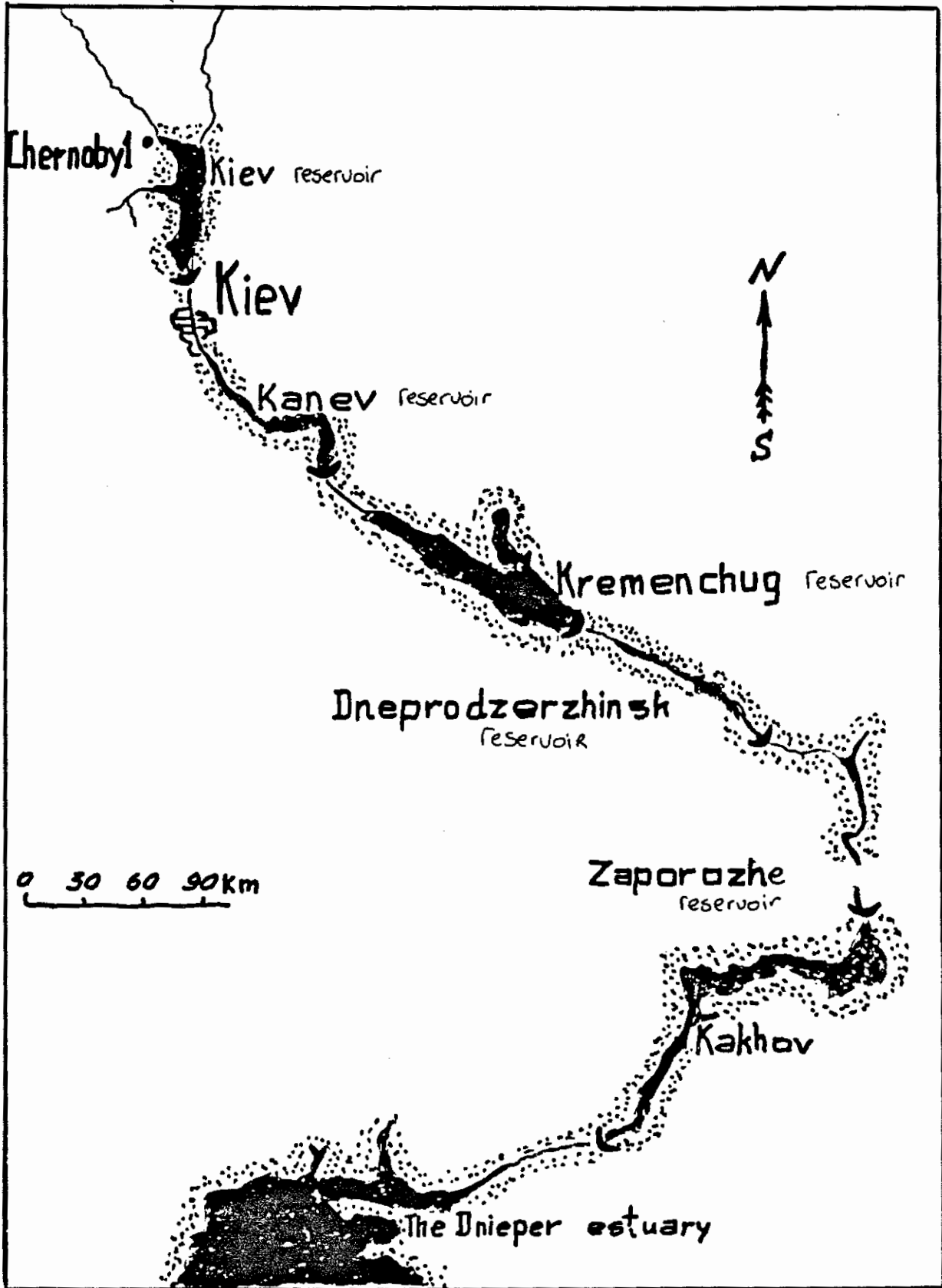


Fig. 7: Cs-137 concentration in the water of Dnieper river between the coling pond of the Chernobyl NPP (1) and the Dnieper-bug estuary (16-18)

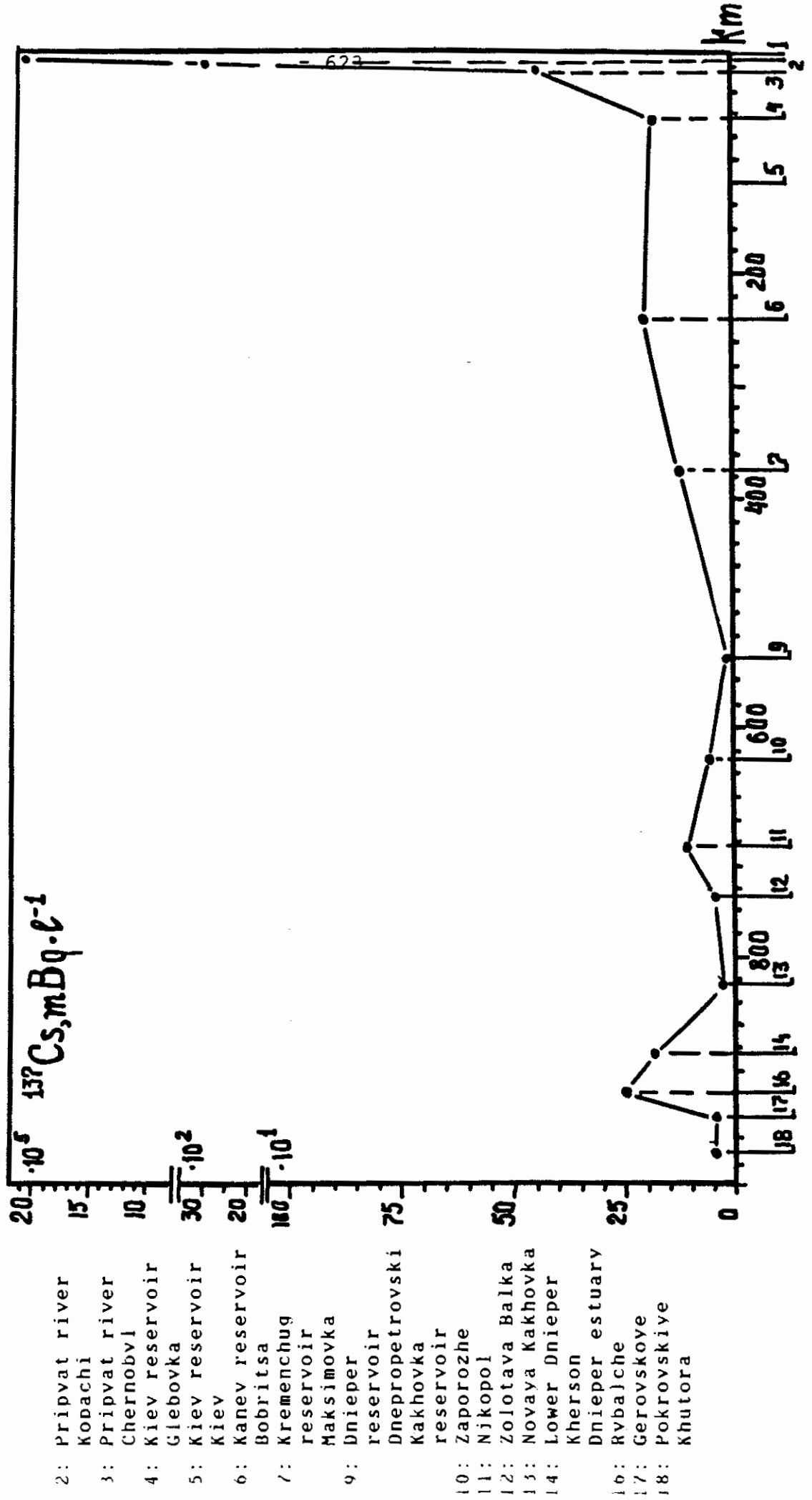




Fig. 8: Relationship between the specific activity of Cs-137 in bed sediments of the shelf area of the Black sea and depth

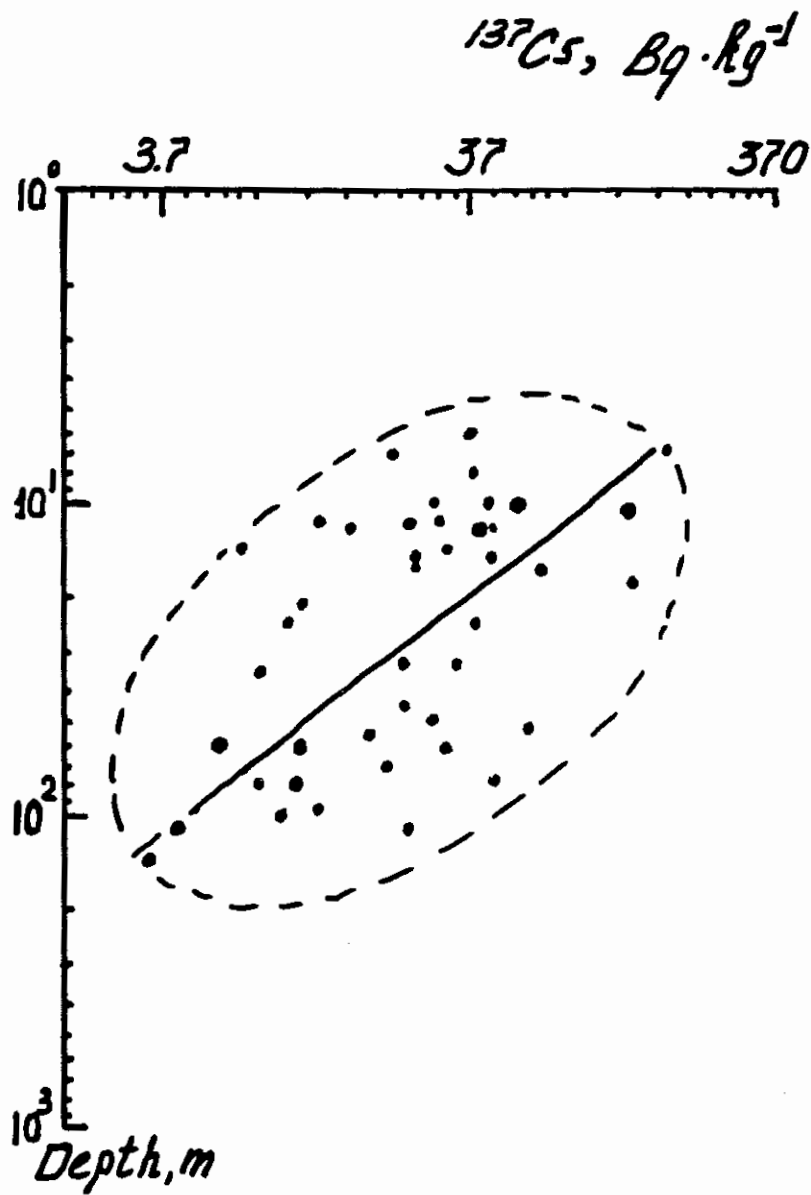


Fig. 9: Dynamics of the concentration of Cs-137 in bed sediments (solid lines) and water (dotted line) in the Dnieper River (Kherson)

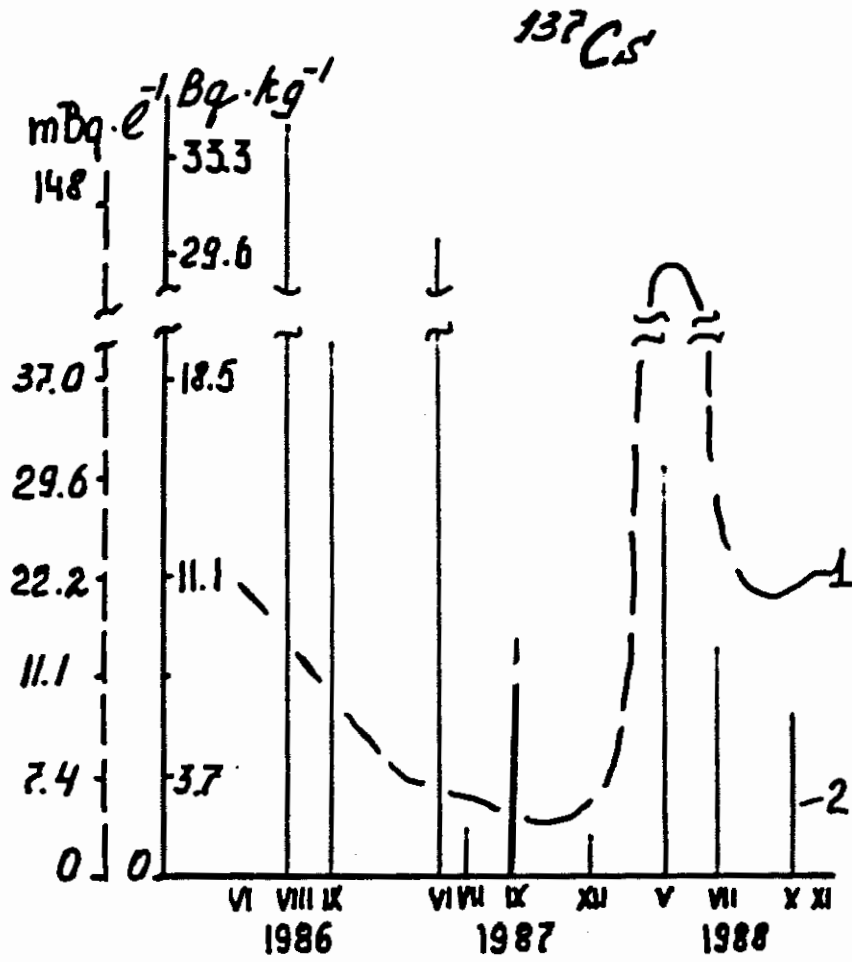


Fig.10: Dynamics of the concentration of Cs-137 in aquatic plants of the Dnieper River (1986-1989)

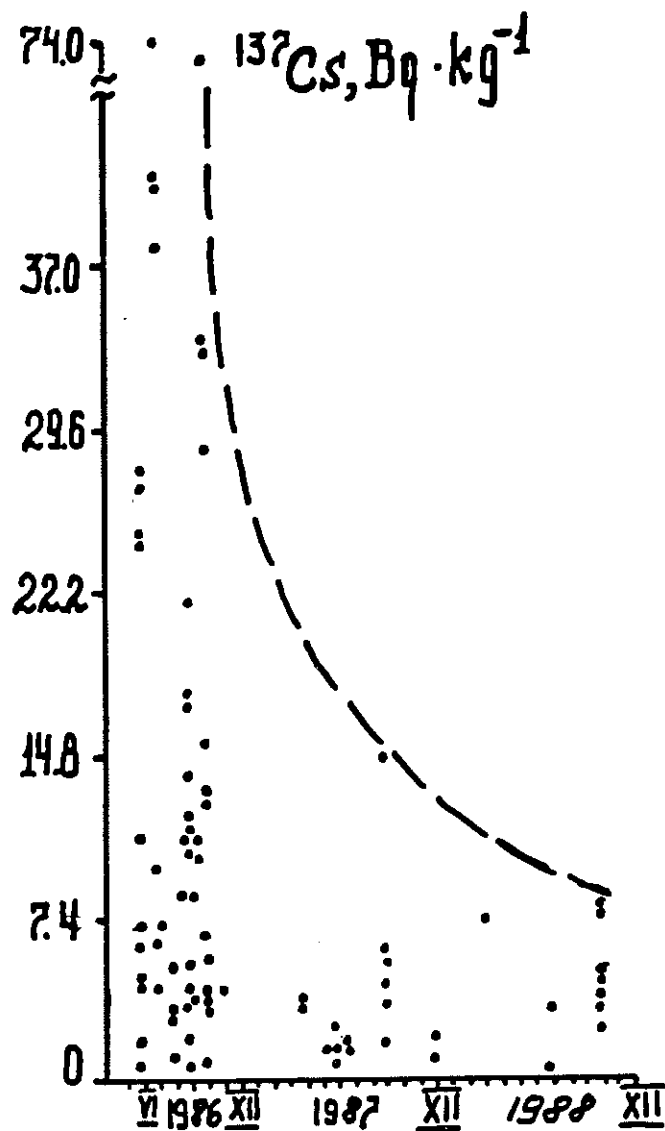
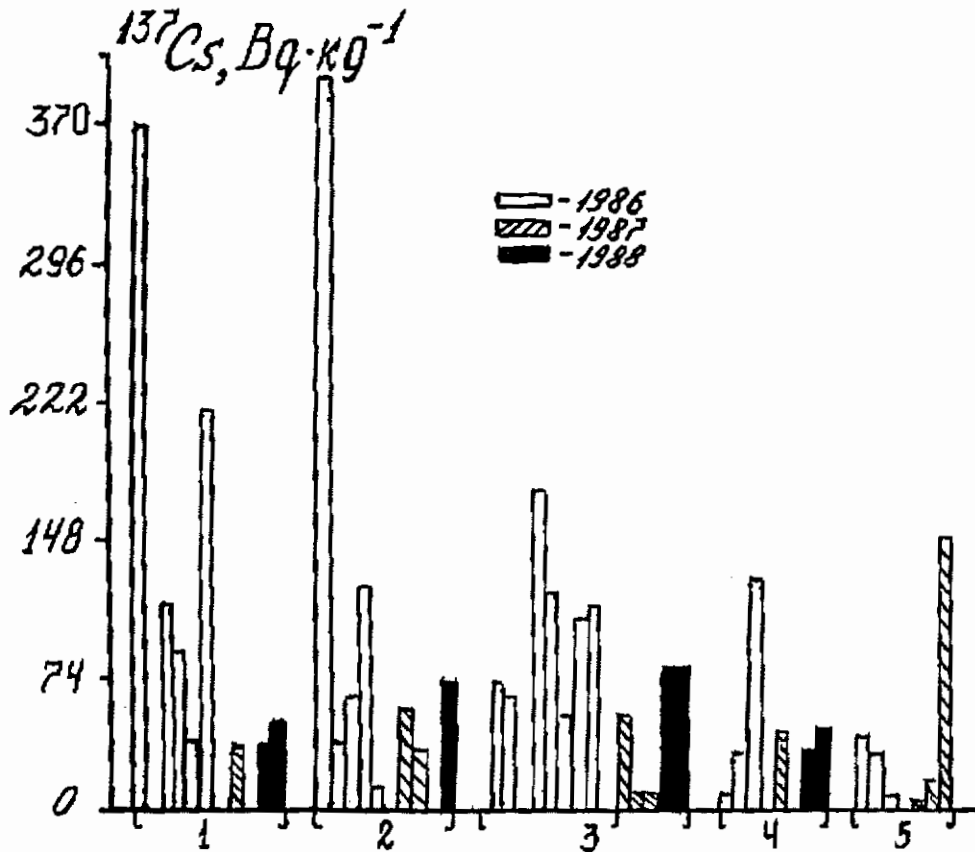


Fig.11: Dynamics of the  $^{137}\text{Cs}$ - $^{627}$  concentration in plants of the Kakhovka reservoir (1-3) and the lower Dnieper (4-5)



- 1 = Canada parrot feather  
(*Myriophyllum verticillatum*)
- 2 = Fennel-leaved pondweed  
(*Potamogeton pectinatus*)
- 3 = Thoroughwort pondweed  
(*Potamogeton perfoliatus*)
- 4 = Canada parrot feather  
(*Myriophyllum verticillatum*)
- 5 = Thoroughwort pondweed  
(*Potamogeton perfoliatus*)

Fig.12: Dynamics of the concentration of Cs-137 in Black sea mussels (1986-1989)

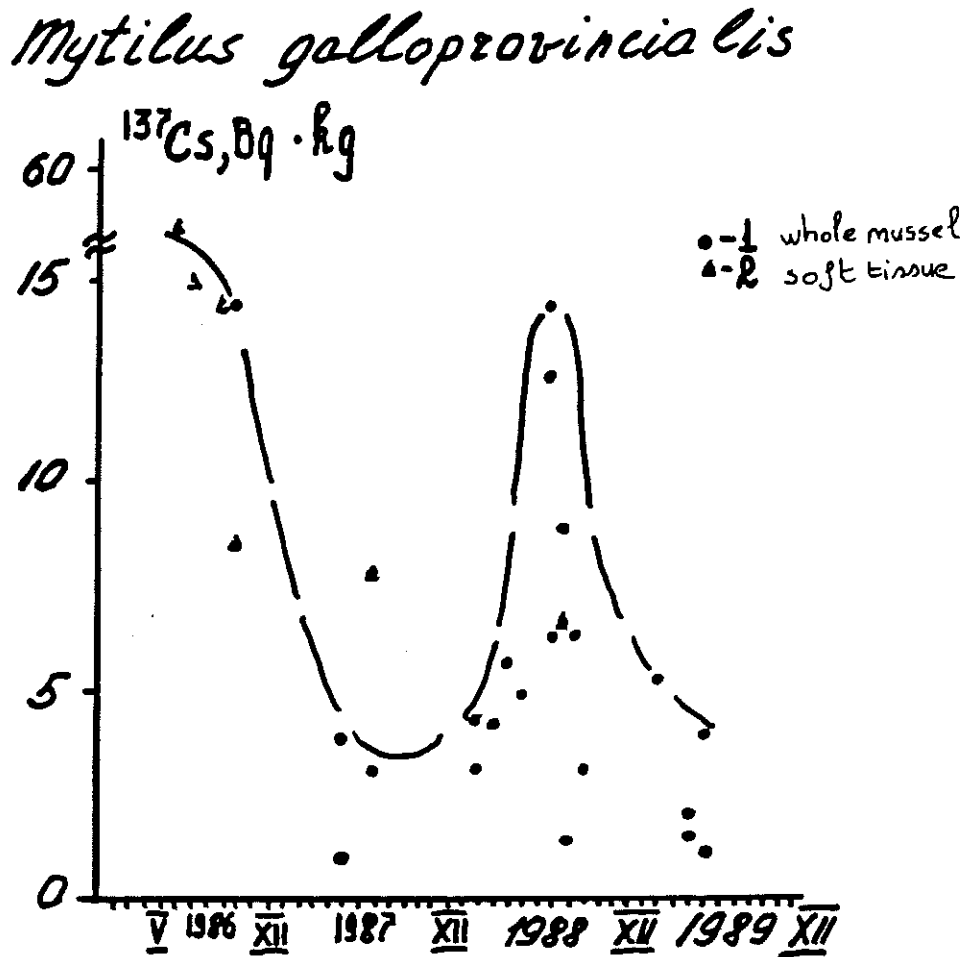




Fig. 14: Dynamics of the Cs-137 concentration in Dnieper molluscs (1986-1988)

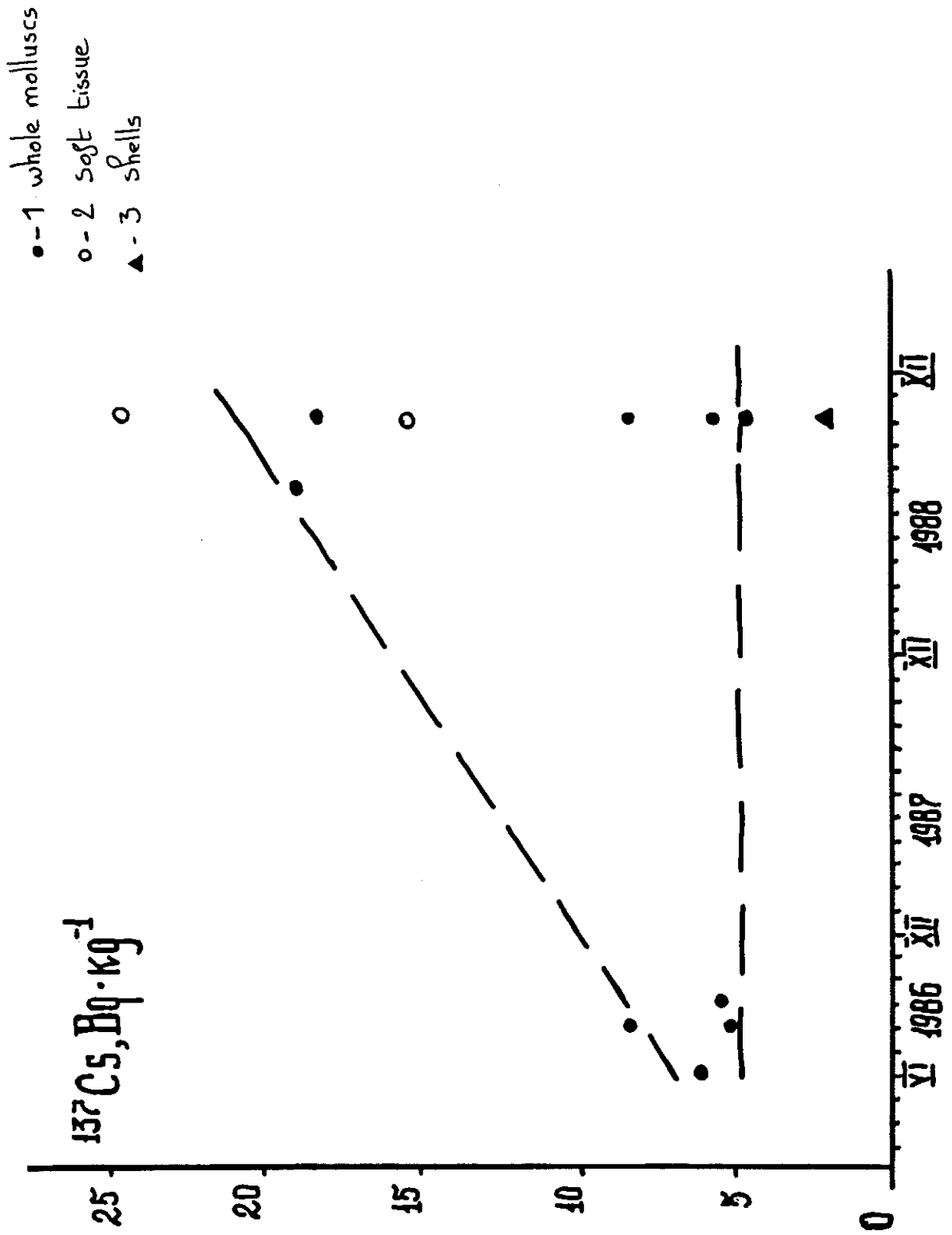


Fig. 15: Dynamics of Cs-137 concentration in Dnieper fish (1986-1988)

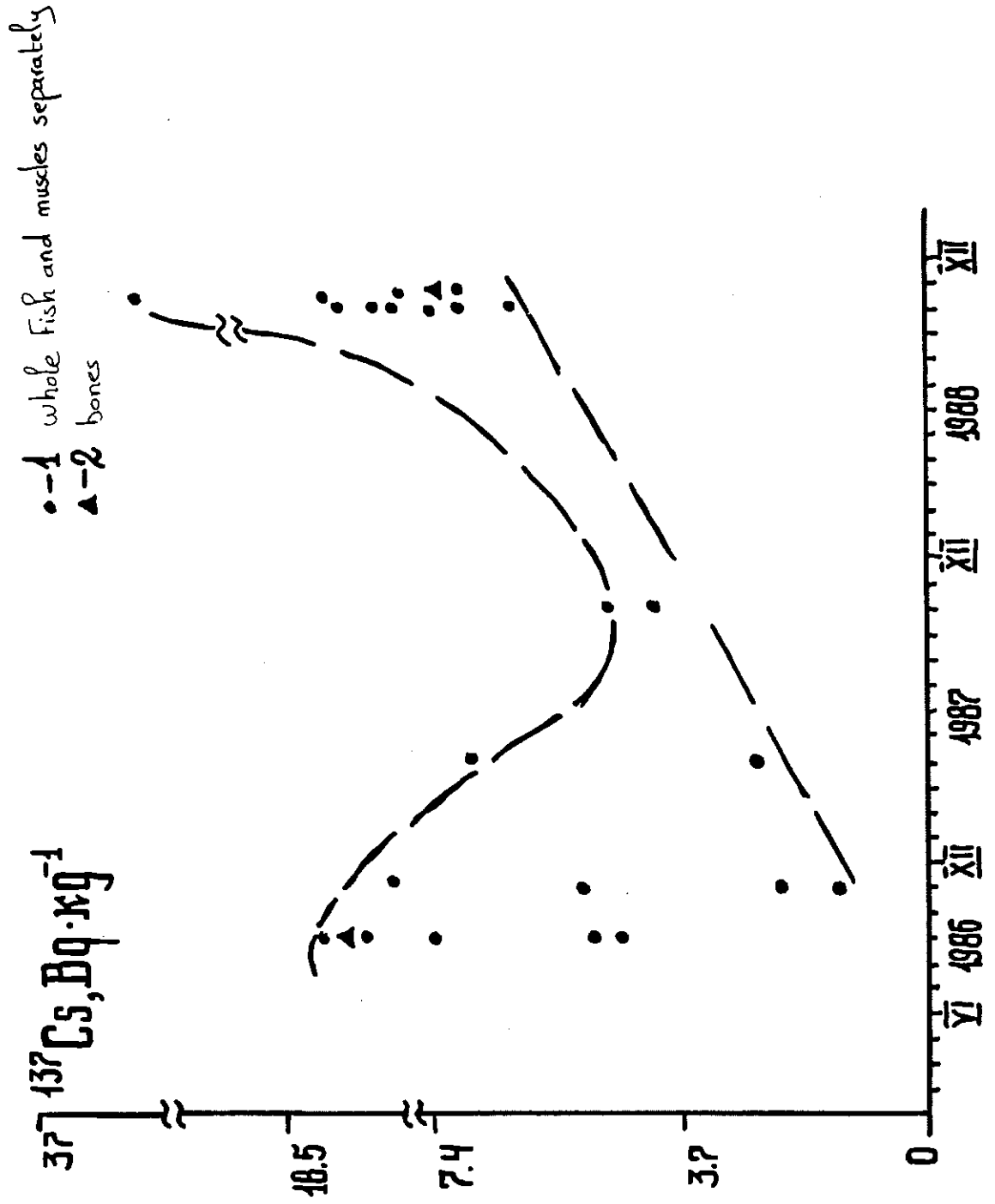




Fig. 16: Cs-137 concentration in Danube fish in 1988 (shaded columns) in comparison with 1978-1979 (unshaded columns)

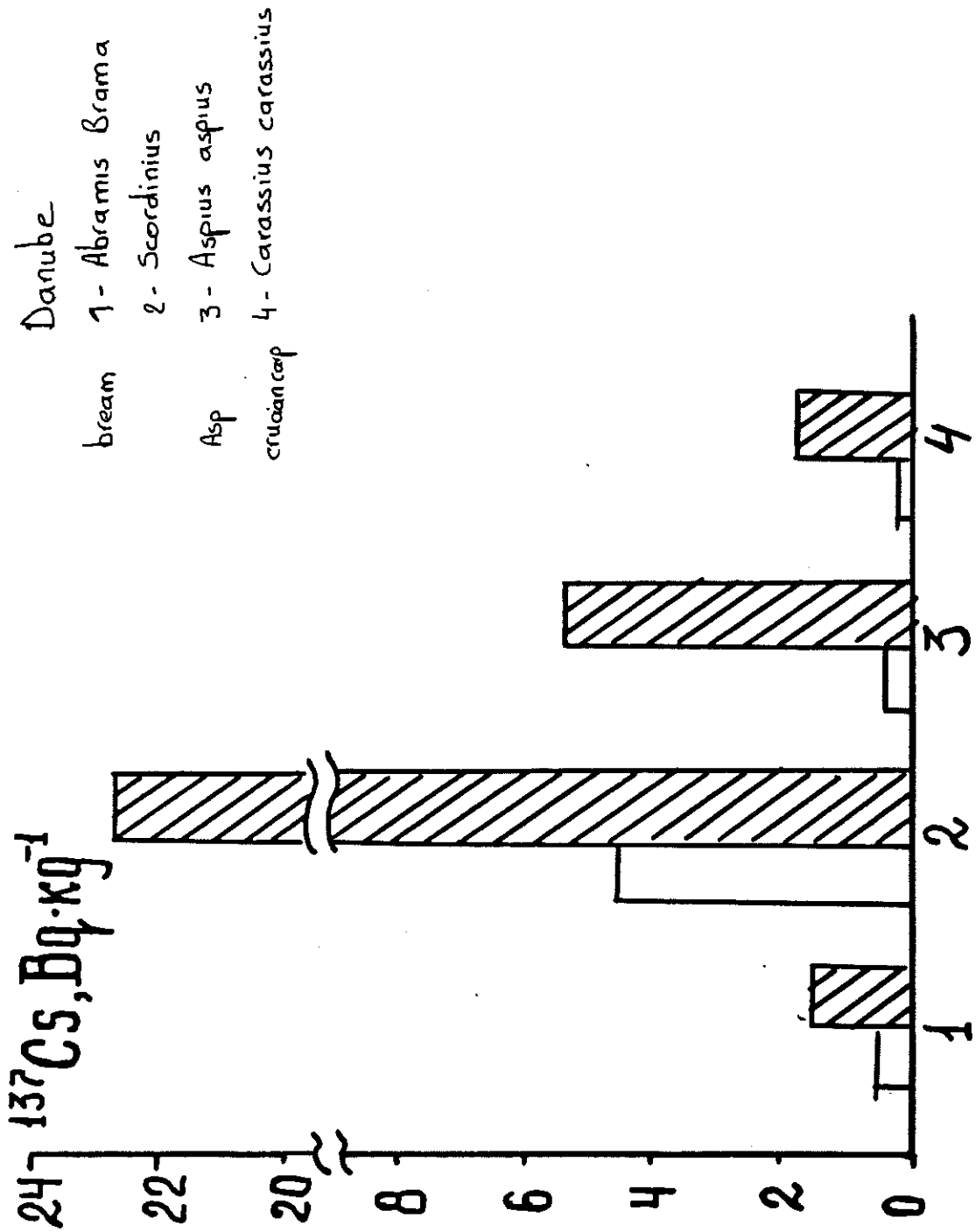


Fig.17: Dynamics of the Sr-90 concentration in surface waters of the Black sea (1986-1988)

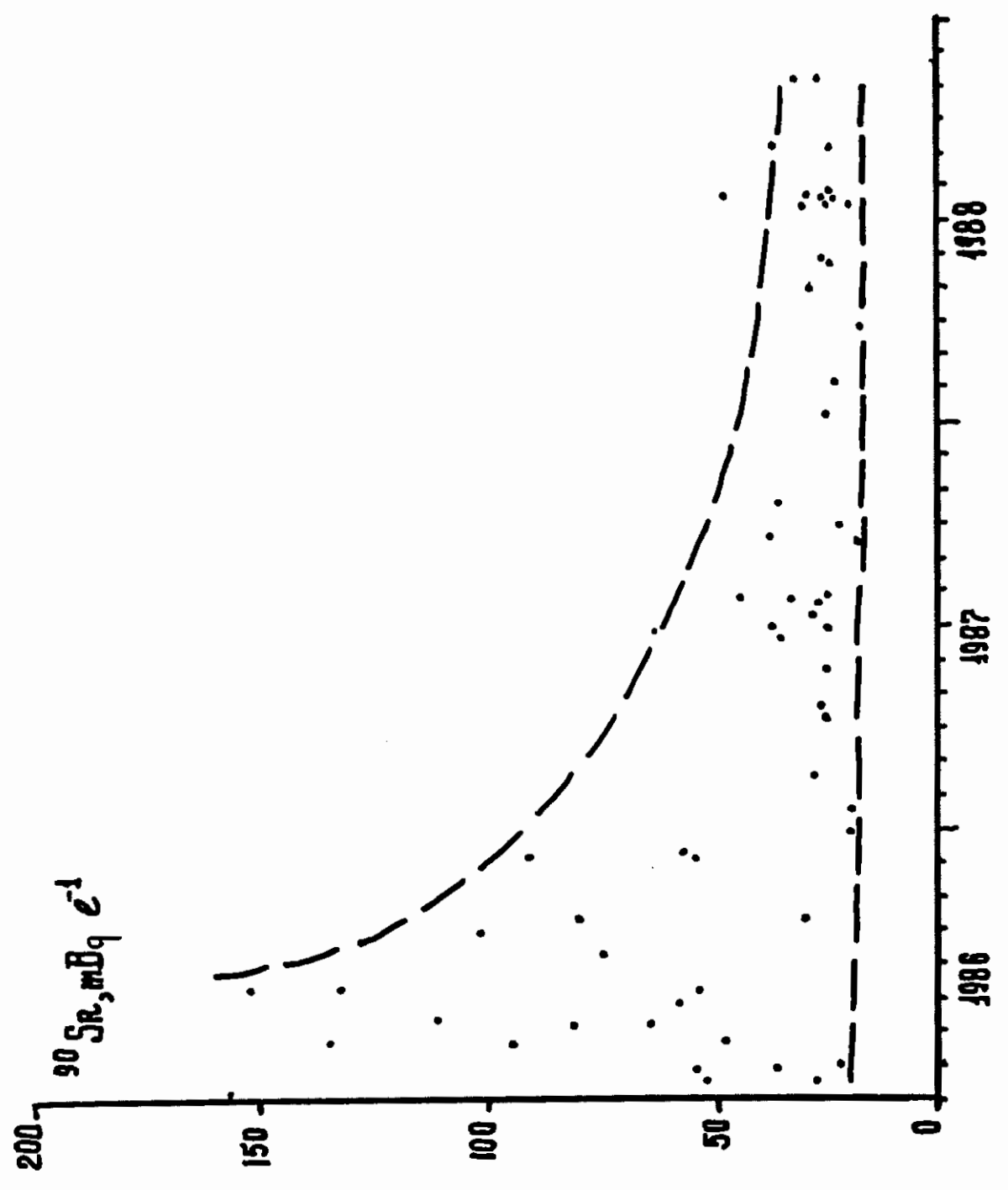


Fig.18: Ratio between the concentrations of Cs-137 and Sr-90 in the water around the Crimean peninsula (1986-1988)

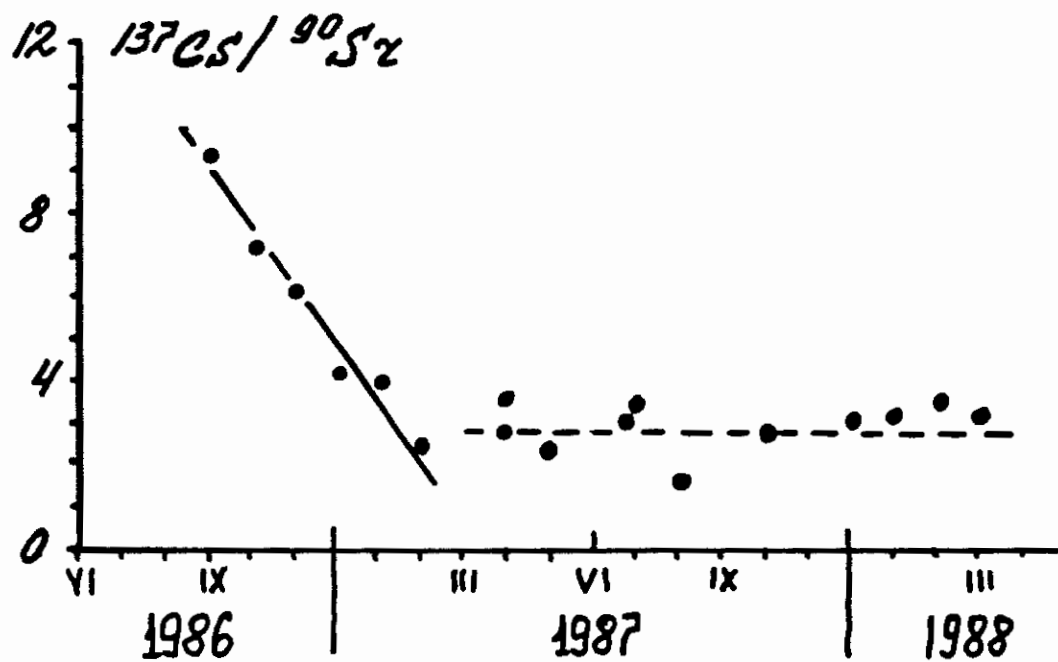


Fig.19: Sr-90 concentration in the Kakhovka reservoir (Dnieper) in 1986

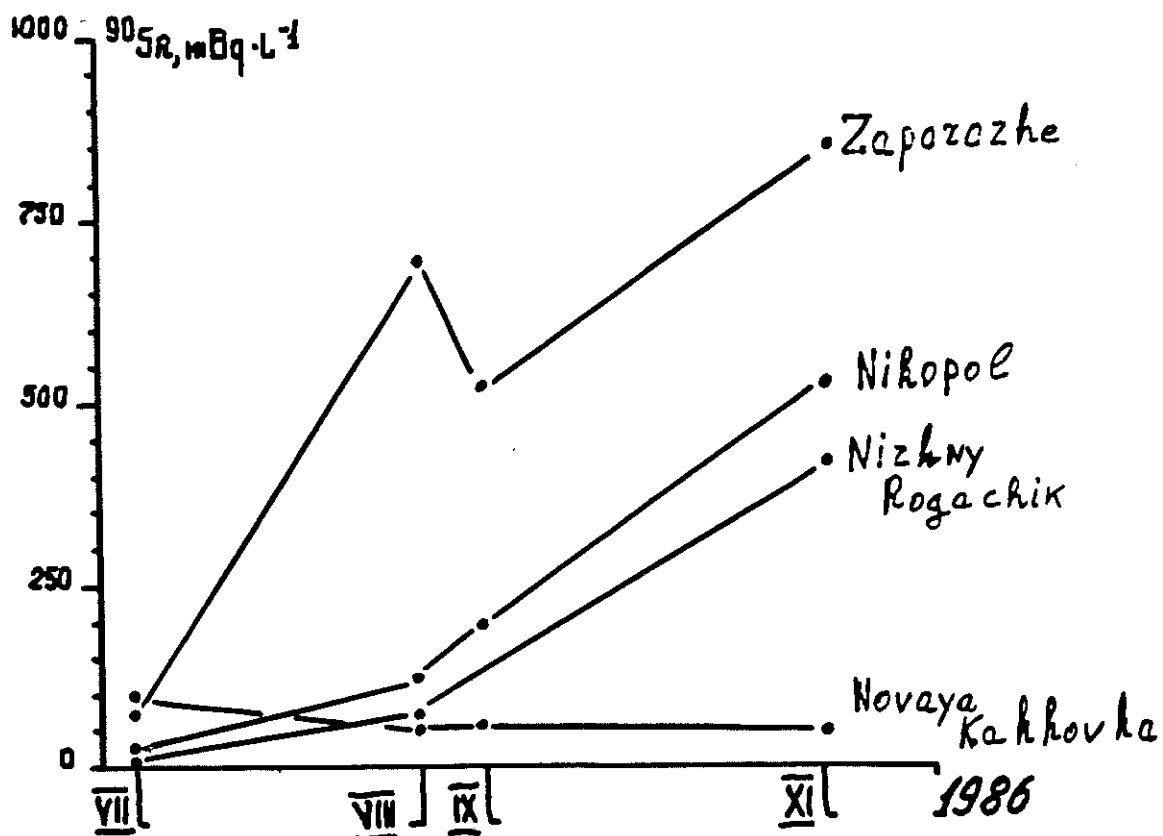


Fig. 20: Distribution of Sr-90 concentrations in the Dnieper river from the cooling pond of the Cher NPP to the Black sea in 1987 (1) and 1988 (2)

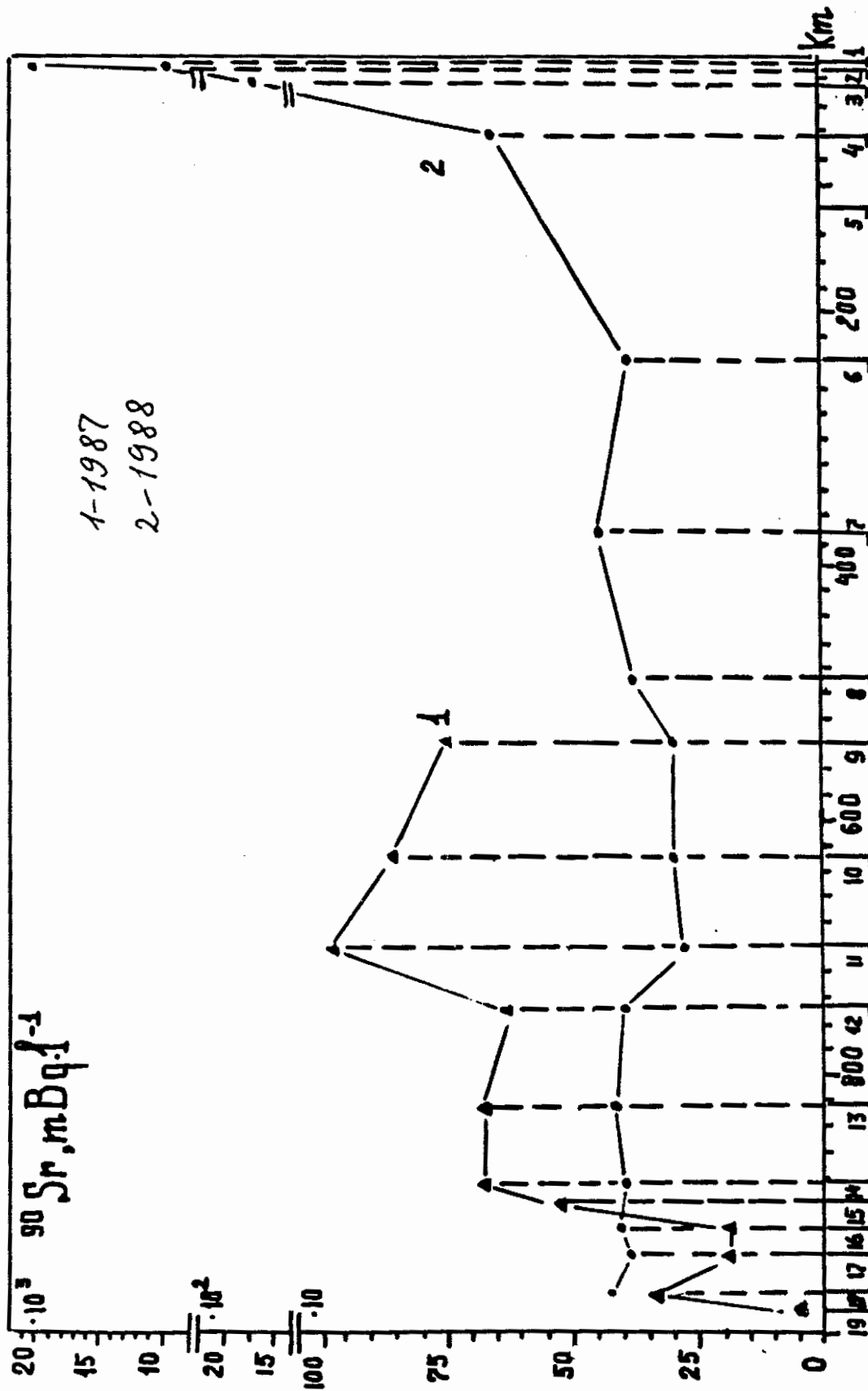


Fig.21: Dynamics of the Sr-90 concentration near the estuaries of the Dnieper, Dniester and Danube rivers and near the Tarkhankut promontory (Crimean peninsula) in 1986-88

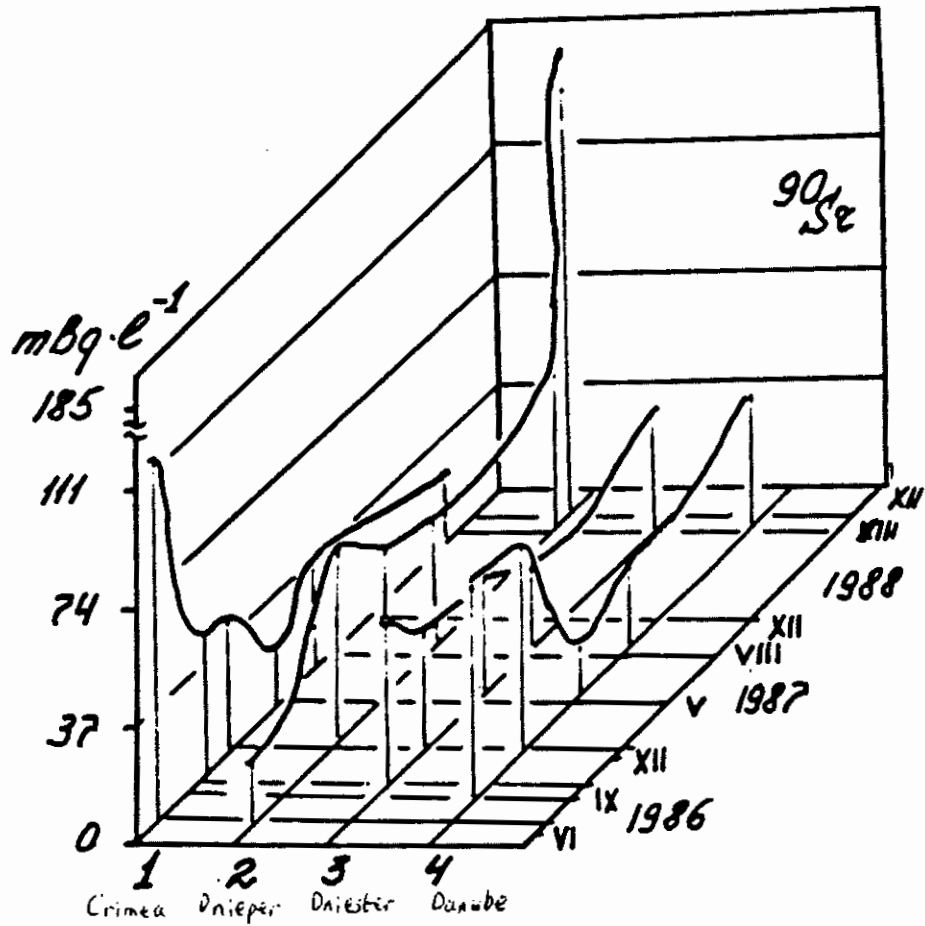


Fig.22: Sr-90 removal into the  $^{638}\text{Black}$  sea with Dnieper Water  
(1986-1989)

Years	$V, \text{km}^3$	concentration $\bar{c}, \text{Bq/m}^3$	Flow $^{90}\text{Sr}$ $T \text{Bq}$
$\frac{1986}{\text{V-XII}}$	21.0	51.8	1.090
1987	37.1	407.0	14.681
1988	45.8	289.0	14.015
1989	33.9	290.0	9.908
Total:			39.694

Fig. 23: Sr-90 removal into the Black sea with Dnieper water (1987)

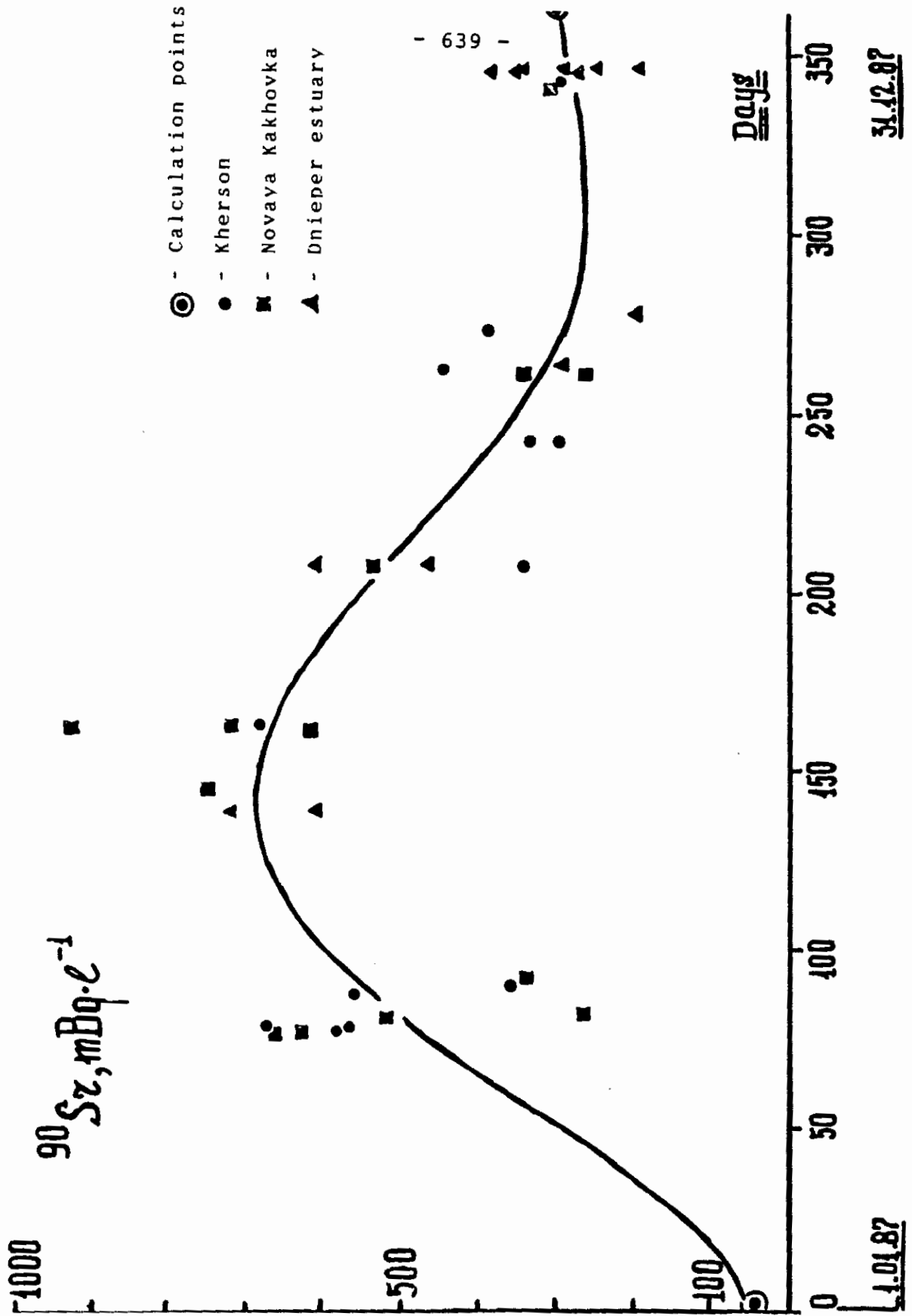




Fig.24: Dynamics of the Sr-90 concentration in bed sediments of the lower Dnieper

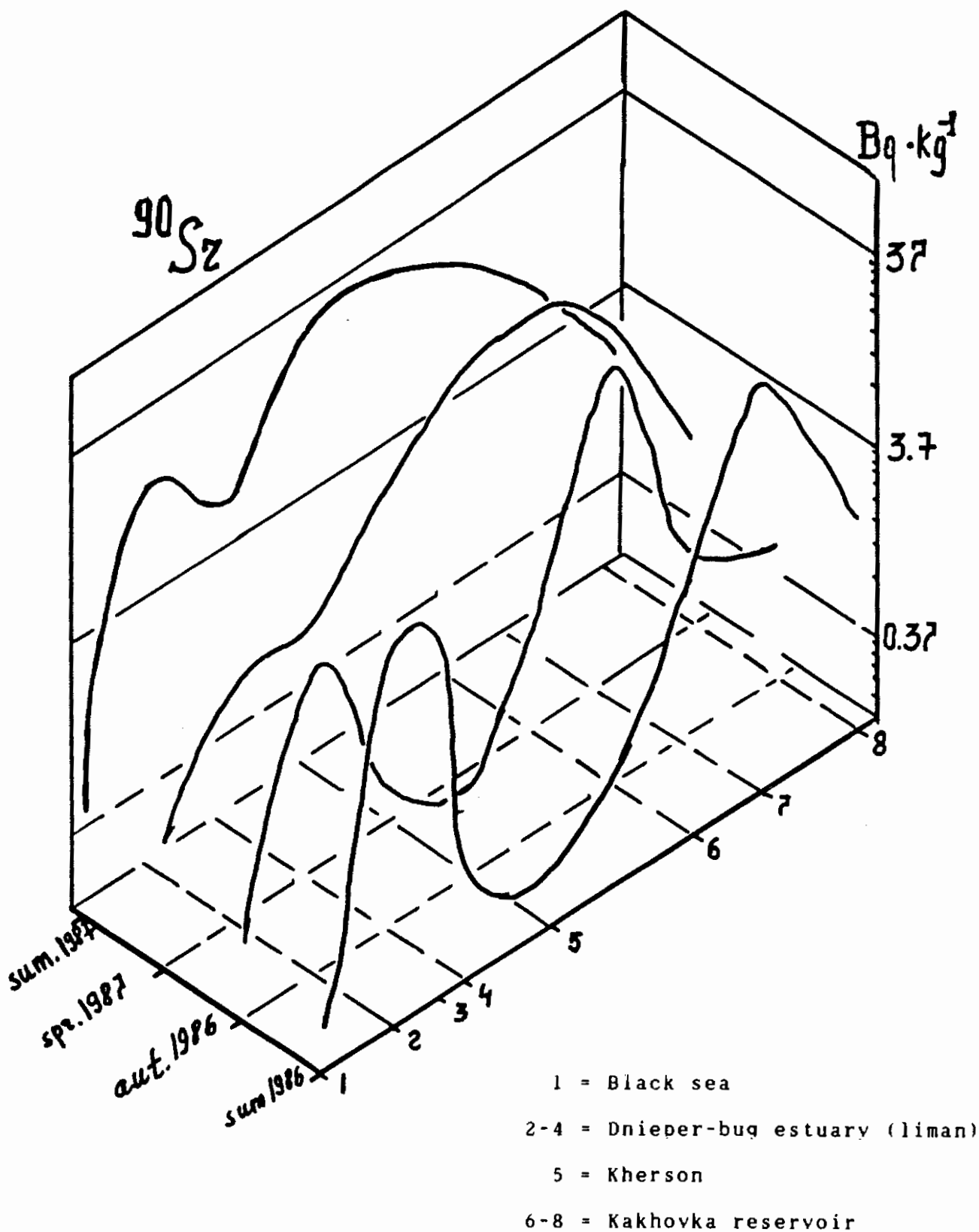


Fig.25: Sr-90 concentration in the water and bed sediments of the Dnieper river at Kherson (1986-1989)

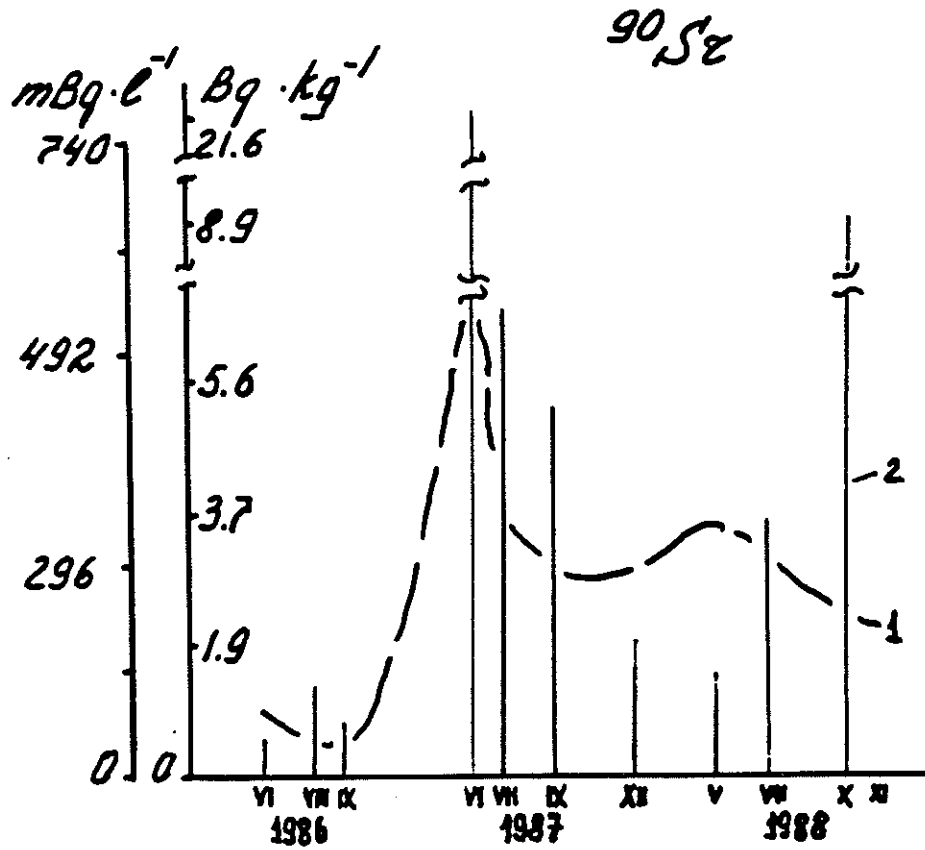


Fig.26: Sr-90 concentration in Dnieper plants (1986-1989)

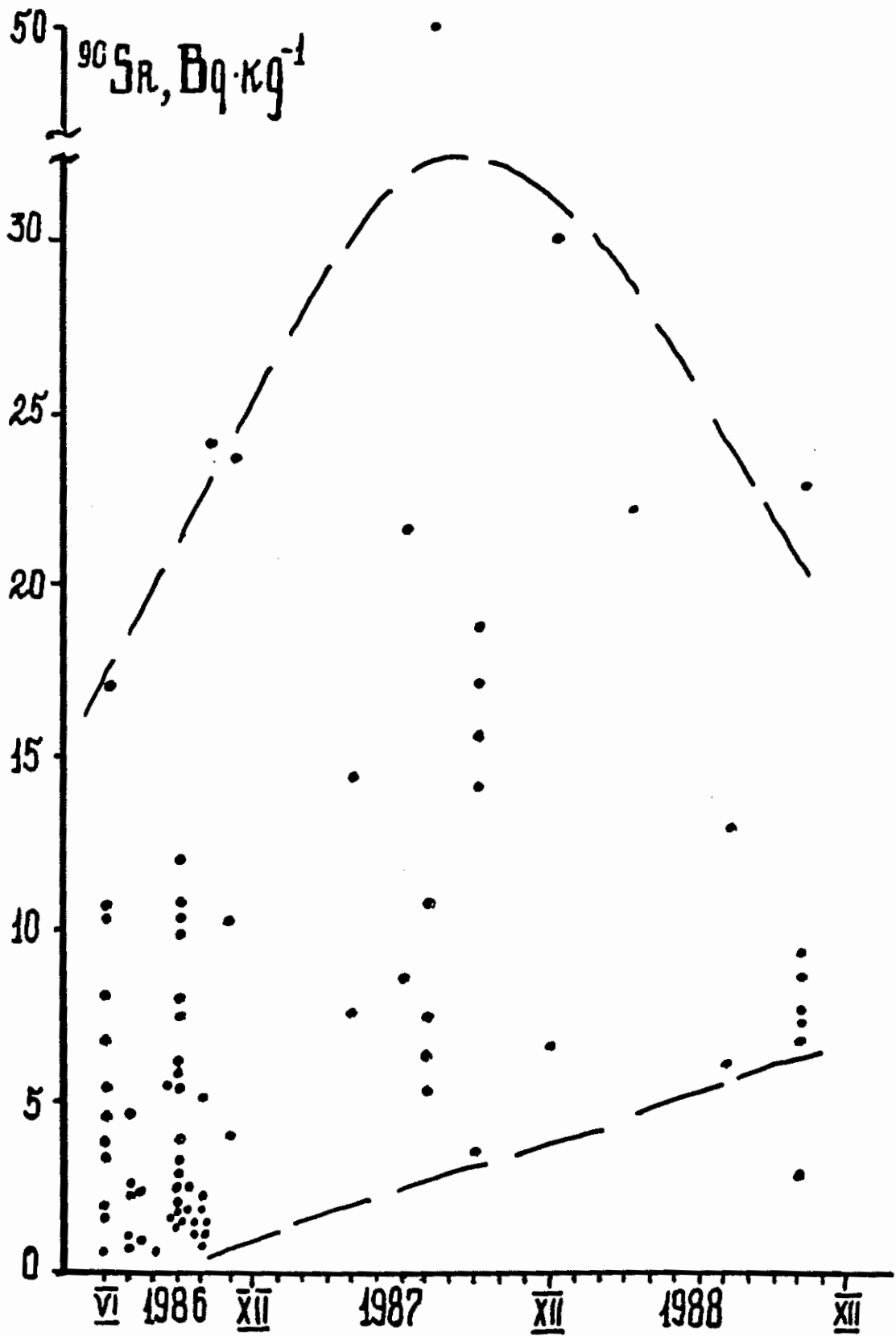


Fig.27: Sr-90 concentration in Black sea mussels (1986-1988)

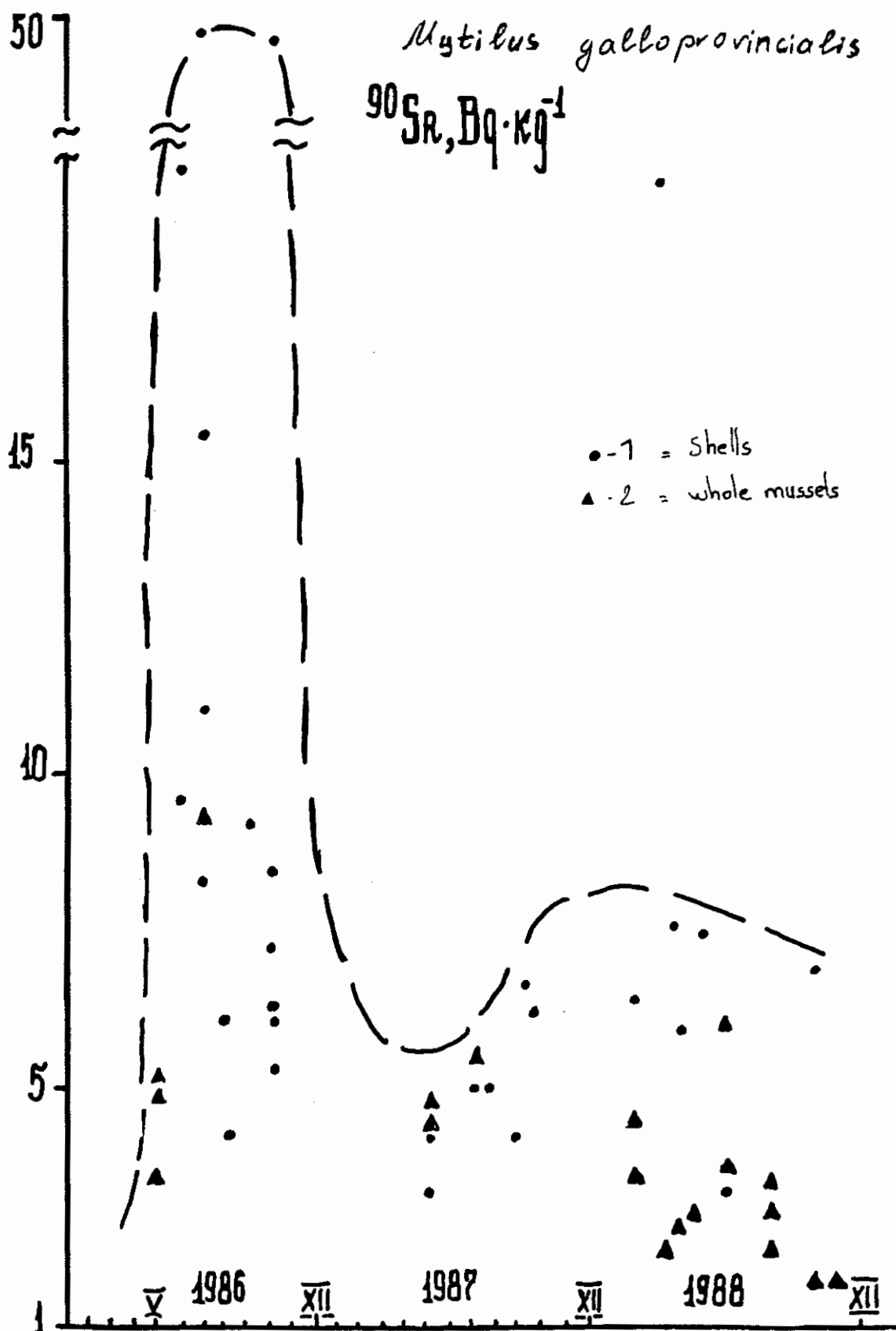


Fig.28 : Sr-90 concentration in Dnieper molluscs (1986-1989)

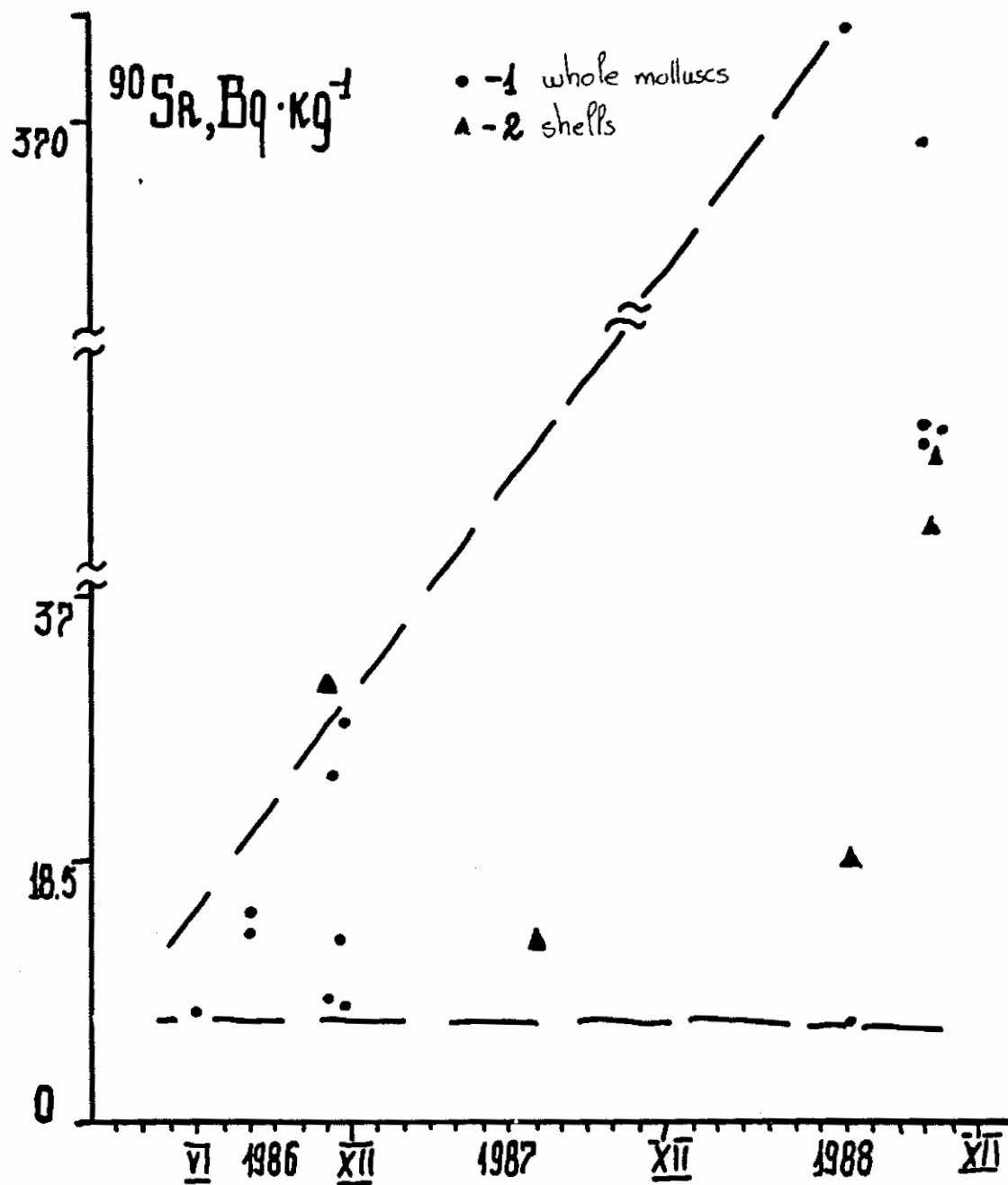


Fig. 29: Dynamics of Sr-90 accumulation in Black sea fish (1986-1988)

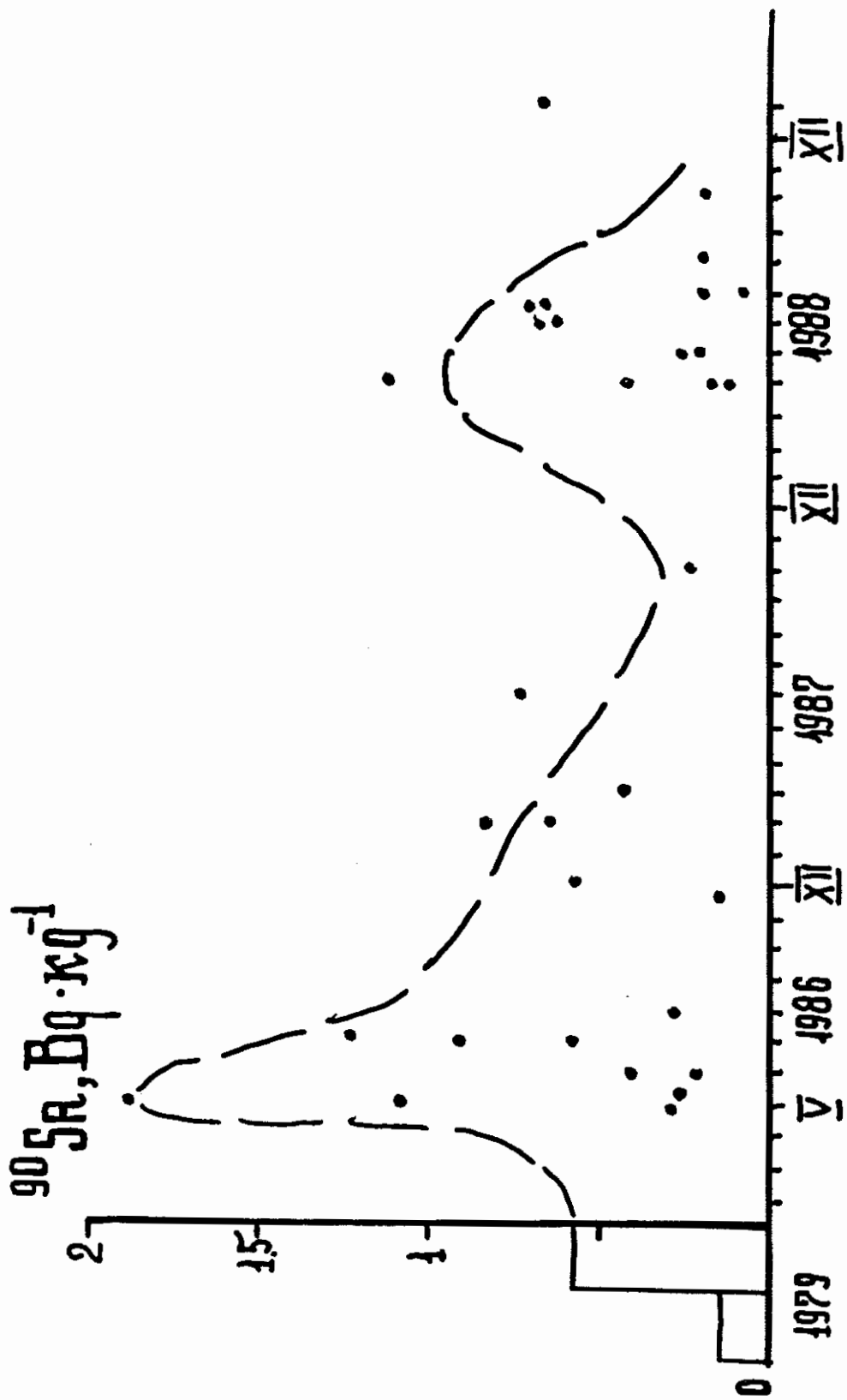
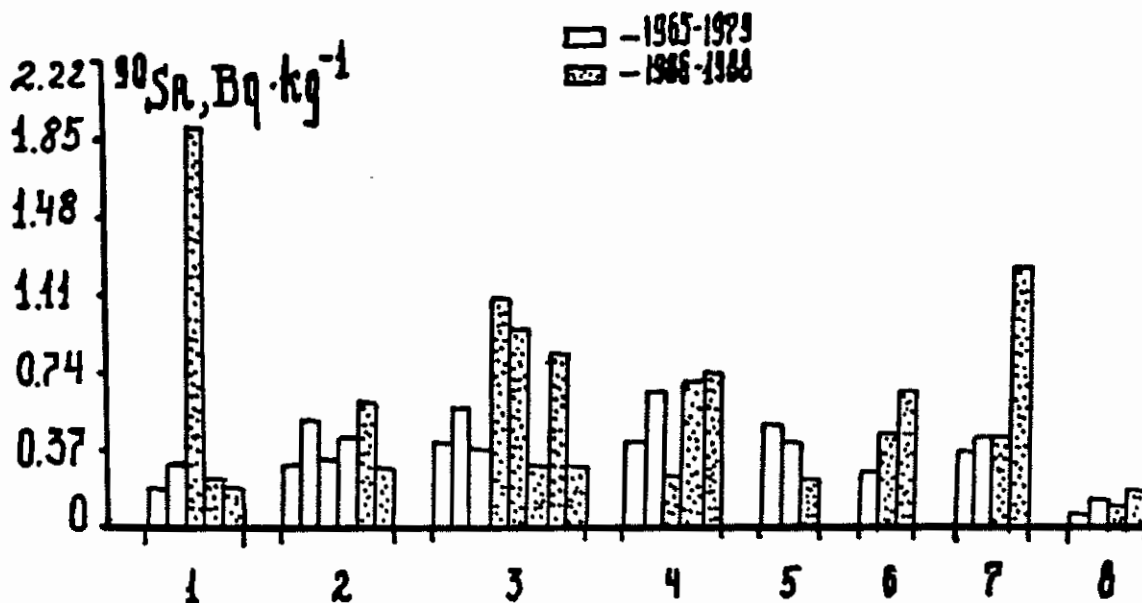
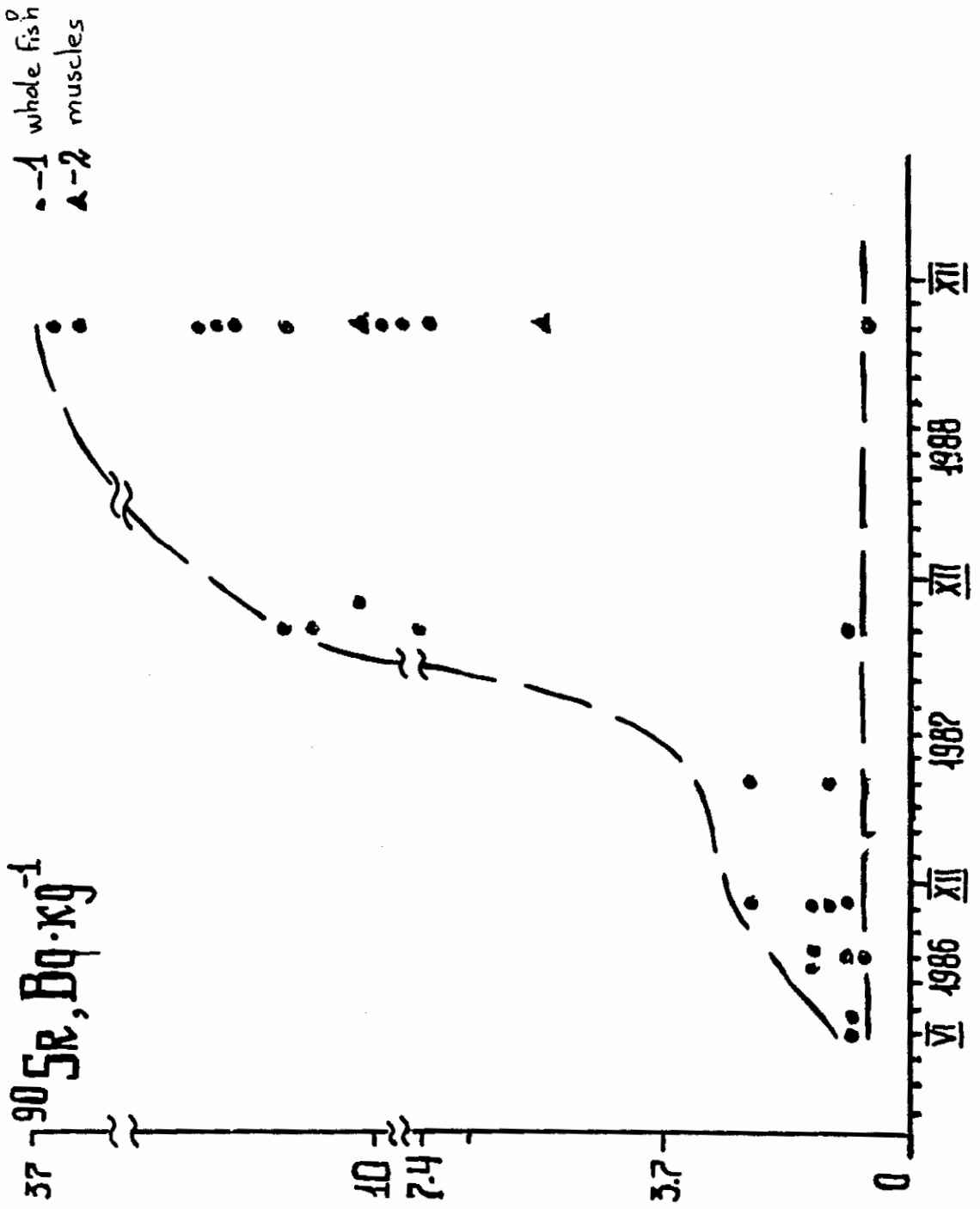


Fig.30: Sr-90 Concentrations in Black sea fish before and after the Chernobyl accident



- 1 = Whiting (*Odontogadus merlangus*)
- 2 = Buckler skate (*Raja clavata*)
- 3 = Blacksea turbot (*Scophthalmus maeoticus maeoticus*)
- 4 = Peacock wrasse (*Crenilabrus tinca*)
- 5 = Mediterranean scad (*Trachurus mediterraneus euxinus*)
- 6 = Black goby (*Gobius sp.*)
- 7 = Sea scorpion (*Scorpaena porcus*)
- 8 = Sprat (*Sprattus sprattus sprattus*)

Fig.31: Dynamics of Sr-90 accumulation in Dnieper fish (1986-1988)







**Modelling Large-scale  
Contamination of the Black Sea  
Caused by Long-lived  
Radionuclides of Cs-137 and  
Sr-90 Following the Chernobyl  
Accident**

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## ABSTRACT

The problem of making predictions was solved using a box balance model of the Black Sea based on average annual parameters, account being taken of precipitation and evaporation above the sea surface, vertical mixing processes, factors linked to radionuclide concentration in aquatic organisms and sorptive interaction at the water/bed-sediment interface. The rate of vertical mixing of waters was evaluated assuming a stationary water salinity profile by depth. The model was verified using the results of measurements of  $^{137}\text{Cs}$  content in the Black Sea between 1986 and 1990. To illustrate how the model is used, the paper presents forecast changes in  $^{137}\text{Cs}$  content in water and bed sediment, including an assessment of the rate of vertical migration and of changes in the amount of  $^{137}\text{Cs}$  stored in the Black Sea over a period corresponding to three half-lives of the radionuclide.

The urgency of evaluating and forecasting the radioactive contamination of the Black Sea following the Chernobyl accident stemmed from the wide-scale utilization of its recreational and biological resources.

According to the latest estimates,  $37 \cdot 10^{15}$  Bq of  $^{137}\text{Cs}$  and  $8.1 \cdot 10^{15}$  Bq of  $^{90}\text{Sr}$  were deposited in the environment after the accident.<sup>4</sup> Some of this fell in river basins, and from 4 to 6.5% of the total release was deposited in the form of aeolian fallout on the Black Sea surface.<sup>6, 9</sup>

Initially, the  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  radionuclides were very unevenly spread in the surface waters, but the differences noted in our observations later decreased due to horizontal mixing and vertical migration of the radionuclides, which mainly depended on the physical processes occurring over a period of time commensurate with the radioactive half-life of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ .

The aim of our study was to devise a mathematical balance model reflecting the large-scale impact of the main abiotic processes on the migration and residence time of long-lived radionuclides (e.g.  $^{137}\text{Cs}$ ) in the Black Sea.

Fig. 1 shows the structure of our model of large-scale contamination of the Black Sea, which is arbitrarily divided into layers in line with the model's box-type structure.

This is a closed model with respect to the water and salt balance and the radioactive contamination of the environment. The equation expressing the water balance is based on the assumption that the level and volume of the Black Sea did not change during the period studied. Therefore,

$$Q_H - Q_B + \int_{KA} - \int_{KA} + F + R - E = 0 \quad (1)$$

The salt balance equation is:

for layer No 1 (surface waters):

$$\frac{dS_1}{dt} = \frac{1}{V_1} \left[ f_{KA} S_K + FS_F + RS_{oc} + f_{21} S_2 - (Q_B + f_{AK} + f_{12}) S_1 \right] \quad (2)$$

for the other layers:

$$\frac{dS_i}{dt} = \frac{1}{V_i} \left[ d_i Q_H S_S + (f_{i,i-1} + f_{i,i+1}) S_i + f_{i-1,i} S_{i-1} + f_{i+1,i} S_{i+1} \right] \quad (3)$$

The rate of change in the amount of radionuclides in the surface layer of the Black Sea was determined as follows:

$$\frac{dq_1}{dt} = C_A (A - A_1) + C_F F + C_R R + W + C_K f_{KA} + f_{21} C_2 - (f_{12} + Q_B + \lambda V_1 + K_b P_{b(1)} + f_{AK}) C_1 \quad (4)$$

The radionuclide balance for the intermediate layers (i) is obtained from the correlation:

$$\begin{aligned} \frac{dq_i}{dt} = & C_{i-1} f_{i-1,i} + C_{i+1} f_{i+1,i} + C_H d_i Q_H + K_b (C_{i-1} P_{b(i-1)} - C_i P_{b(i)}) + \\ & + K_A (C_{i-1} A_{i-1} - C_i A_i) - C_i (f_{i,i+1} + f_{i,i-1} + \lambda V_i) \end{aligned} \quad (5)$$

For the benthic layers:

$$\frac{dq_8}{dt} = C_7 f_{78} + K_b (C_7 P_{b(7)} - C_8 P_{b(8)}) + K_A (C_7 A_7 - C_8 A_8) + C_g m (r + r_{ge}) - C_8 V_8 (r + \lambda) \quad (6)$$

and for the bed sediments:

$$\frac{dq_8}{dt} = C_8 (K_b P_{b(8)} + K_A A_8 + V_8 r_c) - C_g m (r + r_{ge} + \lambda) \quad (7)$$

The symbols used in the above stand for the following:  $S$  with a subscript - salinity of the  $i$ -layer of the Black Sea waters;  $S_K, S_F, S_{OC}$  and  $S_b$  respectively - salinity of the Sea of Azov waters, river flows, precipitation and lower Bosphorus current;  $V$  with subscripts - volume of the  $i$ -layer of the Black Sea waters;  $f_{KA}$  and  $f_{AK}$  respectively - annual ingress of water from the Sea of Azov into the Black Sea, and from the Black Sea into the Sea of Azov via the Strait of Kerch;  $j$  - portion of the average annual flow of the lower Bosphorus current entering the  $i$ -layer;  $f$  with two subscripts - rate of ingress of water from the layer designated by the first subscript into the layer designated by the second subscript;  $q_i$  and  $C_i$  - radionuclide content and concentration in the layer designated by the  $i$ -number;  $C_A, C_F, C_R, C_K$  and  $C_Q$  respectively - radionuclide concentration in allochthonous suspensions, river flow, precipitation, water of the Sea of Azov and Black Sea bed sediments;  $A_i$  and  $P_b(i)$  respectively - allochthonous and biogenic sedimentation flows egressing from the  $i$ -layer;  $K_A$  and  $K_b$  - coefficient of radionuclide accumulation by allochthonous and biogenic suspensions;  $\lambda$  - radionuclide decay constant;  $m$  - bed sediment boundary layer mass;  $r_c, r$  and  $r_{ge}$  respectively - rate constants for sorption, desorption and remobilisation processes during interaction of bed sediments with water masses.

In the above equations, the volumes, areas of layers, water balance components and salinity of layers were determined from published data, while the rate of vertical exchange between the water layers was calculated. The parameters of radionuclide concentration by allochthonous and biogenic suspensions, plus the sorptive interaction of the radionuclides with the bed sediments and the aqueous medium, were obtained from experiments involving radioactive tagging and in-situ observations. The intensity and vertical profiles of sedimentation were obtained from existing literature, with the trophism of the Black Sea waters being taken into account. The radionuclide content of the Black Sea surface and its horizons was obtained from our own data plus existing information.



In specific terms, our research involved the following:

- empirical loading of the model with the findings from hydrological and hydrochemical observations, and estimation of the rates of vertical exchange of the Black Sea waters based on large-scale temporal averaging;
- verification of the model by comparing the results from calculations with observed findings;
- using the model to study and forecast the main features of large-scale distribution, and evaluation of the residence time, of long-lived radionuclides of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  contaminating the Black Sea as a result of the Chernobyl accident.

Table 1 sets out the components making up the Black Sea water balance (for various flow percentiles in the Danube and Dnieper rivers). The relationship between river flow volume and salt content in the Black Sea is illustrated in Fig. 2 (assuming stationary conditions). It shows that the amount of salt currently in the Black Sea corresponds to a mean annual 30 percentile flow for the Danube and Dnieper.

To estimate vertical water exchange it was assumed that ingress of salt waters from the Sea of Marmara into the deep layers of the Black Sea was of equal probability. Assuming this, and that the Black Sea's vertical salinity profiles remained stationary, the Type 1 and 2 equations were solved for the parameters representing the rate of vertical water exchange on the basis of large-scale temporal averaging. Table 2 shows the basic data used in these calculations, the distribution of the lower Bosphorus current in the Black Sea layers and estimates of inter-layer water exchange.

Our analysis showed that, when calculating the intensity of vertical water exchange on the basis of the rate of water flow from the surface to the bed, the period for complete vertical mixing of the Black Sea waters was 101 years, equivalent to  $0.7 \cdot 10^{-4} \text{ cm} \cdot \text{s}^{-1}$  on average. When calculating the period of vertical mixing on the basis of replacement of the water in each layer via exchange with adjacent layers, we arrived at a figure of 51 years, equivalent to  $1.5 \cdot 10^{-4} \text{ cm} \cdot \text{s}^{-1}$ . These values generally tallied with those found in the literature.<sup>1</sup>



Figs. 3 and 4 show the profiles of the vertical mixing coefficients and the stationary profiles of Black Sea salinity for different river-flow percentiles. We can see here that the stationary profile of salinity change by depth tallied with that naturally observed only for a vertical water exchange commensurate with the 30 percentile flow of the Danube and Dnieper combined. Thus, in order to predict the large-scale vertical transport of  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  in the Black Sea, we used the estimates for the water mixing processes (by depth) using the 30 percentile flow.

We verified the degree of model applicability (Fig. 1) in making predictions by comparing computed and observed data on the distribution of  $^{137}\text{Cs}$  in the Black Sea from 1986 to 1990.

Fig. 5 shows the vertical distribution profiles of  $^{137}\text{Cs}$  from 1986 to 1989 and the figures obtained from modelling its vertical migration (starting from the initial radioactive contamination of the waters in 1986). The observed profiles of  $^{137}\text{Cs}$  vertical distribution were obtained from 6, 9 and from our own data gathered during field work between 1986 and 1989. We averaged out the vertical distribution patterns in the various parts of the sea for each corresponding year, and calculated the total and average  $^{137}\text{Cs}$  content in each separate layer. Confidence intervals were calculated for a significance level of 0.1.

As Fig. 5 shows, the model reflected the dynamics of  $^{137}\text{Cs}$  vertical transport in the Black Sea with a sufficient degree of accuracy. Extending the results of these numerical experiments over a longer time interval (Fig. 6) showed that after the Chernobyl accident the amount of  $^{137}\text{Cs}$  in the Black Sea decreases exponentially with a time constant of 17 years (Fig. 6a), and will reach the pre-accident level in 13 years.

Analysis of the radioactive caesium flow, as governed by various mechanisms, revealed that the  $^{137}\text{Cs}$  flow deposited in bed sediments via sedimentation processes did not exceed 8-15% of the  $^{137}\text{Cs}$  radioactive decay rate, even given extreme estimates of the intensity of sedimentation and concentrational ability of the sediments. The amount of  $^{137}\text{Cs}$  in the bed sediments was considerably lower than 1% of the amount in the aqueous medium, and the ingress of radioactive

caesium into the water via remobilization processes was insignificant, even for extreme estimates.

The function relating to changes in the amount of  $^{137}\text{Cs}$  in the biologically active layer (0-200 m) of the Black Sea was also exponential in character (Fig. 6b), the time constant for this process being 12.5 years. In the surface layer (0-50 m), the mechanism of change in  $^{137}\text{Cs}$  content displayed a double exponential character, with time constants of 2.3 and 12.5 years (Fig. 6c). The contribution of the exponent reflecting the mechanism of the faster-attenuating processes was 57%. Annual removal of radioactive caesium through the straits did not exceed 3% of its amount in the 0-50 m layer.

In order to verify the model over a large-scale time interval we compared a) the trends we had obtained with b) already existing data on changes in  $^{137}\text{Cs}$  content in the 0-50 m and 0-200 m water layers of the Black Sea which had been recorded after atmospheric nuclear weapons testing had ended and prior to the Chernobyl accident. Fig. 6 shows the data for 1964<sup>5</sup> and 1977<sup>3</sup> expressed in relative units, adjusted to when readings for the 0-200 m layer began and relating to the exponential function reflecting the slower processes involved in reducing the  $^{137}\text{Cs}$  concentration in the 0-50 m layer. We concluded from this that from 1964 to 1977 the impact of abiotic and biotic processes on the field of radionuclides led to rates of reduction in the amount of  $^{137}\text{Cs}$  similar to the expected change in the content of this radionuclide in the Black Sea after the Chernobyl accident. This proves the adequacy of the model, and allows us to conclude that the mechanisms as predicted by the model reflect the large-scale impact of hydrophysical and biogeochemical processes on the migration of radioactive caesium in the medium, regardless of from where it originates and how much of it enters the Black Sea surface waters.

Thus, our studies using the model showed that as a reaction to the radioactive contamination occurring at individual sites, the combined impact of hydrophysical and biogeochemical processes (plus that of radioactive decay) leads to a time-related exponential reduction in  $^{137}\text{Cs}$  content in the Black Sea and its individual layers. An isotope's residence time in any system is usually taken to be five times its time constant of decay or elimination. Therefore, calculations based on the model led us to conclude that  $^{137}\text{Cs}$

residence time in the 0-50 m and 0-200 m layers will not exceed 63 years, with the corresponding figure for the Black Sea as a whole being 85 years. Comparing these estimates with the "lifetime" of  $^{137}\text{Cs}$  atoms (150 years) showed that hydrophysical and biogeochemical processes halve the residence time of  $^{137}\text{Cs}$  in the Black Sea waters.

We know that the Chernobyl accident contaminated the Black Sea not only with  $^{137}\text{Cs}$  but also  $^{90}\text{Sr}$ . In contrast to caesium, strontium does not become lodged in soils to any great extent. Therefore, in addition to the aeolian deposition which occurred immediately after the accident, river flow is a significant factor in  $^{90}\text{Sr}$  contamination of the Black Sea. Therefore, the mechanisms involved in formation in the Black Sea of the radioactive field caused by  $^{90}\text{Sr}$  radiation depend not only on large-scale processes, but also on geophysical processes occurring on a smaller temporal and spatial scale of averaging.

$^{90}\text{Sr}$  has many features in common with  $^{137}\text{Cs}$ , which make for identical rates of migration in the Black Sea. Both  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  are accumulated by allochthonous and biogenic suspensions, bed sediments and aquatic biota (with low accumulation coefficients).<sup>8</sup> The radioactive half-lives of these radionuclides are practically the same. That hydrophysical and biogeochemical processes have a similar effect on the movement of these radioisotopes, is demonstrated by the fact that the  $^{90}\text{Sr}/^{137}\text{Cs}$  ratio in the depths of the World Ocean does not change.<sup>5</sup> Therefore, we can expect that the large-scale migration trends seen in connection with  $^{137}\text{Cs}$  will fully reflect those of  $^{90}\text{Sr}$  too.

Thus, our research leads us to draw the following conclusions. Following contamination of the Black Sea waters by the Chernobyl accident,  $^{137}\text{Cs}$  content decreases in the 0-50 m layer in line with a double exponential function with time constants of 2.3 and 12.5 years; in the biologically active layer (0-200 m) it changes exponentially with a time constant of 12.5 years; while the decrease in the amount of  $^{137}\text{Cs}$  in the Black Sea as a whole changes exponentially with a time constant of 17 years. Under the impact of hydrophysical and biogeochemical processes the residence time of long-lived  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  radionuclides in the Black Sea waters is 63 to 85 years, i.e. it is halved compared to the average lifetime of the atoms in these radionuclides (150 years).

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Black Sea salt balance equation

for layer No 1 (0-50 m):

$$\frac{dS_1}{dt} = \frac{1}{V_1} \left[ f_{KA} S_K + FS_F + RS_{oc} + f_{21} S_2 - (Q_B + f_{AK} + f_{12}) S_1 \right] \quad (1)$$

for the i-layers (2-8) beneath the surface horizon:

$$\frac{dS_i}{dt} = \frac{1}{V_i} \left[ d_i Q_h S_\delta + f_{i-1,i} S_{i-1} + f_{i+1,i} S_{i+1} - (f_{i,i-1} + f_{i,i+1}) S_i \right] \quad (2)$$

where

- $S_i$  and  $V_i$  - i-layer salinity and volume;
- $f_{KA}$  and  $S_K$  - annual ingress and salinity of water from the Sea of Azov;
- $F$  and  $S_F$  - river flow and salinity;
- $R$  and  $S_{oc}$  - precipitation and salt content in precipitation;
- $f$  with subscripts - water ingress from layer designated by the 1st subscript into the layer designated by the 2nd subscript;
- $Q_b$  and  $Q_h$  - upper and lower Bosphorus currents;
- $d_i$  - portion of flow of the lower Bosphorus current entering the i-layer;
- $S_\delta$  - salinity of the waters of the lower Bosphorus current;
- $f_{AK}$  - flow from Black Sea into Sea of Azov.

Equation covering radionuclide balance in the benthic layer:

$$\frac{dq_8}{dt} = C_1 f_{18} + K_8 (C_1 P_{8(1)} - C_8 P_{8(8)}) + K_A (C_1 A_1 - C_8 A_8) + C_9 m (r - r_{gc}) - C_8 V_8 (r_c + \lambda) \quad (5)$$

Equation covering radionuclide balance in bed sediments:

$$\frac{dq_9}{dt} = C_8 (K_8 P_{8(8)} + K_A A_8 + V_8 r_c) - C_9 m (r + r_{gc} + \lambda) \quad (6)$$

where

- m - bed sediment boundary layer mass;
- C<sub>g</sub> - radionuclide concentration in the bed sediment boundary layer;
- r<sub>c</sub>, r and r<sub>gc</sub> - indicators of rate of sorption, desorption and remobilisation processes during interaction of bed sediments with water masses;
- g<sub>m</sub> - amount of radionuclide in the Black Sea bed sediments.

Equation covering radionuclide balance in the Black Sea waters:

For layer No 1 (0-50 m):

$$\frac{dq_1}{dt} = C_A(A - A_1) + C_F F + C_R R + W + C_K f_{K1} + C_2 f_{21} - (f_{12} + Q_B + \lambda V_1 + K_B P_B + f_{A1}) C_1 \quad (3)$$

For the intermediate i-layers:

$$\frac{dq_i}{dt} = C_{i-1} f_{i-1,i} + C_{i+1} f_{i+1,i} + C_H \alpha_i Q_H + K_B (C_{i-1} P_B(i-1) - C_i P_B(i)) + K_A (C_{i-1} A_{i-1} - C_i A_i) - C_i (f_{i,i-1} + f_{i,i+1} + \lambda V_i) \quad (4)$$

where

- $q_i$  - amount of radionuclide in i-layer;
- $C_i$  - radionuclide concentration in i-layer water;
- $C_A, C_F, C_R$  and  $C_K$  - radionuclide concentration in allochthonous suspensions, in river flow water, in aqueous precipitation over the Black Sea and in the water of the Sea of Azov;
- $\lambda$  - radionuclide decay constant;
- $A$  and  $A_i$  with subscripts - allochthonous flows of radionuclide onto the Black Sea surface and flows of removal from layers indicated by the subscripts;
- $W$  - aeolian fallout of radionuclide over the sea surface;
- $P_B$  with subscripts - rate of elimination of biomass from the layer designated by the subscript;
- $K_A$  and  $K_B$  - coefficients of radionuclide accumulation by allochthonous and biogenic substances;
- $C_H$  - radionuclide concentration in the water of the lower Bosphorus current.

Table 1: Water balance of the Black sea for annual flow percentiles in the Danube and Dnieper ( $\text{km}^3 \cdot \text{year}^{-1}$ )<sup>2,7</sup>

Number of variant	1		2		3		4		5		6		7	
	10 %		20 %		30 %		40 %		50 %		75 %		95 %	
Balance component	Gain	Loss	Gain	Loss	Gain	Loss	Gain	Loss	Gain	Loss	Gain	Loss	Gain	Loss
Danube flow	258		238		225		213		202		176		143	
Dnieper flow	60		55		52		50		47		41		33	
Flow of remaining rivers	97		97		97		97		97		97		97	
Total river flow	415		390		374		360		346		314		273	
Evaporation		330		330		330		330		330		330		330
Inflow from sea of Azov	92		92		92		92		92		92		92	
Precipitation	116		116		116		116		116		116		116	
Inflow into sea of Azov		66		66		66		66		66		66		66
Upper Bosphorus current		406		392		382		375		367		349		326
Lower Bosphorus current	182		192		199		204		210		223		240	
Total	805	802	790	788	781	778	772	771	764	763	745	745	721	722



Table 2: Volumes, areas and salinity of individual layers and characteristics of distribution of lower Bosphorus current and large-scale water exchange.

Layer No	Layer boundaries (m)	Layer volume (km <sup>3</sup> )	Mean salinity of layer (%)	Area bounding layer above (km <sup>2</sup> )	Portion of lower Bosphorus current entering layer (%)	Water exchange with underlying layer (km <sup>3</sup> . year <sup>-1</sup> )
1	0 - 50	20 000	18.27	413 000	0	3740
2	50 - 100	17 900	19.11	380 000	3.4	2402
3	100 - 150	15 818	20.28	344 000	3.0	3878
4	150 - 200	15 793	20.95	312 000	3.0	2887
5	200 - 500	90 805	21.77	299 450	17.0	5051
6	500 - 1000	142 772	22.14	287 000	27.0	7303
7	1000 - 1500	129 096	22.30	265 000	24.6	13 747
8	1500 - 2200	115 532	22.34	219 000	22.0	0

# **Artificial Radionuclides in Aquatic Biota of Major European Rivers**

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In the past decades, one of the major anthropogenic factors with a growing impact on the main European watercourses has been radioactive contamination as a direct consequence of the development of atomic energy. Two nuclear plants - at Balakova and Kalinin - are currently operating in the Volga basin. And while 10 nuclear plants were in operation in the Danube basin in 1978, there will be a total of 29 in the year 2000 with an aggregate output of 14 800 Mwt.<sup>4,9,10</sup>

The Rovensk, Khmel'nits, Zaporozhe, Chernobyl, Kursk and Smolensk nuclear plants are located in the Dnieper basin. The South Ukrainian nuclear plant at Yuzhny Bug continues to increase its output.

Studies on the content of radionuclides in water, suspended particles, bed sediment and aquatic biota of different trophic levels relate to different periods and regions and suggest that our knowledge of the radiological situation in Europe's largest rivers varies considerably.<sup>1,3,5,8,11</sup> Radioactive contamination of the Danube has been studied in greatest detail - a corollary of the intensive development of nuclear energy and of international interest in radioecological problems in this region.

Here we present the results of studies of Sr-90 and Cs-137 - the ecologically most significant artificial radionuclides - in aquatic biota of different trophic levels in the Volga, Danube and Dnieper. The studies were carried out in 1988.

#### Methodology

Samples were taken from the Volga over a distance of 3 085 km ranging from the estuary to the city of Tver during the "Complex Expedition" organized by the Aquatic Problems Institute of the Academy of Sciences of the Ukrainian SSR in August and September 1988 on the research vessel 'Akvatoriya'. During an expedition on the research vessels 'Akademik Vernadsky' and the 'A. V. Topachevsky', belonging to the Institute of Hydrobiology of the Ukrainian Academy of Sciences, samples were taken from the Soviet stretch of the Danube (from the estuary to the city of Reni, a distance of 163 km); samples were also taken from the Dnieper in the Kiev, Kanev and Kremenchug reservoirs and in the Dnieper-Bug Estuary.

In order to determine radionuclide content, the aquatic biota were first ashed at temperatures of not less than 450°C. The radionuclides were separated from the samples using radiochemical methods.<sup>2,6</sup> Sr-90 was determined through its daughter yttrium-90, while Cs-137 was determined through the final product Cs<sup>137</sup>Sb<sup>219</sup> with the aid of a low background UMF-1500M plus SBT-13 end-window counter and an AI-4096/A-90 amplitude analyser with semiconductor detector. Sr-90 and Cs-137 content was determined in green filamentous algae, 20 species of higher aquatic plants, 14 species of molluscs and 13 fish species. Content is expressed in Bq/kg in vegetation (dry mass) and in animals (wet mass).

Note that the investigations were carried out two years after the Chernobyl accident of 26 April 1986. The uranium fission fragments ejected during the reactor explosion were also distributed in the basins of the investigated rivers. Since the accident the short-lived radionuclides had disintegrated and so it was essential to determine the content of the more dangerous, long-lived radionuclides, i.e. Sr-90 and Cs-137, in aquatic biota.

## Findings

The difficulties of a comparative analysis of the level of radioactive contamination of the ecosystems of such major rivers as the Volga, Danube and Dnieper are conditioned by the great disparities in the physical and geographical characteristics of the river basins, the seasonal and long-term dynamics of water runoff, and the peculiarities of their hydrochemical and hydrobiological regimes. Added to this were the complications in synchronizing the collection of samples, determining the ecological classification of the biotopes and the growth characteristics of the aquatic biota. However, radioactive contamination of the river basins is mainly caused by nuclear power plants, and by accidental releases in particular.

Both for the abiotic (water, bed sediment) and biotic (aquatic biota of different trophic levels) components, quantitative estimates of radionuclide content in bed sediment and aquatic biota constitute the most objective indicator of the radioecological situation in aquatic ecosystems. It is this that reflects the intensity of biotic migration of the radionuclides and the risk of radioactive contamination for aquatic ecosystems as a whole.

For comparison purposes let us examine the mean figures for Sr-90 and Cs-137 content in aquatic biota of the three rivers (Table 1).

### Volga:

In the green filiform algae *Cladophora glomerata* the recorded concentration of Sr-90 ranged from 0.4 to 9.7 Bq/kg, with a mean of 6.1 Bq/kg, while for Cs-137 the figures ranged from 9.3 to 24.5 Bq/kg. Mean specific radioactivity of algae was 15.4 Bq/kg for Cs-137 - two and a half times that of Sr-90. Of the 16 species of higher aquatic plants examined, the smallest quantity of Sr-90 was found in *Butomus umbelatus* (0.9 Bq/kg) and *Typha latifolia* (1.5 Bq/kg), while the maximum was found in *Potamogeton natans* (18.5 Bq/kg) and *P. perfoliatus* (32.6 Bq/kg). For Cs-137 the range was somewhat wider - from 0.6 Bq/kg for *B. umbelatus* and 1.6 Bq/kg for *Phragmites australis* to 23.0 Bq/kg for *Potamogeton perfoliatus* and 40.7 Bq/kg for *Spirodella polyrhiza*. Of the 8 species of molluscs, Sr-90 concentration was least in *Lymnaea ovata* (4.9 Bq/kg) and *Unio pictorum* (5.2 Bq/kg), and greatest in *Dreissena polymorpha* (16.5 Bq/kg). For Cs-137, concentrations ranged from 2.2 Bq/kg for *Viviparus viviparus* to 21.6 Bq/kg for *Anodonta* sp. In the 9 fish species investigated, the limit values for Sr-90 were considerably lower than for algae, higher aquatic plants and molluscs (Table 2). Accumulation was lowest in the sturgeon family: *Acipenser guldenstadti* (0.2 Bq/kg), *A. ruthenus* (0.6 Bq/kg), *A. stellatus* (0.7 Bq/kg). Against this, Sr-90 content reached 1.4 Bq/kg in *Perca fluviatilis* and 1.6 Bq/kg in *Cyprinus carpio*.

Scatter was quite low for Cs-137 in fish, the measurements ranging from 1.1 Bq/kg in *Abramis brama* to 3.2 Bq/kg in *Cyprinus carpio*.

### Danube:

Mean concentrations of both radionuclides were higher than in the Volga for green filiform algae, molluscs and fish. Only in the case of higher aquatic plants was the concentration range of Sr-90 lower than in the Volga (Table 2). The mean quantity of Sr-90 in the alga *Cladophora glomerata* was 17.0 Bq/kg, while for Cs-137 the corresponding

figure was 39.6 Bq/kg. Of the 7 higher aquatic plants, Sr-90 was lowest in *Typha latifolia* (0.8 Bq/kg) and highest in *Glyceria* sp. (5.0 Bq/kg). The corresponding figures for Cs-137 were higher, ranging from 6.3 Bq/kg for *Phragmites australis* to 93.6 Bq/kg for *Potamogeton tucens*.

The concentration of Sr-90 in molluscs ranged from 3.5 Bq/kg for *Unio tumidus* to 64.1 Bq/kg for *Lymnaea stagnalis*, and of Cs-137 from 3.3 Bq/kg for *Unio pictorum* and *Viviparus viviparus* to 11.7 Bq/kg for *Unio tumidus* and 11.8 Bq/kg for *Lymnaea stagnalis*. A unique quantity of Sr-90 was measured in the fish species *Rutilus rutilus* and *Cyprinus carpio* (1.3 and 1.6 Bq/kg respectively), fished both in the Volga and in the Danube. And while for *Stizostedion lucioperca* mean Sr-90 concentration was 0.9 Bq/kg in the Volga, it was 2.3 Bq/kg in the Danube.

For all 4 Danube fish species studied, Cs-137 concentrations were higher than Sr-90, ranging from 10.7 Bq/kg to 16.9 Bq/kg. For aquatic biota, therefore, radioactivity in the Danube exceeded levels in the Volga by the following factors:

Sr-90: 2.8 (algae); 4.4 (molluscs); 1.9 (fish) Cs-137: 2.6 (algae); 6.5 (fish).

Dnieper:

Here the impact of radioactive contamination of the river following the Chernobyl accident was clearly visible from the measurements. Accumulations of Sr-90 and Cs-137 in Dnieper populations were far in excess of those found in the corresponding organisms in the Volga and Danube. Cs-134 was constantly identified in samples taken in 1988, while in some samples ruthenium-106 + rhodium-106 and cerium-144 were also recorded. However, a significant contribution of these radionuclides to total radioactivity was established only in aquatic biota from the Kiev reservoir (Table 3).

The mean concentrations of Sr-90 and Cs-137 in green filiform algae were 292.3 Bq/kg and 110.0 Bq/kg respectively. For the higher aquatic species listed in Table 1, Sr-90 concentrations ranged from a minimum of 92.1 Bq/kg in *Ceratophyllum demersum* to a maximum of 440.3 Bq/kg in *Elodea canadensis*. For Cs-137, the concentrations were far higher, ranging from 133.2 Bq/kg in *Typha angustifolia* to 3034.0 Bq/kg in *C. demersum*.

Of all the aquatic flora and fauna studied, the highest concentration of Sr-90 was found in molluscs from the Dnieper. Moreover, Sr-90 levels in these biota are in most cases far higher than those of Cs-137. This is because Sr-90 is absorbed as an analogue to calcium in the structural tissue that forms the shells of these organisms. For Dnieper molluscs, Sr-90 concentrations ranged from 28.0 in *Anodonta* sp. to 4329.0 in *Sphaerium* sp. For Cs-137 the concentrations range from 33.3 for *Anodonta* sp. to 351.5 Bq/kg for *Lymnaea auricularia*. If Sr-90 predominated in the case of molluscs, for fish it was Cs-137, with concentrations ranging from 161.9 Bq/kg in *Rutilus rutilus* to 1478.0 Bq/kg in *Esox lucius*.

The results give us a picture of Sr-90 and Cs-137 concentrations in the aquatic biota of the three rivers at different trophic levels. Table 2 shows that Sr-90 concentrations are lowest in Volga biota, ranging from 0.2 Bq/kg to 32.6 Bq/kg. In the Danube, concentrations of Sr-90 were higher, ranging from 0.2 to 64.0 Bq/kg. The highest concentrations were found in the aquatic organisms of the Dnieper reservoir Cascade -

from 5.9 to 4 144 Bq/kg. In all rivers the concentrations were highest in molluscs, with means of 7.6, 33.7 and 2 075.7 Bq/kg in the Volga, Danube and Dnieper respectively (Table 2). Besides, in the Volga Sr-90 concentrations were particularly high in higher aquatic plants.

Depending on the sampling point and the species-specific characteristics of the organisms, Cs-137 levels ranged from 0.6 to 58.8 Bq/kg (Volga), 2.6 to 93.6 Bq/kg (Danube) and 9.6 to 44 400 Bq/kg (Dnieper). Cs-137 accumulation was highest in green filiform algae and in higher aquatic plants. Specifically, mean concentrations in the Volga were (in descending order) 15.4 Bq/kg (green filiform algae), 9.6 Bq/kg (higher aquatic plants), 7.2 Bq/kg (molluscs) and 2.1 Bq/kg (fish). This regularity in the accumulation of Cs-137 as a correlate of the level of evolutionary development was not as marked in the Danube and Dnieper. For the Danube the sequence was: higher aquatic plants, green filiform algae, fish, molluscs; for the Dnieper it was: green filiform algae, higher aquatic plants, fish, molluscs. A comparison of the results shows that mean Sr-90/Cs-137 contamination of biota in the Danube and Dnieper was higher than in the Volga by factors of 4.5/7 and 21-272/11-250 respectively. However, concentrations of radionuclides in individual samples, particularly in the upper reservoirs of the Dnieper, can exceed the mean by over 4 orders of magnitude.

One distinctive characteristic of radioactive contamination of the Dnieper's ecosystem is the steady decrease in concentrations in all categories of organisms from the Kiev reservoir to the Dnieper-Bug Estuary. Sr-90 accounts for 28% of total radioactivity in green filiform algae in the Volga, 29% in the Danube, and 29% in the Dnieper. For higher aquatic plants the corresponding figures are 50%, 17% and 34% (Table 4). In the case of molluscs, Sr-90 predominates, with mean concentrations ranging from 51% in the Volga to 91% in the Dnieper. In the case of fish, however, Cs-137 was the main radionuclide, accounting for 67% (Volga), 87% (Danube) and 91% (Dnieper) of total radioactivity in these organisms.

#### Conclusions

We studied the content of the artificial radionuclides Sr-90 and Cs-137 in the aquatic biota of different trophic levels in Europe's major rivers: the Volga, Danube and Dnieper. The data indicate that the content of Sr-90 in the Volga's aquatic biota ranges from 0.4 to 9.7 Bq/kg (algae, *Cladophora glomerata*), 0.4 to 32.6 Bq/kg (higher aquatic plants), 2.3 to 28.1 Bq/kg (molluscs) and 0.2 to 2.2 Bq/kg (fish). The corresponding figures for the Danube are: 3.9 to 30.1 Bq/kg (green filiform algae), 0.2 to 5.7 Bq/kg (higher aquatic plants), 3.5 to 64.0 Bq/kg (molluscs) and 1.4 to 2.3 Bq/kg (fish), while for the Dnieper they are: 92.1 to 525.4 Bq/kg (green filiform algae), 25.5 to 1 165.5 Bq/kg (higher aquatic plants), 6.5 to 4 144.0 Bq/kg (molluscs) and 5.9 to 120.6 Bq/kg (fish). For Cs-137 in the Volga the figures were: 9.3 to 24.5 Bq/kg (green filiform algae), 0.6 to 58.8 Bq/kg (higher aquatic plants), 0.9 to 35.6 Bq/kg (molluscs) and 0.8 to 5.1 Bq/kg (fish).

The corresponding figures for the Danube were: 8.9 to 69.6 Bq/kg (green filiform algae), 2.6 to 93.6 Bq/kg (higher aquatic plants), 3.3 to 11.8 Bq/kg (molluscs) and 10.7 to 17.0 Bq/kg (fish). The Dnieper figures were: 259.4 to 2 479.0 Bq/kg (green filiform algae), 10.1 to 5 920.0 Bq/kg (higher aquatic plants), 9.6 to 148.0 Bq/kg (molluscs) and 15.5 to 44 400.0 Bq/kg (fish). The accumulation of artificial radionuclides

in aquatic biota is mainly determined by the quantity of such nuclides in the inorganic components, their uptake via the food chain and the ecological and physiological specifications of the organisms' development.

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Table 1: Sr-90 and Cs-137 content in aquatic biota of the Volga, Danube and Dnieper in 1988. Bq/kg

Species	VOLGA		DANUBE		DNEPER	
	Sr-90	Cs-137	Sr-90	Cs-137	Sr-90	Cs-137
<i>Cladophora plomerata</i>	6,1	15,4	17,0	39,6	292,3	1110,0
<i>Glyceria</i> sp.	x	x	5,0	12,0	x	214,6
<i>Phragmites australis</i>	x	1,6	1,5	6,3	x	x
<i>Typha latifolia</i>	1,5	8,5	0,8	7,0	365,3	341,9
<i>Typha angustifolia</i>	x	x	2,5	12,2	51,8	133,2
<i>Butomus umbellatus</i>	0,9	0,6	x	x	x	x
<i>Sagittaria sagittifolia</i>	5,9	1,9	2,4	29,0	x	x
<i>Salvinia natans</i>	2,6	3,7	x	x	x	x
<i>Hydrocharis morsus ranae</i>	14,1	9,6	x	x	x	x
<i>Spirodela polyrrhiza</i>	17,0	40,7	x	x	214,2	395,9
<i>Nymphaea alba</i>	16,6	20,7	x	x	x	x
<i>Potamogeton natans</i>	18,5	18,5	x	x	x	x
<i>Potamogeton rufescens</i>	32,6	12,9	x	x	x	x
<i>Potamogeton pectinatus</i>	5,1	7,2	x	x	x	x

Table 1 - (Continued)

Species	VOLGA		DANUBE		DNIEPER	
	Sr-90	Cs-137	Sr-90	Cs-137	Sr-90	Cs-137
<i>Potamogeton perfoliatus</i>	11,0	23,0	x	x	296,0	473,6
<i>Potamogeton tucens</i>	10,7	8,5	2,9	93,6	x	x
<i>Ceratophyllum demersum</i>	7,4	11,5	x	x	92,1	3034,0
<i>Myriophyllum spicatum</i>	10,7	8,6	x	x	x	x
<i>Elodea canadensis</i>	x	x	x	x	440,3	1628,0
<i>Trapa natans</i>	0,3	2,0	x	x	x	x
<i>Iris pseudacorus</i>	x	x	2,0	13,7	x	x
<i>Unio tumidus</i>	6,2	5,5	3,5	11,7	132,2	66,3
<i>Unio pictorum</i>	5,2	4,9	7,0	3,3	469,0	42,7
<i>Anodonta piscinalis</i>	6,1	6,4	x	x	175,6	x
<i>Anodonta sp.</i>	7,4	21,6	6,9	10,8	28,0	33,3
<i>Sphaerium sp.</i>	x	x	x	x	4329,0	x
<i>Dreissena polymorpha</i>	16,5	4,9	x	x	x	x
<i>Dreissena bugensis</i>	x	x	x	x	496,2	43,1
<i>Viviparus viviparus</i>	5,3	2,2	46,6	3,3	1322,0	112,8
<i>Lymnaea auricularia</i>	x	x	x	x	138,2	351,5

Table 1 - (Continued)

Species	VOLGA		DANUBE		DNIEPER	
	Sr-90	Cs-137	Sr-90	Cs-137	Sr-90	Cs-137
<i>Lymnaea ovata</i>	4,9	7,8	x	x	155,1	110,1
<i>Lymnaea stagnalis</i>	11,4	3,4	64,1	11,8	358,2	54,4
<i>Planorbis corneus</i>	x	x	15,8	3,2	540,2	105,9
<i>Planorbis</i> sp.	x	x	28,5	x	x	x
<i>Lithoglyphus naticoides</i>	x	x	x	x	1715,0	110,9
<i>Acipenser guldenstadti</i>	0,2	1,4	x	x	x	x
<i>Acipenser ruthenus</i>	0,6	1,3	x	x	x	x
<i>Acipenser stellatus</i>	0,7	2,6	x	x	x	x
<i>Esox lucius</i>	1,0	2,9	x	x	36,8	1478,0
<i>Rutilus rutilus</i>	1,3	3,0	1,4	11,0	21,8	161,9
<i>Aspius aspius</i>	x	x	1,7	15,1	22,9	294,5
<i>Glicca bjoerkna</i>	x	x	x	x	56,3	264,0
<i>Abramis brama</i>	1,1	1,1	1,9	12,8	25,2	298,0
<i>Abramis ballerus</i>	x	x	x	x	25,1	211,1
<i>Pelecus cultratus</i>	x	x	x	x	31,7	498,0
<i>Cyprinus carpio</i>	1,6	3,2	1,6	10,7	x	x
<i>Stizostedion lucioperca</i>	0,9	2,7	2,3	16,9	40,2	656,0
<i>Perca fluviatilis</i>	1,4	1,4	x	x	54,4	616,0

Table 2: Concentration limits and mean content of Sr-90 and Cs-137 in aquatic biota of the Volga, Danube and Dnieper

Groups of Aquatic biota	Sr-90			Cs-137		
	Min.	Max.	Mean	Min.	Max.	Mean
<b>VOLGA</b>						
Green filiiform algae	0.40	9.7	6.1	9.3	24.5	15.4
Higher aquatic plants	0.4	32.6	9.6	0.6	58.8	9.4
Molluscs	2.3	28.1	7.6	0.9	35.6	7.2
Fish	0.2	2.2	1.0	0.8	5.1	2.1
<b>DANUBE</b>						
Green filiiform algae	3.9	30.1	17.0	8.9	69.9	39.6
Higher aquatic plants	0.2	5.7	2.9	2.6	93.6	48.1
Molluscs	3.5	64.0	33.7	3.3	11.8	7.5
Fish	1.4	2.3	1.9	10.7	17.0	13.7
<b>DNIEPER</b>						
Green filiiform algae	92.1	525.4	292.3	259.4	2479.0	1110.0
Higher aquatic plants	25.5	1165.5	356.3	10.1	5920.0	740.0
Molluscs	6.5	4144.0	2075.7	9.6	148.0	78.8
Fish	5.9	120.6	21.8	15.5	44400.0	518.0

Table 3: Contribution (%) of fission products in artificial radioactivity of aquatic biota in the Kiev reservoir (1988)

Aquatic biota	Sr-90	Cs-137	Cs-134	Ce-144	Ru-106+Rh-106
Cladophora glomerata	15.0	39.0	11.0	35.0	x
Viviparus viviparus	91.5	3.0	0.5	3.0	2.0
Dreissens bugensis	94.0	2.5	1.0	2.5	x
Esox lucius					
Edible part	0.5	66.0	20.5	13.0	x
Non-edible part	1.9	67.0	21.6	9.5	x

Table 4: Contribution (%) of Sr-90 and Cs-137 in aquatic biota of the Volga, Danube and Dnieper 1988

Aquatic biota	VOLGA		DANUBE		DNIEPER	
	Sr-90	Cs-137	Sr-90	Cs-137	Sr-90	Cs-137
Green filiform algae	28	72	29	71	29	71
Higher aquatic plants	50	50	17	83	34	66
Molluscs	51	49	65	35	91	9
Fish	33	67	13	87	9	91



**The impact of the Chernobyl  
Accident on Continental  
Aquatic Ecosystems.  
A Literature Review**

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## ABSTRACT

### The impact of the Chernobyl accident on continental aquatic ecosystems.

This synthesis has been produced by contract with the Commission of the European Communities (DG XI) and the International Union of Radioecologists. 117 reports from 19 countries have been studied.

Collecting information on water, sediments, aquatic vegetation and fish provides data concerning transfers in rivers and lakes which must be identified. There is a direct relation between radioactivity and the deposit level which is mainly in wet form. Beyond the areas near the accident, it is particularly noticeable in Scandinavia, Central Europe, northern Italy and in the north-west of the United Kingdom. Wide variations occur from one station to another in a same region. The radioecology of each site must be taken into consideration.

Seven radionuclides have been detected in significant amounts :  $^{132}\text{Te}$ ,  $^{131}\text{I}$ ,  $^{103} + ^{106}\text{Ru}$ ,  $^{134} + ^{137}\text{Cs}$ ,  $^{110\text{m}}\text{Ag}$ .  $^{137}\text{Cs}$  predominated very quickly.

The main characteristic lies in the acute nature of the contamination in comparison with the more chronic one brought about by nuclear testing between 1959 and 1963. The results show the speed of the radionuclide transfer and fixation process and the rapid return to former levels, except for  $^{137}\text{Cs}$ .

Fish provide the best means of comparing radionuclide concentrations through time and space. In rivers, the values are less than or equal to  $400 \text{ Bq kg}^{-1}$  w. w. of  $^{137}\text{Cs}$ . In the lakes of the regions which were affected the most by the fall-out, we find  $1000 \text{ Bq kg}^{-1}$  w. w., and up to 131,000. The effective half-life *in situ* is approximately 200 days in rivers and 500 in lakes.

Some elements will also be given for comparing the impact of Chernobyl to that of fall-out from nuclear tests.

## INTRODUCTION

The aim of this paper is to provide as complete an understanding as possible of the impact of the Chernobyl accident on aquatic ecosystems and a deeper understanding of the radionuclide transfer process. The growing number of publications has created a need for a bibliographical synthesis to make the findings more accessible. To do this, the Commission of the European Communities (DG XI) signed a contract with the International Union of Radioecologists. The first report has been written. 117 documents from 19 countries have been studied. The text is open to all information or corrections. A certain number of indications concerning water, sediments, vegetation and fish are presented here.

Among the radioactive isotopes that were released on April 26, 1986, some are highly visible in aquatic environments. Large amounts of  $^{131}\text{I}$ ,  $^{132}\text{Te}$ ,  $^{103+106}\text{Ru}$  were found, as was  $^{134+137}\text{Cs}$ , which rapidly became predominant everywhere. Strontium and cerium were found in lesser amounts, except for the area immediately surrounding Chernobyl where  $^{241}\text{Am}$ ,  $^{125}\text{Sb}$  and  $^{154+155}\text{Eu}$  were also found. The composition of the radionuclides varies over the affected areas, as do chemical forms, particularly the quantities of particle forms (Cs, Ru, Ag, Ce...) and dissolved forms (Te, I, Ru, Cs...). There are cationic forms, which are not very mobile and are highly fixed ( $\text{Cs}^+$ ,  $\text{RuNO}_3^+$ ...), and anionic forms, which are mobile and only slightly absorbed (I, Sb,  $\text{HTeO}_4$ ,  $\text{RuO}_2$ ...) [1,2].

The ecosystems in which the influence of Chernobyl is most evident are those where the deposits were most intense [3] (fig.1). It is most noticeable in Scandinavia, Central Europe, northern Italy and in Cumbria in the United Kingdom. Fall-out occurred with rainfall and was therefore closely related to atmospheric precipitation. This accounts for major differences in radioactivity levels in adjacent regions. With this variety, a separate study would be needed for each site to be able to understand the kinetics of the phenomena.

A first assessment can be made of the figures obtained, making a distinction between running water and lakes, which have different reactions. An approach comparing the impact of Chernobyl and the impact of nuclear explosions between 1959 and 1963 can be used.

## 1. DATA ON WATER RADIOACTIVITY

Two rainwater fall-out periods can be distinguished, the first on April 26 and 27, followed by a drop in radioactivity before a new increase on May 4 and 5. After Chernobyl, the activity of rainwater reached a level around ten times higher than after the atmospheric weapons tests.

Rainwater measurements generally showed considerable variations. The same phenomena were observed in continental water systems. The amounts which fell directly into surface water through precipitation accounted for most of the pollution during the first few hours following the accident. Later, more appeared through the effects of soil leaching and the movement of suspended matter [4]. Radioactivity was always very low in drinking water supplies due to the decontaminating role of water treatment facilities.

It is difficult to determine precisely how the radioactivity evolved with time in rivers and lakes. Because of the acute contamination mode, continuous and immediate radioactivity measurements were indispensable. This was only possible where permanent measurement stations were in operation. More often, field samples were only taken after the initial radioactivity peak had already subsided. Dilution in flowing water was yet another reason for taking samples immediately. The samples also had to be filtered in order to allow for differences in suspended matter content, making the process even more complicated. Unless it is known whether the measurement concerned raw water or filtered water, it is exceedingly difficult to compare the results obtained at different sites. Finally, because of the low radioactivity level, the measured values were often at or below the detection threshold, and thus of little use in radioecology research.

In lakes the radioactivity level dropped more slowly than in rivers, depending on various factors, including the water renewal rate, the presence or absence of a thermocline or the surface area to volume ratio.

Table 1 shows some typical values for rivers and lakes. Table 2 shows the maximum values observed in west German rivers [15].

Figures 2, 3 and 4 provide examples of the decrease of radioactivity in water. The radioactivity peak in river water occurred very soon after the accident and lasted only a short time, after which the radioactivity level rapidly declined because of dilution. The time it takes to come back to the situation that existed before the accident, however, may be quite long. At the end of 1987, for example, in the Elba, the values were still

higher than those of 1985, which were approximately  $3 \text{ mBq l}^{-1}$  of  $^{137}\text{Cs}$  [4]. Hubel [16] reports that for lakes, the half-life is 1.5 days for filtered surface water and 300 days for the total body of water.

## 2. RADIOACTIVITY IN SEDIMENT

Suspended matter and sediment deposits quickly absorbed contamination from the water because of their high radionuclide fixation capacity, notably for caesium. This accumulation was enhanced as activity settled on the bottom, while new activity reached the surface water as it was leached from the soil. The  $^{137}\text{Cs}$  peak is clearly visible in the top centimeter of sediments on the lake bottom, and can be compared with the activity values in lower layers corresponding to fall-out from atmospheric weapons testing.

The results (table 3) show the often high values observed. However these results must be analysed from the standpoint of hydrographic systems, drainage basins and lake structures. For example, the deposits tend to migrate downstream in rivers. A general process of sediment contamination is suggested by the data collected.

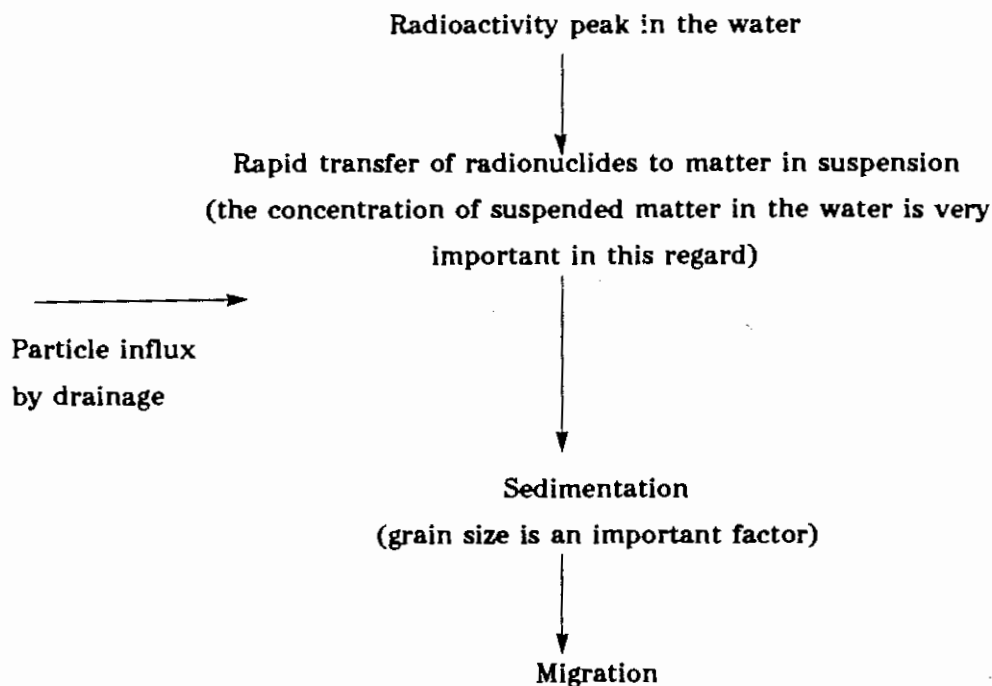


Figure 5 shows the kinetics of the phenomena in the Meuse downstream from nuclear power plants. Chernobyl caused a concentration of  $^{137}\text{Cs}$  which was twenty times higher. The return to the previous situation took about 26 months, with a half-life of 210 days.

Conkic [23] gives a half-life of 234 days for  $^{137}\text{Cs}$  and 144 days for  $^{106}\text{Ru}$  in the Danube.

### 3. RADIOACTIVITY IN AQUATIC PLANTS

Little data is available concerning aquatic plants, although they are excellent radioactivity indicators. They react quickly to radioactivity in the water, providing a relatively accurate estimate of qualitative and quantitative fluctuations (table 4). In particular, note the interest of aquatic mosses and their importance in monitoring pollution (table 5). Algae of the *Cladophora* genus are also very good radioindicators. In the Danube,  $^{137}\text{Cs}$  went from 50 Bq kg<sup>-1</sup> d.w. to 1,900 in May, 1986 [26].

### 4. RADIOACTIVITY IN FISH

Fish play an important role in radioecological research because of their position in the aquatic food chain and in human consumption. Fish provide a satisfactory comparison of radioactivity levels at different stations and monitoring of radioactivity variations in time. This comparison is made easier by the fact that the same species are found in most European rivers and lakes.

#### 4.1. River Fish

Differences were observed among species of river fish, with a strong tendency toward higher values in carnivorous fish. In order to evaluate the Chernobyl effect, river sections unaffected by liquid waste discharges from nuclear power plants had to be examined. After Chernobyl,  $^{110\text{m}}\text{Ag}$ ,  $^{106}\text{Ru}$  and  $^{103}\text{Ru}$  were found in some fish; the increase in the radioactive strontium concentration was slight. The most interesting observation was the rate of contamination by  $^{134+137}\text{Cs}$ . Tables 6, 7, 8 show some examples.

For the reasons already noted concerning the evolution of radioactivity levels in water the  $^{137}\text{Cs}$  concentrations observed in river fish were low in all cases (table 9). The fish were subjected to high radioactivity levels in the water for only a short time. This made it possible to determine *in situ* effective half-lives of the radionuclides involved. Estimates include 1 day for  $^{132}\text{Te}$ , 4 for  $^{131}\text{I}$  and 10 for  $^{103}\text{Ru}$ . Several authors [1, 18, 31, 33, 34] reported a long half-life of 100 to 300 days for  $^{137}\text{Cs}$  in agreement with experimental findings. In the Rhône, the situation returned to normal 2.5 years after the accident [33].

## 4.2. Lake Fish

The problem is different with lake fish. The radioactive contamination levels in the fish differ more sharply among lakes and regions, and even among different species of fish. Each lake is a special case, but everywhere the radioactivity levels depended on the fall-out zones ; this was especially visible for  $^{137}\text{Cs}$ . Table 10 gives a clear view of the maximum values obtained.

The effective *in situ* half-life for  $^{137}\text{Cs}$  is considerably longer than in rivers. It was never less than 100 days in herbivorous fish [2] and reached several years in some carnivorous species. The authors site half-lives of more than one year [1, 4, 6, 35, 43, 44, 45, 46]. Kinetics depend on several factors : level of fall-out, structure of the lake, chemical composition (role of  $\text{K}^+$ ), temperature, nature of the species, age ranges and food chains. In a mountain oligotrophic lake, a half-life of 3.4 years was observed for trout [43]. Carnivorous species have slower contamination and decontamination speeds and higher concentration factors. Fish raised on fish farms have lower concentrations due to their artificial food [4]. Figure 6 gives a diagram of this process.

These phenomena were observed in Finland, Sweden and also in northern Italy. The  $^{137}\text{Cs}$  concentrations in fish were highest in small lakes with concentration factors of 500 to 3,000. In the pelagic or shoreline portions of lakes, plankton-eating or carnivorous species showed the highest concentration factors.

In the Cumbrian district of the United Kingdom, the  $^{137}\text{Cs}$  concentrations varied among the fish species in decreasing order as follows : Perch --> Pike --> Brown trout --> Eels. The main parameters that must be taken into account for radionuclide transfer studies in lakes are summarized in figure 7.

This ecological variability makes it very difficult to give an overall statistical approach. The peak values obtained and the half-lives observed do, however, make it possible to form an evaluation of decontamination speeds and the time needed to return to the former situation.

In rivers where, before the accident, fish contained  $1 \text{ Bq kg}^{-1}$  w.w. of  $^{137}\text{Cs}$  [4, 19] and where the maximum values are from 20 to  $400 \text{ Bq kg}^{-1}$  w.w, it takes 2.5 to 4.5 years to return to the former levels, based on a half-life of 200 days. We have more precise data for lakes which are given in figure 8. There is a gap of at least one year between the peak activity of the fall-out [47] and that of carnivorous fish. In 1964, a maximum concentration of  $880 \text{ Bq kg}^{-1}$  w.w. in perch was observed [48] ; eight years later, the values were between 10 and  $100 \text{ Bq kg}^{-1}$  w.w. [18, 39, 49]. For 1987, Hakanson [33] gave

an average of 9,800. With a half-life of 2 years, it will take approximately 13 years to fall back to former levels. This is an average. It will be longer for cold oligotrophic lakes where the peak concentration for fish is higher ; on the other hand, in lakes which are rich in salts and for plankton-eating fish it will be much shorter.

## DISCUSSION

After the Chernobyl accident, the contamination of aquatic ecosystems could be seen as far away as Western Europe. Its level was directly related to the intensity of the fall-out connected to rainfall, and varied greatly depending on the local ecological conditions. In particular, I, Te, Ru, Sb and Cs were detected.

$^{137}\text{Cs}$  has the most lasting impact. All contamination processes were swift. After a peak of a few hours, activity in water decreased rapidly, but stayed at a level that was higher than before due to soil leaching. Radionuclides are mainly stored in sediment. Plants, in particular mosses and algae, are the best radioindicators. Fish are rapidly contaminated in direct relation to the fall-out. Caesium is transferred through the food chains. The date of contamination is an important parameter, since in winter metabolic processes are slowed. In rivers, activity is weaker because of rapid dilution. In lakes, activity is much stronger because of the time it stays in the lake and the nature of the food chains. The effective half-life of  $^{137}\text{Cs}$  in fish can be as long as three years or more. Carnivorous fish have the highest concentration, concentration factors and half-lives.

The important role of fish in the human diet must be taken into consideration. Fish consumption accounts for up to 40 % of the total quantity of  $^{137}\text{Cs}$  ingested by the human population in Finland. In Sweden radioactive contamination of fish was higher than for other plant or animal species of the region [29].

Observations made after 1963 have proven correct. The main difference lies in the chronic aspect of contamination by fall-out from nuclear tests, and the acuteness of the Chernobyl fall-out. The levels of  $^{137}\text{Cs}$  reached in aerosols, rainwater and fish after the accident were approximately 10 times higher. It may take from 2 to 15 years for the level in fish to return to its former level, depending on the region, the ecosystem and the species.

This synthetic approach allows us to present an initial overview of our knowledge in this area and raises the question of methodology. It is still necessary to ensure that consistent techniques are used throughout the cycle of radioecological studies

(Sampling --> Sample preparation --> Analytical methods --> Presentation of results --> Data interpretation and management). It is often difficult to compare data from one publication to that of another. We should point out the lack of information on strontium and  $\alpha$  emitters.

The availability of reference data is indispensable to assess the radioecological impact of nuclear facilities. Field and laboratory studies are fully complementary and must be further developed. Control measurements are not enough to interpret the mechanisms and intensity radionuclide transfer. The Chernobyl accident demonstrates the importance of water channels in the contamination process.

As the diversity of the bibliographical sources amply demonstrates, this approach requires the development of scientific cooperation. The efforts undertaken by the EEC and the IUR are a step in this direction.

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Table 1 : Examples of some radionuclides concentration in water of different rivers and lakes for 1986

(mBq l<sup>-1</sup>)

	I-131	Cs-137	Ru-103
R Pripiat [5]	16 000	18 000	8 900
I Finland [6]		5 200	2 000
V Denmark [7]		2 800	
E Sweden [8]	6 900	4 100	2 100
R Elbe [9]	22 000	4 000	
S Switzerland [10]	22 200	1 200	1 900
Danube [11]	22 500	500	4 500
Denmark [7]		21 000	
L Bavaria [12]	32 000	10 100	14 100
A Come (Italy) [13]	11 000	2 600	4 800
K Muggelsee (east Germany) [4]	11 000	10 000	
S Esthwaite (United Kingdom) [14]		280	

Table 2 : Radionuclides concentration in raw water of west Germany rivers (2 - 8 May 1986)

(mBq l<sup>-1</sup>)

	I-131	Cs-137	Ru-103
Danube	193 000	6 100	30 000
Innerste	110 000	10 000	30 000
Weser	40 000	8 000	20 000
Elbe	22 000	4 300	
Moselle	20 000	1 000	3 000
Rhine	5 600	2 000	4 000

Table 3 : Maximum values of some radionuclides concentration in sediment of different rivers and lakes for 1986.

(Bq kg<sup>-1</sup> d.w)

		Cs-134	Cs-137	Ru-103	Ru-106
R	French [19]	320	970	400	420
I	Elbe [20]	640	1 570	2 800	
V	Rhine [21]	1 000	2 500	4 700	
E	Danube [20]		7 000	400	2 000
R	Meuse [22]	500	1 100		
S					
L	Chernobyl*	1 600	12 000		900
A	Bavaria [12]	2 120	4 070	2 200	
K	Orta (Italy) [17]	1 200	3 700		
E	Windermere (U.K) [14]	1 620	3 700	7 500	
S	Lowester (U.K) [18]	140	300	30	170

\* Values obtained for May 1990 on a sample taken by ourselves.

Table 4 : Maximum values of some radionuclides concentration in aquatic plants.

(Bq kg<sup>-1</sup> d.w)

Station	Species	Cs-134	Cs-137	Ru-103	Ru-106
Po [24]	Water milfoil	120	240	1 040	450
	Pondweed	100	220	525	180
France [25]					
Tavignano (Corse)	Pondweed	26	52	52	86
Moselle	Pondweed	39	85	95	170
Ruhr [15]	Ranunculus	12	24	2	26
Loweswater U.K. [18]	Pondweed	150	310		

Table 5 : Maximum values of some radionuclides concentration in aquatic mosses (Fontinalis - Cinclidotus) after Chernobyl accident.

(Bq kg<sup>-1</sup> d.w)

Nuclide	South-East France	Ruhr	Cumbria (U.K.)
Cs-134	170	670	2 510
Cs-137	500	1 500	5 420
Ru-103	500	20	1 490
Ru-106	1 010	290	3 730
Ag-110 m	5		
Sb-125	10	230	



Table 6 : Radionuclides concentration in freshwater fish in France before and after Chernobyl accident [19]

(Bq kg<sup>-1</sup> w.w.)

Radionuclides	Before Chernobyl	After Chernobyl		
Cs-134		0.1	to	20
Cs-137	0.05 to 0.5	0.07	"	43
Ru-103		-	"	13
Ru-106		-	"	9
Ag-110m		-	"	2
Ce-144		-	"	0.5
Sr-90	0.4 to 2	0.3	"	2
H-3	27 to 80	5	"	60

Table 7 : Radionuclides concentration in Danube fish before and after Chernobyl accident [27]

(Bq kg<sup>-1</sup> w.w.)

Radionuclides	Before Chernobyl	After Chernobyl
Cs-134		11 to 32
Cs-137	0.2 to 0.5	24 to 71

Table 8 : Radioactivity in Finland river fish - May September 1986 [28]

(Bq kg<sup>-1</sup> w.w.)

Species	Cs-134	Cs-137
Pike : <u>Esox Lucius</u>	1 to 43	5 to 91
Roach : <u>Rutilus rutilus</u>	3 to 27	7 to 53
Perch : <u>Perca fluviatilis</u>	2 to 210	7 to 230

Table 9 : Maximum values for Caesium 137 in rivers fishes after Chernobyl.

(Bq kg<sup>-1</sup> w.w.)

Countries	Concentration
Sweden [29]	≤ 400
United Kingdom = Cumbria [18]	≤ 400
Finland : Loviisa [28]	≤ 230
Germany : South Bavaria [12]	≤ 100
Italy : Po [30]	≤ 80
Czechoslovakia : Danube [27]	≤ 70
Switzerland [1]	≤ 70
France : South-east [19]	≤ 50
Germany : Rhineland - Palatinate [31]	≤ 30
Austria [32]	≤ 20

Table 10 : Maximum values for caesium 137 in lakes fishes after Chernobyl.

(Bq kg<sup>-1</sup> w.w.)

Countries	Concentration
Sweden [35]	131 000
Norway : Eastern [36]	55 000
Finland : Central and eastern [37]	16 000
East Germany [4]	5 400
Italy : Viverone lake [38]	4 000
United Kingdom : Loch Dee Scotland [39]	3 000
Austria : North [40]	2 500
West Germany : Bavaria South [12]	1 600
Hungary : Balaton lake [26]	700
Switzerland : Constance lake [1]	400
Ireland : North [41]	270
France : North Corse [25]	180
Holland : Northern [42]	50



**Fig. 1 : Zones the most affected by the fallout  
after Chernobyl accident**

(Decreasing radioactive density from 1 to 6)

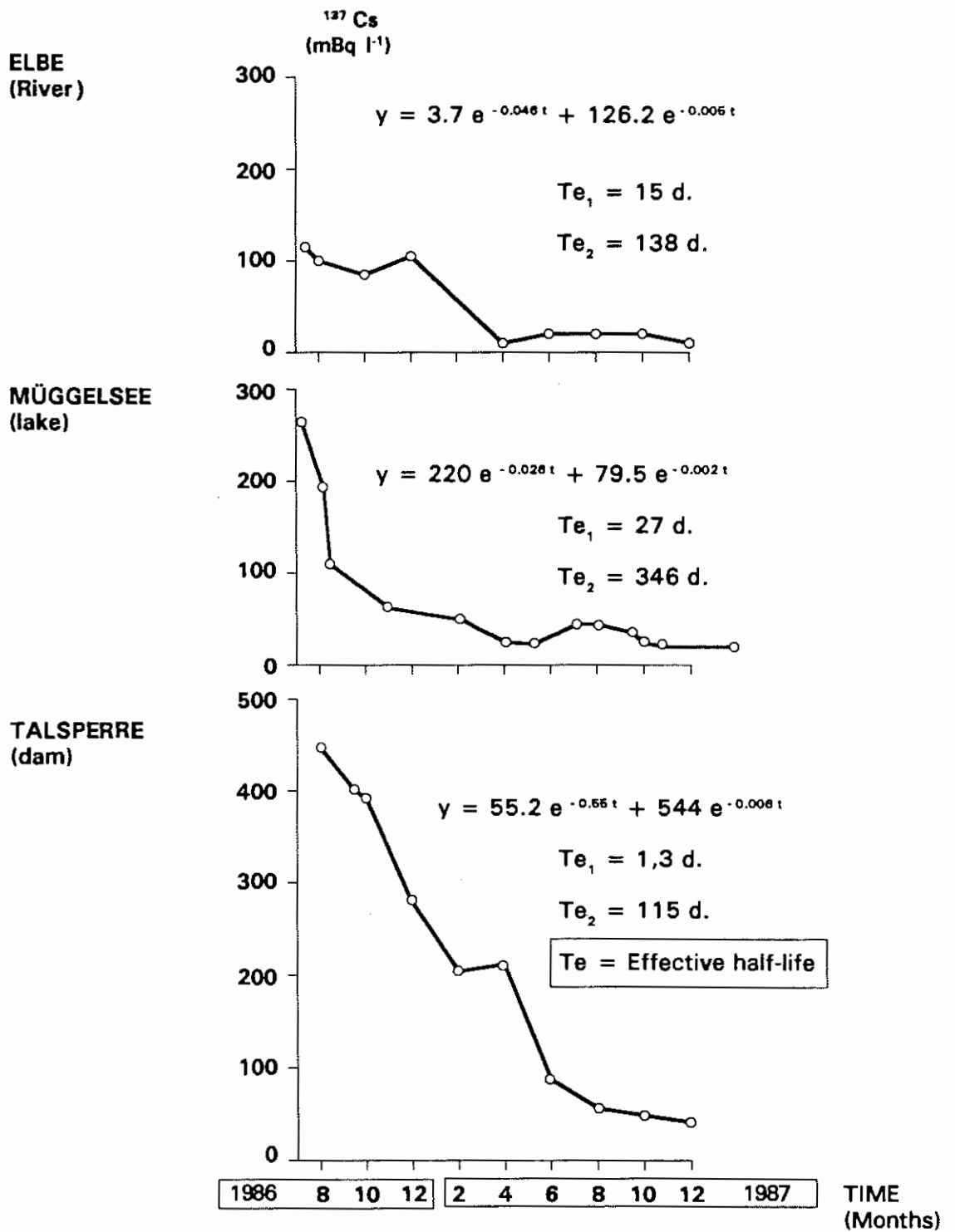


Fig. 2 - Evolution of <sup>137</sup>Cs concentration in water of 3 different ecosystems in East Germany [4]

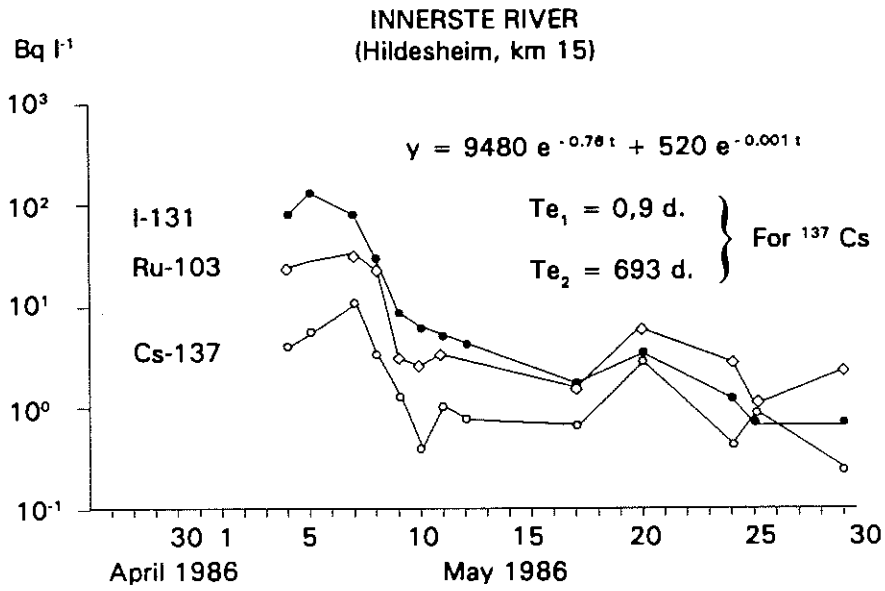


Fig. 3 - Evolution of I-131, Ru-103 and Cs-137 concentration in INNERSTE River [15]

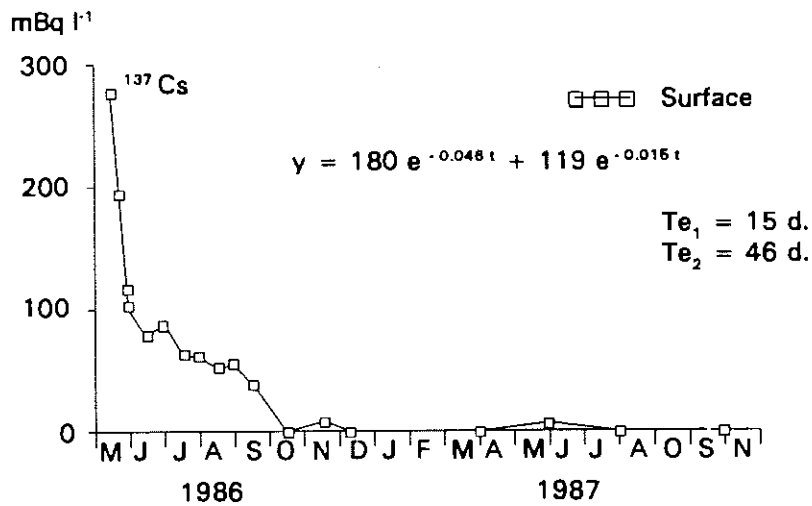


Fig. 4 - Evolution of Cs-137 concentration in water from Esthwaite lake [14]

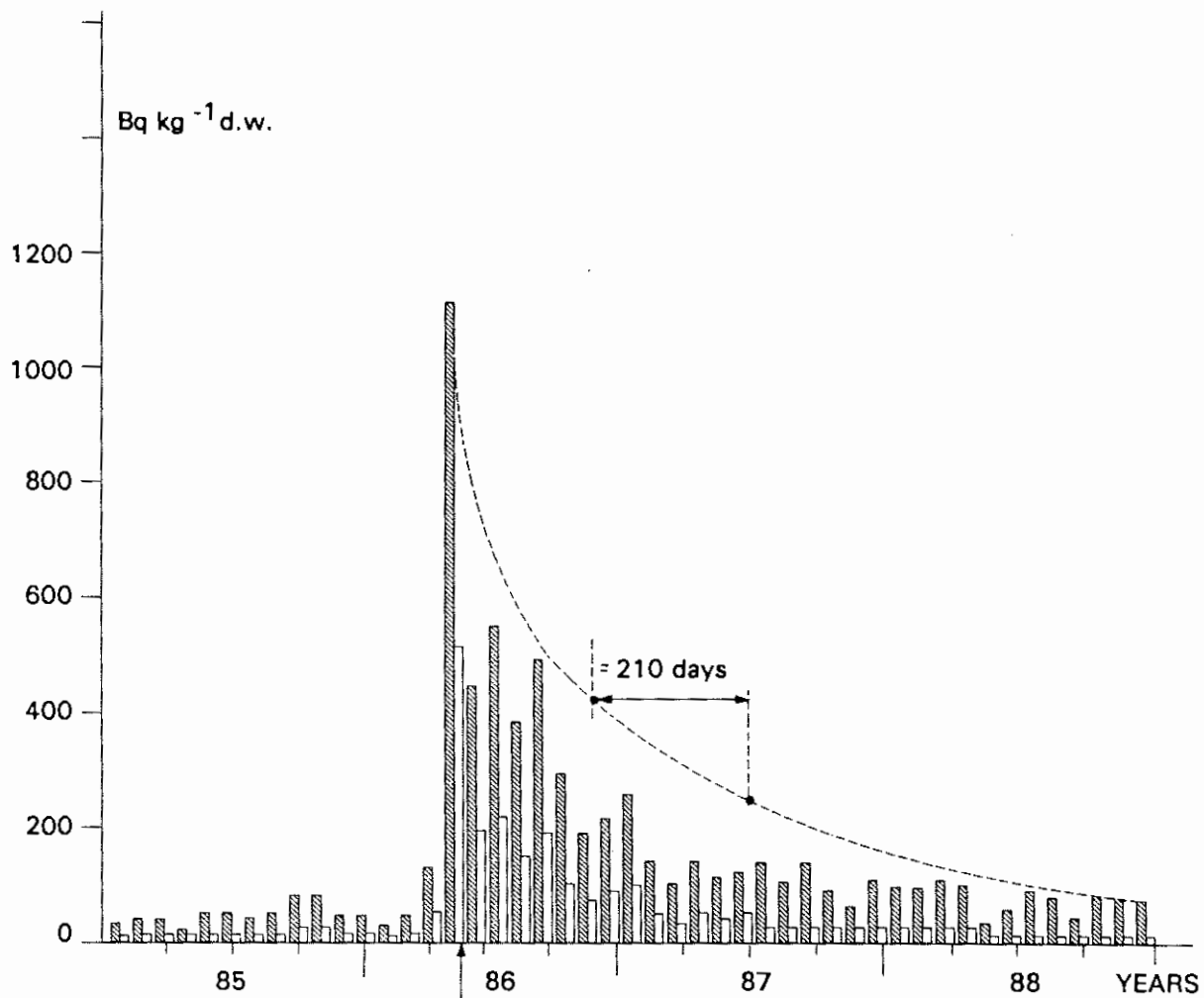


Fig 5 : Evolution of <sup>137</sup>Cs (hatched bars) and <sup>134</sup>Cs (white bars) concentration in suspended matter in Meuse river at TAILFER [22]

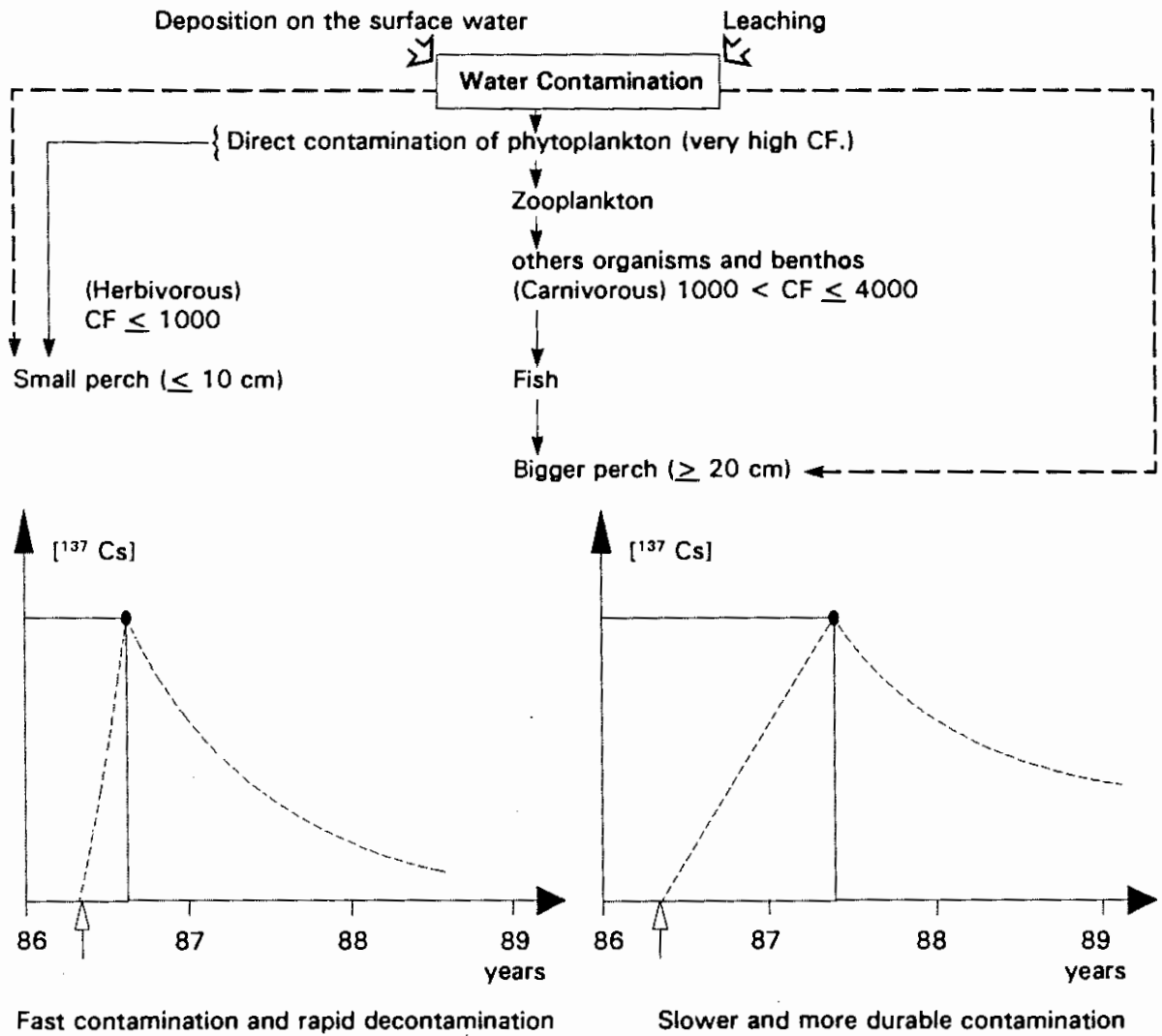


Fig. 6 - Scheme of <sup>137</sup> Cs transfer to lake fish from fallout (contamination arise in spring with phytoplanktonic bloom)

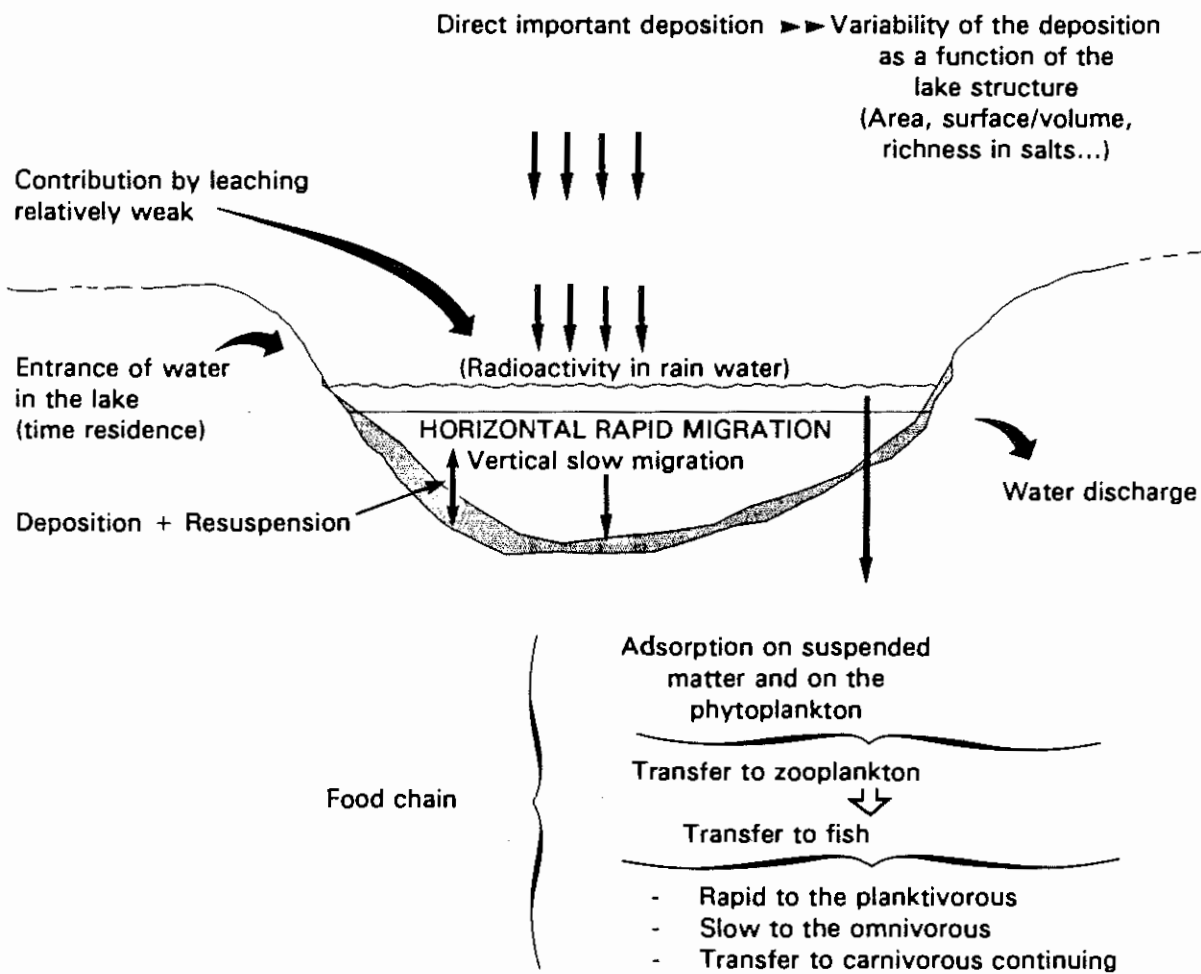


Fig. 7 - Main parameters necessary for studies on the radionuclide transfers in lakes.



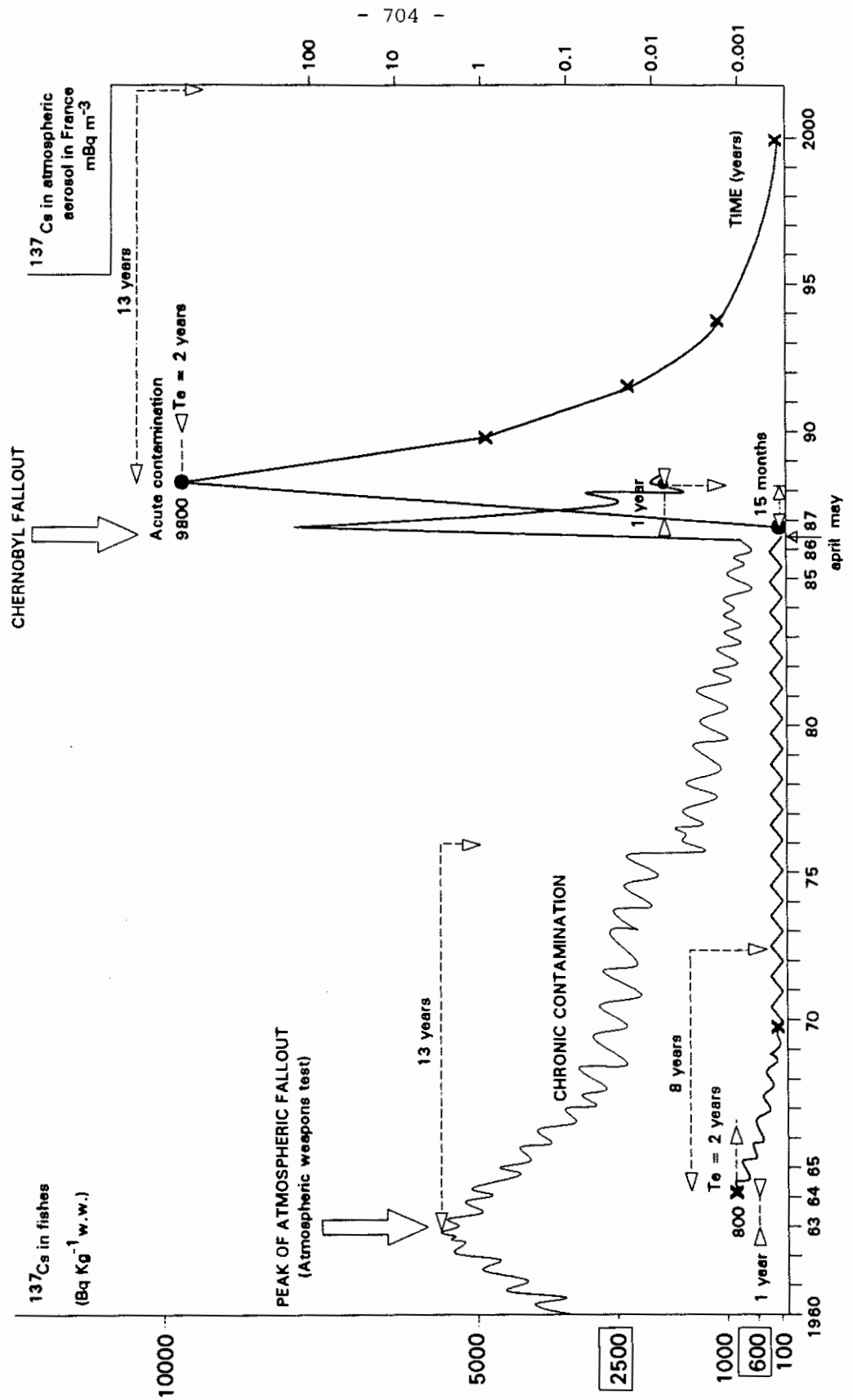


Fig. 8 : Accumulation and depuration of Cs-137 by lakes fishes (example for predators : perch in scandinavia lakes)

# **Cs-137 Releases from Sellafield and Chernobyl: Lake Sediment Based Evidence from West Cumbria, UK**

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Abstract

A study has been made of the  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  content of sediments from Ponsonby Tarn, a shallow nutrient-rich lake situated 1.5 km east of Sellafield in Cumbria. Previous work in the area has estimated the deposition of  $^{137}\text{Cs}$  resulting from the 1957 fire at Windscale (now Sellafield) nuclear weapons tests and the Chernobyl accident in the area. Sediment cores obtained from the tarn in 1988 were dated and cross-correlated using  $^{210}\text{Pb}$  and compared with the results obtained from previous studies in 1980 (Eakins and Cambray, 1985) and 1986 (Bonnnett and Cambray, in press).

Sediment from the tarn displayed marked changes between 1980 and 1988. Chernobyl-derived  $^{134}\text{Cs}$  inventories increased four-fold between 1986 and 1988 whilst the inventory of weapons test and Sellafield derived  $^{137}\text{Cs}$  showed only modest increases.

Radionuclide and palaeoecological (diatom and pollen analytical) data suggest that complex sediment accumulation patterns, hydraulic flushing and sediment focussing may account for some of the features evident in the tarn.

## Introduction

Lake watershed ecosystems and in particular the lake sediment record constitute a convenient geographical unit for examining the localised impact of anthropogenic pollutant deposition and transfer. The surface of a lake and its surrounding catchment area is a system subject to the flux of both natural and artificial radionuclides. In suitable circumstances lake sediments provide an integrated record of direct atmospheric input to a given area (the lake surface) and indirect input via weathering and subsequent transport.

Ponsonby Tarn, a small nutrient rich lake in West Cumbria has been extensively studied due to its proximity to the Sellafield Nuclear Establishment (Eakins and Cambray 1985; Bonnett and Cambray, in press; Bonnett et al., 1990). This area of Cumbria also experienced some of the highest deposition of  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  due to the Chernobyl accident (Clark and Smith, 1988; Jackson, 1989).

The tarn therefore provides a useful opportunity to compare the impact from and significance of releases from Sellafield (formerly Windscale), Chernobyl and nuclear weapons tests.

## Study Area

Ponsonby Tarn (National Grid Reference NY046045) lies on the border of the Lake District National Park ~1.5 km east of Sellafield and ~3 km inland from the Irish Sea (fig.1). The tarn is an artificial lake constructed towards the end of the nineteenth century by the widening of Newmill Beck which rises on Ponsonby Fell ~15 km to the east.

The area is underlain by Permian and Triassic (New Red) sandstone. The tarn is underlain by Quaternary alluvium. On the higher part of the catchment there are peat deposits, and some Pleistocene fluvioglacial sands and gravels are exposed in the northernmost area of the catchment. (Institute of Geological Sciences, 1978). The tarn is surrounded by managed woodland, conifers and rhododendrons with slopes adjoining the tarn being clear felled between 1981 and 1986 (E Y Haworth, 1988 pers. comm.). Other catchment characteristics are given in Bonnett and Cambray (in press).

## Methods

### Sampling

The first sediment core (PT1) was obtained in 1980 at location 1 in Fig. 1 using a Gilson corer (Macan, 1970). Three further cores (PT862, PT863, PT861) were obtained in October 1986 from locations 2, 3 and 4 and one in December (PT882) at location 5 using a short pneumatically driven Mackereth corer (Mackereth, 1969) of diameter 6.3 cm. Sediments from PT862 and PT863 were extruded using a hydraulically driven piston (Mackereth, op cit.) and sectioned at intervals of one to two centimetres. Core PT861 was analysed as a bulk sample. Soil cores to a depth of 15 cm were taken from two separate locations (PS1 and PS2), near the tarn.

### Analysis

$^{137}\text{Cs}$  (half-life 30.0 years) and  $^{134}\text{Cs}$  (half-life 2.06 years) were determined in samples of dried and ground sediment by gamma ray spectrometry. The techniques of analysis used are described by Salmon et al., (1983) in greater detail.

$^{210}\text{Pb}$  and PT and PT863 was determined by dry distillation of its  $^{210}\text{Po}$  daughter using the method set down by Eakins and Morrison (1978).  $^{226}\text{Ra}$  in sediment was determined by a method similar to that described by Pennington et al., (1976) using  $^{133}\text{Ba}$  as a yield tracer.  $^{210}\text{Pb}$  analysis of PT882 was carried out by low background direct gamma assay (Appleby et al., 1986).

Percentage weight loss on ignition was calculated by heating pre-weighed sub-samples in a muffle furnace for two hours at 550°C.

Diatoms in the top 14 cm of core PT862 were analysed using the method described in Batterbee (1986). A Leitz SM-LUX microscope with 1250x magnification was used to identify 200 valves from each sample. Identification was aided by reference to Krammer and Lange-Bertalot (1986). Pollen analysis of PT882 was undertaken using the methods described in Faegri and Iversen (1975) and Moore and Webb (1978).

All pollen and spores were counted using a Nikon (L-Ke model) microscope at a magnification of x400, with x1000 oil immersion for critical

determinations. Counts were made in complete traverses at regular intervals across 22 x 22 mm coverslips. When it was not necessary to count whole slides traverses were spaced to include representative areas of the slide thus minimising problems due to the possible non-random distribution of pollen grains on slides.

The pollen diagram (Faegri and Iversen, 1975) was calculated on a pollen sum of dry land pollen. Pollen of obligate aquatic plants and all spores were excluded in accordance with palynological convention. In addition, taxa of doubtful dry land origin such as Gramineae where there is incomplete identification (only to family level) and those with a wide habitat tolerance such as Alnus, for which unusually high pollen counts were made for each sample, were excluded from the pollen sum. Percentages for pollen types not included in the basic pollen sum were calculated as a percentage of the basic pollen sum plus the sum of that particular type. For example, percentages for Gramineae were calculated as a percentage of the total pollen sum plus the sum of Gramineae pollen.

#### Results and Discussion

The short lived isotope  $^{134}\text{Cs}$  is derived almost entirely from Chernobyl fallout. Using the  $^{134}\text{Cs}/^{137}\text{Cs}$  ratio in Chernobyl fallout of  $\sim 0.6$  (Cambray et al., 1987),  $^{134}\text{Cs}$  activities have been used to partition the total  $^{137}\text{Cs}$  activity into a Chernobyl derived component and a component derived from sources other than Chernobyl.

In order to assess the environmental significance of events such as the Chernobyl and Windscale accidents it is essential to establish the estimated contribution from these sources. From the data quoted in Booker (1962) and Cambray et al. (1987) the notional  $^{137}\text{Cs}$  deposit at Ponsonby Tarn up to October 1986 (corrected for decay) is thought to stem from four major sources namely:-

i)	Sellafield discharges prior to 1957	1530 Bq m <sup>-2</sup>
ii)	Windscale fire of 1957	285 Bq m <sup>-2</sup>
iii)	Fallout derived from atmospheric weapons testing	4320 Bq m <sup>-2</sup>
iv)	Chernobyl-derived $^{137}\text{Cs}$	<u>- 9865 Bq m<sup>-2</sup></u>
	Total	-16000 Bq m <sup>-2</sup>

It is apparent that the  $^{137}\text{Cs}$  content of the sediment in 1986 is dominated by the Chernobyl release with discharges from Sellafield (both routine and accidental) contributing only ca. 12% of the total sediment inventory.

The total deposition of  $^{137}\text{Cs}$  directly onto the surface of the tarn by 1986 is thus estimated at  $\sim 16\,000\text{ Bq m}^{-2}$ . At the time of the previous study of the tarn in 1980 (Eakins and Cambray, 1985) the total deposit was  $\sim 7100\text{ Bq m}^{-2}$  of  $^{137}\text{Cs}$  whilst in 1988 total deposition is estimated at  $\sim 14\,680\text{ Bq m}^{-2}$ .

These figures are in reasonable agreement with other estimates of local deposition based upon soil sampling in the catchment in 1986 (mean value  $17\,400\text{ Bq m}^{-2}$ ) and nearby sampling under deciduous woodland by Hursthouse (pers. comm. 1989).

However, examination of the radionuclide content of the 1980, 1986 and 1988 cores reveals considerable variation from the expected baseline inputs. Radionuclide inventories also vary temporally (Table 1). In core PT1 the total  $^{137}\text{Cs}$  inventory is in close agreement with that expected from weapons fallout and Sellafield discharges. In core PT862 the inventory from these sources is only  $\sim 80\%$  of the expected value. Core PT863 has an inventory from weapons fallout and Sellafield discharges twice the expected value. In contrast, Chernobyl derived  $^{134}\text{Cs}$  inventories are markedly below expected values in cores PT862 and PT863 but  $^{134}\text{Cs}$  values in PT882 are approximately *two* times greater than estimated.

These anomalies may be due to a number of factors:

- 1) Marked changes in the atmospheric flux of radionuclides to the catchment.
- 2) Catchment inputs to the tarn.
- 3) Hydraulic flushing.
- 4) Spatial patchiness of sediment deposition and or sediment focussing.

The variation evident cannot be attributed to changes in atmospheric deposition. During the period 1980-1988 fallout of  $^{137}\text{Cs}$  was negligible. Natural radioactive decay will have reduced the  $^{137}\text{Cs}$  inventory by  $\sim 20\%$ .

Inputs of radionuclides from the catchment in particle associated form possibly resulting from ground disturbance due to tree planting and harvesting might account for the elevated weapons test and Sellafield derived inventories in the 1986 sediment record. However, as can be seen in Fig. 2, the pollen record contained in the tarn appears to describe only minor changes in the surrounding vegetation over the last 150 years. Whilst documented occurrences of afforestation are reflected in slight increases in the pollen frequencies of the taxa involved (eg the increase in Quercus from ~25-30 cm relating to the 1911 planting of oak in the catchment), there are no changes in the pollen record which could account for the large increase in radionuclide inventories evident in core PT882.

A summary total diatom concentration curve calculated on a count of 400 valves is shown in Fig. 3. This shows no evidence of the marked fluctuations in diatom concentrations commonly associated with the episodic inwash of catchment material resulting from catchment disturbance (Battarbee and Flower, 1984) which might, in part, account for the radionuclide inventories seen in the 1986 cores (Kreiser, pers. comm., 1988).

Palaeoecological data in the form of diatom and pollen analysis does not therefore, appear to indicate any significant input of catchment derived material to the tarn. Similarly the stable loss on ignition values of cores PT862, PT882 and PT883 (Fig. 4) indicates that the physical characteristics of the sediments are homogeneous down core and suggests the lack of episodic inwashes of either organic or minerogenic material from the catchment. The balance of evidence would appear to suggest that both the changing radionuclide inventories evident in the 1986 and 1988 cores, together with the marked increase in sediment accumulation rates in 1988 are the result of processes other than catchment derived inputs of material.

Whilst the mean residence time of water within Ponsonby Tarn is not known it is conceivable, given the size and depth of the tarn, that it may be quite short. Hydraulic flushing during high flow conditions would reduce this further and may account for the lower than expected  $^{134}\text{Cs}$  inventories if this process occurred between initial deposition of Chernobyl radionuclides (May, 1986) and sampling of PT862, PT853 (October, 1986). Hilton et al., (1989) estimate that hydraulic flushing of dissolved  $^{134}\text{Cs}$



and  $^{137}\text{Cs}$  resulted in 34-66% of the initial deposition onto the surface of Windermere and Esthwaite Water being transported out of the lake system before it became adsorbed onto the lake sediment. These figures are similar to those quoted by Santschi et al., (in press) for Lake Zurich (<50%) and Lake Constance (<10%) due to such flushing. Some of the Chernobyl derived radiocaesium may, therefore, have been transported out of the system before scavenging by settling particles occurred in 1986.

However, such a flushing process cannot explain the large increase in Chernobyl radiocaesium evident in core PT882. This feature, together with differences in the weapons fallout and Sellafield derived  $^{137}\text{Cs}$  inventories apparent between PT1, PT861, PT862 and PT863 may reflect the complex sedimentation patterns noted in other small lakes, (eg Davis, 1976; Dearing, 1983). Such patterns may, in part, be due to the natural variability in deposition of sediments in lacustrine environments. Downing and Rath (1988) examined the spatial patchiness of sediment characteristics in central, bathymetrically uniform sites in eight lakes and concluded that "sediments are spatially heterogeneous in ways that are unrelated to depth gradients, sediment focusing or other large scale redistribution processes" (p.454).

Heterogeneity may therefore partly account for the variable radionuclide inventories. The complex patterns of sedimentation may also be due to sediment focusing (Lehman, 1975; Hilton, 1985). Such focusing could account for the variable inventories present in the 1980, 1986 and 1988 cores with differences between the cores reflecting differences in the intensity of sediment focusing and redistribution at each site. A variable sediment flux at the different sample sites is also suggested by the differing sediment accumulation rates calculated from different cores (Table 2) and the rapid increase in accumulation rates in core PT882 in recent years.

### Conclusions

The significance of Chernobyl-derived and weapons test/Sellafield derived radiocaesium vary markedly in the sediments of Ponsonby Tarn over time. The radionuclide inventories of the tarn in 1980, 1986 and 1988 may be regarded as being a product of the natural variability of sediment deposited over the tarn basin together with hydraulic flushing and

spatially and temporally variable sediment focusing. Whilst it is tempting to ascribe the increased Chernobyl component in core PT882 to catchment inputs resulting from tree harvesting evident within the catchment, neither the pollen nor the diatom record contained within the sediment shows any evidence of this.

#### Acknowledgements

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Table 1  
Radionuclide inventories in the sediment cores

Core	$^{137}\text{Cs}$ (Bq/m <sup>2</sup> )		$^{134}\text{Cs}$ (Bq/m <sup>2</sup> )	
	Total	Pre-Chernobyl	Chernobyl	Chernobyl
PT1	6100	6100		
PT861	21000	14500	6500	4600
PT862	>9700	>5000	4700	2800
PT863	16800	13000	3800	2300
PT882	26850	12050	14800	8880
Estimated Deposition				
1980	7100	7100		
1986	-15800	6130	9700	5800
1988	15250	6050	9200	5500

">" Figures signify base of core not reached  
Results corrected for decay to 3 May 1986

Table 2  
Sediment accumulation rates in Ponsonby Tarn  
(Derived from  $^{210}\text{Pb}$  dating)

1980 Study

0.60 cm/yr or 0.082 g/cm<sup>2</sup>/yr

1986 Study

1.11 cm/yr or 0.20 g/cm<sup>2</sup>/yr

1988 Study

Year	g/cm <sup>2</sup> /yr	cm/yr
1988	0.40	2.9
1987	0.27	1.7
1986	0.27	1.5
1985	0.26	1.15
1981	0.25	1.1
1976	0.19	0.8
1972	0.24	0.9
1965	0.10	0.4
1957	0.09	0.3
1937	0.09	0.25
1912	0.06	0.15
1866	0.04	0.08

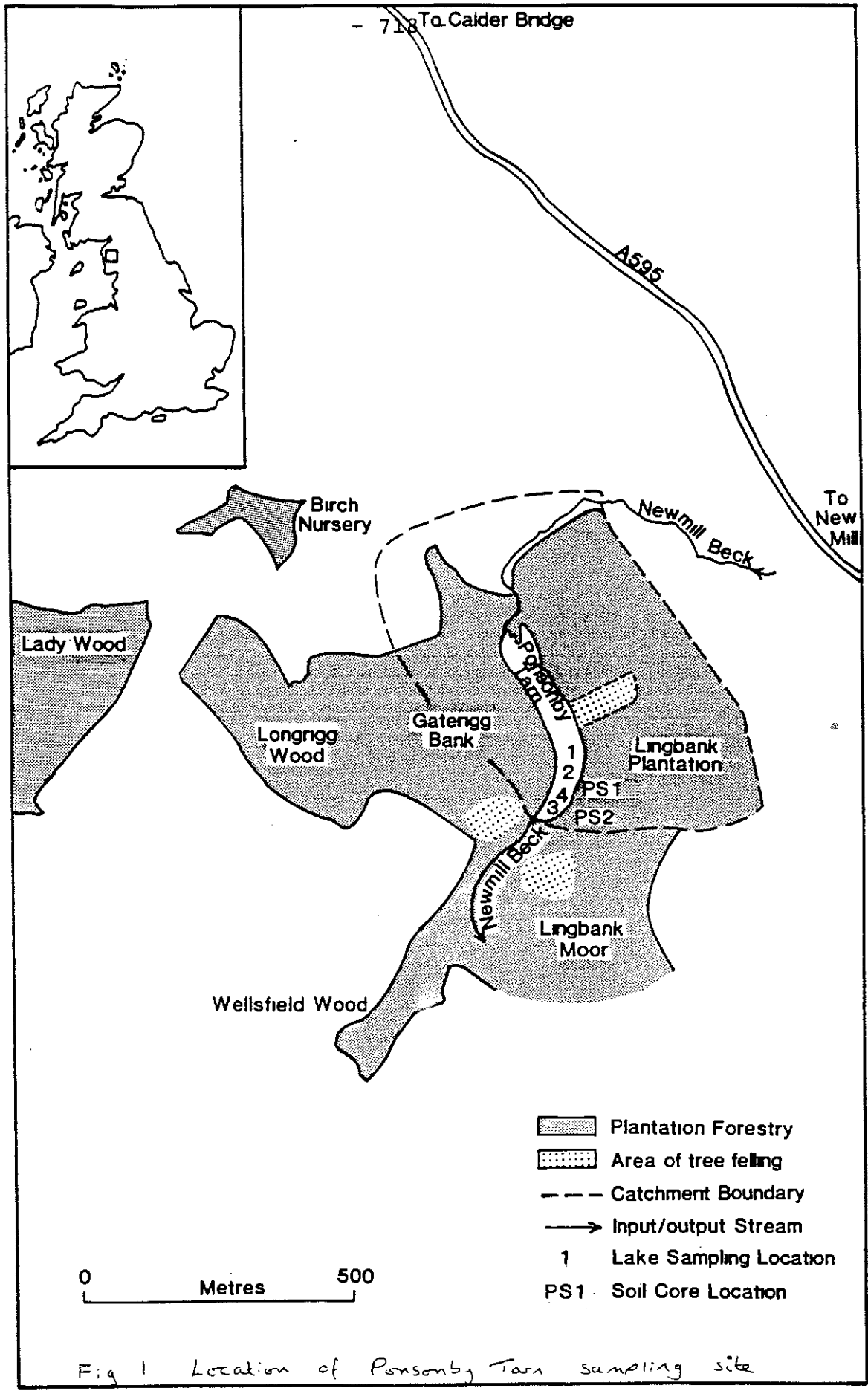


Fig 1 Location of Ponsoby Tarn sampling site

PONSONBY TARN, CUMBRIA (PT88/2)

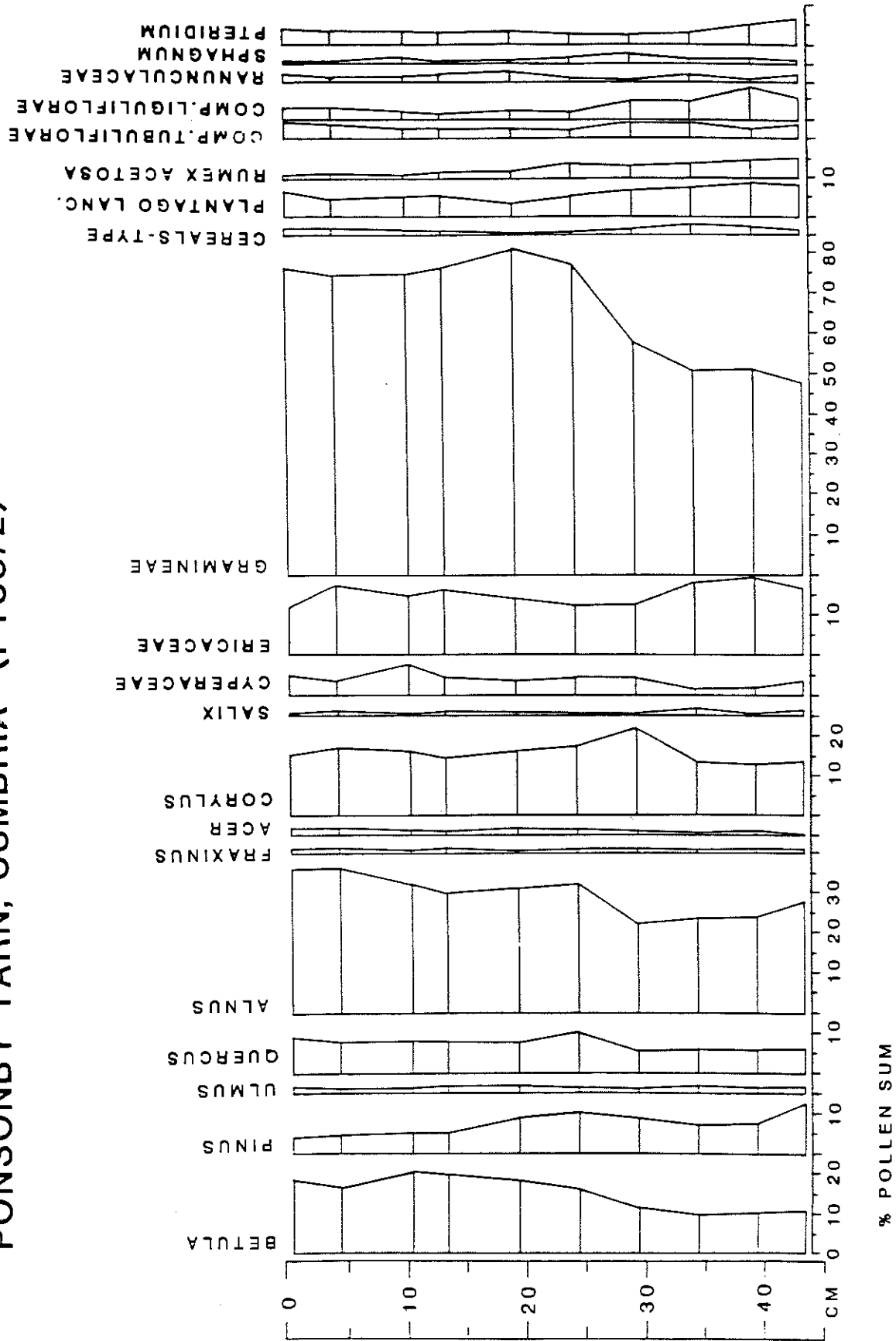
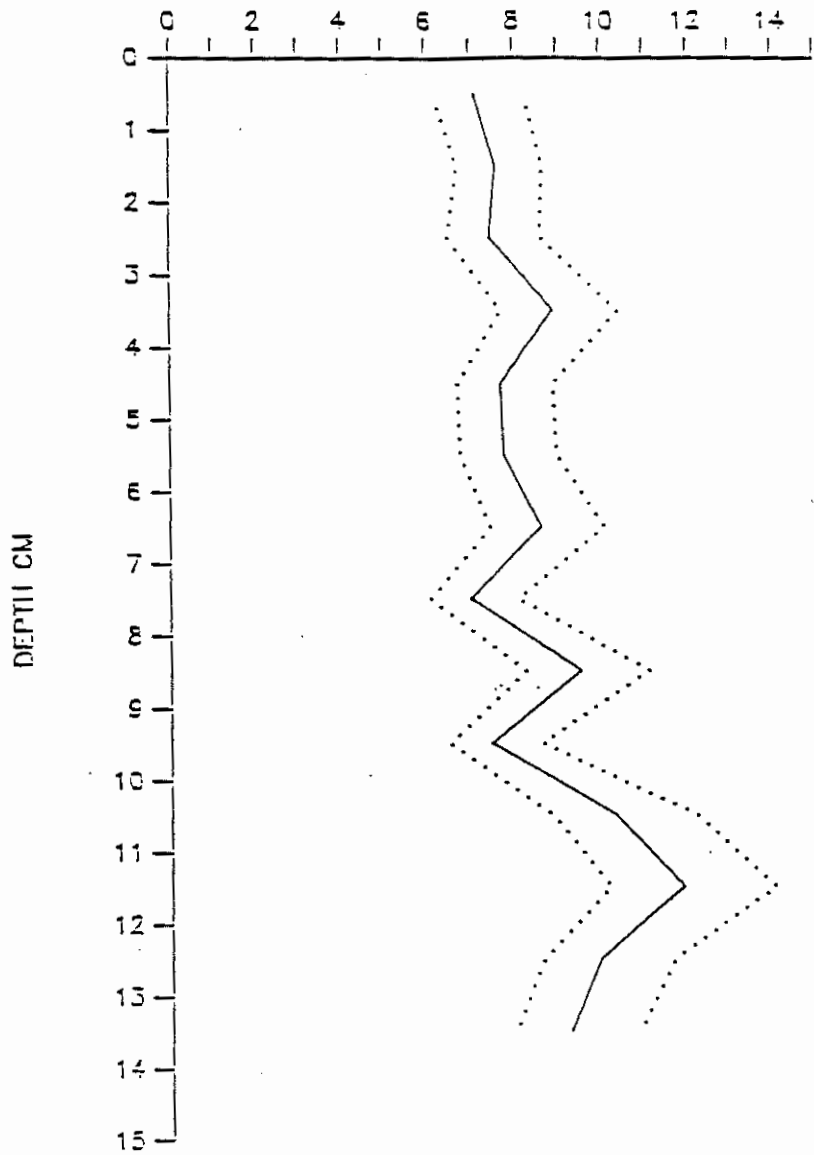


Figure 2. Relative pollen diagram for Ponsonby Tarn core PT88/2. Pollen frequencies are expressed as a percentage of a dry land pollen sum.





..... Upper and lower 95% confidence limits

Fig. 3 Total diatom concentration x 10<sup>7</sup> g<sup>-1</sup> weight (F7862)

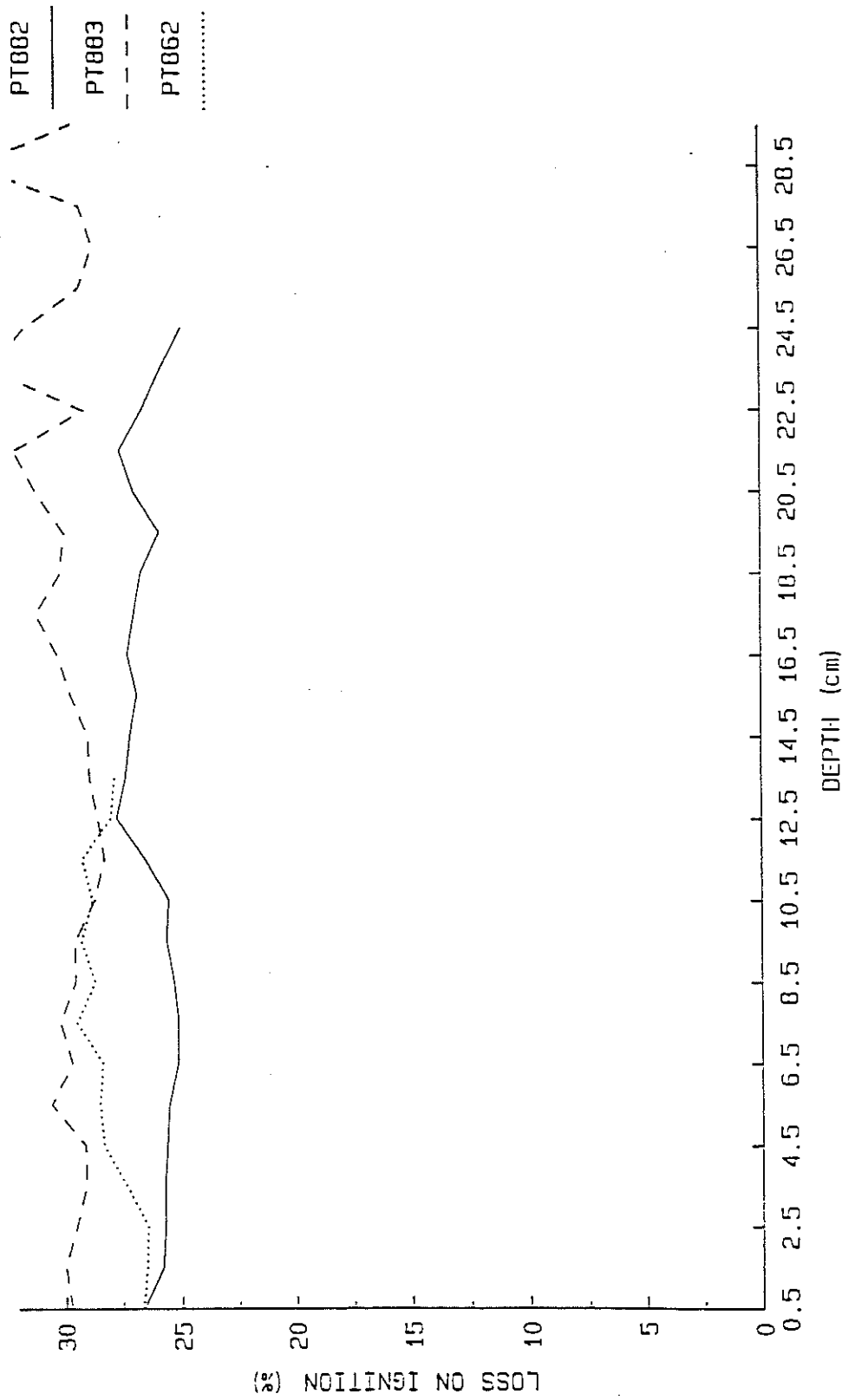


Fig. 4 Loss on Ignition values (PT862, PT882 and PT883)



Session VI

**RADIOLOGICAL IMPLICATIONS  
FOR MAN AND HIS  
ENVIRONMENT**



**Mathematical Modelling of  
Radionuclide Dispersion in  
Surface Waters after the  
Chernobyl Accident to  
Evaluate the Effectiveness of  
Water Protection Measures**

**M.I. ZHELEZNYAK, O.V. VOYTSEKHOVICH**

This report presents the results (obtained by the "V. M. Glushkov" Institute of Cybernetics of the Academy of Sciences of the Ukrainian SSR and the Ukrainian National Hydrometeorological Research Institute of the Soviet State Committee for Hydrometeorology) in assessing the effectiveness of water protection counter-measures in the Chernobyl NPP 30-km zone, using mathematical modelling methods to simulate radionuclide migration in water and drawing on field data and laboratory research.

#### 1. Water protection measures in the Chernobyl NPP zone

Immediately after the accident, work started on devising measures to reduce, and if possible prevent, radioactive fallout products from being washed out into surface and ground waters. Dozens of kilometres of water-protection embankments and earthen dykes were constructed to stop pluvial runoff from contaminated areas entering the rivers around Chernobyl. In summer and autumn 1986 dozens of regulating dams, incorporating filtering sorptive materials, were built on streams and small rivers in the zone, while areas of highly radioactive silt on the bed of Pripyat River channels and bends were covered over. Several traps in the form of pits were dredged in the Pripyat River channel to facilitate sedimentation of radioactive load (suspended and bed load) out of the river flow. Most of this work was done during a very difficult period, when sufficiently reliable data on the main sources of river contamination were not available. Neither did we have a detailed picture of the mechanisms and processes involved in the migration of the various physico-chemical types of radioactive fallout in the catchment areas and water bodies in the radioactively contaminated territories of the combined Pripyat-Dnieper river basin. What is more, the amount of work involved was enormous. Therefore, from as early as the summer of 1986 we started evaluating the measures' effectiveness.<sup>1-4</sup> This included experimental observations on rivers to evaluate the actual impact of such structures on cleansing the waters and reducing the radionuclide migration rate, plus work to optimize the effect of the various water protection measures on the basis of mathematical models simulating radionuclide migration processes in aquatic systems after implementation of the proposed water protection measures.

Experimental and theoretical evaluation of the impact of the water protection measures in river channels, and an analysis of their effectiveness, indicated <sup>1-5</sup> that given the vast scale of the radioactive contamination on the flood-prone territories, local counter-measures had hardly any impact. For instance, assessment of the overall impact of all the measures taken from 1986 to 1989 on small rivers in the Chernobyl NPP 30-km zone and in the Pripyat River to restrict the removal of radioactive caesium, showed that such measures

held back less than 10% of the total amount entering Kiev Reservoir with the river flow. The impact was even less marked for  $^{90}\text{Sr}$ . Thus, the water protection structures - not counting the river channel traps for radioactive load - held back no more than a few percent of the water runoff from the contaminated areas, given that in the past few years up to 70% of the  $^{137}\text{Cs}$  flow (but only 30-40% of the  $^{90}\text{Sr}$  flow) has come from outside the 30-km zone. It was shown experimentally that the only way to significantly change radiostrontium migration conditions in the flood-prone areas of the contaminated zone was either (I) to isolate the landscape source of radiostrontium migration, or (II) slow down the process by which its non-exchangeable forms convert to exchangeable and water-soluble forms. The most effective natural cleansing process for  $^{137}\text{Cs}$ , which binds strongly to soil and load particles, is its co-precipitation with load in sedimentation zones. Therefore, given the different hydrological and geochemical migration conditions, water protection engineering needs to be based on rapid assessment of a contaminated water body's reaction to the introduction of a given technical solution. This report provides examples of such an approach in evaluating the effectiveness of, and optimising, measures taken in river channels (traps dredged in the channel to facilitate co-deposition of radionuclides out of the flow and onto the bed together with load) and on the floodplain landscape (isolating and covering the floodplain area).

## 2. ASSESSMENT OF THE EFFECTIVENESS OF MEASURES TO PREVENT CONTAMINATION BEING WASHED OUT FROM THE PRIPYAT RIVER FLOODPLAIN

Because of the high levels of radioactive contamination found on the Pripjat River floodplain in the area near the Chernobyl NPP (from the village of Benevka to Yanov Station railway bridge), we had to analyse in detail the different degrees of radionuclide washout which might occur during inundation of the floodplain by flood events of various volumes. Experimental research in this area yielded information on the amounts and forms of local radioactive contamination. Analysis of different scenarios relating to the river's hydrological regime in this area indicated there was a real danger of the highly mobile radionuclide  $^{90}\text{Sr}$  being washed out of the floodplain soils into the river waters. The consequences of radionuclide washout from the area were predicted (for various flood-volume scenarios) by mathematically modelling the radionuclide migration processes in the soil-water system for typical water dynamics over the floodplain surface. The initial modelling stages involved a) calculating the flow pattern of river waters in the channel and on the floodplain, and the characteristics of mass exchange between water flow and exchangeable-phase radionuclides in the contact layer, and b) calculating the dispersion of radioactive substances entering the main flow of the river along its whole length to the river mouth, and further on down through the Dnieper Reservoir Cascade.



2.1 Basic equations used for modelling. Over the past decade the main trend in development of mathematical models to simulate the dynamics of radionuclides in water bodies has been a switch from simple dimensionless compartmental models in favour of large models processed by computer and based on finite differences and finite elements.<sup>6-8</sup> A series of models of various dimensions was devised at the "V. M. Glushkov" Institute of Cybernetics of the Ukrainian Academy of Sciences 1,3,9 for use in connection with water contamination problems following the Chernobyl accident. A two-dimensional model, based on flow parameters averaged over depth and on contamination characteristics, was used to study the dynamics of contamination on the inundated floodplain. The model of the task in hand included the main hydrodynamic and hydrophysical factors determining the intensity of radionuclide migration, i.e. flow pattern, transport of suspended load, advective-diffusive movement of the soluble and suspension-sorbable forms of contamination, the adsorptive-desorptive exchange of soluble forms with suspensions and bed load, and redistribution of contamination in the water-bed system resulting from bed erosion and settling of suspensions.

The distribution of the depth-averaged components of the flow velocities on the floodplain -  $u_1$  and  $u_2$  - is represented by Saint Venant's equations

$$\frac{\partial u_j}{\partial t} + u_k \frac{\partial u_j}{\partial x_k} + g \frac{\partial \eta}{\partial x_k} = -\frac{g}{c^2 n} u_j |\vec{u}| \quad (1)$$

$$\frac{\partial \eta}{\partial t} + \frac{\partial (h u_k)}{\partial x_k} = 0 \quad (2)$$

where  $k = 1, 2$ ,  $j = 1, 2$ ,  $\eta$  = free surface elevation above the horizontal level of the smooth water surface,  $H$  = depth at this water level,  $h = H +$  = total depth, and  $c$  = Chezy's coefficient. Here and later we employ summation with regard to the recurrent subscripts. To calculate Chezy's coefficient we use Manning's formula,  $c = h^{1/6}/n$ , where  $n$  = floodplain roughness coefficient.

In calculating steady flows, the stationary form of the continuity equation (2) means we can introduce the stream function  $\Phi$

$$\frac{\partial \Phi}{\partial x} = -V \quad \frac{\partial \Phi}{\partial y} = U \quad (3)$$

where  $x = x_1$ ,  $y = x_2$ ,  $U = hu_1$ , and  $V = hu_2$ .

Equation (2) is then dealt with in the same way. As evaluations of the order of the individual terms in the displacement equations show (1),(2), the convective terms in these equations may be omitted if the condition  $L \gg h/\lambda$  is met, where  $L$  = characteristic scale of change in the datums of the free surface, and  $\lambda = g/c_b h$  = bed friction coefficient.

The above condition is met in most instances for the section of the floodplain under study. If one omits the convective terms, cross differentiation of equations (1) and (2) with respect to  $y$  and  $x$  and subtraction of the former from the latter equate to zero.

$$\frac{\partial}{\partial x} \left( k \frac{\partial \Phi}{\partial x} \right) + \frac{\partial}{\partial y} \left( k \frac{\partial \Phi}{\partial y} \right) = 0 \quad (4)$$

where  $k = |\vec{u}| c^{-2} h^{-3}$

The boundary conditions at solid boundaries are

$$Q = \Phi_x - \Phi_r \quad (5)$$

where  $Q$  = volume flow rate of the particular flood calculated, and  $\Phi_x, \Phi_r$  = stream function values on the left- and right-hand boundaries of the flood flow.

At liquid boundaries

$$\frac{\partial \Phi_{B/H}}{\partial N} = 0 \quad (6)$$

where  $N$  = normal to the boundary, subscript  $B$  is the flow's upper and subscript  $H$  its lower boundary.

Changes in the mean concentration by depth of the suspensions in the flow  $S$  are expressed by the equation for advective-diffusive movement:

$$\frac{\partial (hS)}{\partial t} + \frac{\partial}{\partial x_k} (hS u_k) = \frac{\partial}{\partial x_k} \left( h E_{1k} \frac{\partial S}{\partial x_k} \right) + B W_0 (S_* - S) \quad (7)$$

Here,  $B$  = the relation between the benthic concentration of the suspension and the mean concentration by depth assumed to be similar for all cross-sections of the flow,  $w_0$  = the particles' settling velocity,  $S_*$  = the equilibrium concentration of the suspension in the flow (transporting capacity) determined in the present model by Bijker's formula<sup>10</sup>,  $E_{1k}$  = the coefficient of horizontal dispersion related to the coefficients of longitudinal  $D_e$  and transversal  $D_p$  dispersion by the correlations

$$E_{11} = D_e \cos^2 \psi + D_p \sin^2 \psi, \quad E_{22} = D_e \sin^2 \psi + D_p \cos^2 \psi \quad (8)$$

$$E_{12} = (D_e - D_p) \cos \psi \sin \psi$$

The coefficients  $D_e$  and  $D_p$  are precisely determined subject to a constant across depth  $h$ , and the modulus of dynamic velocity  $V_*$  by Elder's formulae <sup>11</sup>:  $D_e = e_1 V_* h$ ,  $D_p = e_2 V_* h$ , where  $e_1 = 5.93$  and  $e_2 = 0.23$ , the last parameter being determined with significant scatter.

The change in the thickness of the bed sediment layer ( $Z_*$ ) is expressed by the equation for bed deformations

$$(1 - \epsilon) \frac{\partial Z_*}{\partial t} = q^s - q^b \quad (9)$$

Here, the vertical flows of the suspension are determined by the correlations

$$q^s = \begin{cases} BW_0(S - S_*) & \text{where } S > S_* \\ 0 & \text{where } S < S_* \end{cases}; \quad q^b = \begin{cases} BW_0(S - S_*) & \text{where } S < S_* \\ 0 & \text{where } S > S_* \end{cases} \quad (10)$$

The equation for the mean concentrations by depth of soluble radionuclide forms  $C$ , taking into account the main hydrodynamic and hydrophysical processes described above, is given by

$$\frac{\partial (hC)}{\partial t} + \frac{\partial}{\partial x_k} (hC u_k) = \frac{\partial}{\partial x_k} h E_{1k} \frac{\partial C}{\partial x_1} - \lambda h C - a_{1,2} S (K_d C - C^s) + a_{1,3} (K_d C - C^b) \quad (11)$$

where  $C^s$  = mean concentration by depth of radionuclides on suspended load,  $C^b$  = concentration of radionuclides in bed sediment averaged over thickness  $Z_*$  of the active layer,  $K_d$  = coefficient of equilibrium distribution of radionuclides in the water-load system,  $a_{1,2}$  and  $a_{1,3}$  = coefficients of the velocity of the exchange between (I) solution and suspension and (II) solution and bed sediment respectively, and  $\lambda$  = decay constant. The corresponding term in equation (11) is omitted when calculating movement of long-lived radionuclides.

The mean concentration by depth of radionuclides on suspensions is given by the equation

$$\frac{\partial}{\partial t} (hC^s) + \frac{\partial}{\partial x_k} (u_k h C^s) = \frac{\partial}{\partial x_k} h E_{1k} \frac{\partial}{\partial x_1} (C^s) - \lambda h C^s + a_{1,2} S (K_d C - C^s) + C^b q^b - C^s q^s \quad (12)$$

Contamination of the bed sediment layer  $Z_*$  is determined by the equation:

$$(1-\varepsilon) \frac{\partial}{\partial t} (Z_* c^b) = - a_{1,3} (K_d c - c^b) - c^b q^b + c^s q^s \quad (13)$$

The model's radiological block includes four constants -  $K_d$ ,  $a_{1,2}$ ,  $a_{1,3}$ ,  $\lambda$ , determined from experimental data.

For numerical integration of equation (4) we used the finite elements method based on the orthogonalization of the approximation error according to Galerkin's method.<sup>12</sup>

The method used for numerical solution of the equations for advective-diffusion of suspended sediment transport (8) and radionuclides transport (15),(16) had to be designed so that the numerical diffusion conditioned by approximation of advective transport was considerably lower than physical diffusion. This condition can be met by using finite difference schemas of high order of accuracy to describe the advective transport processes. The finite difference approximations of equations (7),(11),(12) are based on splitting by physical processes.<sup>13</sup> For the advective step we use an explicit finite-difference scheme with fourth order of accuracy close to that proposed in <sup>14</sup>, while for the diffusive step we use an explicit schema of second order of accuracy. In the third and final step of splitting the ordinary differential equation is solved for sources terms.

The site covered by our calculations (10 250 m x 3 750 m) is located in the railway bridge backwater zone. The left-hand boundary of the site passes along the dyke protecting the polder network. The right-hand boundary follows a line passing through the built-up area of the town of Pripyat and the settlements of Novo-Shepelichi and Benevka. The inflow boundary runs perpendicular to the dyke 10 km from the axis of the railway bridge, in whose vicinity the outflow boundary of the site under study is located.

We calculated the velocity fields and distribution of contamination on the floodplain for maximum discharges of extremely high floods, i.e. with probabilities of 0.01, 0.1 and 0.25 (6 800, 3 400 and 2 100 m<sup>3</sup>/sec respectively). The roughness coefficient  $n$  of the channel was taken to be 0.013, and that for the floodplain 0.02.

Calculations of the flow velocity pattern showed that, for a probability of exceeding the flood equal to 0.01, flow velocities on most of the floodplain lie in the 0.4-0.8 m/sec range, with the bulk of the total discharge - some 5 000 m<sup>3</sup>/sec - occurring on the right-bank floodplain and in the main channel of the Pripyat River.

For a 0.1 probability flood the section of the floodplain covered by our calculations is also completely inundated by water, with a peak water level 1 m lower than the peak of the 0.01 probability flood. In this case, the pattern of the flow field is considerably more changeable. Compared to a 0.01 probability flood, velocities of up to 0.4 m/sec occur on a larger portion of the floodplain area.

Fig. 1a shows the results of calculating the velocity pattern of the flow on the inundated floodplain for a maximum flood discharge with a 0.25 probability, shown in the form of vector fields of specific discharges  $\vec{U}$ . The left-hand corner of the Fig. gives the scale of a specific discharge, corresponding to  $|\vec{U}| = 5 \text{ m}_2/\text{sec}$ . The  $h = 3.5 \text{ m}$  depth isoline delineates the main elements of the channel network at the site studied. The water level at the inflow station was calculated using backwater curves based on analysis of data accumulated over several years of observations.

Parallel to calculating the fields of flow on the floodplain under natural conditions, we also simulated the hydrodynamic regime on the inundated floodplain after introducing water protection measures. Fig. 1b shows the distribution of the specific discharges for a 0.25 probability flood after construction of a dyke preventing flooding of the left-bank floodplain, which is the worst contaminated. A significant increase is noted in the flow velocities on the right-bank floodplain and in the Pripyat Creek region.

The calculated velocity values were used in computer simulations of removal of radionuclides from the Krasnensky floodplain during high water. The first series of calculations evaluated the impact of only the sorptive exchanges in the water-bed system, neglecting the processes connected with rolling and settling of contaminated load. This representation of the process is admissible for evaluating the removal of  $^{90}\text{Sr}$ , most of which is carried along in dissolved form. Here the concentration can be calculated by equation (11), omitting the term relating to water-suspension interaction and ignoring decay.

To determine the  $C^D$  concentration of radionuclides in the top layer of the inundated soils on the floodplain and in the bed sediment, we used data on the level of radioactive contamination of the floodplain by cerium-144 and strontium-90 radionuclides. The total amount of  $^{90}\text{Sr}$  on the section of the left-bank floodplain studied is some 10 000 Ci, with a maximum contamination of over 1 000 Ci/km $^2$ . Because this zone is 3-15 km north of Unit IV of the Chernobyl NPP the specific composition of the contamination is that of fuel particles formed from uranium oxides. Since the accident we have observed an increase in the exchangeable forms of radionuclides due to transformation of the fuel-type fallout. According to data for 1990, as much as 50-60% of the  $^{90}\text{Sr}$  is in exchangeable form, 95% of the activity being concentrated in the surface layer of thickness  $Z_* = 10 \text{ cm}$ .

In the model adopted (similar to the FETRA model <sup>7</sup>), the intensity of exchanges in the water-bed system is determined by the coefficient of inter-phase exchange  $a_{1,2}$  (referred to below without the subscripts). Its value was calculated from experimental and published data. Study <sup>15</sup> contains a synopsis of data on the velocity of diffusive exchange in the water and bed sediment system and on the exchange velocity coefficient  $f = a K_D$ , and recommends a value for  $f$  of 0.6 m/annum. Correspondingly, in the series of calculations performed, the coefficient was defined as  $f/K_D$ , where  $f = 3 \cdot 10^{-8}$  m/sec. In our case the main variable calculation parameter is the sorptive distribution coefficient  $K_D$ . Measurement of strontium concentration in solution and on suspensions in Pripyat River conditions yields mean  $K_D$  values close to 100. Computer simulations taking into account the possible variability of  $K_D$  (depending on geochemical factors) yielded values for this parameter ranging from 50 to 1 000.

A series of computer simulations was performed for a <sup>90</sup>Sr concentration at the inflow station of approximately 50 pCi/l, which corresponds to the extreme values for <sup>90</sup>Sr washout conditions in the studied scenarios of runoff formation in the Pripyat's catchment areas. The purpose of these experiments was to determine the increment in <sup>90</sup>Sr concentration at the outfall station due to the egress of <sup>90</sup>Sr from the flooded soils of the floodplain. The calculations indicate that in the event of a 100 year flood on the part of the floodplain studied, the strontium concentration in the water near the outfall station increases to 65-70 pCi/l at  $K_D = 100$ , and to 70-80 pCi/l at  $K_D = 50$ .

A decrease in the depth of flooding on the floodplain in the event of a 0.1 probability flood leads to an increase in the concentration in the water. The extreme case for the range under study ( $K_D = 50$ ) is given in Fig. 2a. At the outfall station the strontium concentration exceeds 100 pCi/l. In the event of a further decrease in the level of inundation on the floodplain and a reduction in flow velocity (0.25 probability flood), the strontium concentration in the water on the section studied increases by over threefold.

We concluded from this that, going by the criterion of concentration level in water, the most dangerous floods may be those with low levels of floodplain inundation. These variants should be studied more closely in subsequent computer simulations, as should the effects of sedimentational-erosional exchange of contaminated load with the bed.

Simulation findings, data obtained by the Institute of Experimental Meteorology (of the Soviet State Committee for Hydrometeorology) on the kinetics of <sup>90</sup>Sr egress into water when floodplain soils are inundated, plus research carried out on the site in question in 1990, made it possible to devise a system of water protection measures for

the territory under study. Taking several variants for localization of radionuclides, we evaluated - by mathematical modelling - the effectiveness of partially or totally covering the floodplain area with sand from the river channel bed (by hydraulic placing), as well as the variant involving completely isolating the worst-contaminated part of the floodplain by means of a high, sand dyke.

The simulation results (Figs. 2a and 2b) show the effectiveness of the proposed construction of an earth dyke along the river, thus preventing radionuclide substances from being washed out of the floodplain into the Pripyat River all in one go, and providing an opportunity to regulate the removal of  $^{90}\text{Sr}$  from the floodplain into Kiev Reservoir.

The relative effect of this measure on all the reservoirs in the Dnieper Cascade was evaluated using a compartmental model of radionuclide migration <sup>9,16</sup>, in which the inflow of contamination from the Pripyat River into Kiev Reservoir was calculated on the basis of the above-mentioned computed estimates for the floodplain. The results of the calculations (Figs. 3a and b) indicate that building a dyke along the floodplain in the Chernobyl NPP near zone (where some 20% of the total amount of  $^{90}\text{Sr}$  in the landscapes of the 30-km zone has concentrated) will - in years when the conditions involved in formation of the Pripyat and Dnieper flow into the Kiev Reservoir are the worst possible - reduce the predicted contamination levels by a factor of five or more in the post-highwater period.

### 3. TRAPS FOR CONTAMINATED SUSPENSIONS IN THE PRIPYAT RIVER

As indicated earlier, in order to reduce the intensity of radionuclide flows on suspended matter, pits were dredged in the river bed in the first few months after the accident. It was assumed that the protective impact of this measure would lie in intensive "self-burial" of radioactive load at these sites. Since many contaminants are carried in river flow and deposited with load, experiments to optimize the specific dimensions of such bed traps, justify their construction and evaluate their effectiveness under operational conditions are of interest, both for resolving the water protection tasks in the lower reach of the Pripyat River and for drawing up recommendations for measures connected with accidental contamination of surface waters.

To calculate the dynamics of radionuclide accumulation in bed sediments in areas where there are abrupt changes in depth, we used a vertical two-dimensional model <sup>9</sup> and a one-dimensional model simulating the partial case described in the previous chapter based on the two-dimensional depth-averaged model. Let us now look at the block in the first of these models describing the velocity pattern of the flow and movement of suspended load.

Assuming that pressure is distributed over depth according to the law of hydrostatics, the equations covering displacement and continuity in the zone  $-H(x) < z < \eta(x,t)$  are expressed by

$$\frac{\partial u}{\partial t} + u \frac{\partial u}{\partial x} + w \frac{\partial u}{\partial z} = -g \frac{\partial \eta}{\partial x} + A_1 \frac{\partial^2 u}{\partial x^2} + \frac{\partial}{\partial z} A_3 \frac{\partial u}{\partial z} = 0 \quad (14)$$

$$\frac{\partial u}{\partial x} + \frac{\partial w}{\partial z} = 0 \quad (15)$$

where  $u, w$  = horizontal and vertical components of flow velocity,  $\eta$  = elevation of the free surface above the level of the unperturbed depth of the fluid  $H(x)$ ,  $A_1$  = coefficient of horizontal turbulent viscosity assumed to be constant,  $A_3(x, z, t)$  = coefficient of vertical turbulent viscosity.

To close the system (14), (15) we use Prandtl's hypothesis

$$A_3 = \lambda^2 \left| \frac{\partial u}{\partial z} \right| \quad (16)$$

where  $\lambda$  = scale of turbulence, for which we take Montgomery's formula

$$\lambda = \frac{\kappa}{H} (z + H + z_0) (-z + \eta + z_\eta) \quad (17)$$

where  $\kappa = 0.4$  = Karman's constant,  $h = H + \eta$ ,  $z_0, z_\eta$  = the parameters of roughness and free surface respectively.

In describing the displacement of a two-phase flow, use of the closure method (16), (17) developed for a single-phase flow is based on the assumption that the presence of suspension has little effect on the characteristics of turbulent exchange. As shown in 17, this hypothesis is correct for the concentrations of suspensions observed in river flows.

The boundary conditions at the bed are the conditions of adhesion

$$z = -H + z_*: u = 0, w = 0$$

On the free surface of the fluid  $z = \eta$  we adopt the kinematic condition

$$w = \frac{\partial \eta}{\partial t} + u \frac{\partial \eta}{\partial x} \quad (19)$$

and assume that shear stress equals zero.



The dynamics of the suspension are expressed by the equation for advective-diffusive movement:

$$\frac{\partial s}{\partial t} + u \frac{\partial s}{\partial x} + w \frac{\partial s}{\partial z} = w_0 \frac{\partial s}{\partial z} + A_1 \frac{\partial^2 s}{\partial x^2} + \frac{\partial}{\partial z} \left( A_3 - \frac{\partial s}{\partial z} \right) \quad (20)$$

where  $s(x, z, t)$  = concentration of suspended particles with mean settling velocity  $w_0$ .

For the boundary condition on the free surface it is assumed that the vertical flow of load is equal to zero

$$z = \eta: \quad (w - w_0) s = A_3 \frac{\partial s}{\partial z} \quad (21)$$

The diffusive flow of the suspension on the bed is adopted to be equal to the sedimentational equilibrium flow. Then the boundary condition is expressed as

$$z = -h + z_0: \quad A_3 \frac{\partial s}{\partial z} + w_0 s_* = 0 \quad (22)$$

where  $s_*$  = benthic equilibrium concentration of the suspension corresponding to the transport capacity of the flow.

To calculate  $s_*$  we use methods of describing the suspended load based on linking its benthic concentration with overstated actual values for benthic shear stress  $\tau = \rho u_*^2 = \rho A_3 \frac{\partial u}{\partial z}$  above the threshold value  $\tau_c$  corresponding to the beginning of mass movement of bed particles. We used van Rijn's formula 19, together with Bijker's formula 10 (mentioned earlier) based on some other principles.

Accumulation of radionuclides in bed sediments is influenced by two processes: deposition of contaminated suspensions and diffusive exchanges at the water-bed boundary. The role played by the first of these factors is significantly greater in the bed traps, and therefore this paper looks at their impact only.

For numerical solution of the equations with corresponding boundary conditions, we used an explicit-implicit finite difference schema with a scattered chessboard layout for  $u$  and  $h$  (or  $s$ ) in the  $x$ - $t$  plane. Here the implicit block approximates vertical turbulent mixing. Vertical advective movement was approximated using an explicit schema of directed differences of the first order of accuracy. The resultant computed diffusion for the estimates is far less than physical vertical turbulent diffusion, which makes it possible to use the simplified

schema mentioned earlier. Due to the large vertical gradients of the unknown functions, we used a non-uniform grid for the vertical coordinate.

The models proposed were validated using data from laboratory experiments; we also used measurements of distribution of flow velocities and suspended load above an underwater pit in the sandy bed of a laboratory flume <sup>19</sup> (Fig. 4 shows the pit layout and its vertical dimensions). In this particular experiment the suspension parameters were  $w = 0.013$  m/sec and  $D = 0.00015$  m, while for the flow at the inflow station  $u = 0.5$  m/sec and  $h = 0.4$  m.

Figs. 5a and 5b compare the horizontal velocity distributions obtained from the two-dimensional model (unbroken curve) and the suspension concentrations (broken curve) with experimental data <sup>19</sup> (circles and triangles respectively) for the verticals. The values for suspension concentration are normalized to its value when depth  $z = 0.1H$  at the inflow station. Numerical experiments indicated that horizontal turbulent diffusion has very little impact on the calculation results in this particular study, and therefore the calculations assumed that  $A_1 = 0$ . It should be noted that the calculated curves for the distribution of the suspension concentration along the vertical, obtained by using different formulae for the equilibrium bed concentration  $s$ , are - within the accuracy limits of the graphs - practically indistinguishable at distances from the bed exceeding 0.01-0.02 of the depth, and for this reason they are not given separately in Fig. 5. However, the difference observed in the benthic concentrations of suspensions may produce significant differences when calculating the total vertical flow of suspension  $q$ , which is the main feature of water-bed exchanges.

If we use Bijker's formula in the two-dimensional model, the calculated sedimentation velocities tally more or less with the values computed when determining  $s$  using van Rijn's formula. However, the erosive flow at the pit exit is 50-100% lower in the case of the former.

The models were used to evaluate the deposition of suspensions in two underwater pits in the Pripyat River. The first is near the settlement of Otashev. Our calculations were based on the following pit design parameters: length = 100 m, depth = 12 m at a river depth of 4.8 m in the pit approach. The calculations assumed that the mean diameter of the suspensions was 0.05-0.01 mm. It was also assumed that upstream of the trap turbidity in the flow corresponded to the flow's transport capacity.

Our calculations show that the most active accumulation of sediment takes place on the front slope of the trap, and that the rear slope is slowly eroded, mainly by movement of large suspensions. The length of the active sedimentation zone increases with decreasing particle size, and when  $D = 0.01$  mm and velocity = 0.2 m/sec or over it exceeds the length of the pit. The sharp decline - revealed during simulation experiments - in the pit's effectiveness in capturing suspensions as particle size decreases meant a detailed analysis of the radiological processes was necessary.

A factor determining the effectiveness of the bed traps in capturing contamination is the distribution of the radionuclides in the flow on different suspension fractions. Studies of the Pripyat River showed that over 70% of the caesium radioactivity carried along by suspensions is bound to particles under 0.1 mm in size. It is precisely these fractions which predominate in the granulometric composition of the suspended load in the Pripyat River, with 40-60% of the caesium carried along by suspended load being bound to the fraction under 0.05 mm.

These findings on the transport of radioactivity on suspensions were confirmed by an analysis of radionuclide accumulation in the bed sediments of the largest protective structure in the channel of the Pripyat River - the Ivanovsky pit; this is situated two kilometres downstream of the town of Chernobyl and has the following parameters: length = 850 m, maximum depth = 16 m, maximum width = 400 m, which is twice the mean width of the river channel. It was completed by the time the 1987 spring high water began.

Data from in-situ equilibrium studies covering a year of mean water volume show that the caesium accumulated via sedimentation of contaminated suspended particles is some 10% of the total amount of caesium conveyed past the pit inflow station with Pripyat River sediment discharge. This accumulated activity is reflected in an increased caesium concentration in the silt sediment in the pit (500 - 2 000 pCi/g), which is 1-2 orders of magnitude greater than the contamination found in sandy bed load in adjacent areas of the river channel. It should be noted that in areas of the Pripyat mouth where a natural increase in sedimentation occurs (creeks, deep pools, backwaters), the caesium concentration in bed sediments is the same, and sometimes higher, than in the pit.

### Conclusions

1. Our analysis of measures to reduce the intensity of radionuclide movement with surface waters confirmed the advantages of adopting an approach combining mathematical modelling with a detailed analysis of the hydraulic and physico-chemical processes in water flows in order to select the most effective water protection methods.
2. In the conditions obtaining after the Chernobyl accident the most effective measure may be to build a cut-off dyke to prevent floodwaters from washing radionuclides out of the worst-contaminated area of the Pripyat River floodplain in the Chernobyl NPP near zone.
3. Dredging bed pits to trap radioactive load had no significant effect on overall removal of radionuclides from the Pripyat River into Kiev Reservoir, due to their relative ineffectiveness in capturing the fine fractions of suspended load. The pits did play a decisive role in capturing fuel particles, which are associated with relatively large load fractions.
4. Theoretical and experimental studies should be further pursued along these lines for the purpose of elaborating - on the basis of analysing the Chernobyl accident consequences - recommendations for water protection measures after radionuclides accidentally enter aquatic systems.

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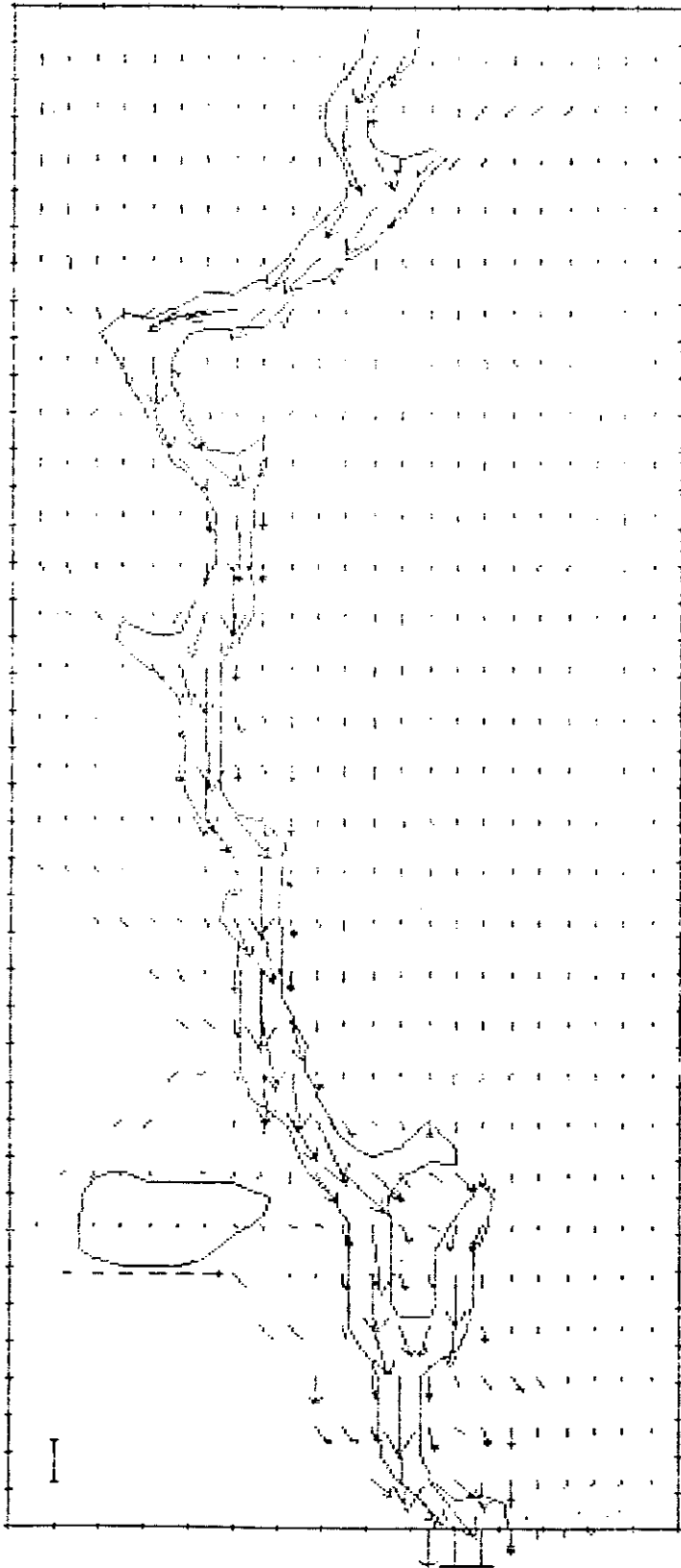


Fig. 1a

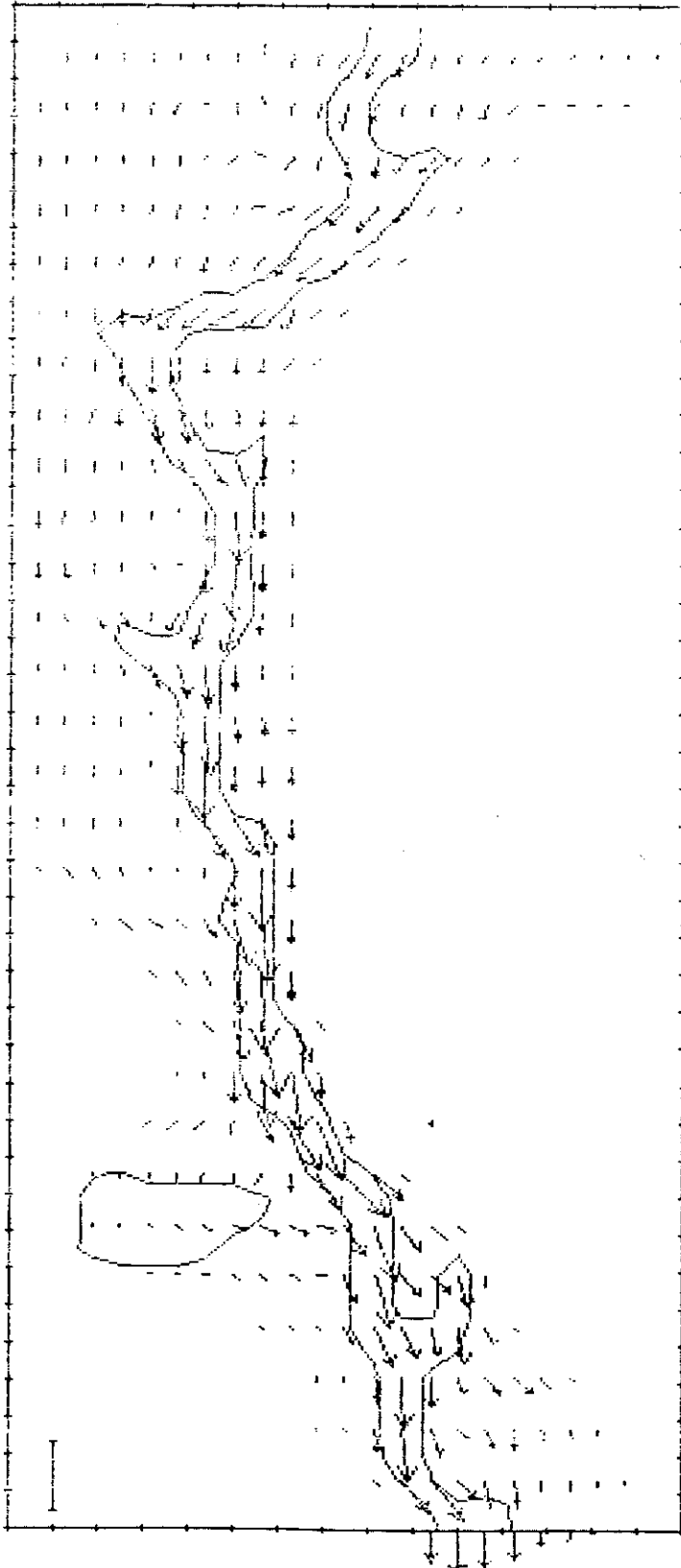
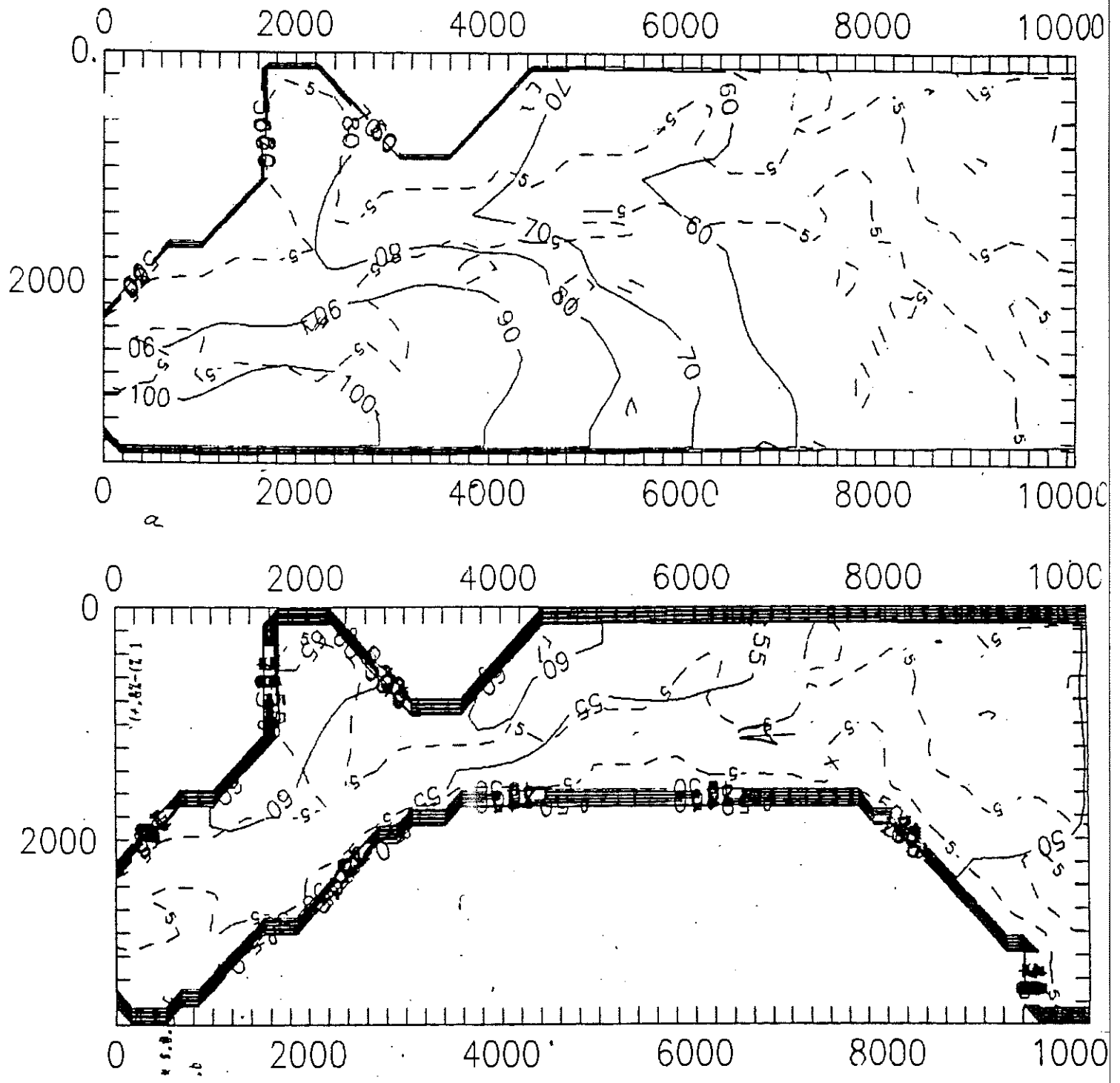


Fig. 1b

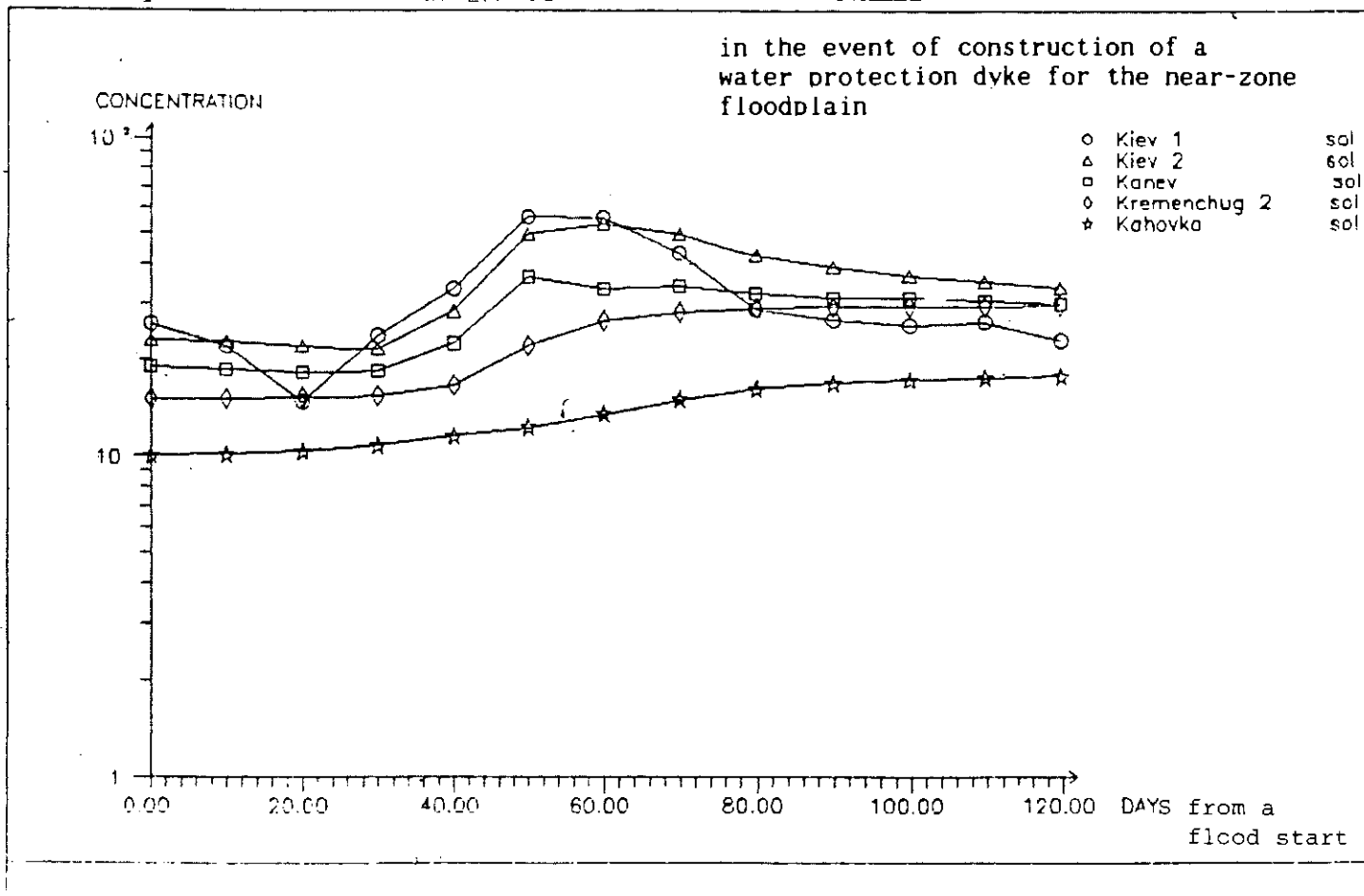
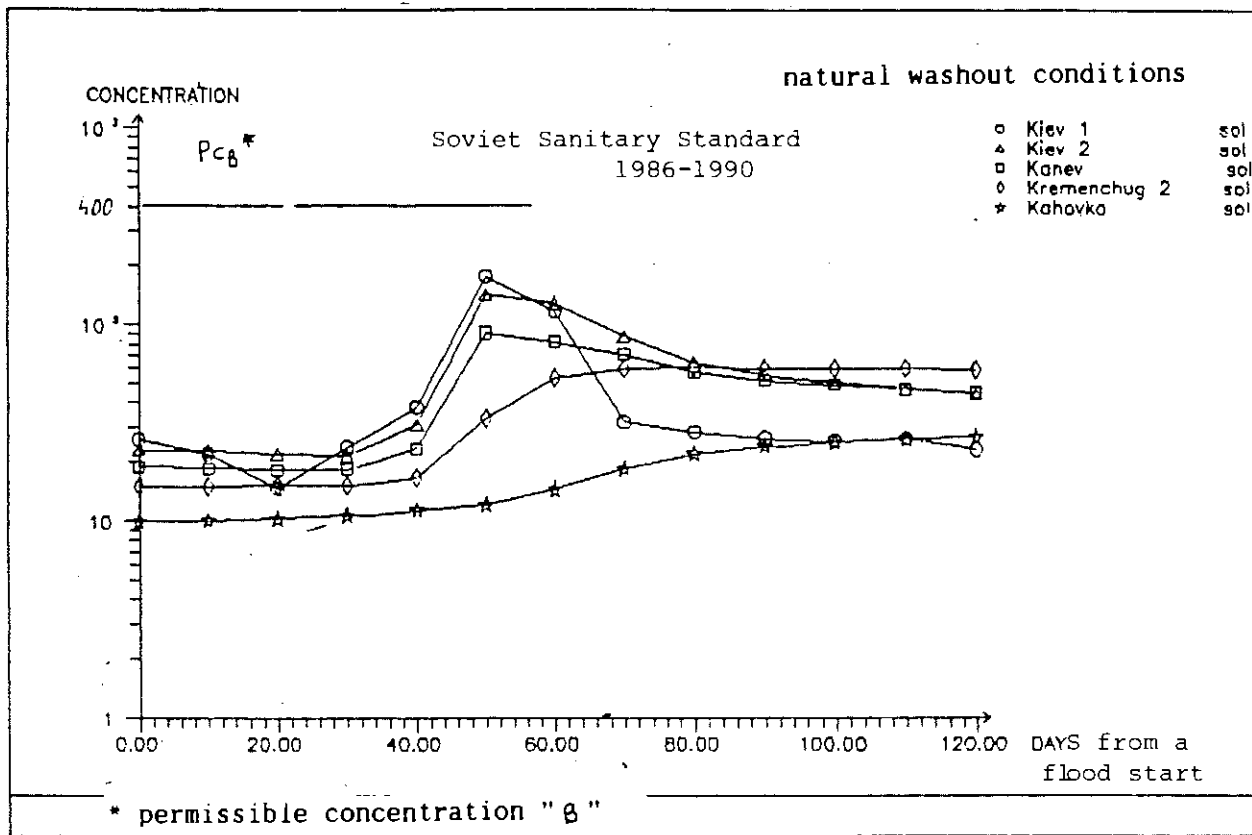


Fig.2: COMPARATIVE ANALYSIS OF Sr-90 WASHOUT FROM THE FLOODPLAIN IN THE CHERNOBYL NPP NEAR ZONE UNDER THE NATURAL CONDITIONS OF EXTREME (10%) HIGH WATER AND IN THE EVENT OF CONSTRUCTION OF A WATER PROTECTION DYKE FOR THE WORST-CONTAMINATED PART OF THE FLOODPLAIN ON THE LEFT BANK OF THE PRIPYAT RIVER



-50- Isolines of Strontium concentration in the water of the Pripyat river

Fig.3: Computed concentrations of <sup>745</sup>Strontium-90 in the reservoirs of the Dnieper cascade in the event of extreme washout of radionuclides from the floodplain in the Chernobyl NPP near zone given a 25 % frequency high water



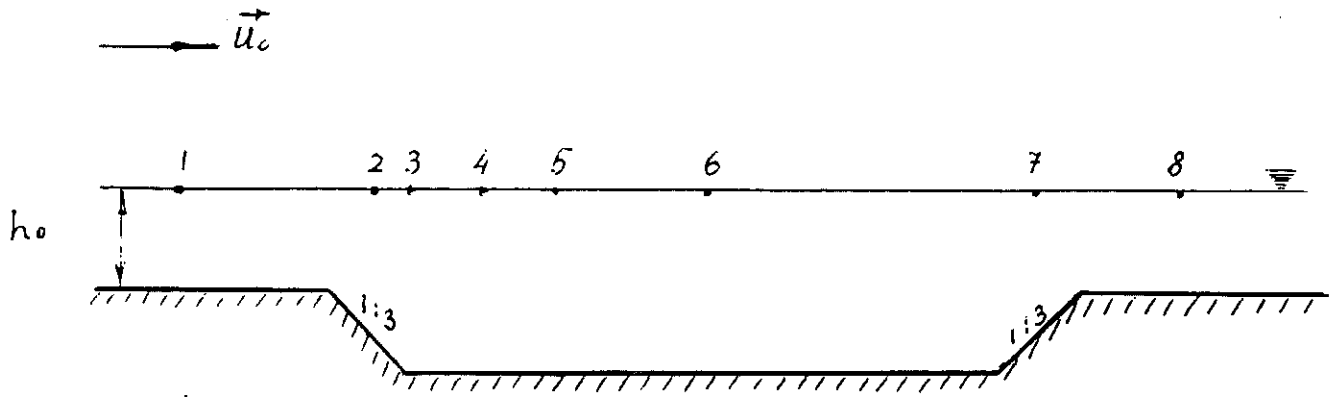


Fig. 4

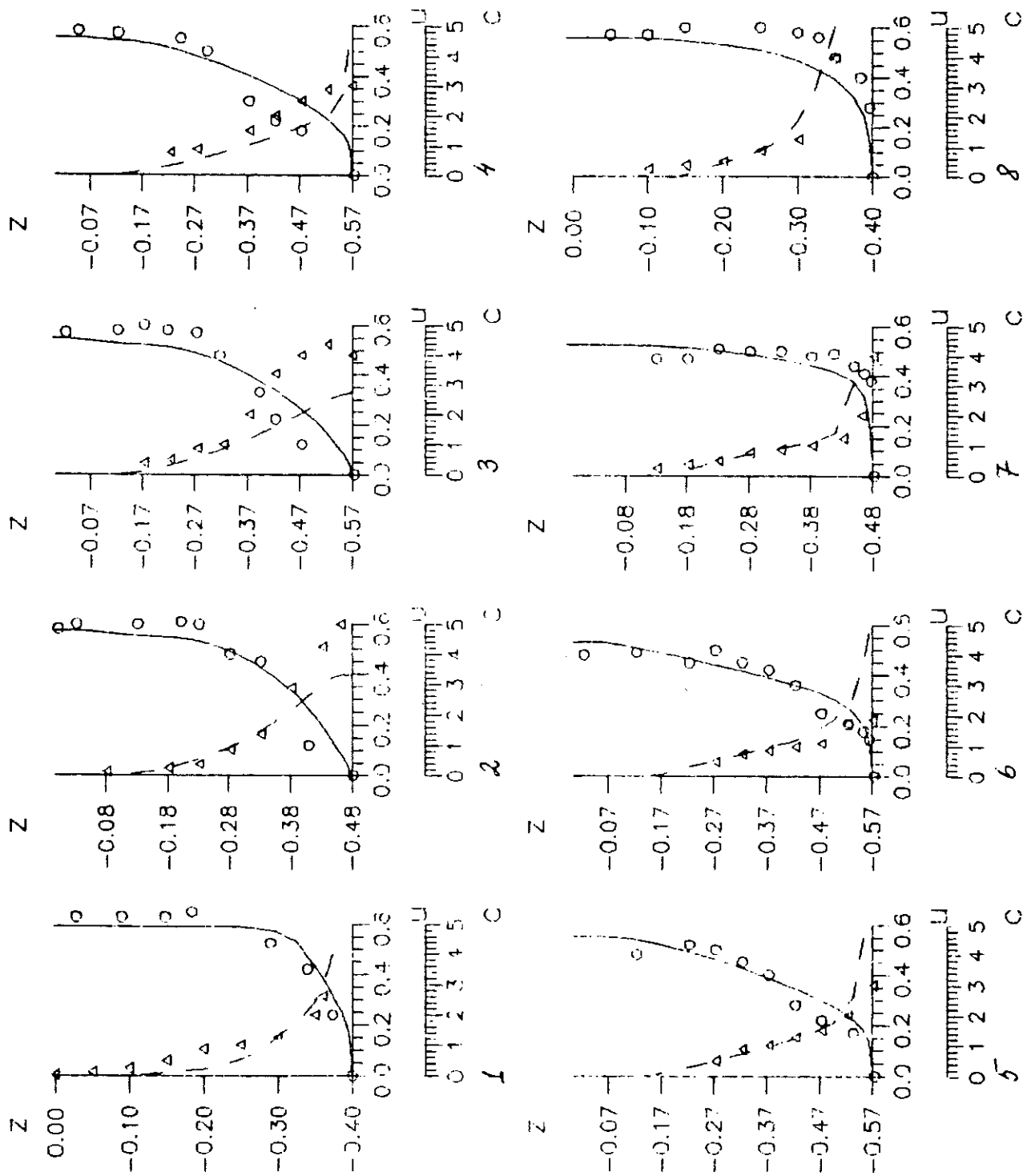


Fig. 5



**Radiological Consequences of  
the Chernobyl NPP Accident in  
Comparison with those of the  
Kyshtym and Windscale  
Radiation Accidents**

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## ABSTRACT

The report briefly sets out the main features involved in the accidental releases of radioactive substances at Chernobyl, Kyshtym and Windscale, and the radiation conditions developing as a result. We outline the dynamics of the transfer of radioactive substances via the "soil-plant-animal-man" food chain, and compare the processes involved in internal and external exposure of the people who stayed on in the radioactively contaminated regions. In the Chernobyl and Windscale nuclear reactor accidents iodine and caesium were mainly responsible for irradiation of the local population. In contrast, the explosion at the storage facility for liquid radioactive waste in the Urals contaminated the area with a mixture of one-year-old fission products, and the long-term ecological effects stem mainly from the presence of strontium-90 in foodstuffs. The report traces the true dynamics of (and predicts future) individual and collective doses to Russia's inhabitants. The latest radiobiological data are used to estimate the incidence of long-term stochastic effects of a carcinogenic, genetic and teratogenic nature.

The one thing common to the worst radiation accidents in the history of mankind - the Kyshtym and Windscale accidents in 1957 and the Chernobyl accident in 1986 - is the fact that they involved medically significant contamination of the environment with radionuclides and consequent irradiation of the population. Large-scale measures were taken to protect the population from radiation after all three accidents. Since two involved destruction of operational nuclear reactors, while the third involved the explosion of a tank containing high-level radioactive waste, the corresponding differences in the isotopic and physico-chemical composition of the radionuclides released also meant there were differences in the way the general public was irradiated. This report compares the irradiation processes affecting the people staying on in the accident zones and estimates individual and collective doses, with most attention being paid to the more recent situation in the controlled zone of the Chernobyl accident, where the author regularly works. It does not examine irradiation of staff working at the facilities involved, nor does it analyse the long-term medical effects of irradiation of the populace, since such effects have not yet appeared among the population, and the coefficients of possible radiation risk required for medical forecasting are under review at this very moment.

Table 1 contains figures, based on data 1-7, indicating the amounts of the nuclear fission products, most significant from a radiation point of view, which were accidentally released into the atmosphere. The Kyshtym cloud contained no short-lived radionuclides of iodine whatsoever and the portion of caesium was insignificant, whereas cerium and zirconium radionuclides, which do not carry very far, and their daughter products dominated. The most ecologically mobile and long-lived radionuclide is strontium-90 (2.7% of the mix), which also determines the radiation situation in the long term 6,5. The Windscale 1 and Chernobyl 2,3,4,7 reactor accidents released a larger amount of volatile radionuclides of iodine and caesium, together with noble radioactive gases (not in the Table). The high-temperature heat-up of the stricken Chernobyl unit also mobilized less volatile isotopes of strontium, cerium and zirconium, etc., which are concentrated more in the area near the Chernobyl NPP. The bottom row shows the amounts released at Chernobyl as estimated by Soviet specialists 2,3,4 and by scientists at the Livermore Laboratory in the USA 7. It is clear from this that the Chernobyl emission of volatile radionuclides of iodine, caesium and ruthenium was three orders of magnitude greater than that at Windscale, despite the similar nature of the accidents. The Kyshtym and Chernobyl releases of non-volatile strontium, zirconium and cerium radionuclides are of comparable magnitude, but in the latter case these nuclides are of secondary importance only.

Tables 2 and 3 set out in qualitative terms the main population irradiation factors in the first year and in the longer term. The number of plusses in the grid expresses the expected individual external or internal irradiation dose to the critical population group and the need to introduce protective measures.

Thus, the inhabitants of the worst affected settlements following the Kyshtym accident were evacuated urgently because of the danger of external gamma radiation (+++). The second most important factor here was irradiation of the gastrointestinal tract (GIT) due to surface contamination of food with radionuclides of low solubility (Table 2). The main long-term factor for individuals remaining in the radioactively contaminated zone is irradiation of the red bone marrow



(RBM) and bone tissue by incorporated strontium-90 (Table 3). In quantitative terms the dose formation process is as follows.

During the first phase of the Windscale accident possible radiologically significant irradiation of the population was associated with the emission of iodine-131 and its accumulation in the thyroid gland, as well as with external gamma-radiation.

The Chernobyl accident involved - to varying degrees - a number of radiation factors, as shown in Tables 2 and 3. The critical factor in the initial phase was external and internal irradiation of the whole body (caesium radionuclides) and, more particularly, of the thyroid gland by radionuclides of iodine. Later on, the dominant factor was external and internal irradiation from caesium-137.

We will now look at the dose formation processes involving the factors mentioned above.

Fig. 1 shows (in relative terms) how the gamma-radiation dose rate in the air depends on time. In the initial period after the Kyshtym accident, 80% of the dose rate was due to radiation from zirconium-95 and niobium-95 and therefore fell quickly - by almost 20 times in the first year. In the following four years the reduction in the dose rate in the air occurred by periods of approximately one year and then slowed down when it reached a level just 0.2% of the initial one. We took the concentration of the most long-lived component in the mix, strontium-90, as an indicator of the level of radioactive contamination of the countryside. Given strontium-90 soil contamination of 37 GBq (1 Ci) per km<sup>2</sup>, the mean dose rate in the air soon after deposition of the cloud from the Kyshtym blast was 1.3 μGy/h.

Following deposition of Chernobyl fallout in the town of Novozybkov, Bryansk oblast (region), 200 km northeast of Chernobyl and with 0.6 TBq/km<sup>2</sup> of caesium-137 on the ground, the initial gamma-radiation dose rate in the air of some 40 μGy/h fell over two months to 2 μGy/h due to decay of the short-lived radionuclides, and then by only one half during the following four years. At present over 90% of the dose rate comes from radionuclides of caesium, and the reduction rate has slowed down.

Fig. 2 shows the dynamics of intake of iodine-131 (in the absence of protective measures) into the body of inhabitants via food and air, as well as the thyroid gland radionuclide content from protracted intake. According to our data, in the Bryansk and other oblasts of Russia the greatest intake was via local milk, while the contribution from also inhaling short-lived isotopes of iodine did not exceed 10% of the dose. I. Zvonova's paper looks at the radioiodine problem in depth.

Fig. 3 shows (in relative unit terms) the dynamics of caesium-137 content in food produced in the radioactively contaminated zone in Bryansk oblast. The specific activity of each product in kBq/kg has been normalized in relation to - i.e. been divided by - soil contamination in TBq/km<sup>2</sup>, to give 10<sup>-9</sup> km<sup>2</sup>/kg.

As the chart shows, the level of aerogenic contamination of leafy vegetables by radionuclides fell by 2-3 orders of magnitude over several months. In contrast, the caesium-137 content in potatoes and root crops is low but changes little in the course of three years. The figures for animal products reflect the processes of initial accumulation of surface contamination on vegetation and soil, metabolic

cleansing and seasonal variations in radionuclide content in feed. The caesium-137 concentration in milk decreases in an undulating manner, with a one-year variation period.

Multiplying the caesium-137 content in food products in each month by the mean consumption of such products by rural inhabitants of the Bryansk oblast prior to the Chernobyl accident <sup>9</sup>, gives the function of alimentary intake of this radionuclide into the body, assuming no changes in diet after the accident (Fig. 4, band 1). In actual fact, thanks to the protective measures taken, consumption of local food products was reduced considerably in the controlled area <sup>9</sup>, as a result of which intake fell to the range shown in Fig. 4, band 2. The upper and lower boundaries of each band correspond to intake in individuals with different eating habits.

The convolution integral - for the intake functions in Fig. 4 and the standard function for caesium retention in the human body (ICRP-30) - predicts the function of radionuclide content in the bodies of rural inhabitants, for both normal and "protective" diets (Fig. 5, bands 1 and 2). Fig. 5 also shows the real caesium-137 content as measured among the inhabitants of three settlements with the aid of transportable whole-body counters. The curves showing actual radionuclide content in the inhabitants' bodies (3) are similar in shape to those calculated (1 and 2), but are at a somewhat lower level. In subsequent years the activity of the incorporated caesium radionuclides falls by a period of 1-2 years in general.

Following one-off contamination of the area by strontium-90, the initial content of this radionuclide in surface-contaminated plant products and in animal products falls very sharply in the course of 1-3 years thanks to migration of the nuclide into the soil and a decrease in resuspension. In keeping with observations made in the Urals <sup>6</sup> three years or more after the accident, intake of strontium-90 into the body with food is associated almost exclusively with root transfer from the soil. The period for subsequent decrease in the content of this nuclide in food is estimated to be around 10 years. The model used in the USSR for strontium-90 metabolism and dosimetry in individuals of different ages, including children, was devised by the Chelyabinsk authors M. O. Degteva and V. A. Kozheurov <sup>8</sup>; it is based on an analysis of world literature on the ecology of global strontium-90 and was verified against Kyshtym accident material.

The structure and dynamics of irradiation of the population living in the Kyshtym accident zone are given in Fig. 6, these being based on data from <sup>5,6</sup> and expanding on Table 2. In line with the isotopic composition of the radioactive contamination in each area given in Table 1, during the first year after the accident the inhabitants experienced whole-body exposure to external gamma-radiation, while the walls of their gastrointestinal tract received beta-radiation from radionuclides ingested with surface-contaminated food products and then weakly absorbed into the blood. Fig. 6 contains quantitative assessments of the dose based on strontium-90 soil contamination of 37 GBq (1 Ci) per km<sup>2</sup> in the absence of active measures to protect the population against radiation. In units of equivalent dose, the GIT irradiation level is almost one order of magnitude higher than that for the whole body. In the very first year after the accident the dose of strontium-90 and yttrium-90 in the red bone marrow and bone tissue cells was similar to the gamma-radiation dose, but after more than three years, when the external radiation dose and the dose to the GIT were more or less finally formed, any further increase in the effective

dose is due to radiation from osteotropic strontium-90 and yttrium-90. The effective whole-life dose for people living in podzolic soil areas with strontium-90 contamination of 37 GBq/km<sup>2</sup>, is put at 10-15 mSv on the basis of the data in 5,6.

The structure and numerical estimates of the irradiation dose to the population in the Chernobyl accident zone are shown in Fig. 7 and Table 4 (on the basis of our own observations). The quantitative assessments expand on Tables 2 and 3 on the basis of caesium-137 soil contamination of 370 GBq (10 Ci) per km<sup>2</sup> in the absence of active protection measures. In the initial one or two months after the accident the most significant dose equivalent was received by the thyroid gland (from 0.1 Sv in adults to 0.5-1 Sv in children up to 7 years of age). During this time a dose of 1-2 mSv accumulated in the whole body from external gamma-radiation, with a somewhat smaller one to the whole body resulting from internal irradiation by caesium radionuclides (caesium-134 and -137). The processes involved in - and the level of - chronic internal irradiation of inhabitants by caesium radionuclides ingested with local food very much depend on the properties of the soil. Thus, on the Bryansk oblast's podzolic soils the internal irradiation dose is similar to, or slightly above, the external irradiation dose (Fig. 7, curve 1), while in Tula oblast, where chernozem soil predominates, this dose ratio lasted for the first year only, when radionuclide surface contamination of food products played a major role. In subsequent years the chernozem soils firmly fixed the caesium (and strontium), as a result of which the internal irradiation dose was several times lower (by as much as an order of magnitude) than in Bryansk oblast (Fig. 7, curve 2). The dose level after 1 600 days is derived from the results of personal dosimetry of external and internal irradiation of the population. We then extrapolated the dose over 70 years on the basis of the following safe-side assumptions: a) the dose rate in the air will decrease mainly as a result of caesium-137 decay over a period of approximately 30 years; b) cleansing of the biosphere and the food of the local population from caesium radionuclides will occur over a period of 10-15 years. On this basis we estimate the mean effective 70-year dose to people in settlements in an area with podzolic soil and caesium-137 contamination of 370 GBq/km<sup>2</sup> to be about 0.1 Sv. In the far-off Chernobyl accident zone, where the mean strontium-90/caesium-137 activity ratio is around 1%, the strontium-90 and yttrium-90 contribution to the effective dose of the critical group - those who were children at the time of the accident - is estimated to be somewhere between 1 and 10% (in Fig. 7 - 3%).

To illustrate the actual irradiation level for the population in the Chernobyl accident zone, Table 4 gives some figures from our institute's data base covering the controlled area (over 0.6 TBq/km<sup>2</sup> caesium-137) in Bryansk oblast. All the mean doses, apart from those predicted, are based on the findings of selected individual measurements of caesium and iodine radionuclide content in inhabitants' bodies, as well as measurements of external gamma-radiation dose using thermoluminescent dosimeters. The dose received by the inhabitants is related to the caesium-137 level on the ground, but also depends on a number of other factors. For example, in the town of Novozybkov over half the population live in brick houses which shield them from gamma-radiation, as a result of which the mean dose is correspondingly lower than in settlements. The consumption of a number of local food products was forbidden sufficiently early throughout the zone, and these were replaced by others imported from the "clean" zone; as a result, over the past four years the contribution of internal irradiation to the effective dose is somewhere between 13-25% instead

of the 50-70% to be expected in the given natural conditions. What is striking is the great dependence of the thyroid gland dose on the age of the inhabitants in one and the same populated area. The ratio between the dose in children up to 7 years of age and that in adults is 6, due to the difference in body mass and milk consumption. In the 1986-1990 period the mean dose did not exceed the established levels in any of the populated areas in Bryansk oblast. However, the inhabitants of the Zaborye and Svyatsk settlements, where the mean whole-life dose might exceed the 35 rem limit at some stage in the future, are currently being resettled to other parts of Bryansk oblast where radiation is at natural levels.

To compare the radiological consequences of the three accidents correctly Table 5 contains separate assessments of the collective dose, i.e. for iodine radionuclides to the thyroid gland and for the effective dose equivalent. The Windscale accident figures are taken from 1; those for Kyshtym are our own assessment based on 5,6; those for Chernobyl covering the controlled area and the European part of the USSR come from us and our co-authors 4, while those for Eurasia are taken from 2 and supplemented by us. We should start by noting the massive scale of the Chernobyl accident compared to the 1957 accidents, the difference in the collective effective dose equivalent being some two orders of magnitude and more. Assuming a linear unlimited "dose-effect" relationship, we can thus expect there to be a correlation with the number of fatal illnesses. Secondly, we would stress the differences in irradiation localization and in the expected medical consequences. In the case of the Kyshtym accident, in which a significant contribution to the effective dose equivalent came from osteotropic radionuclides in addition to whole-body irradiation, increased attention should be paid to the number of malignant diseases of the blood system. The main factor in the Windscale accident was radioactive iodine, which could lead to a higher incidence of tumorous pathologies of the thyroid gland. This factor also played a role in the Chernobyl accident, but to a significantly greater extent and in conjunction with external and internal irradiation of the whole body. Consequently, medical attention should focus on the condition of the thyroid gland and on the whole spectrum of long-term stochastic effects, whether carcinogenic, genetic or teratogenic. We should not forget the enormous psychological impact of the stress to which the accident zone population has been subjected for four years now. This factor, which is playing an increasing role in the USSR at the present time, can quite easily have an adverse affect on the health of the population. This paper does not make any quantitative forecasts about the possible radiogenic medical effects, for the reasons set out in the introduction.

In conclusion, we should note that the analysis, contained in this report, of the radiation factor in major public accidents is not yet finished. The Chernobyl accident revealed gaps in the studies of several major questions concerning ecological dosimetry, such as:

- long-term forecasting of external and internal irradiation of the population, taking the particular soil features into account;
- the radiological role of "hot particles";
- the radiological role of actinides.

Research into these phenomena, plus an assessment of the role played by radon in the accident zone, is currently of vital importance.

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Table 1: Accidental release of some radiologically significant nuclides. PB<sub>1</sub>

Accident & year	Bibliography	I-131	Cs-134	Cs-137	Sr-90	Zr-95	Ru-106	Ce-144
Kvshtym 1957	(5.6)	-	-	0.03	2	9	1.4	24
Windscale 1957	(1.7)	0.7	0.001	0.04	$2 \cdot 10^4$	-	0.01	-
Chernobyl 1986	(2.3.4.7)	600 - 1300 *	40 - 50 *	70 - 90 *	1.3* - 8	8* - 140	6* - 60	5* - 90

(\*) Based on data from the Livermore Laboratory, USA (7); release carried for away

Table 2: Main factors in irradiation of the population in the first year after the accident

Accident & year	External irradiation	Internal irradiation			
		Whole body	thyroid gland	GIT	RBM. bone
Kyshtym 1957	+++	-	-	++	++
Windscale 1957	+	-	+	-	-
Chernobyl 1986	+++	+++	+++	+	+

Table 3: Main factors in long-term irradiation of the population

Accident. & year	External irradiation	Internal irradiation	
		Whole body	RBM. bone
Kyshtym 1957	+	-	++
Windscale 1957	-	-	-
Chernobyl 1986	++	++	+

Table 4: Mean population irradiation doses for some populated areas in the Bryansk oblast after the Chernobyl accident

Town, settlement (population)	Cs-137 (TBq/km <sup>2</sup> )	Dth, cSv (iodine-131)			Dwb, cSv (1986-1989)		Def. cSv (1986-2056)
		< 7 years	7-16 years	> 16 years	ext.	int.	
T. Novozybkov (46 thousands)	0.6	40	20	7	1.5	0.5	5 - 10
T. Zlynka (6 thousands)	1.0	40	20	7	3.0	0.7	10 - 15
S. Svyatsk (632)	1.8	80	40	20	6.6	0.7	30 - 40
S. Zaborye (1065)	2.5	120	70	50	9.4	1.5	40 - 60



Table 5: Expected collective dose following the major radiation accidents  
(thousands of man-Sv)

Accident & year	Area	Population million people	Collective dose	
			I-131 thyroid gland	Effective
Kyshtym 1957	Urals	0.27	-	4 - 6
Windscale 1957	Europe	-	18	1.3
Chernobyl 1986	"Controlled"	0.27	70	56
	European part of USSR	75	460	330
	Europe, Asia	-	1000	600 - 700

Fig.1: Kerma rate in air 1m above virgin area of soil after the kyshtym and Chernobyl accidents.

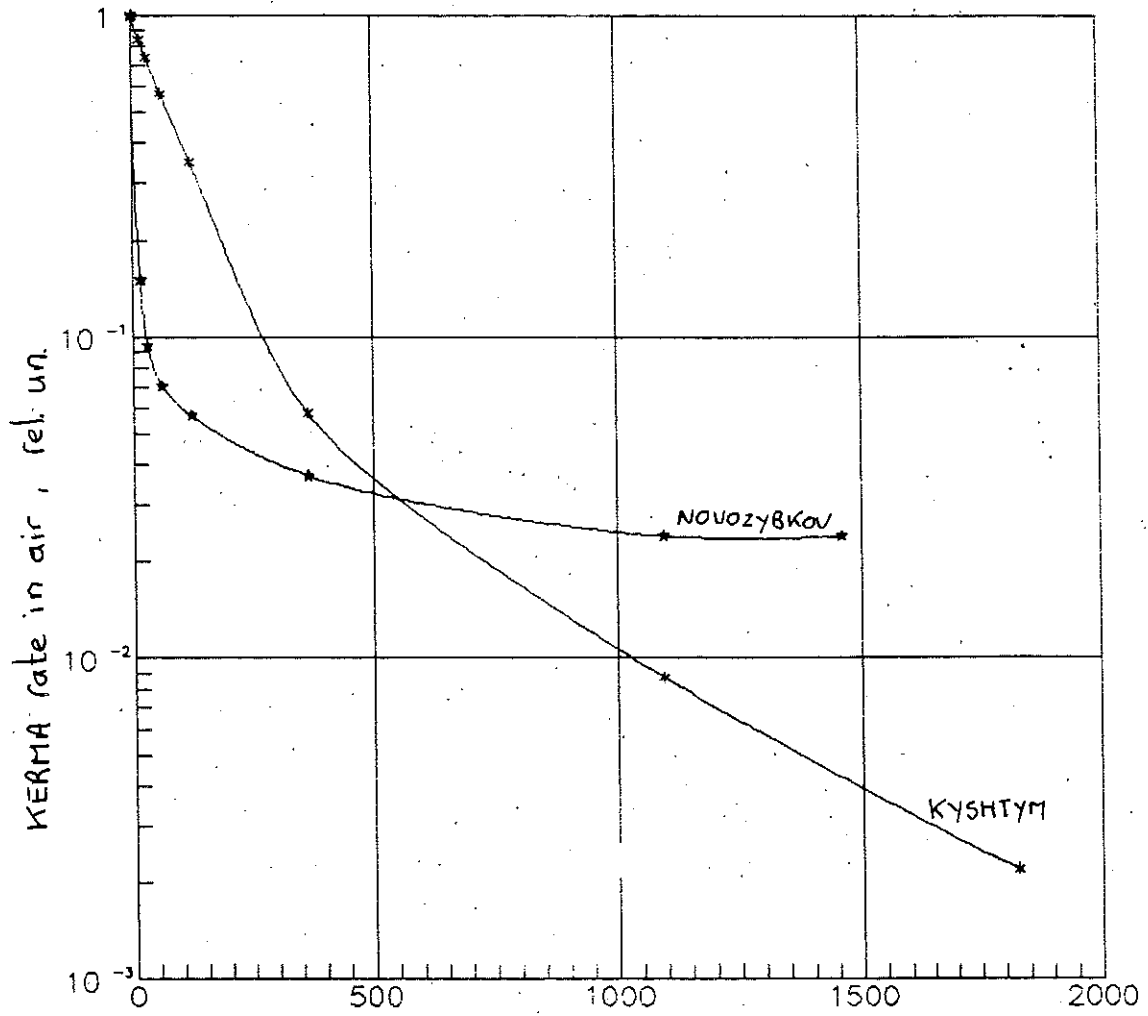


Fig.2: Daily intake (A) and content (B) of iodine-131 in the thyroid gland of residents after the Chernobyl accident (pattern)

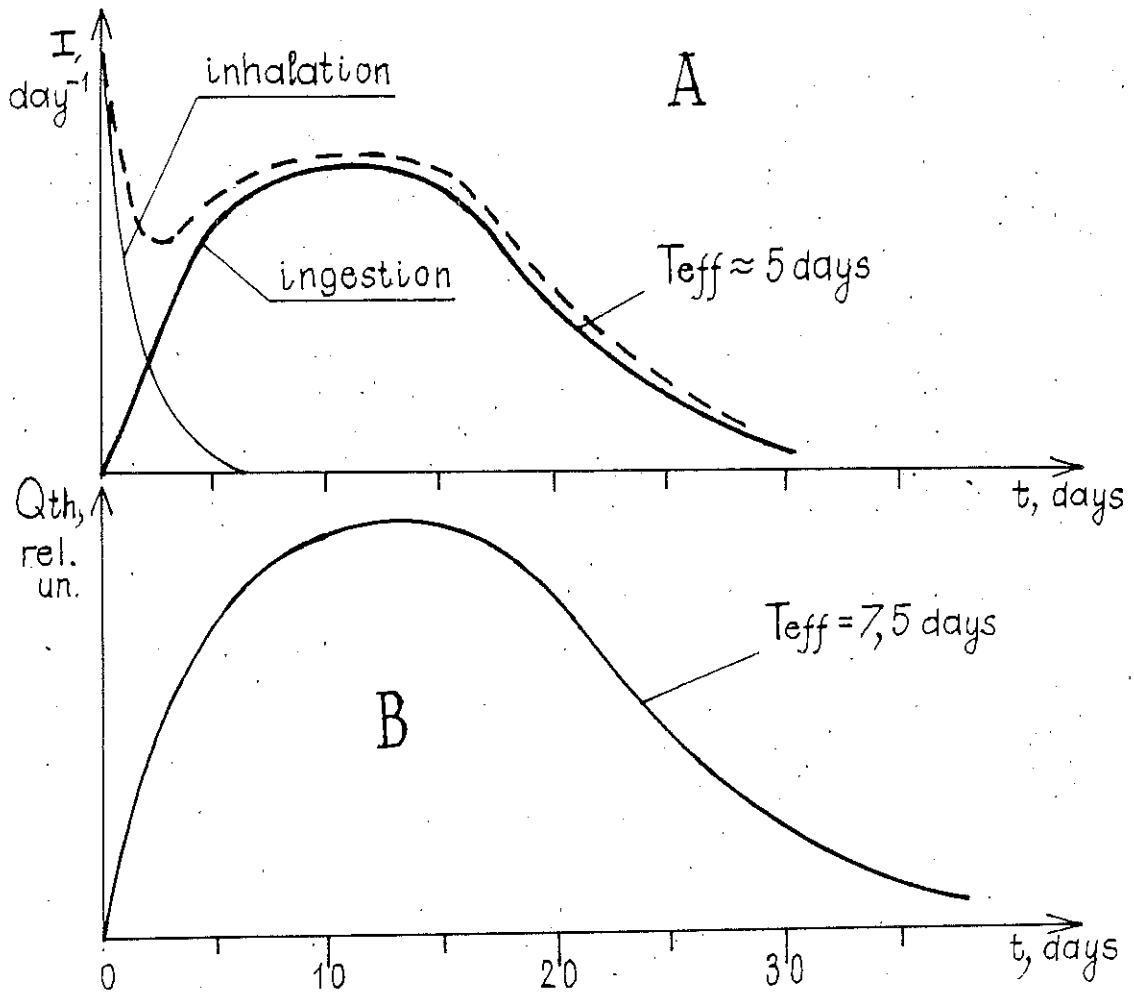


Fig.3: Caesium-137 content in local agricultural produce in Bryansk oblast normalized to soil contamination level  
(Bq/kg)/(TBq/km<sup>2</sup>) = 10<sup>-9</sup> km<sup>2</sup>/kg

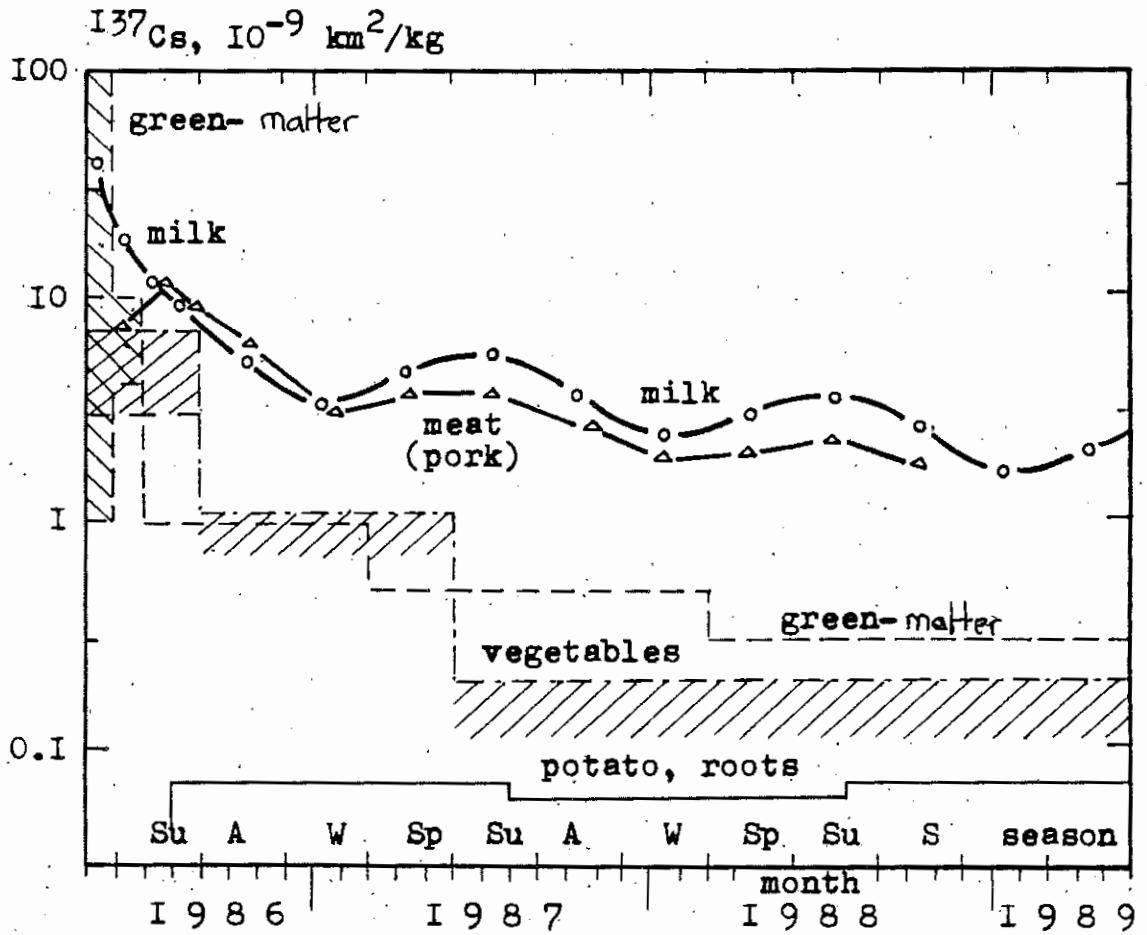


Fig.4: Daily intake of caesium-137 into the body of adult rural residents given usual (A) and protective (2) diet.  
Caesium-137 soil contamination level - 1 Tbq/km<sup>2</sup>

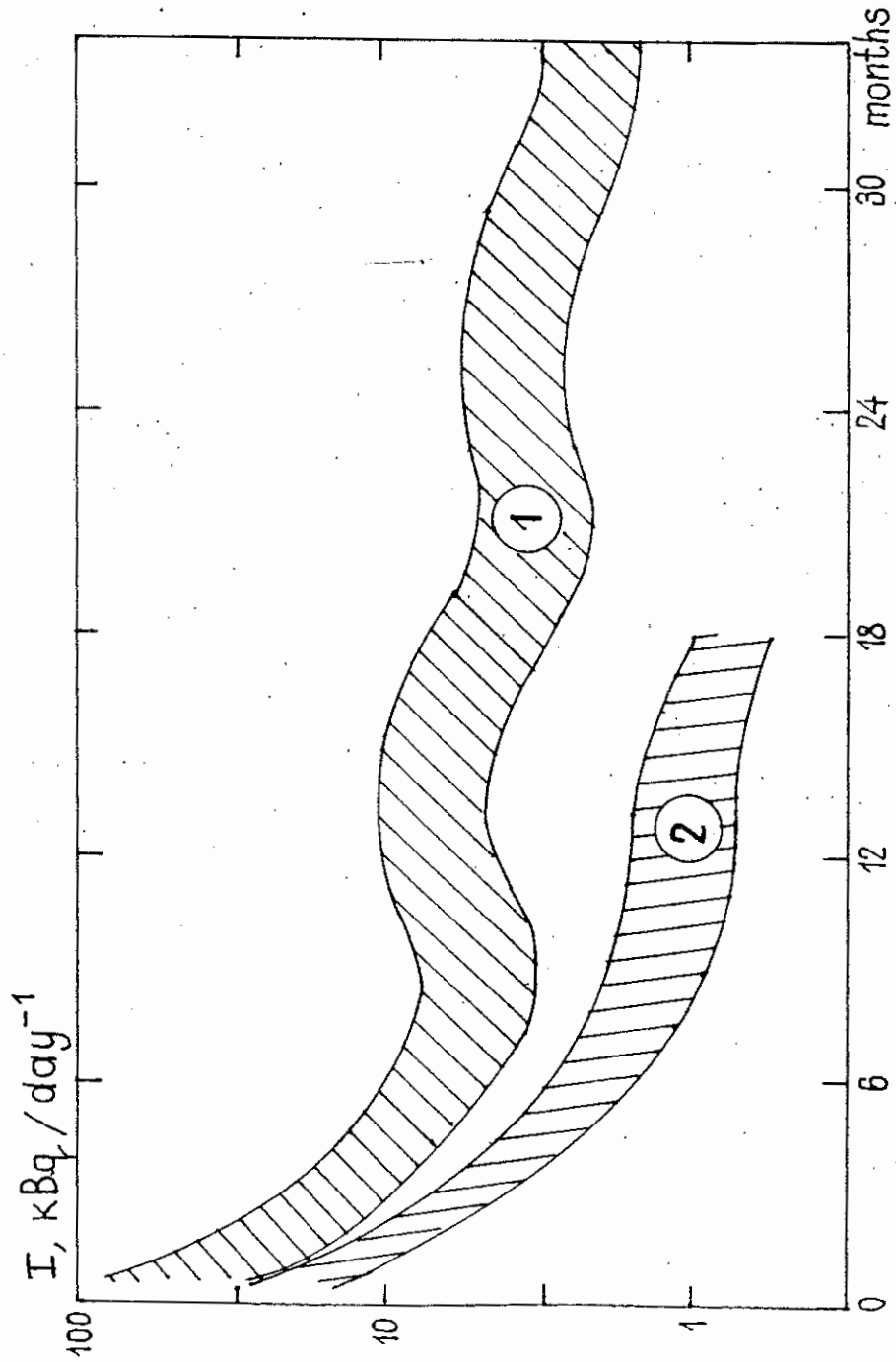


Fig.5: Mean caesium-137 content in the body of adult rural residents calculated for usual (1) and protective (2) diets, plus that actually measured in residents of three settlements in the Bryansk oblast (3).  
Caesium-137 soil contamination level - 1 Tbq/km<sup>2</sup>

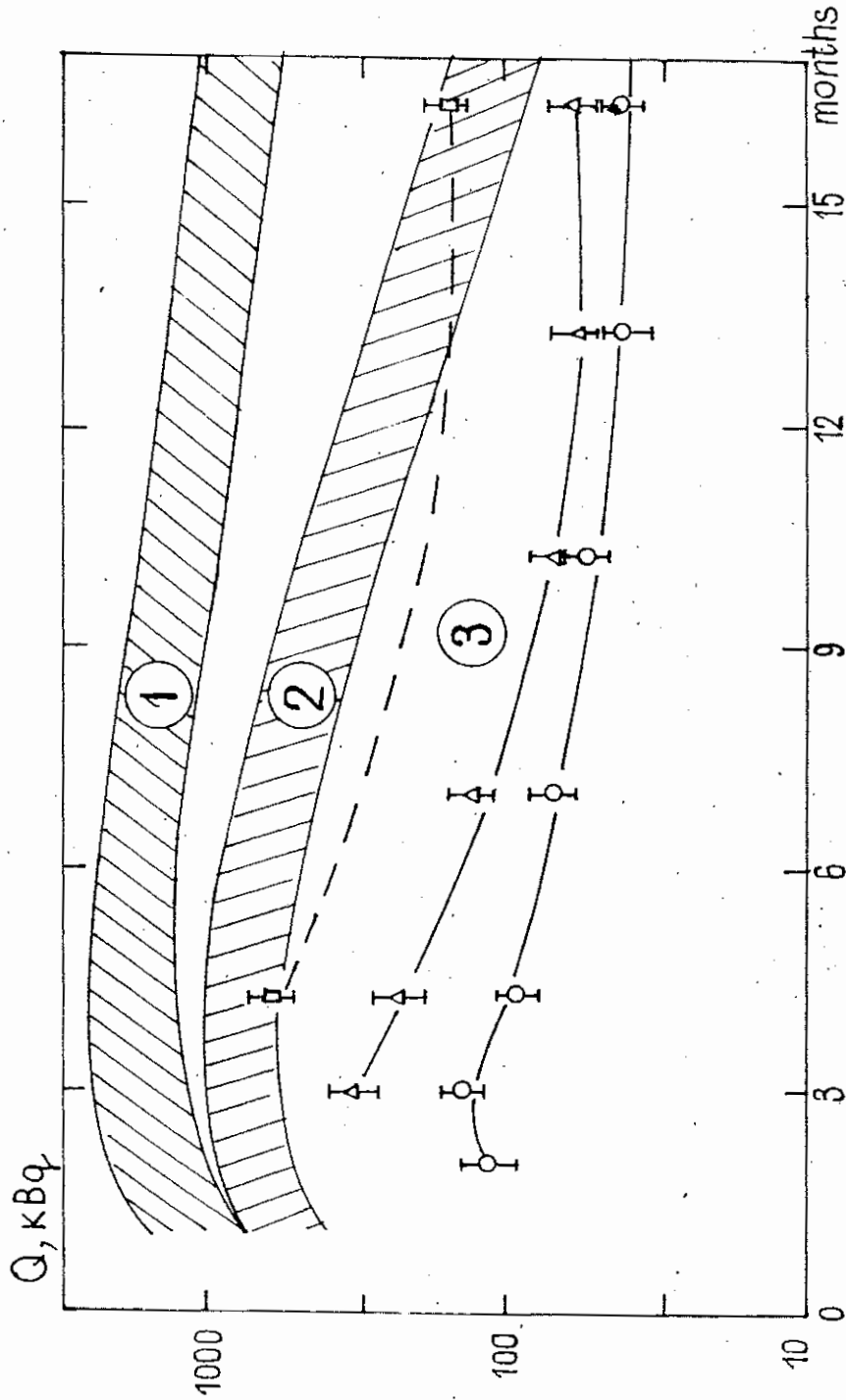


Fig.6: Dynamics of dose accumulation in adult rural residents after the Kyshtym accident.  
Strontium-90 soil contamination level - 1 Ci or 37 Gbq/km<sup>2</sup>

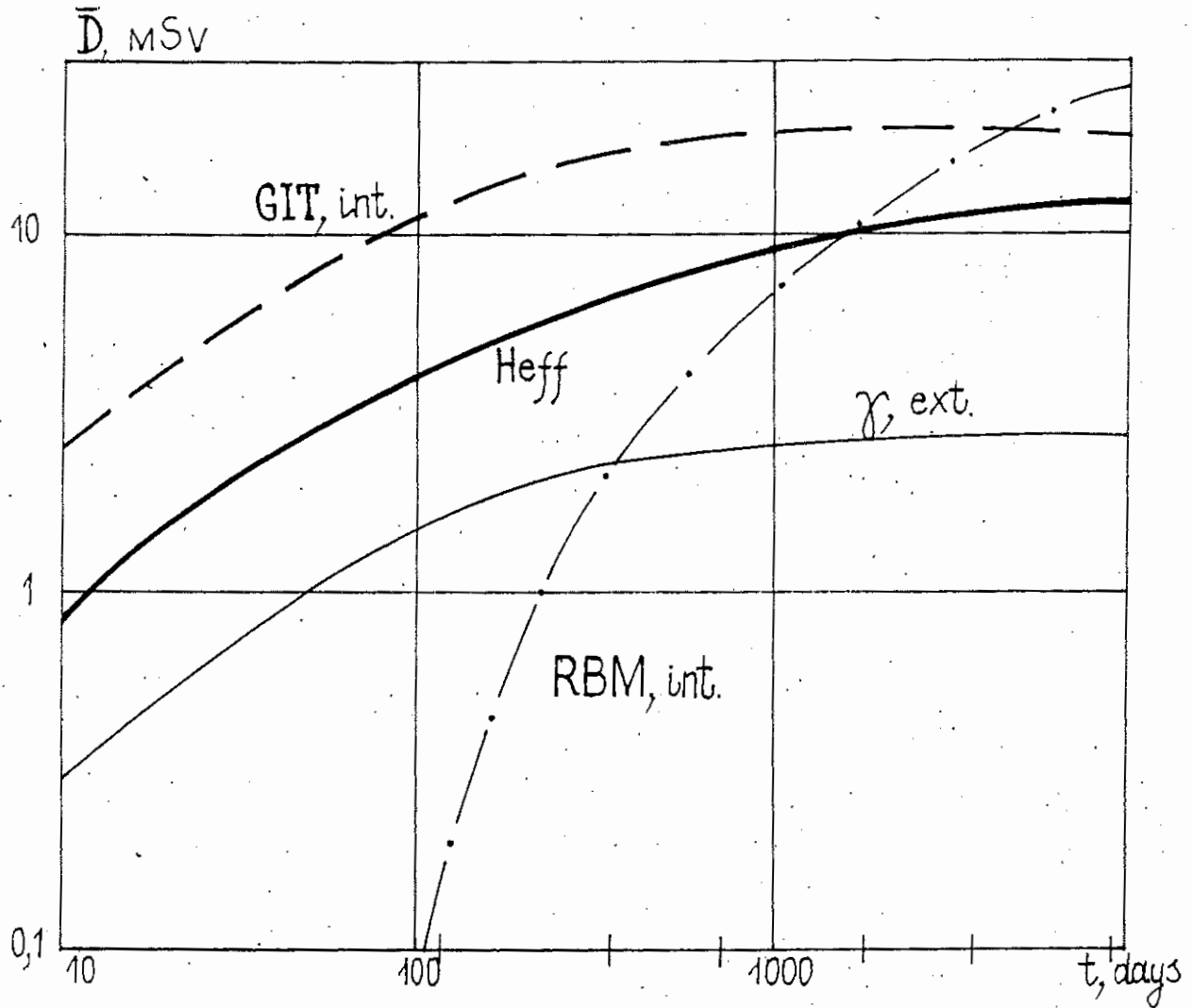
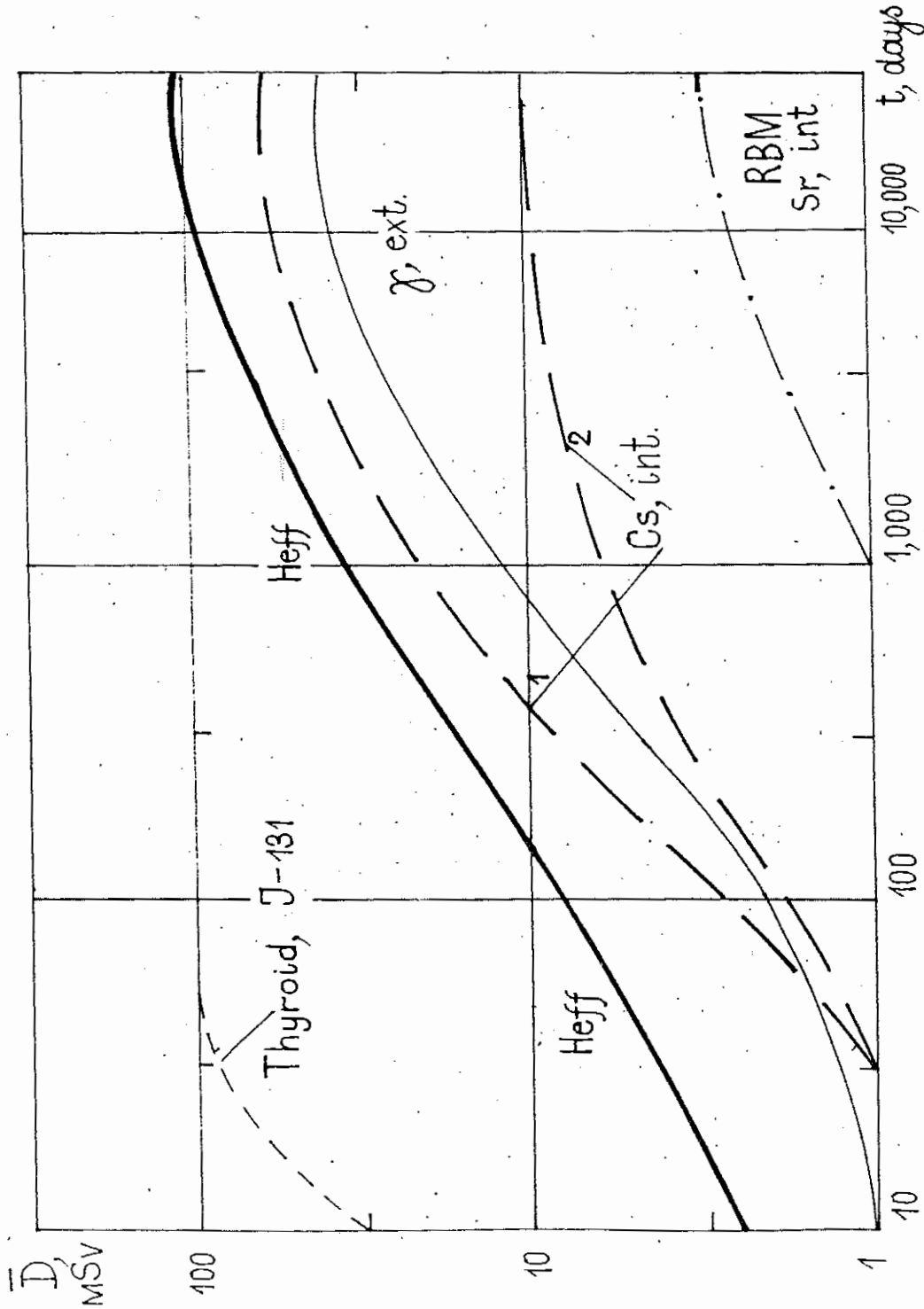


Fig.7: Dynamics of dose accumulation in adult rural residents after the Chernobyl accident.  
Caesium-137 soil contamination level - 10 Ci or 370 Gbq/km<sup>2</sup>







# **Medical Consequences of the Kyshtym Radiation Accident of 29 September 1957**

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## ABSTRACT

As a result of the accidental release of long-lived radionuclides, the gamma-radiation dose rate in the near zone of the trail reached tens of cGy per hour and, in a number of populated areas in the open countryside,  $0.1 \text{ cGy} \times \text{hour}^{-1}$ . The evacuation of 10 730 people reduced the possible radiation doses by 2-24 times.

Examination of people who had received the highest effective dose equivalents prior to evacuation (2.3-52 cSv) revealed, in the first two years, instability in leukocytes and thrombocytes (used as indicators), but this did not exceed normal fluctuations.

The structure of morbidity and mortality among the adult and child populations and the incidence of congenital pathology and infant mortality do not differ from the control. The proportion of families with children born of parents aged between 10 and 30 at the time of the accident does not differ from the same indicators for the whole of the USSR, and, in the case of those aged between 0 and 9 years at the time of the accident, this proportion is 5-10% lower than control values, although the number of people who married is considerably higher than in the control group. In addition, the standardized birthrate coefficients in the study group ( $31.8 \times 10^{-3}$ ) are considerably higher than in the control group ( $18.4 \times 10^{-3}$ ).

On 29 September 1957 a malfunction in the cooling system of a 300 m, concrete tank containing highly active nitrate-acetate waste caused the waste to explode, resulting in the release of radioactive fission products into the atmosphere and their subsequent dispersal and deposition on parts of the Chelyabinsk, Sverdlovsk and Tyumen oblasts (regions). Approximately  $2 \times 10^6$  Ci ( $7.4 \times 10^{16}$  Bq) was released into the atmosphere.<sup>3,4</sup> The composition of the mixture is shown in Table 1.

In the case of  $^{90}\text{Sr}$  contamination of  $0.1 \text{ Ci/km}^2$  (twice the level of global radioactive contamination), the maximum length of the radioactive trail was 300 km, while in the case of  $^{90}\text{Sr}$  contamination of  $2 \text{ Ci/km}^2$ , the length of the trail was 105 km (the width of the trail being 8-9 km).<sup>2,3</sup> The number of square kilometres with the various contamination levels is shown in Table 2.

The presence of gamma-emitting nuclides in the mixture resulted in external irradiation of the population and the environment. In the initial period the dose rate was approximately  $150 \mu\text{R}/\text{hour}$  for  $^{90}\text{Sr}$  contamination of  $1 \text{ Ci/km}^2$ , with maximum values of around  $0.6 \text{ R}/\text{hour}$  in the front part of the trail where the contamination level reached  $500 \text{ Ci/km}^2$  for  $^{90}\text{Sr}$ .

The following measures were taken to protect the population:

- evacuation (resettlement) of the population;
- decontamination of part of the agricultural area concerned;
- monitoring of radioactive contamination levels in agricultural produce and food supplies, and rejection of produce with levels exceeding the permissible limits;
- restricted use of the contaminated area;
- reorganization of agriculture and forestry with the creation of specialized State agricultural farms and forestry enterprises working to special recommendations.

The dynamics of the evacuation of the population and the doses received are shown in Tables 3 and 4.

Immediately after the accident (7-10 days) 1 054 people were evacuated from the nearest populated areas, while a further 9 000 people were evacuated in the following 1½ years. A total of 10 790 people were evacuated. The maximum mean irradiation doses received prior to evacuation were 17 rem for external irradiation and 52 rem for the effective dose equivalent (150 rem to the gastrointestinal tract). These doses may be doubled given the lack of uniformity in the contamination level and radiation conditions.

Mass screening of the local population was conducted for one year after the accident (involving paediatricians, therapists, neuropathologists, gynaecologists and otolaryngologists), and included an analysis of the morphological and biochemical composition of peripheral blood and a measurement of the person's height and weight.

TABLE 1

CHARACTERISTICS OF THE RADIONUCLIDE MIXTURE  
IN THE ACCIDENTAL RELEASE

Radionuclides	Contribution to the activity of the mixture, %	Radioactive half-life	Type of radiation	Nature of radiological hazard
<sup>89</sup> Sr	Traces	51 days		
<sup>90</sup> Sr+ <sup>90</sup> Y	5.4	28.6 years		Internal irradiation (skeleton)
<sup>95</sup> Zr+ <sup>95</sup> Nb	24.9	65 days		External irradiation
<sup>106</sup> Ru+ <sup>106</sup> Rh	3.7	1 year		External irradiation
<sup>137</sup> Cs	0.036	30 years		External and internal irradiation
<sup>144</sup> Ce+ <sup>144</sup> Pr	66	284 days		External irradiation
<sup>147</sup> Pm	Traces	2.6 years		
<sup>155</sup> Eu	Traces	5 years		
<sup>239</sup> Pu	Traces	-		

TABLE 2

Size of the areas affected by different contamination levels<sup>5</sup>

<sup>90</sup> Sr contamination level, Ci/km <sup>2</sup>	Size of area, km <sup>2</sup>
0.1-2	15 000-23 000
of which:	
2-20	600
20-100	280
100-1 000	100
1 000-4 000	17

TABLE 3

DYNAMICS OF EVACUATION OF THE POPULATION AND RADIATION DOSES RECEIVED BY THE POPULATION PRIOR TO EVACUATION<sup>3,4</sup>

Name of each group and number of people concerned (in thousands)	Mean contamination of the area, Ci <sup>90</sup> Sr / km <sup>2</sup>	Evacuation periods	Mean dose received prior to evacuation, cSv	
			External irradiation	Effective dose equivalent
A 1.054	500	7-10 days	17	52
B 0.28	65	250 days	14	44
C 2.0	18	250 days	3.9	12
D 4.2	8.9	330 days	1.9	5.6
E 3.1	3.3	670 days	0.68	2.3
Total: 10.73				

TABLE 4

MEAN RADIATION DOSES AND NUMBER OF PEOPLE IRRADIATED<sup>6</sup>

Group	Number of inhabitants	Duration of irradiation	Mean doses, cSv						
			External radiation	Internal radiation					Effective dose equivalent
				Gastro-intestinal tract	Lungs	Red bone marrow	Bone surfaces		
A	1 054	10 days	17	150	2.7	0.5	0.7	52	
1	10 270	30 years	0.4	2	0.2	3.8	5.2	2	
2	23 230	30 years	0.1	0.7	0.1	0.7	1.0	0.4	
CG	21 537	30 years	-	-	-	-	-	-	

The population which had received the highest level of radiation (group A) was relatively young: 45% between 0 and 17 years, 39% between 18 and 49 years and 16% over 50 years.

Clinical examination of the population did not reveal any cases of radiation sickness. In the initial study period some of those irradiated were found to have lower levels of leukocytes in their peripheral blood. However, the mean levels of thrombocytes ( $236-280 \times 10^9/l$ ), leukocytes ( $7.2-7.5 \times 10^9/l$ ) and neutrophils ( $4.1-4.7 \times 10^9/l$ ) in adults did not differ from the control indicators. The distribution function of these indicators in irradiated subjects in the initial examination period was standard with respect to median values, but nevertheless a considerable percentage displayed double-sigma deviation from the mean. Thus 17-19% of those examined had more than  $9 \times 10^9$  leukocytes per litre and 7-8% had more than  $350 \times 10^9$  thrombocytes per litre.

The reaction of the cardio-vascular system to exposure in the persons examined was assessed in terms of arterial pressure level and heart rate.

Results for 75% of the more or less healthy persons in group A show (Table 5) that there is no systematic increase in the deviation frequency of these indicators from standard distribution and radiation dose.

Of those examined 25% had general somatic diseases, more than half of whom (as can be seen in Table 6) had pathology of the cardio-vascular system and almost 30% a disease of the respiratory organs.

Thus, medical examination of the inhabitants of the populated areas in the front part of the trail did not reveal clinical symptoms of radiation pathology. It can be assumed that a limited deviation in the distribution of blood indicators is connected with a haematological reaction to irradiation observed in the early periods (leukopenia, relative lymphopenia and left shift in neutrophile count).

A considerable time after the accident medical examinations were carried out on people who possibly belonged to the critical group; these were people who received radiation while they were growing, when the radiation levels were at their highest (group A in Table 4).

One third of these people were more or less healthy. A thorough examination of the rest revealed areas of non-aggravated chronic infection (18% - chronic otitis, 13% - chronic tonsillitis, 16% - chronic gastritis and cervicitis). The incidence of osteochondrosis increased with age. Three people had epilepsy because of alcoholism and skull trauma. In comparison with the control group, no peculiarities were discovered in the morbidity of those who had been irradiated.

The peripheral blood indicators fell within generally accepted limits. The older the subject, the higher the frequency of dystrophic changes on the ECG (classes 4, 5 and 9, Minnesota Code). The frequency of class 0 ECGs (no changes) in irradiated subjects was no lower than in the control.



TABLE 5

FREQUENCY OF DEVIATIONS IN ARTERIAL PRESSURE AND  
HEART RATE IN GROUP A SUBJECTS

Indicator	Percentage of subjects with the given indicator
Tachycardia (heart rate of 90 per min.)	(5.5) 4-7.4
Bradycardia (heart rate of 60 per min.)	(8.5) 0-14.1
Arterial hypertension (arterial pressure of 160/96 mm Hg)	(3.3) 1.7-4.0
Marginal hypertension (arterial pressure of 140/90-159/94 mm Hg)	(10) 7.5-14.5
Hypotension (arterial pressure of 100/60 mm Hg)	(16.4) 10.8-24

TABLE 6

NATURE AND INCIDENCE OF DISEASES IN THE SUBJECTS EXAMINED

Class of diseases and nosological units	Percentage of people diagnosed as having this disease	
Parasitical, helminthiasis	0.6	
Gangliform goiter and thyrotoxicosis	0.5	
Mental disorders, neurasthenia	1.9	
<b>Diseases of the blood circulation system:</b>		
Rheumatic heart diseases	1.8	
Hypertonic disease	2.5	
Ischaemic disease	3.3	13.7
Coronary and cerebral arteriosclerosis	5.1	
Varicose veins	1.0	
<b>Diseases of the respiratory organs:</b>		
Acute nasopharyngitis	5.3	
Bronchitis	2.3	8.3
Emphysema	0.7	

The concentration of cholesterol (millimol/l) in the serum of irradiated subjects did not differ from the control, and amounted to  $4.78 \pm 0.1$  for those up to 29 years of age,  $5.25 \pm 0.06$  for those up to 39 years of age,  $5.41 \pm 0.06$  for those up to 49 years of age and  $5.69 \pm 0.06$  for those over 50 years of age.

Certain diseases considered risk factors for the development of oncopathology in irradiated subjects were no more common than in the control. Thus, chronic gastritis, endocervicitis and erosion of the neck of the uterus were detected in 2.3, 11.1 and 11.1% of 28-year-old subjects respectively, and in 9, 20 and 0% of 50-year-old subjects respectively.

Early child mortality and abnormalities in intra-uterine development are two of the most sensitive criteria of damage due to ionizing radiation. 35 cases of death from congenital abnormalities were recorded in descendants of the population in the radioactive trail area over a period of 35 years. In group 1 there were 10 cases among 10 270 people exposed to  $^{90}\text{Sr}$  levels of 1-2 Ci/km<sup>2</sup>, and in group 2 there were 25 cases among 23 230 people exposed to  $^{90}\text{Sr}$  levels of 0.1-1 Ci/km<sup>2</sup>. Among the 21 537 people in the control group (CG) there were 39 deaths (<0.1 Ci/km<sup>2</sup>  $^{90}\text{Sr}$ ). Mortality from development defects accounted for 0.36-0.67% of overall mortality (see Table 7).

As Table 7 shows, the differences in the groups are statistically uncertain. Nor were there any differences in the first two years after the accident.

Highly specific data were obtained from the analysis of early child mortality in the initial years after the accident (Table 8).

Table 8 shows that there is no fundamental difference between the level of early child mortality in the three groups even though total mortality was high in those years. In addition, early child mortality is not linked to the levels of radiation exposure but apparently stems from the differences in medical care provided to infants.

In order to determine the long-term consequences of the irradiation of the population, the irradiated and control groups were studied at the same time. Table 9 shows the results of radiological exposure in the most highly irradiated group.

In the most highly irradiated group, intensive mortality indicators in groups A, 1, 2 and CG were present in 272, 2 760, 6 578 and 5 873 cases respectively, and the corresponding mortality coefficients were 9.5, 11.5, 11.0 and 10.9 (all  $\times 10^{-3}$ ) respectively. There is clearly no difference between the irradiated groups and the control.

In addition, up to the age of 4 and over the age of 60 the age indicators of mortality deviate significantly from the control. However, it is not possible to establish a link between this and the radiation dose received. Thus, in groups A, 1, 2 and CG the mortality coefficients up to the age of 1 year were 91, 32, 63 and 52 respectively, between the ages of 1 and 4 years they were 13.7, 1.7, 5.0 and 3.3 respectively, and over the age of 60 years they were 39.2, 50.4, 43.1 and 46.9 respectively. For all other ages there was no difference between the groups and the control regarding mortality indicators and mortality coefficient.

Table 7

INDICATORS OF MORTALITY DUE TO CONGENITAL DEVELOPMENT ABNORMALITIES

Population group	Extensive indicators	Intensive indicators over 30 years	
		10 <sup>5</sup> population	10 <sup>-3</sup> infants
1 - 10 270 people	0.36	4.2	1.02
2 - 23 230 people	0.38	4.2	1.93
CG - 21 537 people	0.67	7.4	2.66
Chelyabinsk oblast			
1965	0.53	3.6	2.3
1986	0.23	2.2	1.3

Table 8

CHILD MORTALITY UP TO THE AGE OF 1 YEAR FROM 1957 TO 1961  
(PER 1 000 INFANTS)

Cause	Trail zone	Control No. 1 on the boundary of the trail	Control No. 2 at a considerable distance from the trail boundary
All causes	27.7	31.4	38.6
Dietary disorder	15.2±2.8	12.3±3	5±1
Pneumonia	1.7±1.0	3.1±1.5	16.1±1.8
Infections	1.6±0.9	2.3±1.3	3.0±0.8
Infant diseases	8.7±2.2	13.8±3.2	14.5±1.7

It should be noted that, for the 272 deaths in group A, oncological diseases were the third, rather than the second, most frequent cause after cardio-vascular diseases and trauma/accidents. The fact that mortality due to infectious diseases was more common than mortality due to diseases of the respiratory organs is another particular feature.

The analysis of mortality due to malignant tumours is of prime interest, in that over a longer period it is the main symptom of previous irradiation. The most certain mortality indicators were observed in inhabitants of group 1 (see Table 10). However, the sample was not large enough to allow us to confirm that there is a significant difference between the values observed.

Of all the neoplasms detected throughout the observation period, one of the most frequent is cancer of the digestive organs and, in particular, of the oesophagus (see Table 11). Apparently there is a trend towards an increase in the incidence of oesophagus cancer in the population group which received the highest dose (group A) although this increase is not statistically certain.

The fatalities due to neoplasms of the lymphatic and blood-forming tissue should be noted. The mortality coefficient in dose group A was  $13.2 \times 10^{-5}$ , in comparison with  $4.7 \times 10^{-5}$  in the other groups. Although the differences are uncertain (given that they are based on three deaths) the reader should note that the effective dose equivalent in this group was 52 cSv, which is close to the critical dose for the induction of leukoses.

Table 12 shows the level of mortality due to malignant diseases by decade for the zone of influence of the trail, compared with intensive indicators for neighbouring oblasts (No. 1 - Chelyabinsk and No. 2 - Sverdlovsk). The table shows that there is an increase in mortality with each successive decade. In all, mortality increased from 145.8 to  $160.7 \pm 25$  per  $10^5$  people in the zone of influence of the radioactive trail and where nuclear industry enterprises are situated, and to  $167.6 \pm 3.2$  and  $159.4 \pm 6.6$  per  $10^5$  people in oblasts 1 and 2 respectively. Mortality increased to only  $105 \pm 12.7$  in the closest populated area, even though it received a higher dose of radiation. The lower figure is merely a result of the younger age of the population in this area.

Analysis of the causes of morbidity due to malignant neoplasms connected with the accident allowed us to classify the incidence of initially diagnosed tumours in relation to several external exposure factors. Chelyabinsk oblast morbidity data revealed a) no connection between increased morbidity and a rise in radiation dose; b) an overall correlation between the incidence of diseases and releases of  $SO_2$  into the atmosphere. Although  $SO_2$  is not a carcinogen, it is highly suitable as an indicator of general chemical pollution. Actual data show that the annual morbidity level per  $10^5$  people is 225 if no  $SO_2$  is released, whereas it is 250, 275 and 300 for releases of 50, 100 and 150 thousand tonnes of  $SO_2$  per annum. Thus, the incidence of mortality due to malignant diseases on a map of the Chelyabinsk oblast correlates not with the trail of radioactive contamination, but with the location of metallurgical and chemical industry plants.

Considerable attention was paid to the state of the reproductive function in the irradiated subjects of various ages. Table 13 shows that it is not possible to detect systematic deviations of this highly important demographic indicator in subjects who received the highest

Table 9

MORTALITY INDICATORS

	Size of population	Total deaths	x 10 <sup>-3</sup>			
			Total per year	Up to the age of 1 year	between 1 & 4	Over 60
A	1 054	272	9.5	91	13.7	39.2
1	10 270	2 760	11.5	32	1.7	50.4
2	23 200	6 578	11.0	63	5.0	43.1
CG	21 537	5 873	10.9	52	3.3	46.9

Table 10

Extensive (%) and intensive (x 10<sup>-5</sup>) indicators of mortality due to malignant tumours over 30 years

Population group	Number of cases	%	x 10 <sup>-5</sup>	Confidence intervals, 95%
A	25	11.7	115.9	75-165
1	376	13.6	157.4	142-174
2	775	11.8	129.2	120-139
CG	707	12.0	131.9	122-142

Table 11

Structure of mortality due to malignant neoplasms  
(x 10<sup>-5</sup>)

Main locations	International nomenclature code	Irradiated subjects			CG
		A	1	2	
Oesophagus	150	26.5	8.2	11.3	12.1
Stomach	151	35.3	45.1	32.4	44.3
Other digestive organs	152-159	8.8	30.7	20.9	22.4
Respiratory organs	160-163	17.7	29.5	24.9	26.4
Bones	170	0	3.1	0.9	2.4
Skin, oral cavity	140-147 172-173	0	7.5	1.4	4.5
Mammary gland	174	4.4	4.4	2.1	4.2
Body and neck of the uterus	180-182	0	13.1	9.6	10.8
Other urino-genital organs	183-189	4.6	9.4	6.6	7.6
Lymphatic and blood-forming tissue	200-209	13.2	5.0	5.2	4.7

dose of radiation. However, a lower rate of marriage up to the age of 27 (and a corresponding lower number of children in such families) was detected in those people who were infants at the time of the accident. In the case of the older subjects, however, the marriage rate was higher than in the control group, and the number of children either did not differ from that of the control group or was slightly lower (persons up to 9 years of age at the time of the accident). In addition, as Table 14 shows, the dynamics of the birthrate coefficients over time per thousand population are higher than for the oblast as a whole.

It would seem that the living conditions of, and social factors relating to, the population evacuated earlier are somewhat better than for the rest of the rural population in the region. It may be that some other factors, such as ethnic factors, play a role here.

In conclusion, it should be emphasized that our study of the state of health, morbidity and mortality of the population exposed to radiation from the accident (effective dose equivalents of between 1 and 52 cSv, irradiation of individual organs up to 150 cSv) did not reveal any deviations with respect to the same indicators for non-irradiated subjects.



Table 12

Level of mortality due to malignant neoplasms

	For the zone of influence as a whole	In the nearest populated area*	In Chelyabinsk oblast	In Sverdlovsk oblast
1970-1980	145.8	-	146.6	-
1980-1987	160.7 ± 2.5	105 ± 12.7	167.6 ± 3.2	159.4 ± 6.6

\* See Table 8 for explanations

Table 13

Percentage of marriages and births for irradiated parents

Groups	Age at the time of the accident	Number of subjects	Percentage of people who married	Percentage of people who had children
Infants	up to 1 year old	56	91(82-97)	84(73-92)*
Children	1-9 years	295	93(89-96)*	90(86-93)*
Adolescents	10-19 years	203	93(89-96)*	93(89-96)*
Adults	20-29 years	201	95(92-98)*	91(87-94)*
Adults	30-59 years	308	98(96-99)*	98(96-99)*
Control, USSR			81.9-82.6	94.6

\* differences are certain, in comparison with the control

Table 14

Dynamics of birthrate coefficients among the evacuated population. (x 10<sup>-3</sup>)

Years after the accident	1	5	10	15	20	25	30	1-30
Number of children	51	271	491	717	960	1 242	1 586	1 616
Birthrate coefficient	37.4	42.2	30.2	27.5	26.4	27.8	30.0	31.8
Standardized coefficients	40.4	48.7	31.3	25.9	24.8	25.2	25.6	31.8
Birthrate coefficients for the Chelyabinsk oblast	24.1	20.8	14.8	16.0	16.7	19.8	16.7	18.4

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# **The Results of Medical Research in the 30 km Zone of the Chernobyl NPP**

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The Chernobyl NPP accident is regarded as a global disaster by its economic and social damage as well as medical and ecologic consequences.

As is known, during the first three post-accident months the main biologically active dose-contributing radionuclides were the iodine isotopes. The rate of human and animals' thyroid exposure is attributed to these isotopes. The most significant I-131 contamination was detected on vast areas (dozens of thousand square km) in 39 regions of the RSS, UkrSSR and BSSR. This was the reason why effective radimetric and dosimetric surface contamination pattern determination was so difficult at that time.

Later the main dose-contributing agents were the long-lived nuclides.

The activities on the elimination of the Chernobyl NPP accident aftereffects carried out in a series of sequential stages served both for decision of immediate operational tasks and for forming a basis for subsequent activities. Medical and biological research in the alienation area were started from the first steps of the accident aftereffects elimination. The most urgent tasks were :

- acute atomic disease treatment among the operational personnel and firemen;
- general medical examination of the population in the accident area;
- working out of medical recommendations for the evacuees from the contaminated regions;
- detailed study of radioactive fall-out composition and structure;
- study of the degree of radiological contamination of air, water, soil and vegetation;
- study of radionuclide migration paths in soil, water and vegetation;
- study of the main radionuclide accumulation in food;
- study of the immediate radiation effects on flora and fauna and working out of remote aftereffect predictive estimates.

Medical research during the first two years were focussed mainly on population health in the strict control areas. The results of these activities are summarized in the documents of the scientific conference "Medical aspects of the Chernobyl NPP accident"; the results show the absence of any significant changes in health of the "accident" regions population due to the effect of radiation as compared to the control groups (Refs.1,2).

The conference recommended "to pay special attention to working-out and refining the medical and biological concept of long-term living of the population (commensurable with man's lifetime) on contaminated territories without disturbing traditional tenor of life and activities (Ref.3).

Concurrently, activities on medical examination of personnel working on elimination of post-accident consequences in the 30-km area were carried out. this professional group was called afterwards "the

eliminators". Work on the eliminators' health assessment are carried out under the guidance of the Institute of Biophysics (the USSR Ministry of Health) and the all-Union scientific Center of Radiological Medicine (the USSR Academy of Medical Sciences).

The main results of the 4-year work carried out by the Institute of Biophysics and the Scientific Center of Radiological Medicine show the changes in health of the post-accident effects eliminators. The multifactorial pattern of combined effect leads to stress and over-stress of the adaptation mechanisms which results in creation of preconditions for acceleration of the aging process, and increases risk of central nervous, cardiovascular, alimentary and musculoskeletal system diseases. There are direct results showing increased numbers of alimentary and musculoskeletal system diseased among the post-accident effect elimination personnel, among persons older than 40 years in particular.

Remote aftereffects of small ionizing radiation doses for personnel working in the 30-km zone will, evidently, not lead to an increase of oncogenic diseases but to health damage resulting in a decreased capacity to work and shortening of lifetime (Refs. 4,5).

Of course all the research work is based and shall be based on the data collected by Soviet and foreign radiobiologists during the nuclear weapon tests in the years 1940-50 as well as during the nuclear accidents aftereffects elimination in the Southern Urals, Windscale and Three Mile Island NPP.

At the same time direct application of radiobiological findings at the Chernobyl NPP conditions is not possible due to some specific features:

- . first, the environmental contamination has acquired a highly heterogenous character by composition and concentration of the most dangerous radionuclides;
- . second, a wide range of physico-chemical radioactive fall-out properties (dispersivity, solubility, etc.);
- . third, the accident region displays the unconventional geological, mineralogical and meteorological conditions affecting radionuclide migration through the environment.

The complexity of the Chernobyl NPP accident is attributed to the physico-chemical properties of the radionuclides in the fall-out, especially, soluble forms of transuranium elements, which complicates the prediction of its behaviour pattern in the environment. This is the reason why the radiological experience of the Chelyabinsk accident cannot be used in Chernobyl.

To our opinion the Chernobyl NPP 30-km zone is not only a unique testing ground, but a potential source of radiation hazard - there are 4 million m<sup>3</sup> of radioactive waste at 800 sites of temporal radioactive waste immobilization in the 30-km zone; besides, according to our assessments, the total radionuclides stock in the 10-km zone comprises 110.000 Ci of Caesium-137, 100.000 Ci of Strontium-90 and 800 Ci of Plutonium-239, 240.

It is considered expedient to continue work on the assessment of radioactive contamination on environmental objects :

- development and improvement of a system of radiation monitoring of environmental objects in the 30-km zone - air, water, soil, flora, fauna;
- study of morphology and radionuclide composition of fall-out; hot particles included;
- working-out of an integrated system to reduce the exposure doses for the personnel taking part in post-accident effects elimination;
- assessment of the effectiveness of protection means for personnel working for post-accident effects elimination;
- prediction of remote radioactive aftereffect in Polesye biocenosis.

To our mind the top priority trends in medical research during the nearest years should be as follows :

- sanitary assessment of the effectiveness of protective measures for personnel working for post-accident effects elimination, especially on the territories where the radioactivity level exceeds 200 Ci/km<sup>2</sup>;
- improvement of individual radiation monitoring means, creation of new dose-meters, and of an integrated system and data bank of individual radiation monitoring for eliminators;
- health examination of personnel working in the 30-km zone, determination of critical groups, thorough medical examination using immunological, endocrinological, cytogenic and biochemical methods as well as assessment of the risk of possible plutonium penetration;
- working-out of the eliminators rehabilitation methods;
- creation of therapeutic and preventive diets as well as food products with addition of radioprotective agents and absorbents; possible inclusion of transuranium elements should be considered;
- creation of automatic informational system for the eliminators health examination.

It is considered expedient to create a rehabilitation centre with the aim to keep steady capacity for work and emotional comfort of personnel working in 30-km zone.

The main goal of integrated programmes should be the creation of scientifically grounded and practical recommendation for the protection of personnel, population, flora and fauna from the ionizing irradiation effects, full-scale testing of new methods, technologies and means for post-accident effects elimination and area decontamination as well as practical demonstration of the ways to return contaminated areas to normal exploitation.

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**The Radioiodine Problem  
Following the Chernobyl  
Accident: Ecology, Dosimetry  
and Medical Effects**

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## **ABSTRACT**

Following the Chernobyl accident radioisotopes of iodine constituted the main dose-forming factor among the people who stayed on in the radioactively contaminated areas, and in a number of places the effective doses to the thyroid gland were up to two orders of magnitude higher than the whole-body dose stemming from uniform internal and external irradiation. We consider the mechanisms involved in the radiiodine contribution to the doses in the human organism, depending on intake path, life style and social and ecological factors. We illustrate, by means of examples, thyroid gland dose distribution for various age groups in the population, and discuss the medical effects and predict the long-term risks for the population of exposure to radioisotopes of iodine.

Chernobyl showed once again that when a nuclear reactor accident occurs radioisotopes of iodine pose the greatest hazard for man as regards contamination scale, biological availability and size of dose formed. It is estimated that 600-1 300 PBq <sup>1,2</sup> of <sup>131</sup>I - the biologically most significant isotope - were released into the atmosphere as a result of the accident, together with a large amount of short-lived isotopes (<sup>132</sup>I, <sup>133</sup>I, <sup>135</sup>I) and their precursors, including an amount of <sup>132</sup>Te put at 400 PBq.<sup>1</sup>

The complex meteorological conditions - including strong winds which changed direction, turbulence and precipitation - caused the radioactivity to travel great distances and led to the formation of "patches" of radioactive contamination in distant and unexpected places. Nine oblasts (regions) of the Ukraine, Byelorussia and the RSFSR, accounting for a population of some 1.5 million people, were the worst contaminated with radioisotopes of iodine.

Radioiodine entered people's bodies through inhalation in the first few days after the accident, and then via food (mainly milk) in the subsequent weeks. The health and epidemiological services monitored contaminated food products for total beta-activity from early May, when <sup>131</sup>I accounted for some 80-90% of the radionuclide concentration in milk and dairy products. According to the T. Nedvetskayte group from Vilnius, <sup>132</sup>I was not discovered in milk during this time, even when <sup>132</sup>I and <sup>132</sup>Te were recorded in the air.

Fig. 1 shows measurements of radionuclide concentration by total beta-activity in milk in May 1986 in the Bolkhov rayon (district) in Orlov oblast, where low-level activity fallout occurred on 28 and 29 April, mainly in precipitation. Radiochemical analysis of the samples showed that up to about the final week of May no less than 80% of the activity was due to <sup>131</sup>I. The radionuclide concentration in milk remained more or less constant up to 15-20 May and then decreased with an effective period of 5 days.

Intake of <sup>131</sup>I by the public in the areas contaminated after the Chernobyl accident is estimated fairly well by the model of uniform intake of radioiodine over 15 days, with subsequent exponential reduction of the daily activity intake (Fig. 2). For mass calculations we used a simplified model of uniform intake over 20 days whose estimates, in numerical terms, are more or less the same as those calculated using the earlier model.

The dose absorbed by the thyroid gland (TG) was calculated from measurements of <sup>131</sup>I content therein, taking into account age-related parameters for iodine exchange (Table 1) and the length of time people had spent in the contaminated zone.<sup>3</sup>

The contribution of the inhalation and peroral radionuclide intake pathways to formation of TG dose varies according to the amount of time people spent in the contamination zone and the protective measures taken (limiting milk consumption, iodine prophylaxis, etc.). For example, the inhabitants of the town of Pripyat, evacuated on 27 April 1986, received their internal irradiation dose via inhalation only, and their thyroid glands were found to contain <sup>132</sup>I and <sup>135</sup>I, in addition to <sup>131</sup>I. But in people who remained in the contaminated area, inhalation accounted for less than 10% of the TG dose.

In May and June 1986 TG radiometry of the population was carried out in the oblast hospitals' radiodiagnostic laboratories and oncological clinics, as was rapid mass testing using the non-specific radiometers SRP-68, DP-5 and DRG-03. The measurement methods included prior  $^{131}\text{I}$  calibration of the instruments, measurement of background radiation, TG, and thighs or another part of the body in order to take into account extrathyroid exposure to  $^{134,137}\text{Cs}$ , which are uniformly distributed in the human body. The lack of one of these measurements, the presence of surface contamination or the absence of information about the movement of people out of the contaminated area increased the error in dose determination, sometimes making it impossible. All the measurements made in May and June 1986 were subsequently examined by experts to assess their reliability.

The method used to determine TG dose was the same for all the contaminated areas in the Soviet Union, with some insignificant differences in the computer processing methods used.

The largest TG irradiation doses were received by people living in the Khoyniki rayon of Gomel oblast, Byelorussia. In some settlements where the inhabitants had been evacuated by 5 May 1986 mean doses in children of pre-school age were up to 4 Gy, with some individuals receiving doses in the order of 30-40 Gy. Lack of evacuation might have increased these doses two- to threefold. In the unevacuated settlements in the Gomel, Bryansk and Kiev oblasts the mean doses among children in the youngest age group were as high as 150-200 rem. From early May, in all the worst-contaminated areas locally produced milk was banned, establishments for children were supplied with dried milk or milk imported from clean areas, and iodine prophylaxis measures were carried out. It should be noted that, apart from the town of Prip'yat and populated areas in the 30-km zone, these iodine prophylaxis measures were generally carried out with some delay in rural areas - no earlier than 10 May - which reduced their effectiveness, of course.

All the protective measures taken reduced the TG irradiation dose by two- to fivefold on average in comparison with the maximum possible (depending upon contamination level). However, in each area there were individuals who received higher doses, i.e. 3-6 times higher than the mean values.

In the analysis of the distribution of the doses received, the populated areas were divided into urban and rural ones and their inhabitants into six age groups: up to one year, 1-2, 3-6, 7-11, 12-16 and over 16. Dose distribution within each age group was mostly close to the lognormal, but approximated to the normal in some cases, due mainly to consumption of more homogeneous - in terms of radioactive contamination - milk products from milk-processing plants. The ratio of the maximum doses to the mean was 3-6.

The correlations between mean doses in urban and rural inhabitants of various ages are given in Fig. 3. Given identical levels of contamination of the surroundings, the mean doses absorbed by adults in settlements were approximately 2-3 times higher than those absorbed by town dwellers. The largest doses in towns were received by infants. In rural areas the largest doses were observed in children aged from 1 to 3, who had the highest proportion of dairy products in their diet. This dose correlation reflects not only specific age-based metabolic features in dose formation, but also different eating habits and the observed difference between radioiodine concentrations in centrally supplied milk (a blend from various farms) and that from private holdings.

Using the weighted mean values for the dose ratios in the various age groups it is possible to estimate, for each populated area, the mean dose received by inhabitants of all ages once it has been determined for one population age group in the specific town or settlement. By taking into account the age structure of the population in the RSFSR it is easy to calculate that the mean TG dose to the total urban population is twice the dose for adults ( $D = 2 \cdot D(<16)$ ), and 1.5 times higher in settlements. In both categories of population the mean dose is three times lower than that in children from 1 to 6 years of age, whose diet contains more milk:  $\bar{D} = 0.3 \cdot D(1-6)$ .

Given the massive scale of the accident, the inadequate measuring equipment and organizational difficulties, it was not possible to carry out TG radiometry of all inhabitants in the contaminated areas. Methods were devised of reconstructing the mean and individual TG irradiation doses from the obtained thyroid dose correlations with the concentration of  $^{131}\text{I}$  in milk from 10 to 15 May 1986 (Fig. 4) and with the level of background gamma radiation on dairy cattle pasture near a given populated area (Fig. 5). Both correlations were derived from studies in the Tulska, Bryansk and Orlov oblasts of the RSFSR and were statistically certain. In populated areas where consumption of local milk was not restricted and no iodine prophylaxis measures were introduced until mid-May, the mean TG dose in children from 1 to 6 years was calculated as follows:

$$\begin{aligned} \bar{D}(1-6) &= 1.5 \cdot \bar{C}_m(10-15.05), \text{ Gy} & (1) \\ \bar{D}(1-6) &= 0.5 \cdot \bar{P}_\gamma(10-12.05), \text{ Gy} & (2), \end{aligned}$$

where  $\bar{C}_m(10-15.05) \mu\text{Ci/l}$  is the mean concentration of iodine-131 in milk from 10-15 May, and  $\bar{P}_\gamma(10-12.05) \text{ mR/h}$  is the mean level of background gamma radiation near a populated area from 10 to 12 May. For populated areas where protective measures were taken in good time, a corrective coefficient has to be added to equations (1) and (2) to reflect the measures taken in each populated area.

Individual TG doses were established on the basis of the mean-age dose values in a populated area, taking into account the correlation of the doses obtained by measuring  $^{131}\text{I}$  in the TG with the mean daily consumption of milk (in volume terms) in May 1986 or the content of  $^{134,137}\text{Cs}$  in the human body in the summer of 1986. G. Ya. Bruk and N. F. Korelina analysed the TG radiometry measurements of May 1986, the whole-body radiocaesium content measurements of August-September 1986 and the findings of a survey of the way in which over 300 Bryansk oblast inhabitants had lived during this period. On the basis of these data we obtained the following correlations:

$$D_i = \bar{D} \left( a + b \cdot \frac{V_i}{\bar{V}} \cdot \frac{T_i}{\bar{T}} \right) \quad (3)$$

where  $D_i, \bar{D}$  are the individual and mean-age dose values;  
 $V_i, \bar{V}$  are the levels of individual and mean milk consumption in a populated area;  
 $T_i, \bar{T}$  are the individual and mean dates on which consumption of local milk was discontinued.

The coefficient "b" reflects the amount of radioiodine ingested with milk, while "a" represents all other intake pathways (inhalation, with vegetables, etc.). In the western areas of Bryansk oblast no less than 80% of the activity was ingested with milk, the correlation coefficient for the various age groups being between 0.6 and 0.8.

The link between TG dose and caesium content in the body in later measurement periods was statistically certain ( $r = 0.7-0.9$ ) for individuals over 7 years of age:

$$D_i = K(T) \cdot A_{Cs} \quad (4)$$

where  $K(T)$  is the coefficient dependent upon age  $T$ .

The dose estimates reconstructed by both methods have a degree of error in the range of  $1/3 D_i - 3 D_i$ . However, for the purposes of pinpointing groups for medical observation even this level of accuracy helps reduce the uncertainty of dose characteristics as compared to the use of mean indicators.

We evaluated separately the radiation doses to new-born children, irradiated within the womb, and breast-fed children who ingested radioiodine via their mother's milk. By measuring  $^{131}I$  content in the thyroid gland of mothers and of their breast-fed children we established that in May 1986  $^{131}I$  content in the thyroid gland of infants was four times lower, but the absorbed dose three times higher, than in their mothers. This dose correlation was used to evaluate the dose in the thyroid gland of a breast-fed child on the basis of that in its mother.

To estimate the irradiation dose to fetuses we used the intrauterine irradiation model for single  $^{131}I$  intake from the work of J. R. Johnson.<sup>5</sup> Applying this to prolonged intake we obtained correlations between the doses to the foetus and the mother, which rose from 0 in the 11th week of pregnancy to 3 by the 24th-26th week, decreased to 1 by the 35th-37th week, and then increased in the subsequent weeks to 2.5 on account of  $^{131}I$  intake with mother's milk after the child was born.

By using all the methods for estimating individual and mean doses we estimated the collective dose for thyroid gland irradiation of the population in the worst-contaminated oblasts of the RSFSR, the Ukraine and Byelorussia (Table 2). In the nine worst-contaminated oblasts (with a total population of 15.6 million people) the collective dose is estimated at  $35 \cdot 10^4$  man-Sv.<sup>6</sup> In the 27 worst-contaminated districts in these oblasts (with a population of 772 000) the collective dose is equal to  $22 \cdot 10^4$  man-Sv, and in districts under strict control - the "controlled" area, where  $^{137}Cs$  contamination exceeds  $15 \text{ Ci/km}^2$  and where 273 000 people live - the collective dose is around  $8 \cdot 10^4$  man-Sv.

The same table contains estimates of possible long-term consequences of thyroid gland irradiation for the population groups studied. The risk coefficient for contracting cancer of the thyroid gland for the population in the contaminated areas was estimated mainly in line with the assumptions and assessments contained in report No 80 of the USA's NCRP.<sup>4</sup> The whole-life morbidity coefficient for cancer of the thyroid gland for the demographic structure of the population in the irradiated territories, calculated for two different periods of possible manifestation of the radiation risk (40 years and total

remaining life span), is put at 13 and 18 cases per  $10^4$  man-Sv respectively. The spontaneous level of morbidity of the thyroid gland with cancer for the USSR is 25-40 cases per year per million people. On this basis it can be expected that in the controlled territories whole-life morbidity for cancer of the thyroid gland resulting from irradiation will increase by 20-30%. If one considers that children and adolescents (some 25% of the population) are approximately twice as sensitive to irradiation as adults, and that they account for 50-60% of the collective dose, it is quite possible that once the latent period is over we will see an even greater increase (up to 2-3 times) in morbidity in this age group compared to the spontaneous level. Close medical observation of the irradiated individuals and timely treatment of the illnesses arising should lower the harmful effect of thyroid-gland irradiation in specific individuals. If they are treated in good time there will be no change in the life expectancy of this section of the population.



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Table 1: Age-dependent iodine metabolic parameters in human body and thyroid dose due to I-131

Parameters	Age (years)					
	0.5	5	10	15	> 18	
Thyroid mass. (g)	2	5	9	14	20	
Biological half-time of iodine excretion from the thyroid. (days)	20	30	40	50	110	
Thyroid dose due to intake into thyroid 1 $\mu$ Ci I-131 (rem)	42	19	12	8	6	



Table 2: Possible late effects of thyroid exposure among the population in the most radioactively contaminated areas of the USSR

Area	Number of administrative units	Population (millions)	Thyroid collective dose ( $10^4$ man-Sv)	Excess thyroid cancer	Excess over spontaneous level (%)
Most contaminated regions	9	15.62	34.6	400 - 600	1 - 2
Most contaminated districts	27	0.772	22	300 - 400	15
"Controlled" area		0.273	8	100 - 140	20 - 30

Fig.1: Changes in radionuclide concentrations in milk (total beta-activity) in may 1986 in the region with Cs-137 surface contamination in the range of 5-15 Ci/km<sup>2</sup>. Radiochemical analysis of milk showed that about 80 % of beta-activity was due to I-131.

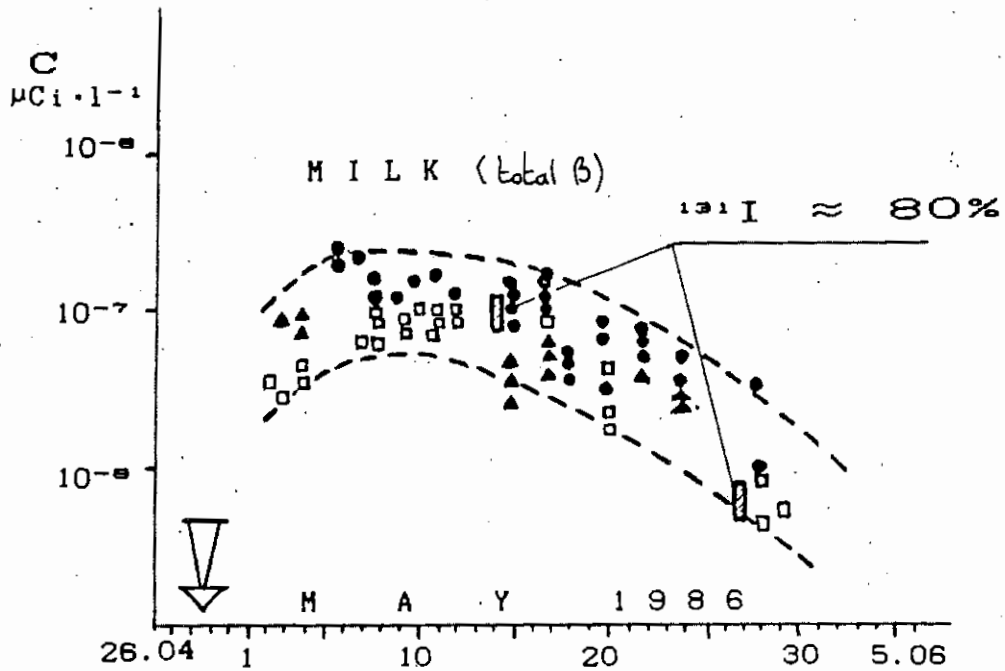


Fig.2: a) Pattern of I-131 intake in inhabitants of the contaminated region.  
b) The I-131 intake models used for thyroid dose estimates after Chernobyl accident.  
c) I-131 thyroid retention according to accepted models of radioiodine intake.

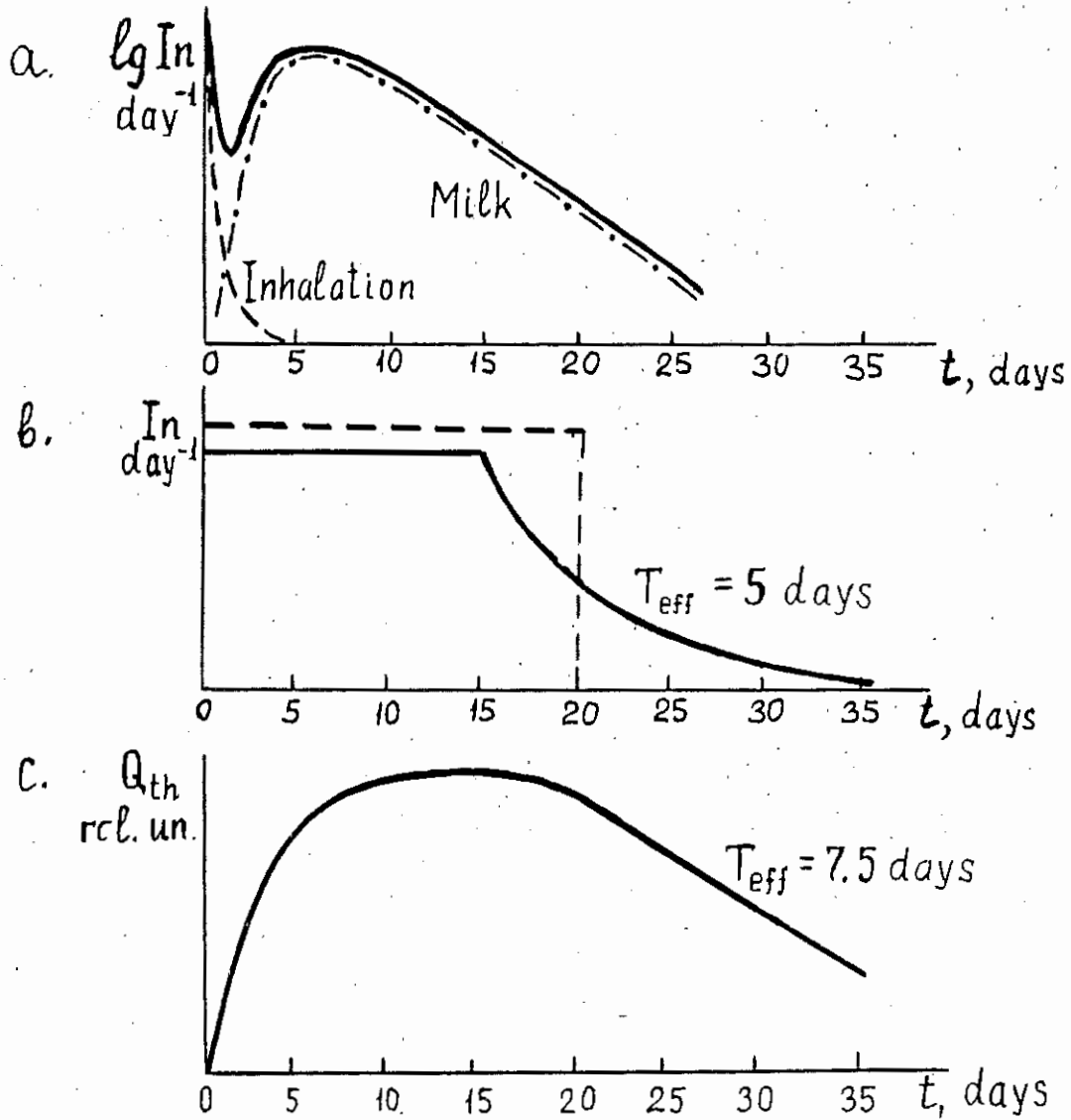


Fig.3: Relative age-dependence of mean thyroid dose for rural and urban population in settlements with the same radioactive contamination. The dose in the group of urban adults is taken as 1.

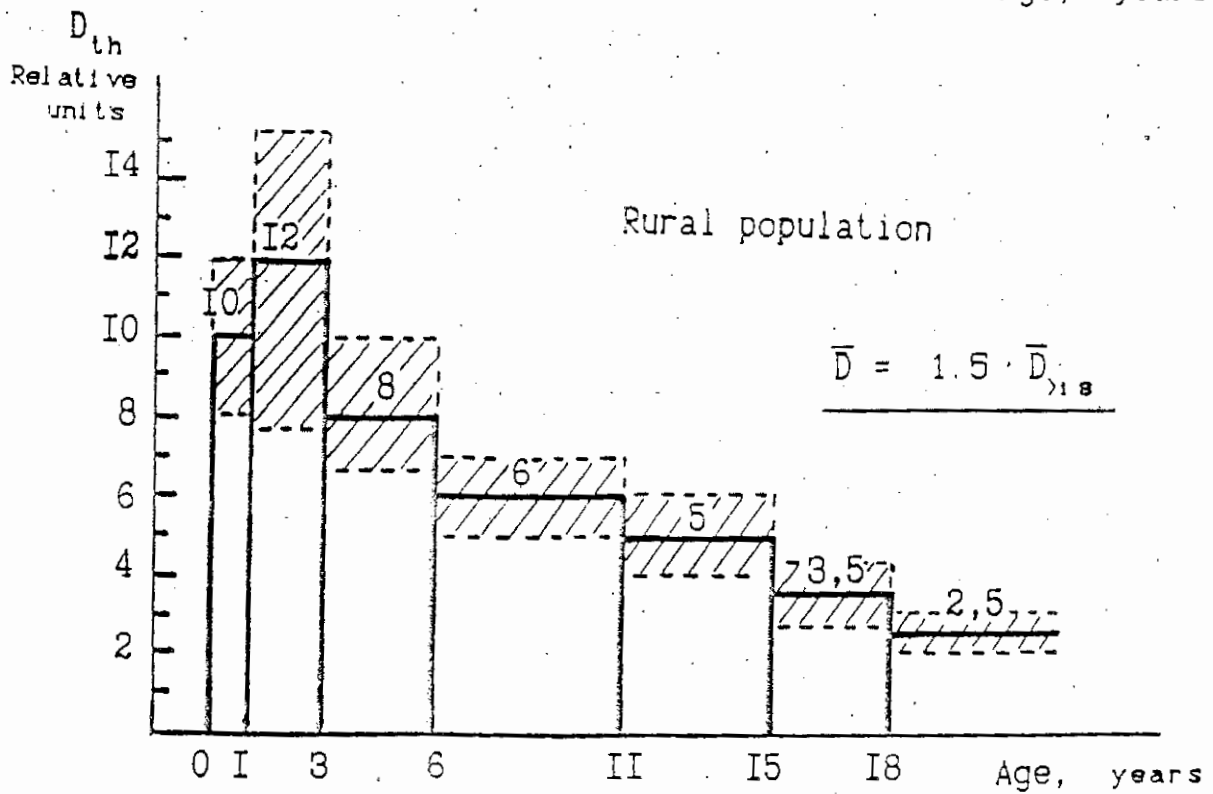
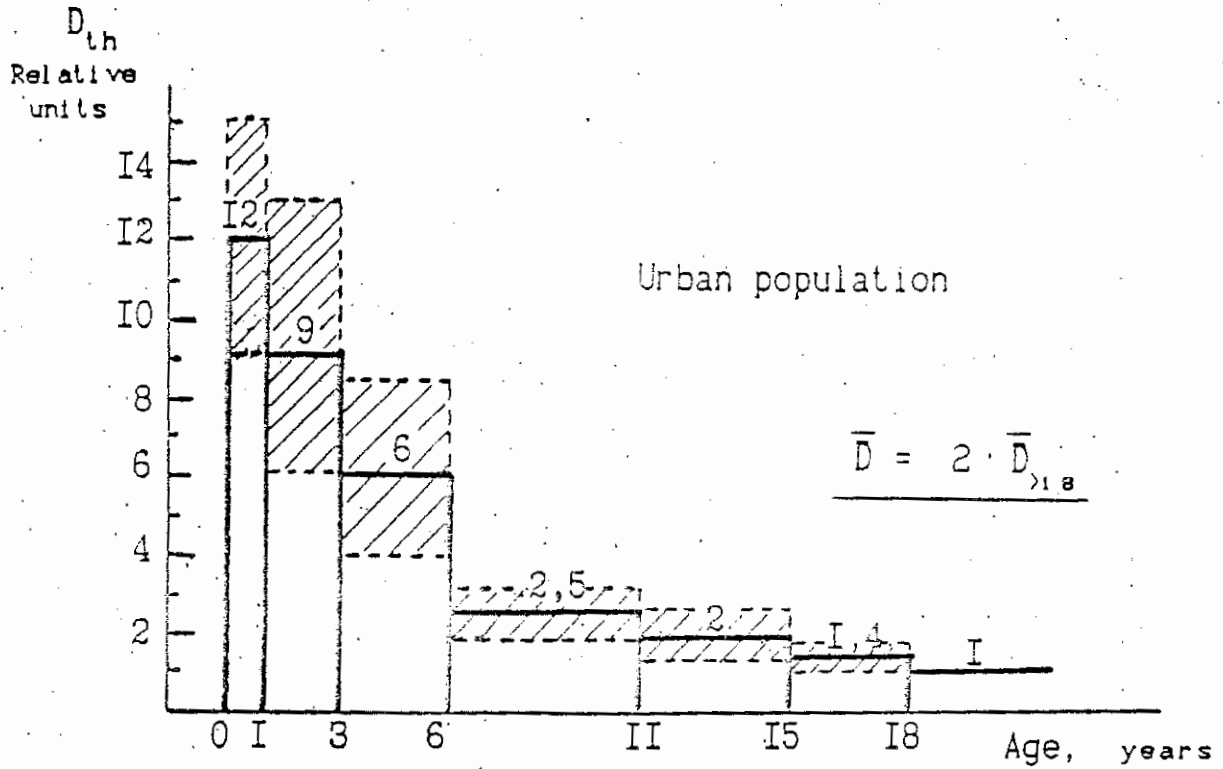


Fig.4: Correlation between mean thyroid dose in children (1-6 years old) and mean <sup>131</sup>I concentration in milk from 10-15 may 1986.

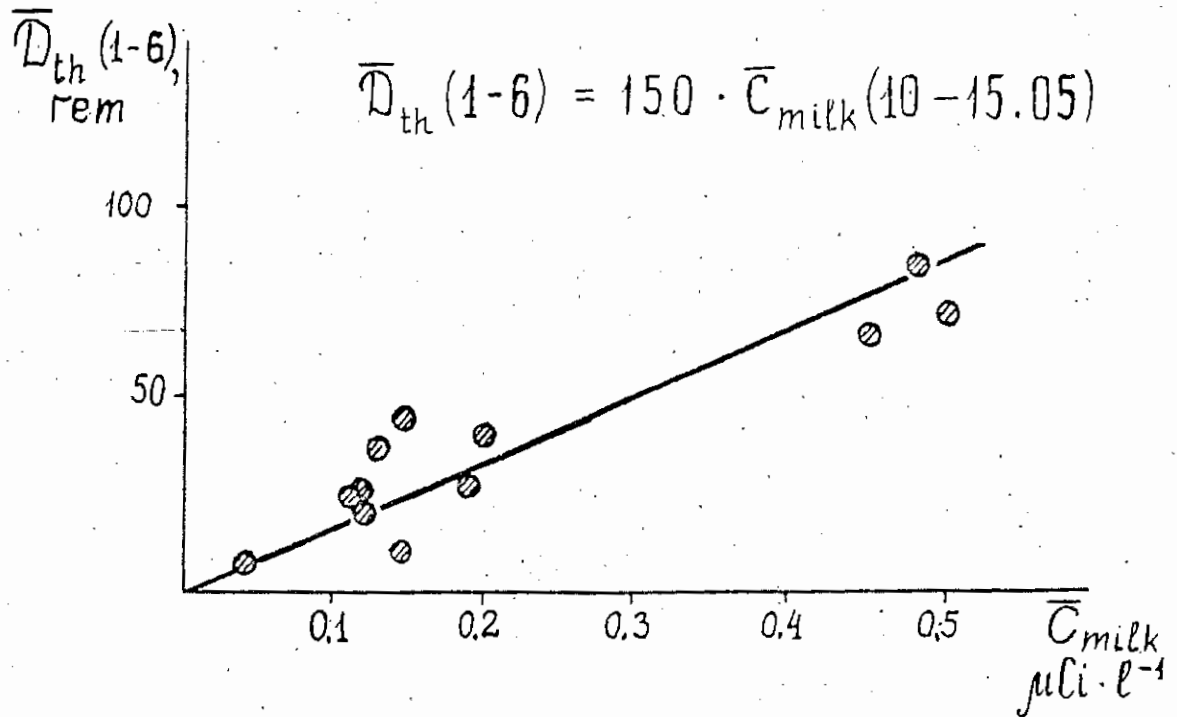
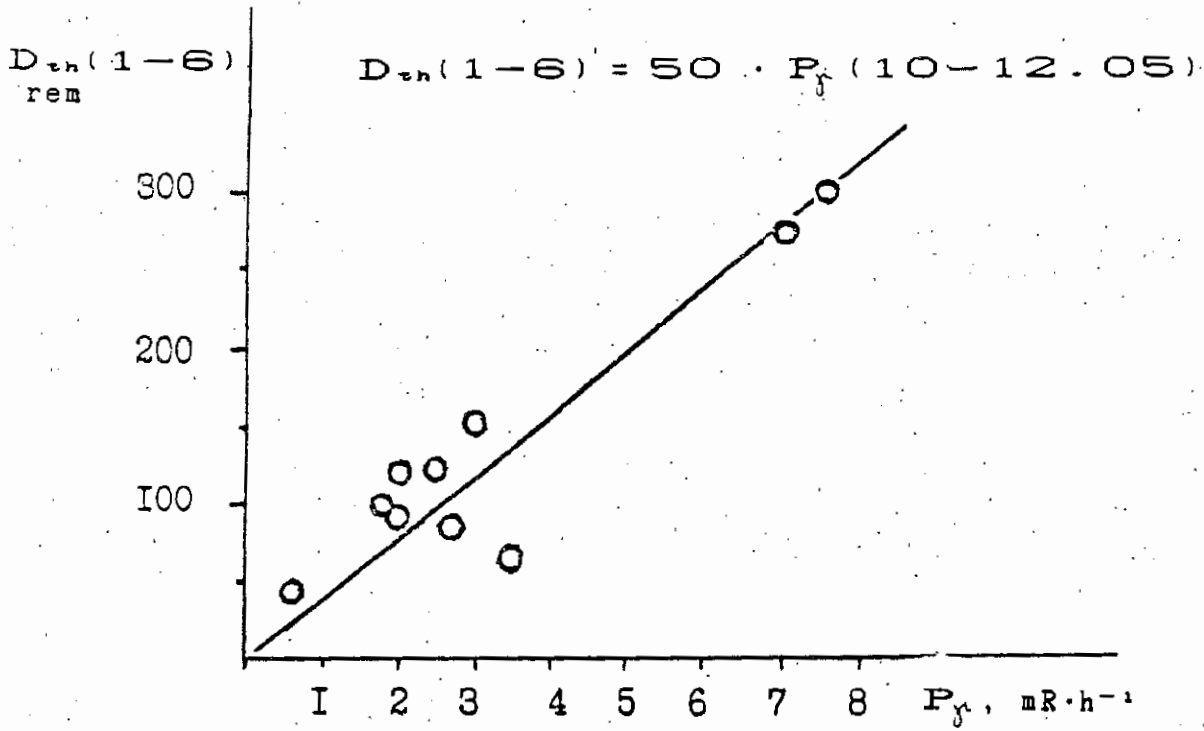




Fig.5: Relationship between mean thyroid dose among children 1-7 years old and the dose rate in the air for a given settlement from 10-12 may 1986.



# **The Consequences of the Kyshtym Accident for Flora and Fauna**

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## ABSTRACT

Flora and fauna irradiated in areas radioactively contaminated by the Kyshtym accident accumulated the bulk of their dose more or less in the first year, with the irradiation being at its most intensive in autumn 1957 and the winter of 1957/58, when plants and many animal species were in the physically dormant state. During the "acute" phase the maximum doses absorbed (at a contamination level of 4 000 Ci  $^{90}\text{Sr}/\text{km}^2$ ) were as follows (in krad): mouse-type rodents and fish 4, birch bud meristems 20, pine bud meristems 40, pine needles 80, dormant leaf buds and gramineae seeds on the soil surface 160. The main radiobiological effects appeared in the spring of 1958 and were observed for several years after; subsequently, in the presence of chronic irradiation at a low dose rate, predominantly genetic effects were observed, conifers being the most radiosensitive among the plants. At doses over 0.5 krad (40 Ci  $^{90}\text{Sr}/\text{km}^2$ ) we observed radiation damage (ranging from partial to severe) in crowns; doses over 3-4 krad (over 180 Ci  $^{90}\text{Sr}/\text{km}^2$ ) led to pine die-back in the autumn of 1959 over a total area of some 20 km<sup>2</sup>. Birch stands died over an area of some 5 km<sup>2</sup> at a dose of 20 krad (4 000 Ci  $^{90}\text{Sr}/\text{km}^2$ ).

Changes in the structure of herbaceous communities - due to the death of perennial species whose leaf buds were located above the soil surface - occurred at doses over 20 krad (1 500 Ci  $^{90}\text{Sr}/\text{km}^2$ ). In subsequent years we observed changes in the structure and numbers of fish and mouse-type rodent populations. The radioactive contamination caused an increase in the rate of mutational processes in plant and animal populations. However, for populations as a whole the increased frequency observed for most mutations (chromosome aberrations, chemical mutations) did not play a major role, since they were speedily eliminated by natural selection. No deformities of a genetic nature - i.e. in progeny, clear-cut pathological deviations from traditional external appearance and behaviour - were found on the contaminated territory. In the 30 years since the accident the biological characteristics of the contaminated area have not differed (except for coniferous forests) from those of the surrounding regions. Natural ecosystems are very radioresistant, and extremely high doses are needed to damage them seriously and irreversibly.

The intensive radioactive contamination of the environment following the 1957 accident had radiobiological consequences for the natural flora and fauna populations and communities. The studies undertaken immediately after the trail of radioactive fallout was deposited in the Eastern Urals showed that irradiation of natural ecosystems involves not only primary radiation effects but also secondary, delayed consequences. The ecosystems' resistance to irradiation largely depends on the relationship between these primary and secondary effects.

### 1. Dose formation characteristics

Three factors determined the doses received by flora and fauna in the contaminated territory of the Eastern Urals:

1. The bulk of the radiation from the fallout mix was beta-radiation (up to 75%) which penetrates several centimetres into biological tissues. Therefore, the distribution of the dose burdens in the different biogeocenoses closely matched the distribution of the radioactive substances therein.

2. Short-lived radionuclides ( $^{95}\text{Zr}$ ,  $^{106}\text{Ru}$ ,  $^{144}\text{Ce}$ ) were the main contributors to the activity in the fallout, thus engendering an 'acute' period of some 1-1.5 years in which most of the absorbed dose formed, followed by a period of long-term or chronic irradiation of considerably lower radiation intensity.

3. The radioactive contamination proper and a large part of the 'acute' phase occurred while plants and many animal species were in the physiologically dormant state, during which the processes of radiation damage and regeneration were slowed down. Therefore, the degree of radiation damage depended not only on the intensity and dynamics of the irradiation processes, but also on the integral dose accumulated by the time the next period of physiological activity began.

With the exception of genetic effects, all the other radiobiological consequences resulted mainly from the dose accumulated in the autumn of 1957 and the winter of 1957/58. Genetic effects may also have been caused by subsequent chronic irradiation at a low dose rate.

The doses received by plants and animals during the 'acute' period are given in Table 1. The maximum absorbed doses correspond to contamination of 4 000 Ci/km<sub>2</sub> of  $^{90}\text{Sr}$  for terrestrial organisms and 1 000 Ci/km<sub>2</sub> of  $^{90}\text{Sr}$  for aquatic organisms.

During the 'acute' period the maximum irradiation was received by pine trees, whose needles retained for a long time the fallout deposited on them, as did the "renewal" (or leaf) buds and generative shoots of plants on or near the soil surface. Maximum doses were absorbed by dormant leaf buds and gramineae seeds (up to 160 krad), pine needles (up to 80 krad), pine bud meristems (up to 10 krad) and birch bud meristems (up to 5 krad). Soil invertebrates received considerable irradiation, with doses ranging from 1-80 krad depending on their habitat (soil or litter). Among mammals and birds the maximum doses were absorbed by those in which a large, if not the main, contribution came from irradiation of the gastro-intestinal tract through eating contaminated food. Mammals and birds can be classified as follows in ascending order of dose received: migratory birds, carnivorous mammals, wintering raptors, wintering granivorous birds, large herbivores, mouse-type rodents. The maximum dose absorbed by mouse-type rodents was

4 krad. Similar irradiation doses were also typically found in fish living in the two most contaminated bodies of water.

Fig. 1 shows the dynamics - over 30 years - of irradiation dose formation in biota in the contaminated territory of the Eastern Urals. Almost all the irradiation dose was formed by 1960, the highest doses being absorbed by organisms located on or inhabiting the surface of the soil-vegetation cover or of bed sediments, and the lowest by those living in water.

## 11. Effects of radiation on flora

Conifers were the most radiosensitive species, and this was later confirmed after the Chernobyl accident. The first radiation effects were observed in pine trees in spring 1958, i.e. yellowing of needle tips and withering of the top and lateral buds. This led to the death of some or all of the needles and to the transformation of the surviving buds into short and thick bundles of shoots with elongated needles. Pine die-back was observed in the autumn of 1959 and occurred at an absorbed dose in the needles of 3-4 krad (over 180 Ci/km<sub>2</sub> of <sup>90</sup>Sr). Some 20 km<sub>2</sub> of pine trees died in all (Table 2).

The following effects were observed over two years at doses of between 0.5 and 3 krad (over 40 Ci/km<sub>2</sub> of <sup>90</sup>Sr): yellowing, drying-up and fall of needles, defects in the development of new needles, reduced growth of shoots and stems, various physiological and morphological irregularities, reduction in the viability of seeds and pollens, and phenological changes.

Birch forests were much more resistant to radioactive contamination. They totally died out only in areas with a maximum contamination level of 4 000 Ci/km<sub>2</sub> where bud meristems received a dose exceeding 20 krad during the 'acute' phase. At lower doses the top layer of the birch crowns dried out and the leaves were underdeveloped, and phenological changes were recorded in the first four years after the accident (retarded appearance of leaves and delayed flowering, premature leaf-fall). Radiation damage to birches was observed over an area of 17 km<sub>2</sub>.

The different levels of damage recorded in pine and birch forests due to radioactive contamination can be explained by the higher radiosensitivity of pines, and by the large dose absorbed by their crowns due to prolonged residence of radioactive materials in the needles. In general, birch die-back was observed when contamination was 70 times higher than that at which pines died.

Among gramineae, those that suffered most were perennials with leaf buds located not far above the soil surface. In areas with contamination of over 1 500 Ci/km<sub>2</sub> (dose absorbed in buds during the 'acute' period exceeding 20 krad), these species disappeared and were replaced by plants with leaf buds in the soil. This process lasted 3 to 4 years, and was then followed by a slow reverse process.

At contamination levels below 1 000 Ci/km<sub>2</sub> adult plants did not die, but the seeds of many species had a reduced germinating capacity. Moreover, morphological changes were observed in certain plant species over 2-3 years: gigantism, chlorosis, blueing and twisting of leaves, and reduction in the number of seed grains in the ear.

Whereas in meadow communities of a simple structure the changes resulted mainly from the direct impact of irradiation (primary effects), in forest communities they resulted from the combined action of primary and secondary effects. In areas where trees died after their crowns had dried out, the microclimate beneath their canopy changed, with more light and moisture getting through. Therefore, in severely damaged mixed forests there was a five-fold increase in the amount of light and a 1.5-2-fold increase in the amount of moisture reaching the soil. Furthermore, in spring the daylight period was considerably lengthened in the lower layer of the forest due to the retarded appearance of birch leaves. All this led to intensive multiplication of the herbaceous vegetation, its mass increasing by a factor of 3-5 compared to uncontaminated forests.

Delayed effects occurred in plant communities also in the form of replacement of the customary herbaceous plants by others (e.g. weeds growing in meadows or photophilic plants developing under the canopy of thinned-out forest). Two years after the accident we noticed an explosion in the proliferation of entomic pests in damaged forests (e.g. gypsy moth). Except for the dead coniferous stands, practically all the plant communities managed to regenerate in the course of time, and today their succession and productivity are no different from those of neighbouring communities.

### III. Effects of radiation on fauna

Among the fauna in the contaminated territory invertebrates constitute the most numerous group. A reduction in population and death following irradiation were noted only in species with a long life cycle and an extended phase of development in the forest litter, turf or top soil layer. These effects were at their strongest in earthworms, myriapoda and mites with hardened plates at contamination levels above 100 Ci/km<sub>2</sub> (dose exceeding 600 rad during the 'acute' period).

Significantly less marked radiation-induced changes were observed in flying insects capable of quickly dispersing over a vast territory, and in invertebrates with a casing. In particular, we did not note any adverse radiation effect on ants, although they spend most of their life on the surface of the forest litter.

It is calculated that birds and mammals received lethal doses in autumn and winter 1957/58 only if they lived permanently in areas where contamination exceeded 1 000 Ci/km<sub>2</sub>.

Since the vast majority of birds in the contaminated territory are migratory species and the accident occurred in autumn, it can be assumed that the radioactive contamination did not start to affect them until spring 1958 when the dose rate in tree crowns had already decreased by a factor of 10. It is calculated that in the summer of 1958 and of 1959 the dose absorbed by birds did not exceed 100-200 rad, i.e. considerably less than the lethal doses (800-1 000 rad). In these and following years we did not record any bird deaths, nor did their numbers depend on contamination level.

As for mammals living in the contaminated territory, the most variegated radiation effects were observed in mouse-type rodents. At contamination levels exceeding 1 000 Ci/km<sub>2</sub> (dose rate in the 'acute' period above 10 rad/day) mortality increased and life expectancy

decreased in individual cases. These primary irradiation effects, observed over 10-15 years, induced secondary irregularities, e.g. changes in population structure and weakening of defence mechanisms. For example, in the first few years following the accident there was an increase in the number of blood-sucking exoparasites carried by mouse-like rodents, and the animals' mobility decreased.

However, after 15 years - i.e. some 30 animal generations later - their populations in the contaminated territory were comparable to those of all other animals, judged by all indicators, and the radioresistance of the populations living in the contaminated territory of the Eastern Urals had increased (due to 'radioadaptation'), as demonstrated in special experiments involving additional irradiation. It was observed that the lethal doses for animals living in the radioactive territory were 1.3 times higher than for the controls.

No similar radiation effects were observed in other mammal populations (elk, deer, wolf, lynx, hare). It is possible that their numbers decreased during the 'acute' period in highly contaminated areas, but this supposition has not been confirmed experimentally.

The most vulnerable link in aquatic ecosystems are herbivorous fish (carp, crucian): they spend the winter in the silt, which leads to additional irradiation of their bodies. Dose burdens observed in the roe of such fish reached lethal levels ( $> 1$  krad), causing a temporary drop (2-3 years) in their reproduction. Control catches of carp and crucians, started in 1960, did not reveal any decrease in their numbers or any significant irregularities in their population structure.

No deleterious irradiation impact was found among the remaining, less sensitive, links in the aquatic ecosystem (plankton, vegetation, invertebrates).

#### IV. Genetic effects

The studies undertaken convincingly showed that the radioactive contamination provoked an increase in the rates of mutational processes, both in plant and animal populations. As the absorbed dose rate decreased, the slower were the mutational processes, some of which - in particular chromosomal aberrations (structural damage) - soon stabilized at a stationary level, whereas others - e.g. biochemical mutations (changes in the structure of individual proteins) - have not yet reached the above-mentioned level to this date, which confirms the lengthy nature of genetic restructuring processes.

For the population in general the increased frequency of most mutations is not significant, given their rapid disappearance in the course of natural selection. However, some genetic changes may accumulate from one generation to another, particularly in the case of prolonged chronic irradiation at low doses. In the contaminated territory this phenomenon was discovered in two types of plants: greater knapweed (*Centaurea scabiosa*) and Scotch pine (*Pinus sylvestris*) growing in areas where contamination ranged from one Ci/km<sub>2</sub> to several dozen curies per km<sub>2</sub>. At sufficiently high dose rates in the generative organs of these plants (up to several tenths of a mrad/day) the frequency of biochemical mutations increased several-fold compared to the natural level, although this frequency did not increase in direct proportion to the absorbed dose. Increased frequency was observed at the smallest dose rates in the range studied. It is difficult for the

moment to give a definitive explanation for this phenomenon. In animals with a quick reproduction rate (e.g. field mice) living in areas with contamination of from 100 to 1 000 Ci/km<sub>2</sub>, the mutation frequency also increased by a factor of 1.5-2 compared to spontaneous levels.

The question remains open as to the consequences of the biochemical mutations still accumulating in flora and fauna even today in some parts of the radioactively contaminated area, since population genetics has not yet provided an answer. All we can say is that in the contaminated territory we did not detect any deformities of a genetic nature, i.e. in progeny, clear-cut pathological deviations from traditional external appearance and behaviour.

#### V. Radioresistance of the environment

Data from many years spent observing the reactions and condition of ecosystems irradiated after the Kyshtym and Chernobyl accidents, as well as additional ecosystem-level experiments, enable us to evaluate the quantitative criteria relating to the radioresistance of typical ecosystems. Given the need for the integrity of all the ecosystems studied to be preserved, i.e. for the course of natural succession to be maintained and continued in conditions of radiation exposure, we can assume that the integrity of an ecosystem depends mainly on the wellbeing of the dominant component therein. Therefore, it is suggested that the radiosensitivity of the dominant be used to measure the radioresistance of a given system. Table 3 gives the Ecological Dose Limits (EDLs) for individual ecosystems, expressed in the form of the corresponding doses determining the radiosensitivity of the dominant.

If the EDLs are reached, not only is the dominant component of an ecosystem damaged and killed, but the ecosystem's net primary productivity is altered too, reflecting a change in efficiency in harnessing the solar energy absorbed. Each phase of development in an ecosystem depends on a relatively stable level of net primary productivity; if this is reduced by the effects of radiation, then the integrity of the ecosystem is disrupted. Analysis of the available data shows that the highest mean value for the dose absorbed by an ecosystem in one year, equal to 1 krad, is a minimum limit which does not cause a reduction in net primary productivity in cases of uniform chronic irradiation during a given stage of succession.

In practice it is quite difficult to evaluate the irradiation doses received by flora and fauna. Therefore, where well-founded, sound estimates exist for irradiation doses to the human population in accidents, these can be used to forecast irradiation of plants and animals. An example is given in Table 4; the main conclusion to be drawn from it is that the dose limit used to protect humans is sufficient to ensure protection of flora and fauna.

The results obtained during 30 years of research in the radioactively contaminated territory of the Eastern Urals show that natural ecosystems possess a high degree of radioresistance. Serious and irreversible disruption of communities requires doses far above those which are lethal to any type of individual organism. The total regeneration of all the damaged communities and ecosystems (coniferous forests excepted) in the contaminated territory is direct proof of the environment's strong regenerative capacity.



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Table 1

Doses absorbed by plants and animals in the radioactively contaminated territory of the Eastern Urals during the first 1-1.5 years

Organism	Maximum absorbed dose, krad	Normalized absorbed dose, $\frac{\text{rad}}{\text{Ci } ^{90}\text{Sr}/\text{km}_2}$
Plants:		
dormant leaf buds and gramineae seeds on soil surface	160	max. 40
pine needles	80	max. 20
pine bud meristem	40	max. 10
birch bud meristem	20	max. 5
tree seeds in crowns	16	max. 4
Animals:		
soil invertebrates	1-80	0.25-20
fish	4	4
fish roe	> 1	> 1
birds	0.1-0.2	0.025-0.050
mouse-type rodents	~4	~1

Table 2

Radiation-induced demise of plant communities

Community	Contamination level, $\text{Ci}/\text{km}_2$ $^{90}\text{Sr}$	Lethal dose absorbed by buds in 'acute' period	Area, $\text{km}_2$
Pine forest	180	4	20
Birch forest	4 000	20	5
Meadows (in part)	1 500	20	15
Meadows (total)	4 000	150-200	5

Table 3

Ecological Dose Limits (EDLs) for typical ecosystems

Ecosystem	Dominant	EDL, krad
- Coniferous forest	Trees	2-4
- Broad-leaved forest	Trees	30-40
- Herbaceous (meadow)	Mixed gramineae	40-50
- Pasture	Sown gramineae	8-10
- Crops	Monocultures	5-6
- Freshwater	Phytoplankton	30-50

Table 4

Ratios between doses absorbed by ecosystem components and the effective dose equivalent of irradiation of the human population during the first year following accidental contamination of the environment, rad/rem

Ecosystems, components	Kyshtym accident	Chernobyl accident
Coniferous forest trees (crowns)	27	47
Broad-leaved forest trees (crowns)	14	40
grass cover	30	50
Herbaceous (meadow) vegetative phytomass	17	45
leaf buds	77	92
Ground zocenososes small rodents	28	30
large herbivores	15	-
Freshwater bed-dwelling aquatic biota	2.6	-
ichthyofauna	1.4	-
fish roe	0.4	-

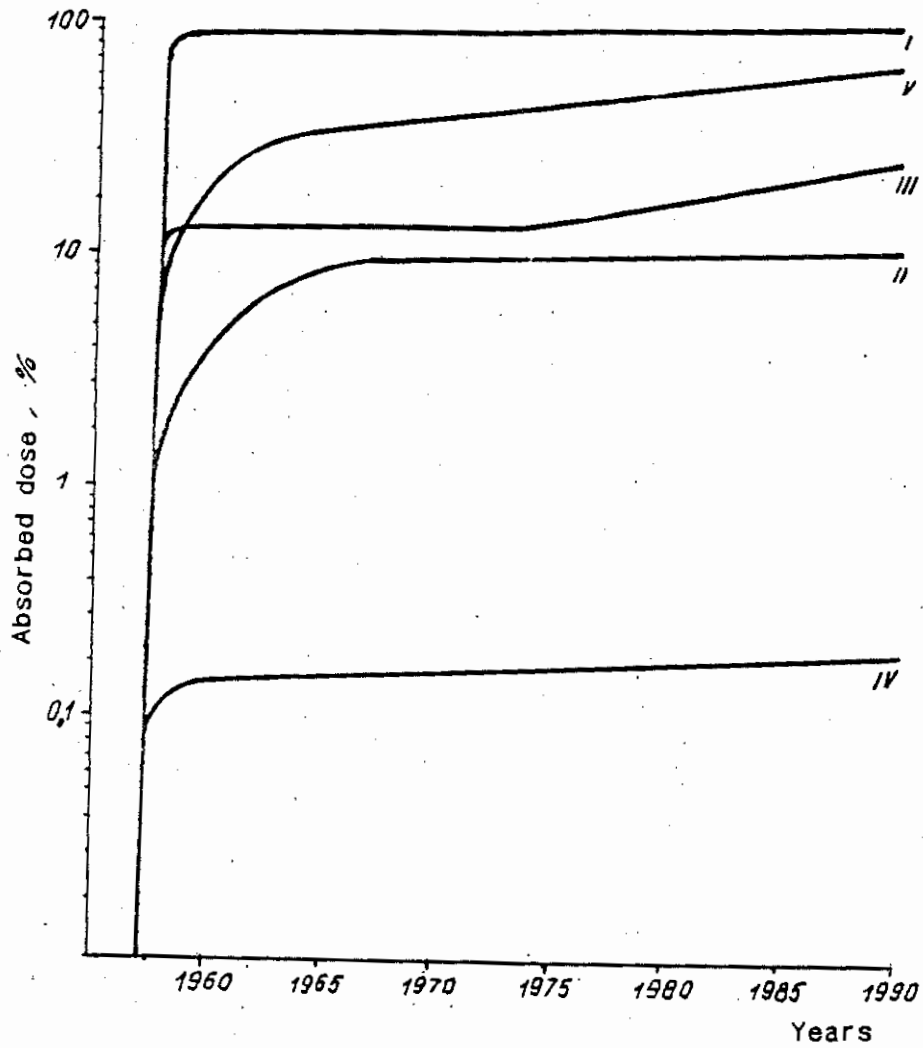


Figure 1: Dynamics of irradiation dose formation for various components of the environment

- I. On the surface of the soil-vegetation cover in meadows
- II. Under tree canopy
- III. In mixed forest crowns
- IV. In the water of lakes
- V. In bed sediments



**Genetic Consequences of the  
Action of Ionizing Radiations on  
Natural Populations After the  
Kyshtym Accident**

**V.A. SHEVCHENKO**

### Introduction

1. The problems concerning the action of ionizing radiations on natural populations and communities are highly topical. The topicality of these problems is dictated by the development of the entire complex of mining and milling of uranium ores, processing, utilization and storage of nuclear fuels and nuclear fission products. The problems of the action of natural increased radioactivity on plant and animal populations are not of minor importance either. Clearing up of these problems can be obtained by studying of ecological consequences of major nuclear accidents for example Kyshtym accident.

2. The strategy of studying ecological consequences of the action of ionizing radiations should include the analysis of genetic processes in natural populations as one of the main elements. Such analysis involves a number of key tasks and has methodological features, the knowledge of the latter being necessary to estimate the risk of environmental pollution and to forecast its genetic consequences.

The object of the present review is to consider some problems in radiation genetics of natural populations reported earlier /1-5/ and to reveal problems which are of key importance, in the author's opinion, in monitoring irradiated natural populations.

In connection with this object, the following problems should be considered:

- a) to forecast remote genetic consequences of the action of ionizing radiations on communities and biocenoses, evidence on radioresistance of different species composing communities is required as well as information on factors responsible for different radioresistance of species; it is necessary to carry out the purposeful search of natural objects characterized by high sensitivity to mutagenic factors and suitable to be used as test systems in monitoring genetic processes in plant and animal populations;
- b) it is necessary to study the dependence of the yield of genetic changes on radiation dose and rate under conditions maximally approximating to the real situations of irradiation of natural objects; special attention should be paid to estimation of genetic efficiency of low doses and dose rates of ionizing radiations;
- c) the mutation level in irradiated populations and their structure should be studied as important genetic criteria for estimating the effect of ionizing radiations on natural populations; it is necessary to study the mutation dynamics in successive generations of irradiated populations and the process of genetic load formation in populations of species differing in the degree of panmixia and in populations cycle characteristics;

- d) it is important to study possible ways of population adaptation to the chronic action of ionizing radiations, the degree of expression and the tempo of adaptation changes in different species depending on their initial radioresistance, modes of reproduction, ploidity and other biological characteristics;
- e) the role of the most significant ecological factors in mutation formation in irradiated natural populations of different species should be analysed;
- f) it is necessary to assess the role of radiation quality and irradiation geometry in the formation of absorbed doses received by critical organs of plants and animals due to the action of incorporated radionuclides;
- h) it is necessary to carry out a comparative study of microevolutionary processes in populations of different plant and animal species exposed chronically to mutagenic factors. Differences in the mutation dynamics and in the tempo of adaptation changes in populations of different species can serve as one of the main genetic criteria in estimating possible consequences of environmental pollution. The most important problem is the study of the relationship between genetic changes induced in populations under the chronic action of mutagens and possible ecological shifts. To reveal ecological shifts determined by genetic processes is one of the bases for ecological rationing of environmental pollution.

#### On Comparative Radioresistance of Species

3. Determination of comparative radioresistance of species is important in estimating biological consequences of the action of ionizing radiations on natural populations of different species and their communities. Comparative radioresistance of species can be determined using a number of criteria, such as the survival rate of microorganisms, plants and animals exposed to acute irradiation, and cytogenetic tests. The factors affecting species radiosensitivity include cell ploidity, nucleus size, chromosome number, DNA amount in nuclei, effective number of chromosome arms, repair efficiency, level of endogenous radioprotectors, etc. Thus, numerous studies on radioresistance of different objects show that the use of the target theory /6,7/ has proved to be fruitful in estimating radiosensitivity of species. This is most completely demonstrated in the works of Sparrow et al. /8,9/. They have shown that cell radiosensitivity ( $D_0$ ) is inversely proportional to the chromosome volume in interphase. This has been proved for a large group of organisms, from the most elementary to the most complex forms. The relationship between  $D_0$  and an average chromosome volume in these organisms is graphically expressed in the logarithmic scale by series of eight regression lines (radiotaxa) with a slope of  $-1$ . Preobrazhenskaya and Timofeyev-Ressovsky /10,11/ point out to another important aspect of the problem of comparative radioresistance of organisms.



Having studied radiosensitivity of seeds of 660 plant species the authors note that the degree of species radiosensitivity depends on the position of species in the phylogenetic system: all coniferous plants studied are radiosensitive; over half of species among monocotyledonous plants are radiosensitive and only one species is radioresistant; among dicotyledonous plants about half of species are radioresistant and only several are radiosensitive.

#### Estimation of Genetic Efficiency of Low Dose Irradiation

4. The dose of ionizing radiation ( $D$ ) is equal to  $I \cdot t$ , where  $I$  is the dose rate and  $t$  is the irradiation time.  
In its documents /12,13/ UNSCEAR assumes the linear character of the induction of one-shot genetic damages in case the dose is the function of irradiation time. Proceeding from this assumption, an expected damage per gamete per dose unit is estimated (direct method) and a dose doubling the natural mutation level is calculated (indirect method).  
It is emphasized in the UNSCEAR documents that the hypothesis of linear dependence is an unproved assumption which is in agreement with a vast body of experimental and epidemiological material. It should be added that most of the material has been obtained provided the dose is the function of irradiation time.
5. In case of radioactive environmental contamination populations are exposed to different radiation dose rates. Thus, total absorbed doses in the cellular ontogenetic and populational cycles of development are the function of irradiation intensity. There are data indicating that the yield of mutations per cGy is higher at low intensities of ionizing radiations (about several cGy per day) than at higher dose rates i.e. there is no linear dose response relationship. This has been demonstrated in experiments with chlorella (*Chlorella vulgaris* Beijer., strain Larg-1), beans (*Vicia faba* Russian Black variety), wheat (*Triticum aestivum*, Skala variety), barley (*Hordeum vulgare*, Maya variety), wild herbaceous plants (*Plantago media* L., *Veronica tenerium* L., *Vicia cracca* L., *Centaurea scabiosa* L.) and loach (*Missquarnus fossilus* L.) exposed to radionuclides of strontium-90, yttrium-90 and promethium-147 /14-16/. It follows that the analysis of the mutation process in chronically irradiated natural populations requires an additional study of dose-response relationships.
6. The experimental materials have been reported in detail in another review. Here, the typical results are presented. Thus, in particular, the frequency of chromosome aberrations in roots of beans (*Vicia faba*) grown for 10 days in regions with different concentrations of strontium-90 - yttrium-90 has been determined (Fig.1). The prevailing types of chromosome aberrations in anaphases of mitosis under the mentioned irradiation regime were single fragments and bridges. The portion of these types of aberrations did not change significantly with the dose rate. In other words, one-shot genetic changes were mainly observed at the dose rates used in the experiment.

Fig.1 shows in the logarithmic scale the data with and without (induced mutagenesis) regard for the natural mutation level. Taking into account the strictly linear character of the curve for induced mutagenesis it is possible to determine an expected number of induced mutations at dose rates lower than 0.005 Gy/day (i.e. at doses lower than 0.05 Gy) by extrapolating the regression line towards low irradiation intensities. In this case an "observed effect" curve in the range of low irradiation intensities can be built up by summing up an expected induced effect and the natural mutation level. This curve will asymptotically approximate to the natural mutation level. The results obtained permit a conclusion to be made that there is a linear relationship between the logarithm of the number of induced mutations and the dose logarithm (Fig.2). This relationship can be expressed by the equation:

$$y = \alpha \left( \frac{D}{D_0} \right)^n \quad (1),$$

where  $D_0$  is a starting point, for example the point of crossing of the regression line and the line corresponding to the natural mutation level;  $D$  - a dose (dose rate x irradiation time);  $n$  - an index describing the regression line slope.

In the above case the slope angle of the regression line is less than 45°, i.e.  $n < 1$ . It follows that radiation efficiency per dose unit reduces with increasing dose rate. As a result, an inverse correlation is observed between the mutation frequency and the dose rate at comparatively low irradiation intensities (0.001-0.1 Gy/day for plant populations).

7. In another experiment the  $\beta$ -radiation (yttrium-90) dose influence on the chromosome aberration frequency in the first and second anaphases of meiosis in *Hordeum vulgare* was studied. On an area of 0.8 hectares an experiment has been performed in which barley crops (Maya variety) were sprayed with yttrium-90 solutions during the main eight phases of plant development [16].  $18.5 \cdot 10^{12}$  Bq  $^{90}\text{Y}$  was used in the experiment. The formation of absorbed radiation doses was studied using thermoluminescent LiF-dosimeters. Dosimeters were placed inside plants directly in the point of a critical organ (in the point of growth, ovary, etc). Heterogeneity of dose distribution over the dosimeter volume (phantom of a critical organ) did not exceed  $\pm 7\%$ . The variance between the values shown by dosimeters at each plot was  $\pm 10\%$ .

Fig.3 presents the relationship between the chromosome aberration frequency in the first and second anaphases of meiosis and  $\beta$ -radiation doses absorbed by the vegetative cone in different developmental phases. This relationship is linear in the logarithmic scale, which permits its approximation by the power function equation (1). The use of the equation (1) makes it possible to describe satisfactorily the dose response relationship for a wide dose range from several cGy to several hundreds Gy (the initial dose rate varying from  $n \cdot 10^{-4}$  to  $n \cdot \text{Gy/h}$ ).

8. The dose-response relationship suggests that a dose doubling the natural mutation level is not constant for a given object and depends on the dose rate. The doubling dose grows with increasing irradiation intensity. This conclusion is supported by the experimental data obtained in the zone of Kyshtym accident presented in Fig.4. In all cases a common regularity has been revealed: the doubling dose is the increasing function of irradiation intensity. It follows that the "doubling dose" criterion can be used to estimate genetic consequences of exposures of natural populations to ionizing radiations only if all complexity of an observed situation is fully understood.
  
9. The data presented in Fig.3 indicate that the same relationship between genetic effects and dose rates will be always observed when populations are exposed to increased irradiation levels due to environmental contamination by radionuclides. In case the dose is the function of irradiation intensity the hypothesis of non-linear dose-response relationship may be suggested. It should be noted that the material presented in Fig.4 was analysed at different periods since the beginning of the experiments. This period was equal to one day for *Chlorella*, and to several days and months for loach, *Vicia faba*, and barley. For natural populations of wild herbaceous plants it was equal to several months. In view of sufficiently long radiation exposures in these experiments, the following explanation can be put forward for the observed relationship: the dose rate increase is accompanied by activation of repair systems reducing the yield of genetic damages. This inducible repair system apparently works as SOS-repair, its activity being the dose rate function.

#### Mutation Dynamics in Irradiated Populations

10. In the previous chapter data for comparatively short-term exposures of different objects to  $\beta$ -radionuclides have been considered. A question arises what the dynamics of induced genetic changes in growing populations is. Since a detailed study of the mutation dynamics in natural irradiated populations is impeded, such studies are carried out on model objects, mainly on *Drosophila* /17/. In our experiments we analysed the mutation dynamics in laboratory and natural populations of unicellular green algae /18-21/. The experiments have shown that the dynamics of induced mutation process in populations of *Drosophila* and unicellular algae is similar in general. At the same time, the short reproduction cycle of algae permits us to study the mutation dynamics in many generations for a sufficiently short period of time. The studies have shown that laboratory populations of unicellular green algae (lower plants) are the simplest model objects for studying the mutation process in populations of higher plants and other organisms.  
A series of laboratory experiments was carried out with populations of *Chlorella vulgaris* Beijer, Larg-1 and *Chlamydomonas reinhardtii*, strain pf(+) from the collection of Dr. R.P. Lewin (USA).

The mutation process was studied in algal populations (an aggregate of pigmental and morphological mutants) in which the exponential growth phase was maintained. The following irradiation regimes were used: acute ( $\gamma$ -rays), chronic ( $\beta$ -particles of  $^{90}\text{Sr}$ - $^{90}\text{Y}$ ,  $^{147}\text{Pm}$ ) and prolonged (a mixture of radionuclides imitating  $^{235}\text{U}$  fission products of different age). These experiments have revealed the main parameters of the mutation dynamics which are necessary for mathematical modelling.

11. Experiments with acute irradiation of continuously exponentially growing algal populations have permitted an estimation of the selection pressure against induced mutant clones (population genetic load). Each mutant clone  $M_i$  arising in a population due to the mutagenic activity and subjected to negative selection has been proved to be eliminated from a constantly reproducing population in successive generations according to the exponential law /18/. An aggregate of visible mutants resulting from acute X-ray irradiation of cultures is eliminated from a population according to the power function law:

$$f(x) = M \cdot x^{-p}$$

where  $f(x)$  is the law of elimination of mutant individuals,  $M$  - the initial number of induced mutants,  $p$  - a coefficient characterizing the slope of the elimination curve in case of graphical representation of  $\lg M$  as the function of  $\lg x$  (the number of generations). This can be illustrated by the data on the tempo of elimination of X-ray-induced mutants from a population of haploid *Chlamydomonas reinhardtii* cells (Fig.5).

A total number of mutant cells in a population in a given generation after irradiation  $\frac{f(x)}{1 - f(x)}$  is equal to the number of

of cells of mutant clones remaining in the population by this generation, i.e.:

$$\sum_{i=1}^n M_{i0} e^{-(\kappa_0 - \kappa_i)x} = \frac{f(x)}{1 - f(x)}$$

With  $n \rightarrow \infty$  (the number of mutant clones is rather high) let us pass on to  $\kappa_0 - \kappa_i \rightarrow \nu$ , where  $0 \leq \nu \leq \kappa_0$  ( $\kappa_i$ ; and  $\kappa_0$  - growth rate coefficients for mutant and normal clones). Thus,

$$\int_0^{\kappa_0} M_0(\nu) e^{-\nu x} \cdot d\nu = \frac{M_x^{-p}}{1 - \eta_x^{-p}}$$

An important conclusion follows from this equation:  $M_0(\nu)$  has the exponential character, i.e. in real populations there is an exponential relationship between the relative growth rate of mutant clones arising in a population after irradiation and the probability of occurrence of such clones /18/.

12. Chronic irradiation represents an additional environmental factor in a complex of factors influencing a population through induction of a number of mutations in each generation. After their phenotypic expression these mutations are subjected to the action of selection; as a result, we have curves describing accumulation of mutations in a series of successive generations.

In the case of irradiation at a constant dose rate an equilibrium between mutation pressure and selection is established. Due to this, the level of mutations in a population is stabilized. Such results have been obtained using irradiated laboratory populations of *Drosophila* /17/. Similar data have been obtained in the experiments with chronically irradiated algal populations (Fig.6). On the basis of such experiments we can study the relationship between the mutation level and irradiation intensity. The number of mutant individuals in a population has been proved to increase in proportion to the dose rate. It is possible to present a mathematical description of the mutation dynamics in populations of haploid microorganisms chronically exposed to ionizing radiations /18,21/.

13. The data available make it possible to determine the greatest lethal damage with which an algal population can still exist. The maximum level of lethally damaged cells in a population with which it can withstand the damage under chronic exposure conditions makes up about 90%, the maximum level of mutant cells being about 35% /21/. It should be noted that in contrast to these data the level of recessive lethals in irradiated *Drosophila* populations can amount to 100%, the viability of the populations being practically similar to the control /17/. So, there are essential differences in the mutation capacity of irradiated haploid and diploid organisms.
14. When populations are irradiated for a sufficiently long time their radioresistance increases /2-4,22/. In our model experiments the resistance to chronic UV-ray exposures increased in the 40- 70<sup>th</sup> cycles of culture density doubling, the level of mutant cells being reduced and the level of viable cells being increased. The period of transition from one resistance level to another covers 30-40 cycles of density doubling.

The stage at which the population resistance to chronic radiation exposures is increased is followed by the stage of population stabilization on a new radioresistance level. The amount of mutant cells in the population is also stabilized on a new level. In radioresistant populations visible mutations occur also in radioresistant cells, which determines quantitative and qualitative changes in the mutation process at this stage. In the case of chronic exposures to ionizing radiations the population resistance is increased later than under the action of UV-rays: about 150 cycles of density doubling are required since the beginning of the experiment for a fraction of radioresistant clones to appear in a population /23/.

15. To give a complete picture, the case should be considered when irradiation intensity reduces in time - such regime of population irradiation is most probable in real situations, for instance, after nuclear explosions. The mutation process in *Chlorella* populations exposed to a mixture imitating fresh <sup>235</sup>U fission products ( $\bar{E}_\beta = 0.33$  Mev) was studied. Mixtures containing nuclear fission products (NFP) of different age were used: 10-14-hour-old, 2- and 20-days-old /19/.

The radioactivity of the products decreased in time according to the power function law providing for different tempos of dose rate reduction for fission products of different ages.

The level of mutant cells in the populations exposed to a NFP mixture is proportional to the dose rate (Fig.7). When a mixture of 20-day-old nuclear fission products, in which mainly long-lived radionuclides remain by this time, is used in experiments the character of mutation dynamics is similar to that at chronic exposures of *Chlorella* cultures to radionuclides with a long half-life ( $^{90}\text{Sr}$ - $^{90}\text{Y}$  and  $^{147}\text{Pm}$ ) /19/.

Some experiments with NFP mixtures dealt with populations exposed to irradiation with a complex character of dose rate formation in time due to the presence in a mixture of numerous radionuclides with different half-life lengths. In this connection, it was of interest to study the mutation dynamics under the action of a radionuclide with a comparatively short half-life. Therefore experiments with  $^{90}\text{Y}$  ( $T_{1/2} = 2.5$  days,  $E_{\beta} = 0.95$  Mev) were carried out. The character of population damage induced by  $^{90}\text{Y}$  is similar to the mutation dynamics for 2-day-old NFPs /19/. It follows that  $^{90}\text{Y}$  as the first approximation can be a NFP imitator in studying the mutation process in populations.

16. Let us compare effects induced in the populations of unicellular algae by ionizing radiations from different sources. Taking into account the fact that the character of dose rate formation in time for the radiation sources used is different, the comparison will be based on the maximal levels of mutant and lethally damaged cells observed in the first phase of the experiments: these maximal levels have place with all sources of ionizing radiation.

A reduction in genetic efficiency is observed as radiation energy is increased:  $\beta$ -radiation from  $^{147}\text{Pm}$  ( $\bar{E}_{\beta} = 0.06$  Mev) has the highest efficiency,  $\beta$ -radiation from  $^{90}\text{Y}$  ( $\bar{E}_{\beta} = 0.93$  Mev) has the lowest efficiency. This observation is most obvious if the relationship between maximal levels of mutant and lethally damaged cells and an average energy of  $\beta$ -particles is considered (Fig.8). The results of different experimental variants are presented for a dose rate of 1 Gy/day. It can be asserted that mutagenic and lethal effects of radiations in the populations of unicellular algae per equal dose rates and therefore per equal accumulated doses per volume unit are reduced as an average energy of  $\beta$ -radiation is increased. The level of lethally damaged cells for all  $\beta$ -radiation sources is on the average 4-6 times higher than the level of mutant cells /21/. This regularity in the general form will be probably observed under the action of other kinds of ionizing radiation. It may be expected that this regularity is specific of not only unicellular algae.

Genetic Processes in Chronically Irradiated

Natural Populations after Kyshtym accident

17. Using natural populations of microorganisms, plants and animals chronically exposed to  $\beta$ -radiation from  $^{90}\text{Sr}$ - $^{90}\text{Y}$  after Kyshtym accident contaminated areas the effect of ionizing radiations on the mutation level in these populations was studied /2,4,5,15,21,22/. The experiments with chronic irradiation of natural populations have been carried out for about 20 years. During this period  $^{90}\text{Sr}$ - $^{90}\text{Y}$  have joined in the turnover of substances in biocenosis and the concentration parameters of these radionuclides in different elements of biogeocenosis have reached the stationary level. Accumulated doses are received by various biological objects from the external exposure of these objects to the contaminated soil and vegetation and from incorporated radionuclides. Irradiation intensity reduces in time due to  $^{90}\text{Sr}$  decay. Accumulated doses were determined by means of thermoluminescent dosimetry, photocassettes and estimated proceeding from the concentration of the radionuclides in various biological tissues. No essential differences between dose values obtained by these methods have been revealed.

Below there are data on genetic variability and radioresistance of natural populations of higher and lower plants and several animal species in areas with increased environmental radioactivity. It can be believed that the results of analysis of dose-response curves for a number of objects and the data on the mutation dynamics in model populations presented in the preceding chapters will serve to more profound understanding of genetic processes in chronically exposed natural populations in which a detailed analysis of the events observed is not always possible.

a) Increase in radioresistance of chronically irradiated *Chlorella vulgaris* populations

18. Various forms of *Chlorella vulgaris* were obtained from soil samples collected in isolated experimental areas in the zone of Kyshtym accident.  $^{90}\text{Sr}$ - $^{90}\text{Y}$  concentrations in these areas were in the range of  $3.7 \cdot 10^4$  -  $13 \cdot 10^{-7}$  Bq/m<sup>2</sup> permitting exposures of algal populations at dose rates from  $1 \cdot 10^{-4}$  to 0.13 Gy/day. The material was collected 5, 6 and 11 years following the contamination of the soil. The samples were placed in a nutrient medium, the isolated *Chlorella* clones were purified of the concomitant microflora, and radioresistance of the control and chronically exposed algal populations was analysed /2/. The cell survival rate in *Chlorella* strains additionally exposed to X- or  $\gamma$ -rays at a dose of 300 Gy was estimated. In all, 316 *Chlorella* strains were isolated. The calculations show that after the accident over 100 generations had passed in the *Chlorella* populations by the last time the natural material was collected /21/.

The *Chlorella* forms obtained from samples taken in areas with different  $^{90}\text{Sr}$ - $^{90}\text{Y}$  contents are samples from isolated populations, therefore an average radioresistance of such groups of clones adequately reflects radioresistance of the populations under study.

The *Chlorella* forms isolated 5 and 6 years after the accident were additionally exposed to X-rays at a dose of 300 Gy. The data on the average radioresistance of the strains isolated in areas with different concentrations of the radionuclides in the soil are presented in Fig.9. The data for all isolated forms including those that are relatively nonresistant to radiation (for instance, pigmental mutants) have been included in the analysis.

A noteworthy fact is an increase in radioresistance of the *Chlorella* forms at all levels of environmental radioactivity ; maximum is observed at medium levels of soil radioactivity. At the highest levels of environmental radioactivity the radioresistance of *Chlorella* is gradually reduced. Thus, there is an optimal level of environmental radioactivity for radioresistance selection in *Chlorella*. At radioactivity levels exceeding the optimal one the genetic load in the population probably increases due to accumulation of a great number of mutations reducing the general viability of the population.

LD<sub>50</sub> calculations indicate that radioresistance of *Chlorella* populations under increased radiation conditions is 1.5-2.0 time higher on the whole than that of the control population.

The irradiated populations show an increased yield of visible mutations (mainly pigmental mutations) which is proportional to the dose rate of chronic irradiation. Pigmental mutants isolated from areas with high  $^{90}\text{Sr}$ - $^{90}\text{Y}$  concentrations in the soil are, as a rule, more radioresistant than the control *Chlorella* strains but less resistant as compared to the *Chlorella* strains with the normal phenotype isolated from the same areas. This indicates that pigmental mutants in areas with high  $^{90}\text{Sr}$ - $^{90}\text{Y}$  concentrations take their origin from those radioresistant forms which have already occupied these areas.

19. The same *Chlorella* populations were examined 11 years after accident /21/. The isolated algae forms were additionally exposed to  $^{137}\text{Cs}$   $\gamma$ -rays at a dose of 300 Gy and the survival rate of *Chlorella* cells at this dose was determined (Table 1). Again all irradiated populations displayed increased radioresistance. Along with the increase in the average survival rate in the irradiated populations there is an increase in population radioresistance on the whole with increasing  $^{90}\text{Sr}$ - $^{90}\text{Y}$  concentration; it manifests itself in an increase of the minimal and maximal survival rates in the strains of the irradiated populations at an acute dose of 300 Gy. No marked increase in the survival variance ( $\sigma$ ) is observed, and thus the variation coefficient reduces from 84.5 to 45 as the level of radioactive environmental contamination is increased. These data suggest the selective action of increased radiation levels.



The fact that there are similar results for all three periods of material collection indicates to radioresistance stabilization in the populations already by the 5th years after the accident.

The data presented show that there is an exponential increase in radioresistance of the populations with increasing level of environmental radioactivity. The curve slopes are similar and correspond to a 25% increase in population resistance with a 10-fold increase in the dose rate.

20. It was of interest to find out if radioresistance of the chronically exposed algae populations changes depending on the season. *Chlorella* forms were isolated in two seasons - in June when intensive reproduction of algae begins, and in October when algae populations practically stop reproducing /20/. It has turned out that populations essentially change their radioresistance in autumn as compared to that in summer (Fig.10). The following conclusions can be derived from the data considered: 1) almost all pigmental mutants as well as phenotypically normal but radiononresistant forms disappear from populations in autumn; 2) an autumn population represents an aggregate of phenotypically normal radioresistant form. The observed cycle in the mutation intensity in *Chlorella* populations is related with periodicity of the reproduction of the algae in nature. In the winter period, due to the absence of reproduction, the population accumulates a large dose (up to 200 Gy) during the cell cycle in areas with the maximal content of  $^{90}\text{Sr}$ - $^{90}\text{Y}$ . Therefore examination of the populations in spring reveals a much higher yield of mutants than in autumn when a large part of mutants is already eliminated in accordance with the dynamic regularities considered above. The cycle observed in the mutation load level in natural populations probably plays a decisive role in acceleration of selection of radioresistant forms. A critical moment is the winter period when accumulated doses per cell cycle can achieve the level which is sufficient for lethal damages of most individuals in the population, particularly of radiosensitive ones.

21. Experiments have been carried out to study the nature of increased radioresistance of *Chlorella* forms from the exposed populations. Experiments with repair inhibitors, analysis of dose-response curves for low-ionizing and high-ionizing radiations, as well as direct estimation of repair activity have led to the conclusion that radioresistant forms have more active repair systems as compared to the control /21/.

b) Effect of ionizing radiations on natural populations of herbaceous plants after Kyshtym accident

22. A high content of radionuclides in the environment induces the mutation process in populations of various organisms, the intensity of this process being proportional to the dose rate of chronic irradiation. As a result, a high mutation level is constantly maintained in populations /4,15,22/.

Increased frequencies of genetic changes have been revealed using the following tests: chromosome aberration frequency in mitotic divisions in root meristems, chromosome aberration frequency in anaphases of meiosis, frequencies of chlorophyll mutations, embryonal lethals and biochemical mutations.

23. The data on *Draba nemorosa* L. can give an idea of the damaging effect of chronic irradiation in natural plant populations after Kyshtym accident. The germinating ability of seeds, fertility of pods, the level of chlorophyll mutations and embryonal lethals in plants grown from seeds collected in the control and several chronically exposed populations were determined (Table 2). It is evident that the experimental populations, except one, do not differ by the germinating ability of seeds. However, the chronically irradiated populations are characterized by increased levels of chlorophyll mutations and embryonal lethals.
  
24. *Centaurea scabiosa* L. populations in the zone of Kyshtym accident were analysed for variations in the electrophoretic mobility of leucinoaminopeptidase (Lap), a highly polymorphic enzyme (Shevchenko et al., 1982). In addition to the analysis of isoenzymes in plants growing in radiocontaminated areas, isoenzymes in the offsprings of these plants were analysed. This permitted the genotype of the parental forms to be specified and the character of inheritance of identified alleles to be studied. Special studies revealed invariability of the plant genotype in morphogenesis as well as identity of allelic combinations in tissues of different organs of the same plant /24/.

Two experimental and one control populations of *Centaurea scabiosa* with similar ecological conditions were examined. The dose rates of chronic irradiation were  $0.6 \cdot 10^{-2}$  Gy/day (population II) and  $1.2 \cdot 10^{-2}$  Gy/day (population III), respectively. All three populations have proved to be polymorphic for the locus under study and characterized by equal levels of homozygosity (about 0.5). The Lap synthesis has been found to be controlled by one locus with three basic alleles. There are also mutant alleles. Their frequency was 0.4% in the control population, 6.6% in population II, and 4.5% in population III. Thus, the frequency of mutation events in the control population was by an order of 1 lower than in the chronically exposed populations ( $P < 0.001$ ). The same situation is observed by the test of chlorophyll mutations detected in seedlings grown in a thermostat in Petri dishes (control - 0; population II - 0.52; population III - 1.56%). It should be noted that the variability related to a whole class of chlorophyll mutations controlled by hundreds of loci. This is probably due to the fact that mutant Lap variants do not cause essential changes in the viability of individuals. It is also probable that new Lap variants result from recombination processes (due to intragenic non-reciprocal recombinations). It should be noted in this connection that the variability for monomorphic loci in *C. scabiosa* and in other chronically to an expected effect calculated from the standard frequency of gene mutations per dose unit.

25. To test regions with populations of wild herbaceous plants chronically exposed to  $^{90}\text{Sr}$ - $^{90}\text{Y}$  in the zone of Kyshtym accident, the *Tradescantia* clone 02 was used. This test system is known to be one of the most sensitive to mutagenic factors [25,26]. Pots with *Tradescantia* plants were placed on sites contaminated with the radionuclides, and the mutation level in stamenfilament hairs (SFH) was studied for 30 days. Dose measurements were made with thermoluminescent dosimeters placed on a level with *Tradescantia* floscules. In the present experiment only external exposure of plants to  $\beta$ -rays was performed. Fig.11 presents summarized data for all experiments. The experimental materials permit a conclusion that in the SFH system there is significant increase in mutagenesis beginning from a dose rate of  $1.3 \cdot 10^{-2}$  Gy/day. Lower dose rates do not allow a significant mutagenic effect to be revealed. It should be noted that in these areas wild herbaceous plants are exposed to irradiation by incorporated radionuclides and to external irradiation, the specific weight of the internal exposure being higher than that of the external one. This is probably why mutagenic effects in wild plant populations are higher than in *Tradescantia*.

26. At a sufficiently high dose rate the radioactive radiation becomes an effective factor of selection acting in favour of radioresistant forms of organisms. In the final analysis, a dynamic equilibrium is established in populations between the mutation process and selection determined by usual ecological conditions and by conditions of radioactive contamination.

It has been demonstrated that plants growing in areas with  $^{90}\text{Sr}$ - $^{90}\text{Y}$  radioactive contamination become more radioresistant [15,22]. Radioresistance was determined by the level of chromosome aberrations in root meristem cells of seedlings from the contaminated and control areas after an additional gamma-irradiation of the seeds.

Higher radioresistance manifests itself in the fact that in plants from the contaminated areas the level of chromosome aberrations at the same irradiation dose is lower than in plants from the control areas (Fig.12). This phenomenon, called radioadaptation, is observed in many species of wild herbaceous plants.

27. Let us consider some features of the radioadaptation phenomenon. The level of radioresistance in plant populations growing for a long time in the environment with a high concentration of  $^{90}\text{Sr}$ - $^{90}\text{Y}$  depends on the content of the radionuclides in the environment (Fig.13). The highest radioresistance is displayed by plants growing in areas with the maximal  $^{90}\text{Sr}$ - $^{90}\text{Y}$  concentration. An increased radioresistance is detected over the whole dose range of additional gamma-irradiation (Fig.14).

28. The structure of *Centaurea scabiosa* L. populations from two areas with a high concentration of  $^{90}\text{Sr}$ - $^{90}\text{Y}$  in the soil ( $3.7 \cdot 10^7$  and  $13.7 \cdot 10^7$  Bq/m<sup>2</sup>) and from one control area was studied with respect to the radioresistance character. Radioresistance was estimated by the level of chromosome aberrations in anaphase of the first division of embryo root meristem cells after an additional exposure to  $\gamma$ -rays at a dose of 20 Gy. The results are presented in Fig.15.

It is evident that the distribution of plants for radioresistance in the experimental and control populations is different (30-35 plants from each population and 10 seedlings from seeds of each plant were analysed). In plants from the experimental populations an average level of radioresistance is higher about 2-fold as compared to the control. Moreover, the range of plant distribution for radioresistance in the experiment is wider than in the control. The range of variability in the experimental populations was extended due to the appearance of new classes of radiosensitive and radioresistant individuals. No such individuals were found in the control.

29. A more detailed study of the increase in radioresistance under conditions of chronic irradiation has been carried out using pure barley lines, Maya variety /24/. To this end, plants 250 closely related families were grown in Kyshtym areas contaminated with  $^{90}\text{Sr}$ - $^{90}\text{Y}$  ( $5.6 \cdot 10^7$  Bq/m<sup>2</sup>, 0.6 c Gy/day) and in the control area. The families were reseeded during several years. A random sample from the yield of each family was taken in each reseeded.

Fig.16 presents the levels of chromosome aberrations in anaphase I of meiosis in a series of generations of the reseeded barley families. It is seen that in  $M_1$  the frequency of chromosome aberrations in plants from the chronically exposed families is significantly higher as compared to that in the control. In  $M_2$  differences are smaller, and in  $M_4$  the frequency of structural chromosome mutations in the experimental material is reduced to the natural level. In  $M_2$  the frequency of visible mutations (chlorophyll and morphological ones) was analysed. The level of mutant plants in the chronically irradiated families exceeded by 3 times that in the control families. In the subsequent generations the level of mutant plants in the irradiated families was not higher as compared to that in the control families.

Seeds from  $M_4$  plants were additionally exposed to  $\gamma$ -rays at different doses to determine radioresistance by the following criteria: survival rate and chromosome aberration level in the 1st mitosis. The material from the irradiated families has proved to be more resistant as compared to the control by the both criteria.

30. The results of examination of the irradiated populations of wild herbaceous plants and the reseeded barley families show that the levels of radioactive contamination considered here are an efficient factor of selection for radioresistance and that the dynamic equilibrium between the mutation process and selection established after the accident in the plant populations growing on the contaminated soil is shifted towards the reduction of the natural level of mutagenesis.

31. It is important to find out if increased radioresistance in chronically irradiated natural plant populations is a stable character. To determine heritability of increased radioresistance, the frequency of chromosome aberrations was analysed after irradiation of plants from the control and irradiated populations grown for 2 years in the control area /4/. The data for frequencies of chromosome aberrations in *Agrimonia eupatoria* L. leaf meristems obtained in this experiment are presented in Fig.17. The same figure shows the level of chromosome aberrations in the same material but when it is grown for 2 years in the area with a high content of  $^{90}\text{Sr}$ - $^{90}\text{Y}$ . The level of chromosome aberrations was estimated after an additional exposure of the plants to  $^{137}\text{Cs}$

$\gamma$ -rays at a dose of 15 Gy. It is seen that the level of chromosome aberrations in leaf meristems of plants grown from seeds collected in the experimental areas is lower than in plants from the control area. Hence, the level of radioresistance established in plants due to selection under chronic irradiation conditions does not change when the same material is grown for 2 years in the control area or in the area with the highest  $^{90}\text{Sr}$ - $^{90}\text{Y}$  concentration in the soil. Thus, genetic stability of a newly acquired character - increased radioresistance of plants - can be asserted.

32. It may be assumed that the intensity of selection for radioresistance will be different for different species depending on their initial radioresistance. This assumption is confirmed by the experiments with populations of wild herbaceous plants /4/. It has been demonstrated that the highest radiosensitivity is displayed by plant species whose cells have the largest nuclei. An additional exposure to  $\gamma$ -rays induces more chromosome aberrations per 1 Gy in such plants than in plants with small cell nuclei (Fig.18). Similar results by the "survival rate" test have been obtained in the works of Sparrow et al. /8,9/.

In populations of radiosensitive species the selection for increased radioresistance is more intensive than in populations of radioresistant species, the result being a more essential increase in radioresistance in the former than in the latter ones. The most radiosensitive species, such as *Vicia angustifolia*, growing under high radiation condition after Kyshtym accident increase their radioresistance 3.4-fold. Resistant clover *T. montanum*, on the contrary, has not practically changed the degree of radioresistance (only 1.2-fold increase). In the most radioresistant plant species the selection for increased radioresistance will be, probably, more intensive at higher dose rates than those used in the experiment.

33. To explain the nature of radioadaptation, a suggestion has been put forward that higher radioresistance of the experimental plants is determined by the more efficient work of their repair systems. To check this suggestion, experiments with *Centaurea scabiosa* were performed in our laboratory to study the influence of acriflavin, a repair inhibitor, on the effect of additional irradiation of seeds from the control and experimental areas. Seeds of several tens of plants from the experimental ( $11.1 \cdot 10^7$  Bq/m<sup>2</sup>,  $^{90}\text{Sr}$ - $^{90}\text{Y}$ ) and control areas were used in the experiments.

Seedlings from seeds of irradiated plants appeared to be twice more resistant to ionizing radiation as compared to the control. Moreover, the relative effect of acriflavin in the case of irradiation of experimental seeds was higher than in the control families. The level of chromosome aberration under the action of acriflavin by 40% in the control material and by 109% in the experimental material (irradiation dose - 30 Gy). These data permit a conclusion that in radioresistant forms the repair systems function more efficiently.

c) Effect of ionizing radiation on animal populations living in the zone of Kyshtym accident

34. The influence of radioactive  $^{90}\text{Sr}$ - $^{90}\text{Y}$  contamination and natural selection factors on *Lymnea stagnalis* freshwater gasteropod mollusk populations has been studied /15/. Three isolated populations were used: I - a control population of large size; II - a control population living in a shallow lake lagoon which is periodically dried up and frozen through to the bottom; III - an experimental population living in an undrying and unfreezing water basin contaminated with  $^{90}\text{Sr}$ - $^{90}\text{Y}$  ( $11.5 \cdot 10^3$  Bq/l) (irradiation for about 20 years). All populations were studied by polyacrylamide gel electrophoresis. An analysis for 13 enzyme systems was carried out. Allelic frequencies for each enzyme system were calculated. The frequencies of mutant alleles were 0.01 (I), 0.01 (II) and 0.02 (III), respectively. It is seen that there are no essential differences in the mutation level between the populations. An average level of heterozygosity was: I - 0.38; II - 0.24; III - 0.27. A deficit of heterozygotes is observed, especially in population structure more efficiently than radioactive environmental contamination, if the intensity of their action is sufficiently high.
35. A cytogenetic analysis of pike (*Esox lucius* L.), perch (*Perca fluviatilis* L.), roach (*Rutilus rutilus lacustris* L.) and mollusk (*Lymnea stagnalis* L.) developing eggs was carried out in a series of generations in an experimental reservoir in the zone of Kyshtym in which water activity for  $^{90}\text{Sr}$ - $^{90}\text{Y}$  was equal to  $5.9 \cdot 10^3$  Bq/l/28/. Eggs were collected on spawning sites in the closely located experimental and control (water activity  $n \cdot 10^{-12}$  Ci/l) reservoirs. The eggs were incubated to the larval stage and the cytogenetic analysis was carried out with the larvae. The results of the cytogenetic analysis are given in Table 3. It is seen that a significant increase in the yield of aberrant anaphases as compared to the control is observed only in the pike larvae. However, the significance of differences with the control may be explained by the fact that the spawning of pike proceeded in the experimental reservoir at 16°C and in the control reservoir at 5-10°C, and this could result in an increased yield of aberrant anaphases as shown in experiments on loach eggs with temperature modifications. The authors make a conclusion that a maximum permissible concentration of  $^{90}\text{Sr}$ - $^{90}\text{Y}$  in water reservoirs is  $n \cdot 10^2$  Bq/l for herbivorous fishes and  $n \cdot 10^4$  Bq/l for predators, but these values will vary depending on hydrochemical characteristics of reservoirs.

36. Populations of redbacked vole *Clethrionomys rutilus* and common field mouse *Apodemus sylvaticus* were studied in regions of Kyshtym accident with artificially added  $^{90}\text{Sr}$ - $^{90}\text{Y}$  ( $3.7$ - $5.6$  and  $6.7$ - $12.6 \cdot 10^7$  Bq/m<sup>2</sup>) and in a control region /4/. 25-30 generations passed in the populations in the experimental areas from the time of contamination till the start of the studies. Increased frequencies of chromosome aberrations and aneuploid karyotypes were observed in the irradiated animals (Table 4). No karyotypes with hereditary changes were found in the animals examined, which would be an indication of the cumulation of genetic effect in the generations. The data show that a chronic exposure suppresses proliferation of bone marrow cells. The mutation level in bone marrow cells of the chronically irradiated animals is higher than in the control. Genetic damages increase with increasing irradiation intensity.
37. To study comparative radioresistance of animals from the control and chronically irradiated populations, groups of animals from these populations were given single doses of  $^{90}\text{Sr}$ - $^{90}\text{Y}$  ( $14.8$ ;  $26.9$  or  $37 \cdot 10^4$  Bq/g b.w.) and the survival rate of the animals was analysed for 30 days /4/. An injection of  $37 \cdot 10^4$  Bq/g b.w. caused the death of all experimental voles from the irradiated population 1 day after the isotope injection. All animals in the group from the control population also died but during 25 days. An injection of  $26.9 \cdot 10^4$  Bq/g caused 10% death in the group of control animals and 60% death in the group of chronically irradiated animals. Common field mice are characterized by higher resistance to  $\beta$ -irradiation as compared to voles. An injection of  $^{90}\text{Sr}$ - $^{90}\text{Y}$  at a concentration of  $37 \cdot 10^4$  Bq/g b.w. resulted in 70% survival rate in the group of control animals and 30% survival rate in the group of irradiated animals. At a concentration  $26.9 \cdot 10^4$  Bq/g b.w. the survival rate was 100% and 50%, respectively. Thus, animals from the control populations are more radioresistant.
38. To study comparative radioresistance of bone marrow cells in voles from the irradiated and control populations, experiments were carried out for studying mutation dynamics in bone marrow cells after a single injection of  $14.8 \cdot 10^4$  Bq/g. In animals from the chronically irradiated population there was a shift in the dynamics of  $^{90}\text{Sr}$ - $^{90}\text{Y}$  accumulation in bone tissue towards reduction of the time of accumulation and towards a significant increase in the level of isotope concentration in the skeleton. These differences are indicative of higher reactivity of metabolic processes in the organism of voles from the irradiated population. At the same time, the effect of additional  $^{90}\text{Sr}$ - $^{90}\text{Y}$  irradiation in bone marrow cells of these voles is weaker as compared to that in the control animals (Table 5).

A relative mutation tempo, determined by a ratio between the percentage of chromosome damages in the experiment and that in the control, in voles from the control population is higher than in animals from the irradiated population. There is no correlation between general radiosensitivity of animals and radiosensitivity of the chromosome apparatus of bone marrow cells.

This fact suggests, that the problem of animal adaptation to chronic irradiation is very complicated and that radioresistance of individual tissues of the organism is not always indicative of general adaptation of animals to irradiation.

Criteria for estimating genetic consequences  
of irradiation of natural populations

39. The results presented and their analysis permit us to draw some general conclusions characterizing the damaging effect of ionizing radiation on natural populations of microorganisms, plants and animals. These general conclusions follow from the study of the relationship between the yield of genetic changes and the irradiation dose and intensity as well as from the analysis of the mutation dynamics in chronically irradiated populations. Besides, the comparison of a number of objects for radiosensitivity and a number of genetic tests for their resolving ability plays an important role.
40. Dose-response curves for one-shot genetic changes (point mutations, chromosome breaks) induced by irradiation of human, plant and animal populations at a constant dose rate (dose - the function of irradiation time) are linear /6,7/. Correspondingly, the frequency of mutations per dose unit and the doubling dose level are constants. UNSCEAR shares these conclusions /12,13/ which, probably, reflect the true picture of mutagenesis at such irradiation regime.

Dose-response curves for the same genetic changes under another irradiation regime, when a dose is the function of irradiation intensity, are non-linear. As the first approximation, this relationship can be expressed by a power function equation with an index  $n < 1$ . As a result, there is an inverse correlation between the mutation frequency and dose rate at comparatively low irradiation intensities (0.001-1.0 Gy/day) for plant populations.

This conclusion is confirmed by the literature data on mice /29/ and human peripheral lymphocytes /30/. Therefore, low irradiation intensities induce more point mutations and chromosome aberrations per 1 gy as compared to high irradiation intensities. When the damage is significant, the inducible repair system is switched on reducing the relative efficiency (per dose unit) of radiation as the dose rate is increased.

41. It follows from the above-said that a hypothesis of power function relationship between the yield of one-shot genetic changes and radiation dose can be formulated on the basis of the dose-response curves presented by us for cases when a dose is the dose rate function. The mathematical aspect of the hypothesis is thus expressed.



According to the essence of the occurring and assumed processes the hypotheses may be called activational implying that within a sufficiently wide range of dose rates, as they are increased, the activation of repair systems increases, the degree of activation being the dose rate function (up to a dose rate at which maximal activation is observed).

42. It is necessary to emphasize an important role of the natural mutation level in determining the resolution of a test system. An increase in natural variability of a research object is equivalent to a reduction in its radiosensitivity since the natural mutation level determines the size of a sample from a population required for statistical estimation of an induced genetic effect. This is true of low irradiation intensities at which the level of an induced effect is comparable to the natural mutation level.

Taking into consideration that natural populations are limited in size, it is always possible to determine a dose rate at which the genetic effect in a given limited population is practically undetectable. Undetectability of a genetic effect in natural populations is aggravated by the fact that samples used to estimate genetic efficiency of ionizing radiations are also characterized by an increased level of information noise due to intrapopulation variability and differences in ecological conditions.

43. In experiments with chronically irradiated populations the detectability of a genetic effect depends on a test used. The methods most frequently used in radiation genetic include the tests of recessive lethals, dominant lethals, visible mutations, chromosome aberrations in anaphase or metaphase of meiosis and mitosis, specific-locus point mutations. In the recent years works have appeared in which induced mutations were detected by electrophoresis of proteins and enzymes in polyacrylamide and other gels. Special experiments of Valkovick /12/ on detection of mutations in offsprings of irradiated male mice by protein and enzyme electrophoresis have shown that in this case the frequency of mutations per locus per rad is the same as in Russell's experiments using the test of specific locus mutations. It follows that these two methods of detection of point mutations result in no principal differences and may be considered together.

44. Proceeding from the target theory /6,7/ one may expect that the resolutions of the chromosome mutation method and the method of specific locus mutations will be different. Indeed, the break of any chromosome in the cell is enough to cause chromosome aberrations, i.e. in this case the target for radiation is the entire genome, while the induction of a mutation at a specific locus requires the damage of this very locus, i.e. a rather small part of the genome. It is assumed at present that there are about  $1 \cdot 10^4 - 1 \cdot 10^5$  structural genes in the animal genome. Assuming that the probability of induction of a chromosome break and gene mutation by ionizing radiation is approximately equal, the probability of detection a gene mutation at a specific locus is lower by  $1 \cdot 10^4 - 1 \cdot 10^5$  times than the probability of detection a chromosome aberration.

This can be proved by comparing experimental data on induction of chromosome aberrations and gene mutations in mice. As it follows from the materials in the UNSCEAR 1977 report /12/, dominant mutations in mice resulting mainly from translocations occur with a frequency of  $1.1 \cdot 10^{-4}$  per gamete per c Gy in males (spermatogonia) and  $1.5 \cdot 10^{-3}$  in females (oocytes). The frequency of induced specific locus mutations is equal to  $0.3-3.0 \cdot 10^{-7}$  locus/c Gy (the data of several authors). It follows that chromosome aberrations occur at least  $10^3-10^4$  times more often than specific locus mutations. If 10 loci are analysed, the difference is, accordingly, reduced by an order of 1. An analysis of recessive mutations (lethals) in autosomes and sex chromosomes involves thousands of genes, therefore the frequency of recessive lethals is high enough ( $\approx 1 \cdot 10^{-4}$  per gamete per c Gy).

Several conclusions follow from the presented calculations which could be supplemented with the materials for other objects, such as *Drosophila*, fishes and plants. Thus, it can be concluded that the probability of detecting chromosome aberrations induced by radiation (for example, in sex cells) is higher by  $10^{-3}-10^{-4}$  times than the probability of detecting a specific locus mutation in the same sample. In other words, specific locus mutations can be detected in a sufficient number of cases only at doses (sublethal or even lethal) inducing a great number of chromosome aberrations. This conclusion can be illustrated by our data on natural populations of *Lymnea stagnalis* living in an experimental reservoir artificially contaminated with  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  /15/. While no increased frequency of new protein variants is observed in the contaminated reservoir as compared to the control ones, the number of chromosome aberrations in cells of developing *Lymnea stagnalis* embryos tends to increase in this reservoir (Table 3).

45. However, the data on high induced variability of leucinaminopeptidase in *C. scabiosa* seem to refute this thesis. The polyporphic locus Lap displays the same variability level as that revealed for a whole class of chlorophyll mutations. It may be supposed that the high variability for Lap locus is due to intragenic recombinations. As a result of intragenic recombinations between several alleles of the locus, the probability of occurrence of mutation-like events in heterozygotes is essentially increased /31/. The principle "polymorphism generates higher polymorphism" is realized.

With a sufficiently low intensity of selection against new biochemical variants in successive generations the level of such variants may be substantially increased. As for monomorphic loci, their variability due to chronic irradiation will be on the level of standard gene frequencies.

It is important that cross-pollinating plants have a high percentage of polymorphic loci, whereas self-pollinating and self-pollinating plants will be different. Variability for monomorphic loci will include exclusively gene mutations, while in variability for polymorphic loci a great role, growing with accumulation of gene mutations, is played by intragenic recombinations which markedly increase variability of polymorphic loci.

46. As it follows from the above discussion, the most sensitive criterion of detecting the effect of ionizing radiation (by the frequency per dose unit and per biological unit - cell, organism) is the chromosome aberration test. This criterion permits estimating the effect of ionizing radiations on such complex characters as productivity and sterility. There is a high negative correlation between the chromosome aberration frequency in vegetating barley plants exposed to  $^{90}\text{Y}$   $\beta$ -particles and the crop capacity of these plants. The methods of recessive lethal mutations, embryo lethals, and visible mutations (e.g. pigmental mutations in barley) also have sufficiently high resolutions. Analysis of genetic changes by these methods involves hundreds and thousands of genes constituting a large part of the genome.

It is possible at present to forecast radiosensitivity of a species on the basis of its genome characteristics (the number of nucleotides per genome, an average chromosome volume). This follows from the works of Sparrow et al. /8/. Frequencies of gene mutations in species can be also forecasted on the basis of the genome size, as it follows from the works of Abrachamson et al. /32/.

47. We have analysed the materials on the mutation dynamics in irradiated populations, on dose-response relationships and on genetic effects of irradiation in chronically exposed populations. Now the criteria for estimation of genetic effects in populations should be considered. The main attention should be paid to the choice of the most universal criteria permitting a maximally adequate estimation of genetic events in irradiated populations. We suppose that the criteria used by UNSCEAR to estimate the risk of irradiation of human populations /13/ should be taken as a basis. These criteria have been approbated, they proceed directly from the main quantitative regularities of natural and induced mutagenesis. The criteria are as follows.

1. Mutation frequency per dose unit.
2. A dose doubling the natural mutation level.

Taking into account the fact that the dose-response relationships in the experiments considered here are non-linear, the mutation frequency per dose unit and the doubling dose are variable values depending on the dose rate.

The both criteria concern mutagenesis within one generation. When considering population processes, a stationary mutagenesis level observed under the chronic action of ionizing radiation and other mutagenic factors may be used as the main parameter. As we have shown, the stationary level varies in proportion to the dose rate. A detailed analysis shows that there is a non-linear relationship between the stationary level of mutagenesis and the dose, i.e. the same non-linear dose-response relationship is observed with respect to the stationary level of mutagenesis as for the yield of one-shot genetic effects in irradiated populations. Thus, the following criteria may be used to estimate genetic consequences of irradiation when population events are considered.

3. The change in the level of mutagenesis related to the dose rate change by a certain magnitude taken as a unit.
  4. A dose rate at which the natural mutation level in a population is doubled.
48. The criteria presented point out changes in the yield of genetic damages but do not permit us to estimate a total detriment to which these changes lead in a population. To estimate ecological consequences of the action of different factors it is necessary to have a criterion which makes it possible to assess a total detriment in populations from induced mutations. In this connection, of high importance is the question of long-term maintenance of some mutations in populations and of potential hazard of some mutations for viability of the organism. Naturally, chromosome aberrations in root meristems are much less dangerous for viability of a species than, for example, mutations leading to sterility and malformations. Proceeding from the abovesaid, it can be considered expedient to use the following criterion of estimating genetic consequences of irradiation:

5. Genetic detriment from irradiation. The criterion of genetic detriment integrally includes the product of frequencies of all types of mutation multiplied by their relative viability.

The genetic detriment from irradiation for populations of higher plants will be determined by a portion of embryo lethals, chlorophyll mutations, mutations causing sterility, visible mutations, recessive and other mutations and by a relative (as compared to the norm) contribution of these mutant forms to the formation of future generations. The genetic detriment caused by irradiation is an induced population genetic load which is realized phenotypically in successive generations under particular ecological conditions. It should be assumed that estimation of ecological shifts in irradiated communities must take into account genetic detriments of irradiation for different species composing a community.

49. It becomes clear that various ecological factors play a very important role in the formation of a definite mutation level in natural irradiated populations. Here the influence of seasonal conditions on the intensity of mutation process in natural *Chlorella vulgaris* populations and the influence of drying up of a reservoir on *Lymnaea stagnalis* populations is considered. In the natural environment ionizing radiation affects populations together with other extreme factors determining in complex the action of natural selection. It is important to know the specific weight of ionizing radiations in the total influence of various ecological factors on natural populations. It is clear that over a sufficiently wide dose rate range radiation-induced effects may be "hidden" due to the action of other strong environmental factors.

50. In this connection, it would be useful to consider the problem of the effect of low ionizing radiation doses on natural populations. It follows from the above analysis of dose-response relationships that at low irradiation intensities the yield of genetic changes per dose unit may be higher than at high dose rates. This conclusion is of primary importance for human populations. For most natural plant and animal populations of large size an increased efficiency of low radiation doses does not seem to be of great practical value. At dose rates at which an increase of radiation efficiency is observed the contribution of induced mutations to the general variation of the population size and viability (on the background of a colossal reproductive capacity of natural populations and on the background of a strong modifying influence of various ecological factors) is insignificant.
51. In analysing the influence of an increased radiation level on populations and biocenoses it is difficult to estimate radioresistance of populations of different species without the concept of integral population radioresistance. Populations consist of individuals at different stages of ontogenesis. Moreover, populations undergo qualitative changes related to seasonal cycles. Sensitivity of cells and organisms also changes with variation in the degree of influence of ecological factors. Finally, each cell, organism or population has natural limits of radioresistance variability which can be considered as the radioresistance norm (Shevchenko, 1967, 1979). The radioresistance norm is an integral sum of possible radioresistance levels of a given genotype which depend on environmental conditions and on cellular, ontogenetic and other cycles. In course of chronic irradiation of populations of different species their fate in the community will depend on integral radioresistance determined by radiosensitivity of all phases of ontogenesis and by the specific weight of the phases in the formation of succeeding generations.
52. In analysing genetic processes in chronically irradiated natural populations it is necessary to take into account that in addition to the damaging effect on populations ionizing radiations induce in populations the process of adaptation to this new environmental factor. The phenomenon of radioadaptation is regarded as an increase in radioresistance of individuals composing chronically irradiated populations to additional irradiation by high doses.

Microorganisms, plants and animals are all involved in the process of adaptation to chronic irradiation. There are two ways to increase population radioresistance. First, it may be the result of selection in an irradiated population of radioresistant forms occurring spontaneously or due to irradiation. Such way of increasing population radioresistance has been demonstrated for natural *Chlorella* populations. An increase in radioresistance is accompanied by a reduction in the level of induced mutations, which has been demonstrated for chromosome aberrations in developing cells of barley plants and some wild herbaceous plants growing for several years under chronic irradiation conditions. Thus, it can be considered that the mutation dynamics in chronically irradiated populations consists of 4 stages: 1) an increase in the mutation level at the start of the experiment; 2) stabilization of the level of mutant individuals;

3) rearrangement of the population structure due to an increase in the concentration of radioresistant forms; 4) stabilization of the population at a new level of mutagenesis and radioresistance. Second, in addition to selection of radioresistant forms, population radioresistance can increase due to activation of repair processes which has a modifying nature. As a result, of repair activation, organisms may display increased radioresistance for a rather long period of time. Moreover, this effect may be expressed in future generations, but in a weakened form. It is difficult to separate these two ways of radioresistance increase in higher organisms - this is a task of future studies. In our analysis it is important that this phenomenon causes 2-fold reduction of the radiation-genetic damage in populations. Two other ways of population adaptation to chronic irradiation are possible. Adaptation may be achieved through selection of preexistent forms characterized by higher radioresistance, for example, of biennial forms against annual ones. Adaptation may be also the result of compensation of a lethal damage of a part of the population due to an increase in fertility. Such way, called ecological radioadaptation, was observed in mammals (wild mice) /34/ and gambusia /35/.

An increase in radioresistance should be considered as an important element in forecasting remote consequences of exposures of natural plant and animal populations to ionizing radiation. It may be suggested that selection of radioresistant forms in populations of different species will be most efficient under chronic irradiation conditions which lead to an optimal for a given species combination of the induced mutation pressure and the pressure of selection against the genetic load. It should be pointed out that selection of radioresistant forms proceeds on the background of strong genetic damage of populations.

53. On the basis of Kyshtym accident it is possible to try to present a general picture of observed genetic and ecological changes in irradiated communities when the dose rate of chronic irradiation is changed (Fig.19). A dose rate range in which irradiation of natural communities is really possible covers 9 orders of magnitude. As the dose rate increases, we see changes in plant and animal populations including chromosome aberrations, visible mutations, biochemical mutations, elimination of sensitive species, and, as the most serious result, - degradation of the community. The beginning of the arrow in Fig.21 corresponds to a minimal dose rate at which a given effect is first observed (the length of each arrow is of no real importance). A dose rate zone ( $10^{-6}$ - $10^{-4}$  Gy/day) characterized by irregular detection of genetic effects can be distinguished. At higher dose rates some effects are registered clearly enough. An increase in species radioresistance (according to the data for unicellular algae and higher plants) is observed beginning from a dose rate of  $5 \cdot 10^{-4}$  Gy/day. Detection of genetic effects of short-term (for several days or weeks) exposure to an increased ionizing radiation level (for example, in Tradescantia, beans, barley, etc) requires dose rate which are 1-2 orders of magnitude higher than those required for detection of mutagenesis under the action of radiation in a series of generations.

Elimination of sensitive species is observed beginning from a dose rate of  $\approx 10^{-2}$  Gy/day. A further dose rate increase is accompanied by gradual aggravation of the damage in the community. The data presented agree, in principle, with the data of US researchers studying consequences of irradiation of various communities from

$\gamma$ -ray point sources /36-38/, although the main attention in our experiments is paid to genetic effects and in the studies of the authors cited - to problems of general species radiosensitivity. There are differences, however, in absolute values of dose rates at which different effects are revealed. The study of genetic effects in chronically irradiated natural populations permits using tests which are by several orders of magnitude higher in sensitivity (judging by a dose rate scale) than the criterion of general species radiosensitivity determined by the lethal apparently begin from a dose rate causing elimination of the most radiosensitive species. Beginning from a dose rate, so to say, an ecological shift is observed in the ecosystem. According to our data, such ecological shift in case of chronic irradiation of communities may occur at dose rates over  $10^{-2}$  Gy/day.

54. Under chronic irradiation conditions, biocenoses (on the level of which the evolution in the biosphere is going on) will undergo essential changes resulting in biomass reduction, complete or almost complete elimination of some radiosensitive species from the community, substitution of basic species and, due to this, rearrangement of the biocenosis structure. In course of these changes, the intensity and degree of which will depend on the irradiation dose rate, the mutagenic action of ionizing radiations will cause radioresistance mutations in populations, and the selective action of the radiation background will promote selection of such mutations, formation of new genetic systems with new forms of gene coadaptation, new balanced polymorphism systems, etc. The succession of such changes will be related with a gradual increase of the norms of population radioresistance. The mutation process for each of the species composing a community will be characterized in successive generations by specific time parameters of the succession of the main phases of the mutation dynamics, which will manifest itself in the change of the structure of irradiated communities as compared to the control ones (Fig.19). A new structure of biocenosis will depend on what the reserves of evolutionary homeostatic reactions in individual species of the community are.

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T a b l e 1.  
Survival rate of *Chlorella* strains isolated in areas with different  $^{90}\text{Sr}$ - $^{90}\text{Y}$  concentrations in the soil in the case of  $\gamma$ -ray irradiation at a dose of 300 Gy

Dose rate Gy/day $\cdot 10^{-2}$	Number of strains	Average sur- vival rate	lim	P	ev	$\bar{x}$	Significance level with respect to the control
> 0,01	23	12,2 $\pm$ 2,2	0,5-35,8	35,8	84,5	10,3	-
0,06-0,13	21	16,1 $\pm$ 2,6	1,2-36,0	33,8	69,0	11,1	P > 0,1
0,13-0,63	25	18,4 $\pm$ 2,3	1,3-39,9	36,3	62,0	11,4	P > 0,05
0,63-1,26	21	20,3 $\pm$ 2,6	6,9-40,2	33,0	52,0	11,9	P < 0,05
1,26-6,3	17	23,1 $\pm$ 3,0	9,2-42,0	33,0	52,0	12,01	P < 0,001
6,3 >	17	30,1 $\pm$ 3,4	6,7-58,4	41,0	45,0	13,9	P < 0,001

T a b l e 2.

Germinating ability of seeds, fertility of pods, level of chlorophyll mutations and embryonal lethals in *Draba nemorosa* L. populations

Variants	Dose rate Gy/day	Number of plants examined	Germinating ability		Chlorophyll mutations		Average number of embryos per pod	P	Embryonal lethals, %
			num-ber	%	num-ber	%			
K		13557	5470	40,3±1,9	-	1	0,02±0,02	-	3,1±0,05
07	0,1	6690	1333	18,4±1,7	< 0,001	6	0,49±0,20	< 0,01	9,6±0,1
05	0,2	12068	5193	43,0±1,9	> 0,05	2	0,04±0,03	< 0,05	4,3±0,1
01	0,3	2820	1368	48,5±4,4	> 0,05	11	0,80±0,20	< 0,001	
04	0,4	7603	3103	40,8±2,5	> 0,05	14	0,45±0,10	< 0,001	
03	0,9	1646	626	38,0±7,2	> 0,05	3	0,48±0,30	< 0,01	6,0±0,1 <sup>05</sup>

K - control population,

0 - experimental populations

Table 3.

The yield of aberrant anaphases upon incubation of eggs of some hydrobionts from the experimental and control reservoirs

Species	Dose rate Gy/day·10 <sup>-4</sup>	Number of - particle hits in chromosomes per 1 hour	Number of aber- rant anaphases, %*
Pike	13,0	3,4·10 <sup>-1</sup>	5,4/2,1
Perch	9,7	1,6·10 <sup>-1</sup>	2,6/2,0
Roach	8,7	1,5·10 <sup>-1</sup>	6,2/5,6
Lymnea stagnalis	10,0	-	3,2/1,9

\* Numerator - experimental reservoir  
Denominator - control reservoir

T a b l e 4.

Frequencies of aneuploid cells and chromosome breaks in bone marrow cells of redbacked voles from the irradiated and control populations

Variants	Number of animals	Total number of metaphases	% of aberrant metaphases	% of breaks	Number of aneuploid cells	
					absolute number	%
Control	20	1172	0,08±0,007	0,08±0,0077	29	2,47±0,14
Experiment	13	789	2,15±0,26 P=0,001	2,66±0,57 P=0,001	91	11,5±1,14

T a b l e 5.

Relative mutation tempo in bone marrow cells of redbacked voles from the irradiated and control populations after a single injection

Time af- ter $^{90}\text{Sr}$ - $^{90}\text{Y}$ in- jection	Control area		Time after $^{90}\text{Sr}$ - $^{90}\text{Y}$ injection	Experimental area	
	% of aber- rations	% Exp. % Control		% of aber- rations	% Exp. % Control
10 h	17,96	2,2	10 h	8,69	0,9
1 day	16,27	2,07	1 day	9,0	0,84
2 days	17,88	2,21	2 days	14,68	1,37
7 days	24,13	2,98	5 days	17,0	1,58
14 days	23,3	2,88	6 days	14,6	1,36
21 days	16,11	1,99	7 days	12,13	1,15
28 days	11,76	1,45	8 days	11,65	1,08
35 days	11,35	1,40	9 days	9,73	0,95
42 days	7,38	0,92	10 days	9,64	0,89
Control	8,08		Control	10,73	

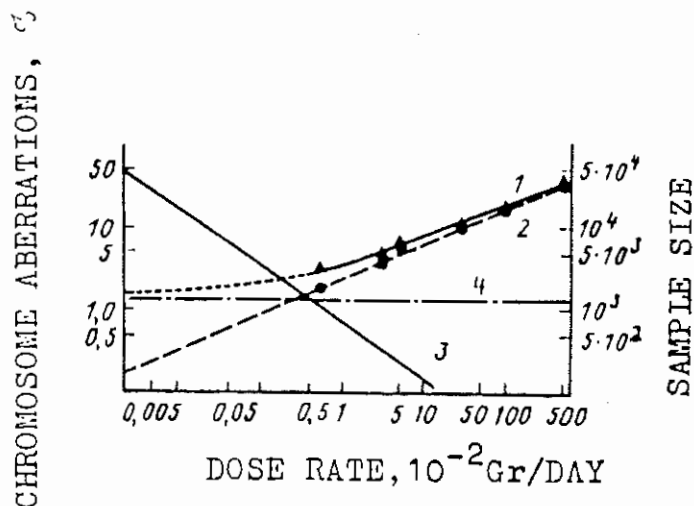


Fig. 1

Relationship between the frequency of chromosome aberrations in cells of *Vicia faba* growing points and the dose rate of  $^{90}\text{Sr}$ - $^{90}\text{Y}$   $\beta$ -radiation.

1 - observed chromosome aberrations; 2 - induced mutagenesis; 3 - cell sample required (99% confidence interval); 4 - control.

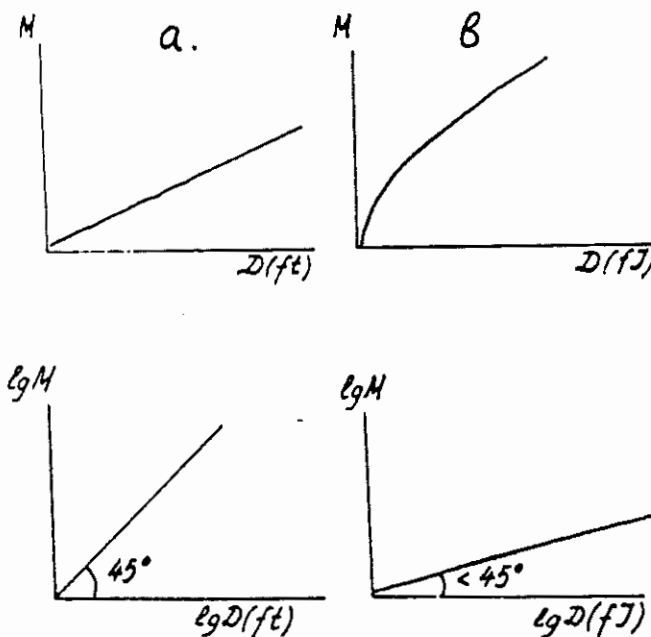


Fig. 2

Relationship between the frequency of one-shot genetic changes (point mutations, chromosome breaks) and the ionizing radiation dose (scheme). Two irradiation regimes used: dose as the function of irradiation time (a) and dose as the function of irradiation intensity (b).



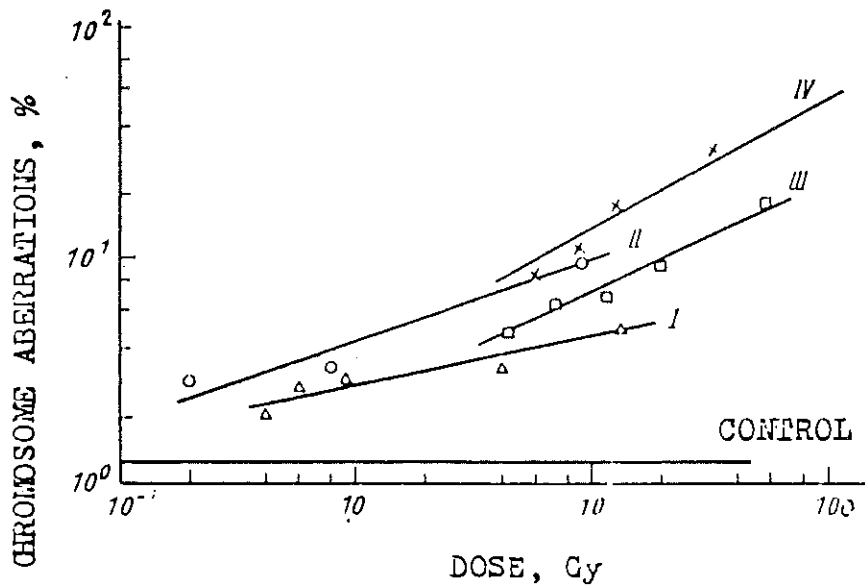


Fig. 3. Chromosome aberrations in anaphase I of meiosis in barley after irradiation with strontium-90 beta-particles at the stages of germination (I), 3 - 4 leaves (II), bushing (III) and tubulation (IV).

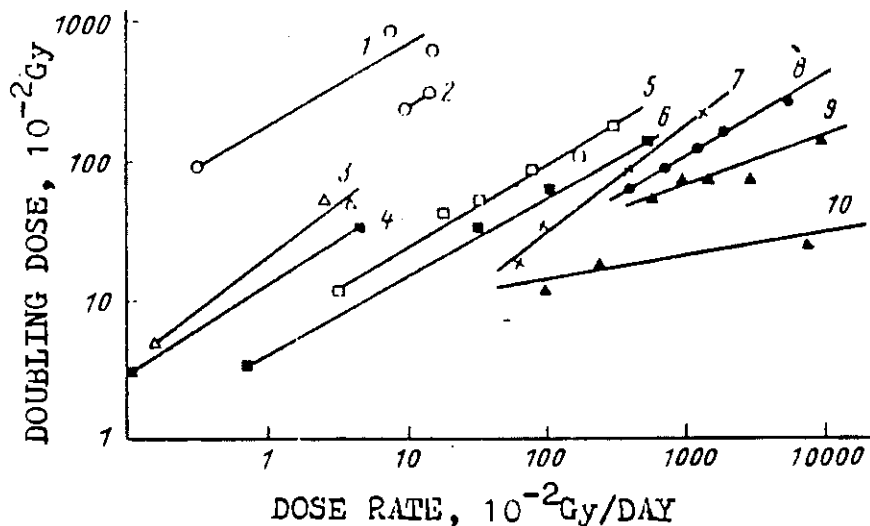


Fig. 4. The doubling dose for natural mutability as a function of dose rate for various biological systems: 1 - Plantago media ( $A_I$  of meiosis), 2 - Veronica tenerium ( $A_I$  of meiosis), 3 - Vicia cracca ( $A_I$  of meiosis), 4 - Centaurea scabiosa ( $A_I$  of meiosis), 5 - Chlorella (visible mutations), Vicia faba (roots, chromosome aberrations), 7 - barley (shoots,  $A_I$  of meiosis), 8 - barley, bushing,  $A_I$  of meiosis), 10 - loach (embryos, chromosome aberrations).

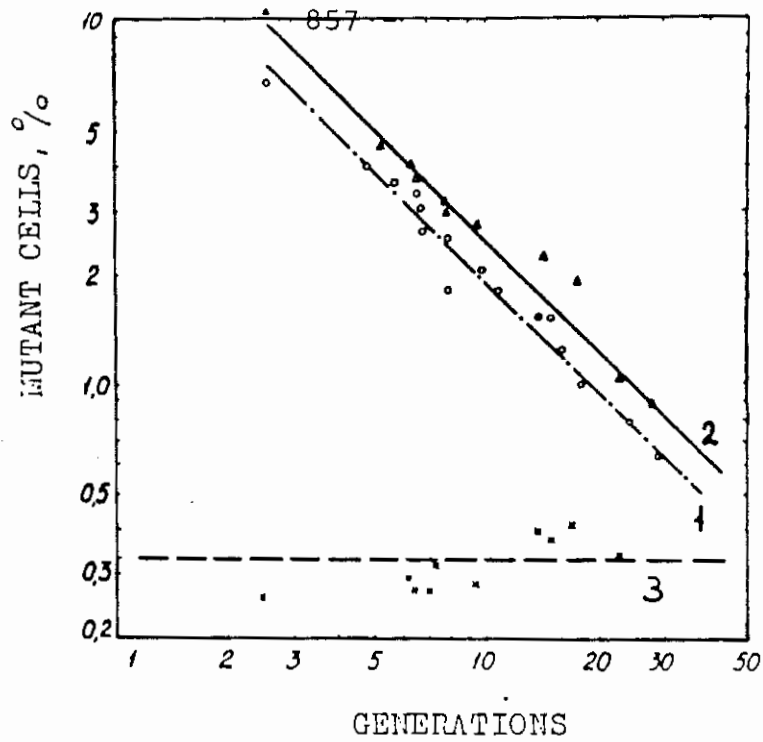


Fig.5. Dynamics of elimination of mutant cells from the Chlamydomonas culture after acute X-ray irradiation at a dose of 100 (O) and 200 Gy ( $\Delta$ ).

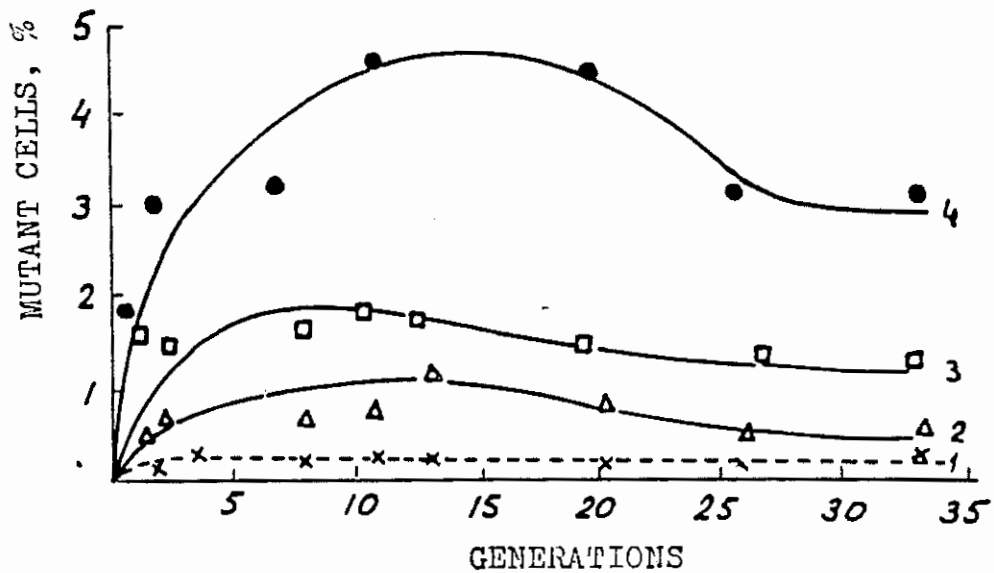


Fig.6. Relationship between the number of mutant cells in Chlorella cultures and the number of density doubling cycles under chronic  $\beta$ -irradiation in  $^{90}\text{Sr}$ - $^{90}\text{Y}$  solutions of different activity:  $\times$  - control;  $\Delta$  - 2.85 Gy/day;  $\square$  - 5.7 Gy/day;  $\bullet$  - 14.25 Gy/day.

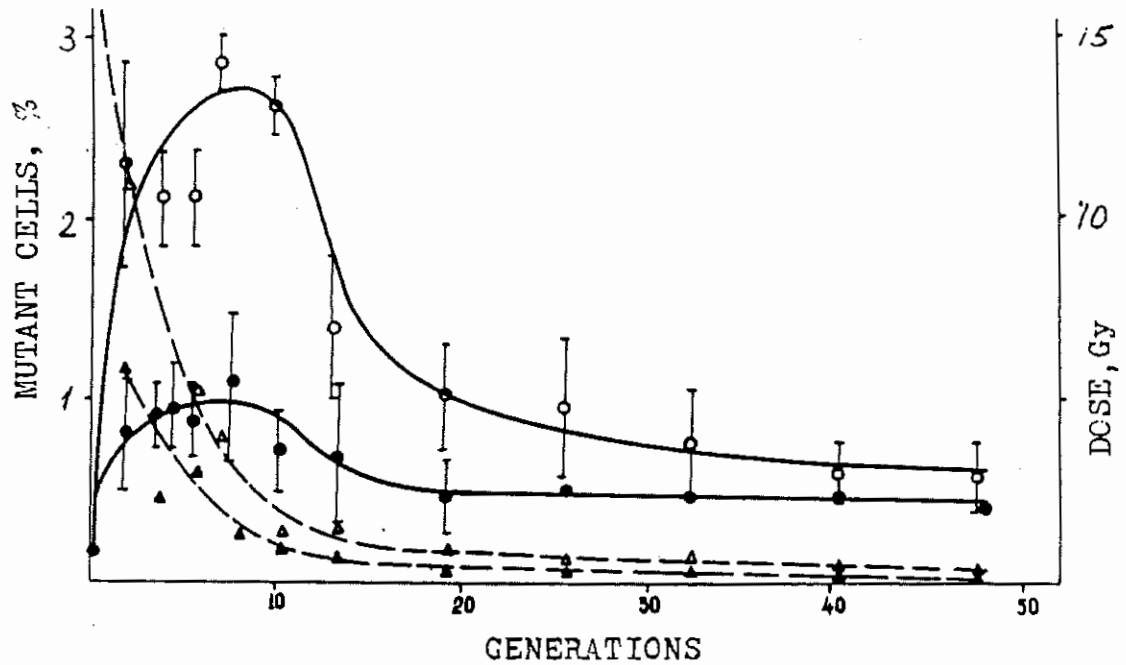


Fig.7. Mutation dynamics in Chlorella cultures under the action of nuclear fission products with an initial activity of  $37 \cdot 10^5$  Bq/ml ( ● ) and  $185 \cdot 10^5$  Bq/ml ( ○ ) (the age of nuclear fission products - 10 h); ▲ - an accumulated dose per density doubling cycle in the culture with an initial activity of  $37 \cdot 10^5$  Bq/ml; Δ - the same for the culture with an initial activity of  $185 \cdot 10^5$  Bq/ml.

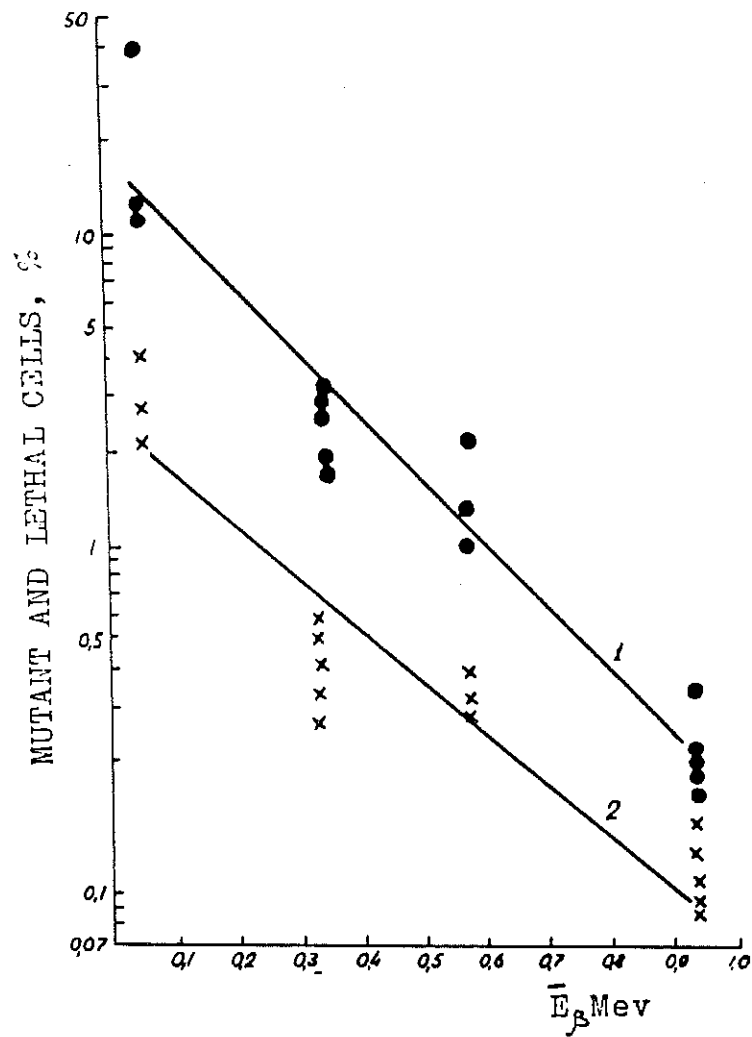


Fig.8. Maximum numbers of mutations (1) and lethally damaged cells (2) depending on the energy of  $\beta$ -particles from different radiation sources (data of experiments with different concentrations of radionucleotides in the environment are presented).

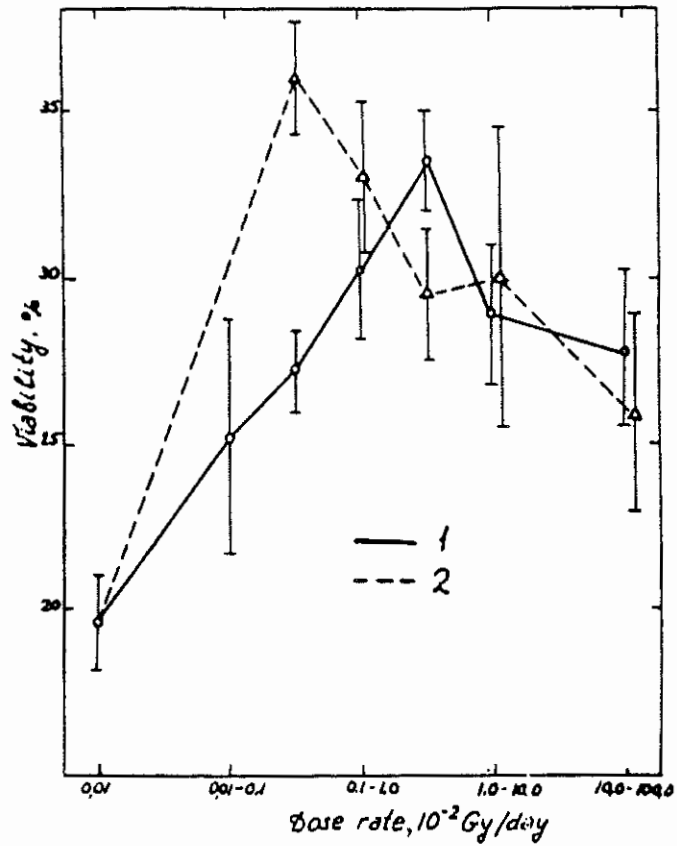


Fig.9. Relationship between radioresistance of natural Chlorella populations 5 years (1, X-ray) and 6 years (2, X-rays) after the start of the experiment and the level of soil radioactivity in the areas of inhabitation of isolated offsprings. Radioresistance of the strains was determined at a dose of 300 Gy.

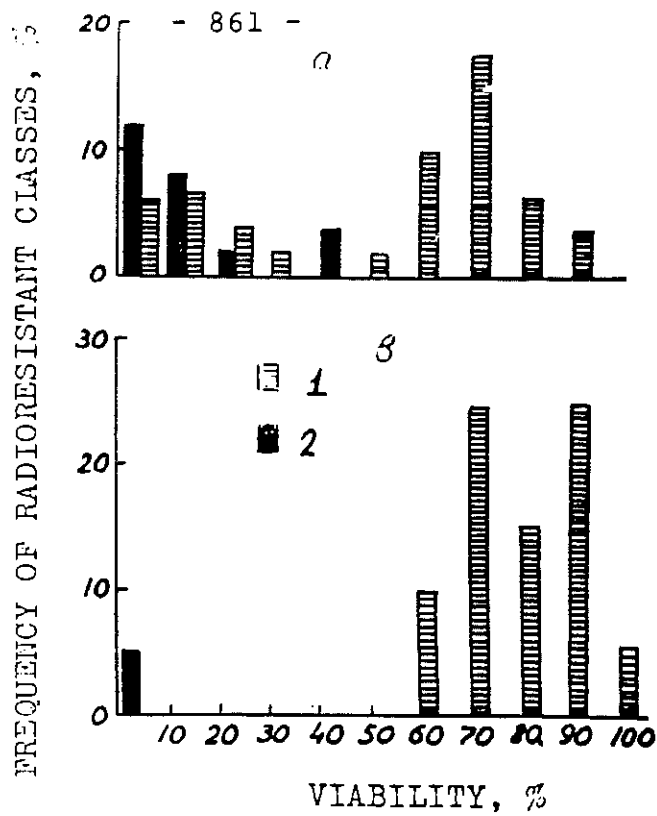


Fig.10. The results of tests of *Chlorella vulgaris* strains isolated in the summer (a) and autumn (b) periods for resistance to  $^{90}\text{Sr}$ - $^{90}\text{Y}$   $\beta$ -radiation.  
 1 - phenotypically normal forms;  
 2 - pigmental mutants.

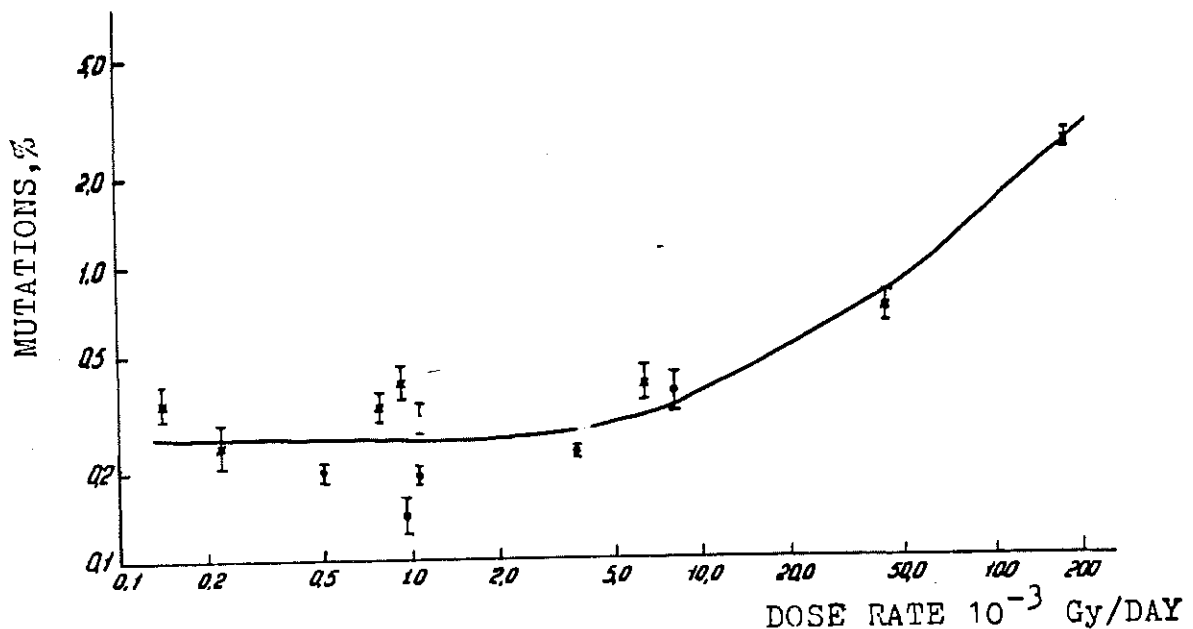


Fig.11. Relationship between the mutation frequency in stamen filament hairs of *Tradescantia* clone O2 and the dose rate of external  $^{90}\text{Sr}$ - $^{90}\text{Y}$   $\beta$ -radiation. Exposure duration - 30 days.

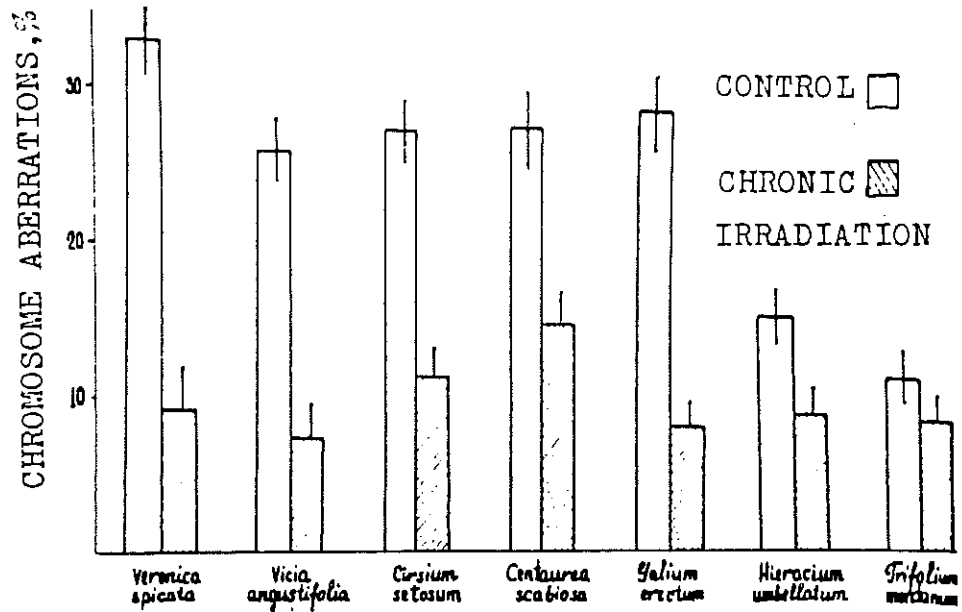


Fig.12. Comparative radioresistance (by the chromosome aberration frequency in meristems of growing points) in a number of wild plant species growing in areas with an increased  $^{90}\text{Sr}$ - $^{90}\text{Y}$  concentration in the soil.

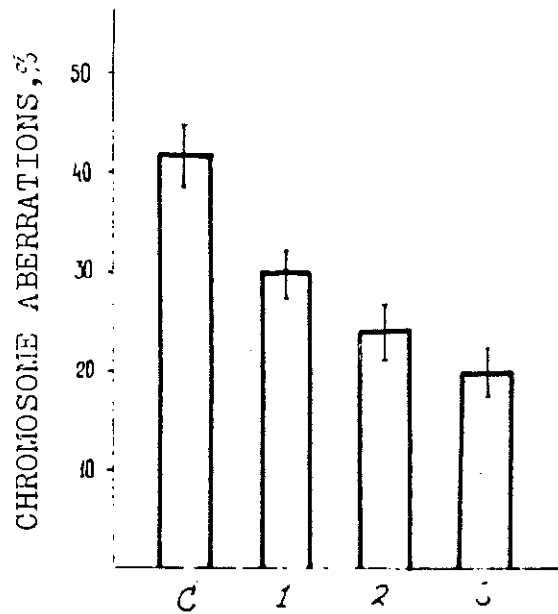


Fig.13. The number of chromosome aberrations in the first mitosis of *Centaurea scabiosa* rootlets in areas with different <sup>90</sup>Sr-<sup>90</sup>Y concentrations after an additional exposure to  $\gamma$ -radiation at a dose of 10 Gy. C - control; 1 -  $3.7 \cdot 10^4$  Bq/m<sup>2</sup>; 2 -  $5.6 \cdot 10^4$  Bq/m<sup>2</sup>; 3 -  $13.7 \cdot 10^4$  Bq/m<sup>2</sup>.

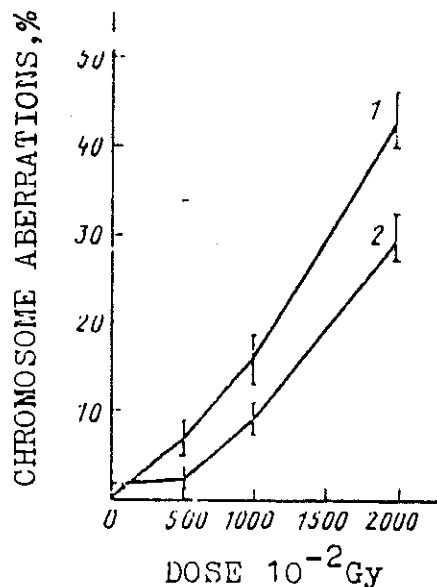


Fig.14. Dose-response relationship for the level of chromosome aberrations in the first mitosis of *Centaurea scabiosa* L. rootlets from the control (1) and chronically irradiated (2) populations.



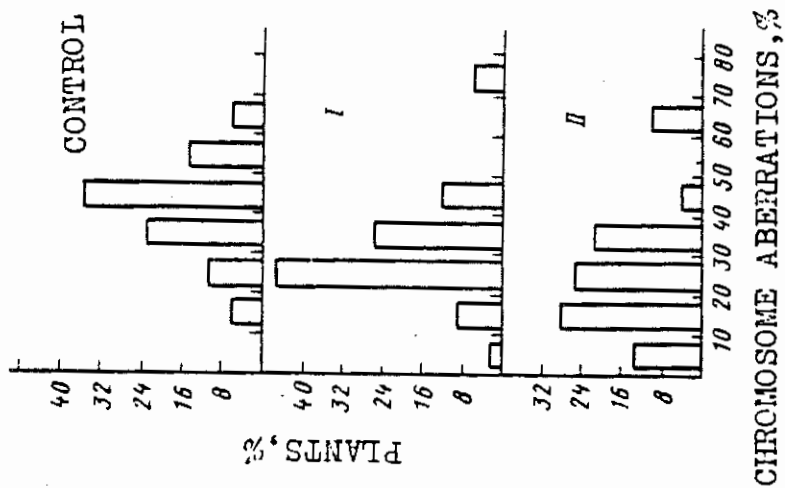


Fig.15. The structure of *Centaurea scabiosa* populations for radioresistance (the test - chromosome aberration frequency in the first mitosis of embryo root meristems). Experimental population I - dose 20 Gy,  $M=30.4 \pm 0.96\%$ . Experimental population II - dose 20 Gy,  $M=19.7 \pm 0.90\%$ . Control population - dose 20 Gy,  $M=42.9 \pm 1.3\%$ .

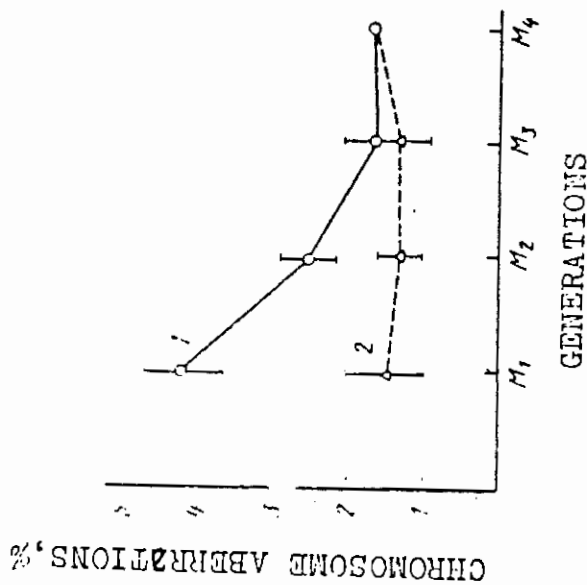


Fig.16. Variation in the chromosome aberration level in meiosis ( $A_1$ ) in a series of generations of reseeded barley families in an area with a  $^{90}\text{Sr}$ - $^{90}\text{Y}$  concentration of  $5.6 \cdot 10^4 \text{ Bq/m}^2$  (1) and in a control area (2).

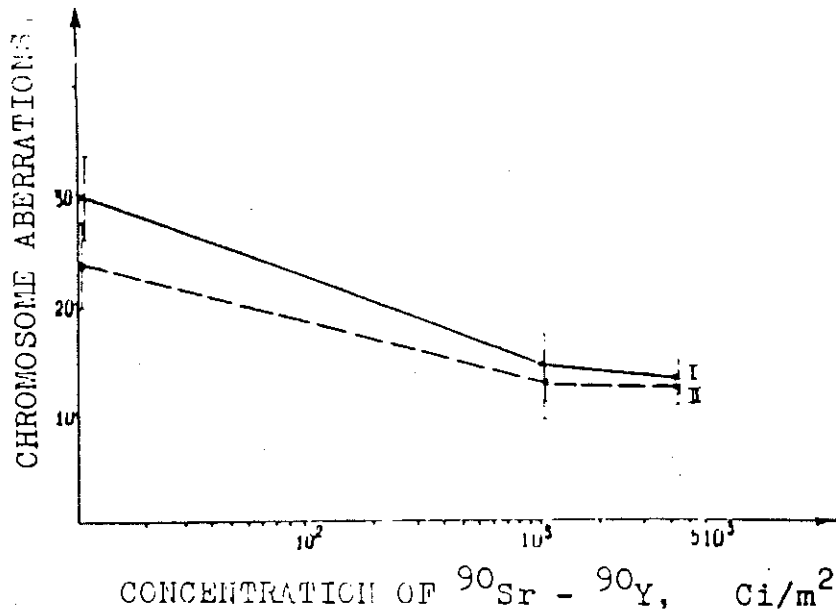


Fig. 17  
Frequency of chromosome aberrations in *Argimonia eupatoria* L. leaf meristems after irradiation (a dose of 15 Gy) of plants from the control (---) and irradiated (—) populations grown for 2 years in the control area and in the material grown for 2 years in an area with a high  $^{90}\text{Sr}$ - $^{90}\text{Y}$  content.

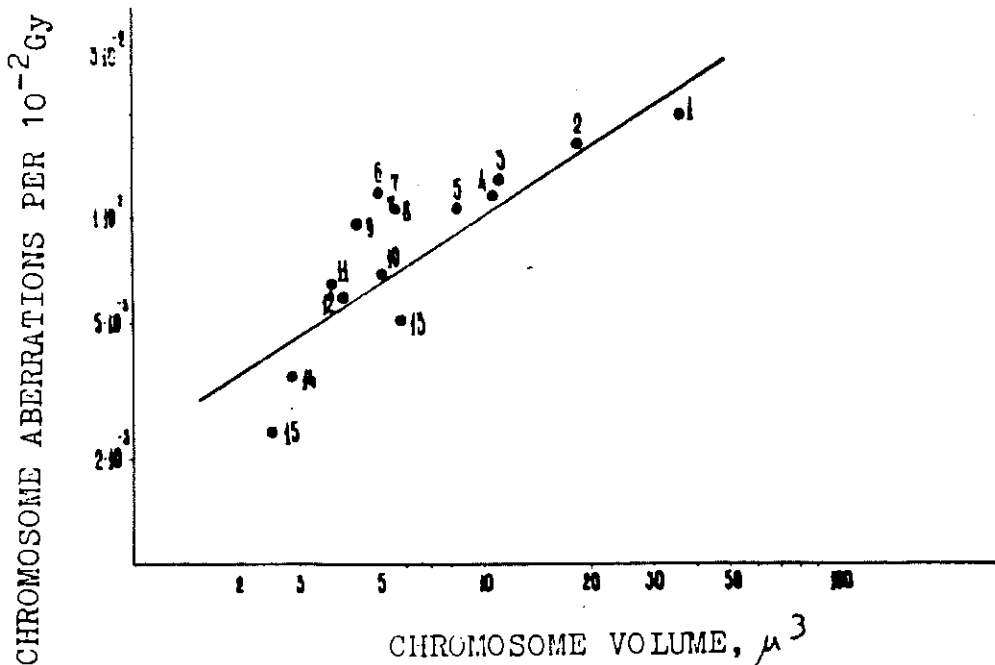
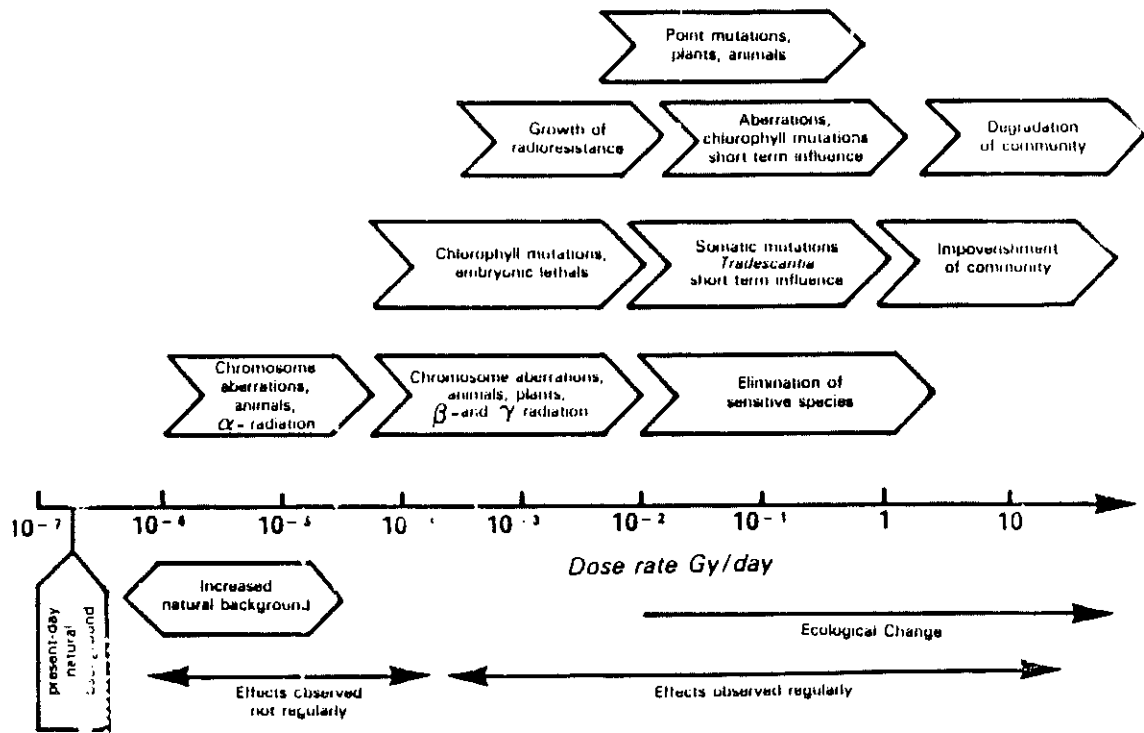


Fig. 18  
Relationship between radioresistance of different plant species (the criterion - chromosome aberration frequency per dose unit) and the nucleus size in cells of these plant species.

Fig. 13. Genetical and ecological consequences of populational exposure to chronic irradiation as a function of dose rate (a scheme). The starting point of the arrow corresponds to the minimum dose rate at which the effect can be observed.



**The Radioecological  
Consequences of the Kyshtym  
and Chernobyl Radiation  
Accidents for Forest Ecosystems**

**F.A. TIKHOMIROV, A.I. SHCHEGLOV**

## ABSTRACT

Following the Urals and Chernobyl accidents 60 to 90% of the radioactive fallout was retained by the above-ground part of forest stands. In the Urals the period for semi-removal of contamination from crowns ranged from 6 to 8 months, compared to around one month in the Chernobyl region - due to different seasonal conditions during the fallout period. The bulk of the dose burden in woody plants' critical organs built up over one to six months. The minimum lethal dose for pine tree needles in the Urals was around 50 Gy, and 25 Gy for the apical meristem; the corresponding figures for Chernobyl were 100 Gy and 25-30 Gy. At lower doses we observed morphological disturbances, reduced growth and suppressed reproductive capability in pines. The resistance to radioactive contamination of deciduous forest was 10-20 times greater than that of conifers.

We studied the irradiation doses of the different groups of organisms living in the various forest storeys, and the effects of irradiation (changes in species composition, prevalence and productivity) in communities of herbaceous plants and soil invertebrates. Specific examples are given to highlight the secondary changes in these communities stemming from radiation damage in species sensitive to radioactive contamination.

We studied the dynamics of dispersion and migration of the long-lived radionuclides  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  in the various components of the biogeocenoses and in the network of geochemically interconnected forest landscapes, and their content in forestry produce.

Some six to ten years after the deposition of radioactive fallout in forest ecosystems the radionuclides were more or less evenly spread throughout the soil-woody plant system. Thus, overall  $^{90}\text{Sr}$  content in the arboreal storey amounts to 1-2% in coniferous forests, and 5-10% in deciduous forests (Urals accident), while the corresponding figures for  $^{137}\text{Cs}$  (Chernobyl accident) are 2 to 3 times higher.

There are three main tasks involved in studying the consequences of radioactive contamination in forest ecosystems:

1. Determining the quantitative indices for primary distribution and subsequent migration of radionuclides in forests;
2. Assessing the influence of dose on the various ecosystem components;
3. Identifying the effects of irradiation at various levels in the biological hierarchy.

A primary requirement for these tasks is information on radionuclide distribution and migration. This is essential for assessing absorbed doses in the various ecosystem components. This in turn makes it easier to interpret the observed effects of radiation and permits forecasting of the long-term consequences of forest contamination. Such information also provides a scientific basis for developing a forestry management system for areas with high levels of radioactive contamination.

Our investigations, involving a comprehensive selection of soil and forest vegetation samples from sites in the Kyshtym accident zone, began in the summer of 1959. Thus, the initial period, when radionuclide migration and redistribution processes were more intensive, was excluded from the observations, but we have reconstructed this initial phase on the basis of data from other researchers.

We have been conducting our work in the forests in the Chernobyl accident zone since July 1986.

#### 1. INITIAL RADIONUCLIDE DISTRIBUTION IN FORESTED AREAS

A radiometric survey of areas within the zones contaminated by the Kyshtym and Chernobyl accidents, carried out directly after radioactive fallout occurred, showed that most of the radionuclide fallout over the forest was retained by the crowns of trees. The radionuclide retention coefficient for trees varied as follows, depending on actual conditions<sup>1</sup>:

Pine forest, 25 years old, crown density 0.8	Kyshtym accident 1957, fallout of particles up to and including 100 $\mu\text{m}$ in diameter	70-90%
Coniferous and deciduous forest, 50-60 years old, crown density 0.7-0.9	Radioactive fallout, particles up to and including 20 $\mu\text{m}$ , Chernobyl accident, 1986	60-90%

Arboreal vegetation has a greater capacity for retaining radioactive fallout than does herbaceous vegetation, the mean retention coefficient for which is 25%.<sup>2</sup> Overall, the retention coefficient for radioactive fallout in the arboreal storey may be regarded as equal to the crown density. The exception to this is deciduous forest during the period in which the trees have no leaves. At such times the retention capacity of the arboreal storey is approximately 3 times less.

Within a few kilometres of the radioactive contamination release point the "forest edge" effect, as it is known, is well pronounced. This is characterized by an increase (2-5 times) in radionuclide deposits in the crowns of trees along forest edges on the windward side facing the contamination source, while forest edges on the leeward side shield adjacent treeless areas from radioactive fallout. After the Kyshtym and Chernobyl accidents this effect was observed some 20-50 m from the forest margin. At greater distances from the release point, where fallout consisted of finely dispersed particles deposited mainly as the result of turbulent diffusion, the forest edge effect was not observed. In general, a tendency was noted for increased deposition of radionuclides over tracts of forest in comparison with adjacent unforested areas. However, the difference does not exceed 20-30%.

## 2. VERTICAL MIGRATION OF RADIONUCLIDES

Vertical migration of radioactive fallout in forests begins immediately after deposition, with the result that radionuclides move from the upper sections of the crown to the lower sections, then down below the forest canopy onto the surface of the forest litter and finally down into the soil. Experiments involving the spraying of woody plants with solutions of radioactive  $^{89}\text{Sr}$  established that migration speed was generally linked to speed of growth.<sup>3</sup> The latter depends primarily on seasonal conditions and, to a lesser degree, on the amount of atmospheric precipitation. In spring, i.e. during the active growth period, this process accelerates; during the dormant phase (autumn and winter) it slows down markedly. This shows that radionuclides migrate down below the forest canopy mainly in solid form, in waxy leaf scales, in the covering scales and caps of buds, and in bark. Thus, the more active the growth process, the faster the rate of transfer of radioactivity from the crowns. This is what occurred after the Chernobyl accident. The time required for semi-removal of above-ground forest contamination, i.e. the time taken for 50% of the total amount of radionuclides to transfer from the crowns to the forest litter surface, was 3-4 weeks in the Chernobyl NPP area, or approximately 2 weeks if radioactive decay is taken into account. Three months after the accident, some 80% of the radionuclides had migrated down below the forest canopy (Fig. 1).

After the Kyshtym accident, in contrast, the autumn conditions resulted in a slower rate of transfer from the crowns. Under these conditions, the period required for semi-removal of contamination from the above-ground pine forest stratum was no less than 6 months. In deciduous forest, leaf-shedding meant that over 50% of the radioactivity in the crowns migrated to the surface of the forest litter during the first two weeks following the accident.

In general, the migration and redistribution of radionuclides among forest stand components may be divided into two stages. In the first stage, lasting up to four years, contamination of the various forest components was mainly linked to the initial deposition of airborne radionuclides onto tree crowns. During this stage, the highest contamination levels were characteristically found in the structural elements having an exposed surface - branches, bark, perennial needles (Fig. 2). The radionuclide content of their phytomass was identical to that of the radioactive fallout. Structural elements protected from radioactive fallout (wood, phloem) contained significantly lower levels of contamination, their radionuclide composition depending on the initial composition of the radioactive fallout.

In the area affected by the Urals accident the radioactive fallout contained radionuclides with a low capacity for basipetal movement in plants ( $^{90}\text{Sr}$ ,  $^{95}\text{Zr}$ ,  $^{144}\text{Ce}$  and their daughter products). Thus, contamination levels in wood and phloem in the years immediately following the accident were relatively low. In contrast, the most significant long-lived radionuclide in the fallout in the Chernobyl accident zone was  $^{137}\text{Cs}$ , which is capable of penetrating the surface of leaves and other organs and contaminating the vascular system, where it can then move easily in any direction within the plant. This was the reason for the appearance immediately after the Chernobyl accident of caesium radionuclide contamination not only externally but also within trees, including in the phloem and the wood (Table 1).

The subsequent radionuclide distribution dynamics, traced over four years, revealed a correlation between the deactivation processes in the above-ground section of tree stands and radionuclide redistribution among their structural elements. Figure 2 shows that radioactivity in branches, pith, needles, leaves and wood declined during the first four years following the Kyshtym accident, reflecting the trend towards vertical movement of radionuclides from the arboreal storey to the surface of the forest litter. Beginning from the third year, however, there was an increase in the concentration of  $^{90}\text{Sr}$  in the vascular layer (phloem) of bark as the result of uptake from the soil. Root uptake increased as the result of decomposition of the forest litter and entry of  $^{90}\text{Sr}$  into the mineral fraction of soil, and the fifth year after the accident saw the beginning of the second stage, in which root uptake of this radionuclide played an increasingly significant role. The result was that, 10-12 years after the accident, equilibrium had been reached between the processes of deactivation of tree stand components and root assimilation of  $^{90}\text{Sr}$ , and its concentration in the structural elements of forest stands became stable.

Under these conditions, the total content of  $^{90}\text{Sr}$  in the above-ground phytomass of the arboreal storey in the Kyshtym accident zone was approximately 1% in coniferous forests and up to 5% in deciduous forests.

A similar pattern was observed in the Chernobyl accident zone. Less time was required (some two years) to achieve near-equilibrium, owing to the high mobility of caesium-137 in woody plants. Deactivation of forest stands was especially rapid in the summer of 1986. During the three months following the accident, the radionuclide content in the arboreal storey fell from 60-90% of overall contamination to 13-15% (Fig. 1). After a year it had fallen to 6-7%, where it remained for the following two years, although there was also some redistribution of caesium radionuclides between the various structural elements of woody plants. Figure 3 shows the distribution and flows of caesium-137 between forest ecosystem components in the Chernobyl NPP zone in 1989.

Table 2 shows that  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  content in various forestry products differs substantially. The lowest contamination levels are normally found in the stem timber and products derived from it (turpentine, resin), while the highest levels are found in mushrooms. There are significant interspecies differences in radionuclide concentrations in phytomasses of similar types. These vary by a factor of 10-15 for the overground phytomass of herbaceous plants, and by a factor of 100 for mushrooms;  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  concentrations in the wood of various woody species differ by 2-3 times. It has been established that, in the Chernobyl accident zone, caesium-137



contamination of all types of forestry products in hydromorphic landscapes is approximately one order of magnitude greater than for eluvial landscapes (Table 3).

### 3. Forest damage by radiation

Analysis of the radioecological information we obtained shows that, of all natural ecosystems, coniferous forests are the most sensitive to radioactive contamination. After the Kyshtym accident pine trees in mixed pine-and-birch forest declined and died over an area of approximately 20 km<sup>2</sup>. Deciduous species covering a total area of 2-5 km<sup>2</sup> were badly damaged. In the Chernobyl NPP zone the corresponding areas were twice as big. The actual amount of dead forest was significantly smaller, however, particularly in the Chernobyl NPP zone, since in many areas which received lethal levels of contamination there were no forests (translator's note: sentence as in original).

The low resistance of coniferous forest to radioactive contamination is due to a combination of factors. Firstly, it is linked to the extremely high sensitivity of conifers to radiation, and secondly to the high dose burden in their vital organs - needles, reproductive system and apical meristem tissue.<sup>1</sup> The latter is due to the large capacity of the crowns of woody plants to retain radioactive and other types of fallout. The essential factor in the formation of dose burdens is the long period of time radionuclides spend in the crowns, together with their relatively slow decontamination rate (particularly during the physiological rest period). Another important factor in dose burden formation in plants is the particular morphological characteristics of their overground organs, particularly the dimensions of needles and leaves. They are commensurate with the penetration range of the beta particles emitted by radionuclides, and all tissues in these organs are available for beta uptake. They absorb a significant amount of beta radiation. Thus, it is beta rather than gamma radiation which is the dominant factor in the formation of external irradiation doses in the vital organs of plants (in contrast to large animals and human beings) in radioactive fallout conditions (Fig. 4). Where the fallout is a mixture of fission products, the contribution of beta radiation to the dose burden in needles amounts to 90-99% of the full absorbed dose, and the size of the absorbed dose itself in these organs, other conditions being equal, is 1-2 orders of magnitude greater than the external irradiation dose for large mammals. In external irradiation of the cambial meristem, dormant buds and root system, which are shielded from radioactive fallout by the phellogen or by soil, the beta radiation contribution is considerably lower.

In the Urals accident, our estimate of the minimum lethal dose for a moribund pine tree was 50 Gy in the needles and 15-25 Gy in the apical meristem tissues. The values given are close to the harmful doses associated with acute irradiation. In the particular case we studied, these doses were accumulated over the seven winter months following the accident, i.e. the irradiation was chronic in nature. It is known that, when subjected to chronic irradiation, growing plants withstand dose burdens an order of magnitude higher than those resulting from acute irradiation.<sup>5</sup> The question therefore arises as to the degree of correspondence between the above-mentioned lethal dose values and those given in the literature. The answer is that after the Kyshtym accident, the chronic irradiation of woody plants took place during their physiological rest period, when their metabolism was suspended and

healing processes were at a low level. The dose therefore accumulated in the plant tissues throughout the winter. Upon the resumption of metabolic activity in the spring, the entire accumulated dose had an impact similar to that of an acute irradiation dose. The main damage was to the apex, this being the most radiation-sensitive organ. Seedlings, protected by the crowns of their parent trees, suffered less damage.

In the Chernobyl NPP zone, where radioactive fallout occurred during the physiologically active phase, the lethal effects of pine forest irradiation became apparent at doses of 10 Gy and above.<sup>6</sup> According to our assessment, however, based on the results of field observations and calculations, the data on lethal doses set out in <sup>6</sup> evidently take into account only external gamma radiation and are understated by several times. If one includes the contribution of beta radiation in the absorbed dose, the lethal dose in the pine needles of moribund pine forests was approximately 100 Gy; the corresponding dose accumulated in the apical meristems of buds was 25-30 Gy.

Thus, the lethal effects in pine forests in the Chernobyl NNP zone occurred in growth sites (the apical meristem) at approximately the same absorbed dose values as in the Urals accident. In the former, absorbed doses in the needles of moribund pine forests were approximately twice as high as in the latter. This indicates that coniferous forest is more resistant to radioactive fallout during the growth period than during the physiological rest period, despite the fact that its radiosensitivity as such is significantly higher in the former.

It was approximately two years before the full effects of radioactive contamination in forests in the Kyshtym and Chernobyl accident zones became apparent. The first signs of radiation damage to pine crowns in the Chernobyl NPP zone appeared in the first few weeks following the accident over an area of approximately 100 hectares close to the reactor, where dose burdens in the needles and apical meristems exceeded 500 Gy. The area of forest destroyed by radiation subsequently continued to expand, although the situation stabilized towards the end of the summer of 1987. Helicopter and ground observations of the condition of pine forests allowed us to classify them into four zones as regards radioactive damage (Table 4).

Where there was partial damage to crowns caused by sub-lethal irradiation doses, irradiation gave rise in all observed cases to physiological and morphological disturbance in newly formed shoots. The frequency of double growth increased - buds forming on shoots began to grow during that same year, and shoots grew twice within a year. Also, new shoots formed not only at the end of the secondary shoot but also in the needle axils. During the following growth period, these shoots formed so-called "witch's brooms" - bunches of up to 40 shoots. In addition, some of the newly formed buds changed their shape and orientation with regard to the shoot axis. By the beginning of the next growth period some of these buds had died, and the survivors put out bunches of needles or short thick shoots with twisted and enlarged needles; many of the bunches had three needles instead of the usual two. No new buds formed on these shoots. Sub-lethal irradiation doses also retarded the growth of needles and shoots, and restricted the number of lateral shoots (particularly in the lower part of the crown) formed in the year after the fallout occurred. Such morphological disturbances were only temporary, however. Two years later, these trees were growing completely normally and there was almost no difference between them and the controls.

Comparing the radioresistance of coniferous and deciduous trees showed that when fallout occurred during the spring and summer (Chernobyl), the differences stemmed mainly from the species' different levels of sensitivity (deciduous species being 5-10 times more resistant than coniferous ones). When fallout occurred in the autumn (Urals), the difference was approximately 20 times greater; this was linked to the reduced retention capacity in the crowns of deciduous trees (and consequently lower irradiation doses) where fallout occurred after defoliation.

A comparison of the extent of radiation damage shows that the area of forest damaged in the Chernobyl zone was approximately twice the size of the area affected in the Kyshtym accident zone, although in terms of radioactive releases the corresponding ratio was 25 to 1. This discrepancy may be explained by the more compact distribution of fallout resulting from a single (momentary) release of radionuclides in the Urals, as compared with the protracted release in the Chernobyl zone and the consequent distribution of radionuclides over a wider area. Another reason is that longer-lived radionuclides were present in the material released at Kyshtym, and their effect has been felt over a longer period.

A comparison of our and other published data shows that beta radiation from radioactive contamination damages woody plants at approximately the same doses as caused by gamma radiation from pinpoint sources.<sup>1</sup> In addition, a map of radiation damage to woody plants from fallout shows some specific characteristics, caused by changes over time and space in the distribution of radiation sources and, in consequence, a corresponding change in dose fields. A particular example of this is the greater resistance to radioactive fallout of the topmost shoot in comparison with the remainder of the crown. In addition, when fallout occurs in winter, the differences in resistance to radioactive contamination between coniferous and deciduous species are, as noted above, significantly greater than the difference in their radiosensitivity, due to the fact that in winter deciduous tree crowns retain fewer of the atmospheric contaminants deposited..

#### 4. THE EFFECTS OF RADIATION IN HERBACEOUS COMMUNITIES

The radiobiological reactions of different species of herbaceous plants to radioactive contamination vary widely, depending on the radiosensitivity of the individual species, their vegetative propagation capacity and the location of young buds with regard to the soil surface as an irradiation source.

From the results of geobotanical observations in the Kyshtym zone and dosimetric assessments<sup>8</sup>, it can be taken as proven that, where dose burdens in seeds on the soil surface exceed 200 Gy, their germinative capacity is, in many species, lost. Thus many therophytes disappeared from the plant community. A similar development was observed among the hemicryptophyte and chamaephyte groups, which have their buds near the soil surface. Their place was taken by plants whose buds are buried in the soil (Fig. 5). This process continued over 3-4 years. Subsequently, the process slowly reversed itself. These changes in species composition appeared not only in communities of meadow plants but also in herbaceous forest plants. However, while changes in the species composition of meadow communities were caused by direct irradiation only (primary effects), secondary effects played an important role in

forest communities. What happened was that in forests, where irradiation damage to tree crowns was severe, there were significant changes in microclimatic conditions underneath the forest canopy. The combination of irradiation and changed microclimate brought about a change not only in the species composition but also in primary productivity. In forest stands where the pine trees died and the birches were damaged, the overground phytomass of the herbaceous community increased by 3 - 5 times as compared to uncontaminated forests of similar type (Fig. 6).

Thus, in the Kyshtym accident zone, changes in the species composition of herbaceous plants in forest and meadow communities initially (i.e. during the first 3 - 4 years) took the form of a decline in the number of species of hemicryptophytes and the complete disappearance of chamaephytes. Their places were taken by cryptophytes such as *Calamagrostis epigeios*, which regenerates by means of rhizomes protected from beta radiation by a thick layer of soil. Photophilic species such as *Cirsium setosum* and *Chamaenerion angustifolium*, (pioneers with powerful root systems, a significant proportion of which occupies uncontaminated soil layers) became dominant. Although related to the hemicryptophytes, these species have a short life span (two years), and they propagate mainly by means of seeds, which may be carried by the wind over long distances and can therefore be brought in from uncontaminated territory.

Four to six years after formation of the radioactive trail, herbaceous communities began to return to their former state in terms of species composition as the irradiation dose fell to 10% of its original level. These changes were reflected in an increase in the proportion of hemicryptophytes and a decline in cryptophytes. However, at least 20 years were required for the plant community to be completely restored to its original state.

In the Chernobyl zone, on the other hand, no substantial disturbances in herbaceous plant communities were observed, with the exception of a small area in the immediate vicinity of the reactor, where irradiation doses exceeded 500 Gy. According to A. I. Taskayev, the first seeds produced after the accident in this area were virtually no different in terms of viability from the seeds of control populations. The explanation for the high radioresistance of herbaceous plants in the Chernobyl NPP zone in comparison with the Urals may be that the initial irradiation of the mother plants occurred during the active growth phase. In this situation, where irradiation took place over a long period of time, the effects (at equal doses) were less apparent than those arising from irradiation during the physiological rest phase, which in terms of impact is equivalent to acute irradiation.

#### CONCLUSION

The Kyshtym and Chernobyl accidents differ in many aspects which determine the ecological consequences of radioactive contamination of adjoining forest areas, such as the overall quantity of radionuclides in the materials released, their radionuclide composition and persistence and the seasonal, soil and ecological conditions (Table 5). Differences in radionuclide composition and, consequently, in the rate of radioactive decay, meant that the dynamics of absorbed dose formation in the crowns of trees differed over time. Their physiological status, linked to seasonal conditions at the time the main dose burdens were formed, influenced the radiosensitivity of critical organs and the radiobiological impact of the dose during

chronic irradiation. The particular persistence and amount of radionuclides released in either case determined the extent of areas with levels of contamination lethal to forest stands.

The soils in the Kyshtym zone were mainly semi-loamy leached chernozems, while in the Chernobyl zone they were sandy, soddy-podzolic and peat-bog soils. In the former case the long-term factor limiting the use of forestry products is  $^{90}\text{Sr}$ , while in the latter it is  $^{137}\text{Cs}$ .

Despite the differences mentioned, however, the radioecological consequences of the Kyshtym and Chernobyl accidents for forest ecosystems were in many respects similar. The areas with radioactive contamination levels lethal to trees are similar in extent. The dose burden values causing the various degrees of tree damage also coincide, as do the trees' reactions to irradiation as a function of the dose absorbed. There are similarities in the dynamics of the appearance of radiation effects over time and in the subsequent recovery of forest stands. The concentration values for the critical radionuclides ( $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ ) in the main types of forestry products are also similar. This is largely due to the fact that, although many different factors are involved inducing manifold effects, they give rise to similar results. In particular, we find similar levels of contamination (strontium-90 in the Kyshtym zone, caesium-137 in the Chernobyl zone), in identical types of forestry products, since the mobility of caesium-137 in sandy soils is comparable to the mobility of strontium-90 in chernozem. Other radioecological effects were explained in the text.

On the whole, the negative ecological consequences of radioactive contamination for forest ecosystems were confined to relatively small areas in both the Kyshtym and Chernobyl accident zones. Forest damage on this scale is not uncommon, often being observed in the vicinity of major industrial complexes which are sources of chemical pollution. The full radioecological effects on forest ecosystems only become apparent after several years. Subsequently, the processes of recovery and adaptation to increased background radiation come to dominate. Removing the anthropogenic pressures on ecosystems in areas taken out of economic and other types of use due to their high levels of radioactive contamination leads to an improvement in ecological conditions for many plant and animal species. This helps to restore the diversity and size of populations and the appearance, in radioactively contaminated areas, of rare and vanishing plant and animal species.

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Table 1: Dynamics of Cs-137 distribution among forestry stand components in the Chernobyl accident zone (eluvial landscape)

Time	Cs-137 (% of total)					
	Needles + leaves	Wood	Bark	Branches	All over-ground phytomass	Soil 0-10 cm
1986 (August)	5.2	1.3	3.8	3.3	13.6	86.4
1987	0.8	1.3	2.6	2.5	7.2	92.8
1988	0.9	0.8	3.0	1.5	6.2	93.8
1989	0.5	0.7	3.3	1.1	5.6	94.4

Table 2: Normalized concentrations of Sr-90 and Cs-137 in forestry products in the Kyshtym and Chernobyl accident zones 4-6 years after fallout

$10^{-3} \text{ m}^2/\text{kg}$

Product	Sr-90 Kyshtym	Cs-137 Chernobyl
Dry wood: -pine	0.5	0.2 - 1.5
Dry wood: -birch	1.5	0.5 - 3.5
Herbaceous plants (dry phytomass)	3 - 50	3 - 20
Mushrooms (fresh)	1.5 - 4.0	2 - 200
Berries (fresh)	0.6 - 0.8	1 - 4
Pine gum	-	0.8



Table 3: Effect of landscape conditions on the level of Cs-137 contamination in forestry products in the Chernobyl accident zone, 1989

Product	Normalized concentration of Cs-137 ( $10^{-3}$ m <sup>2</sup> /kg)	
	eluvial landscape, soddy-podzolic soil	Accumulative landscape peat-bog soil
Dry wood: -pine	0.2	1.5
-birch	0.5	3.5
Herbaceous plants (dry phytomass)	3 - 20	40 - 190
Birch sap	1	5
Mushrooms (fresh)	2 - 17	6 - 220

Table 4

Effects of radiation in contaminated forests, summer 1987

Absorbed dose, Gy		Effects of radiation
Needles, leaves	Apical meristem	
≥ 500	≥ 100	Severe crown damage in deciduous trees, die-back of pines
≥ 100	≥ 25	Die-back of pine forests (some 500 ha). Morphological disturbances in deciduous species
20-100	15-25	Severe damage to pine forests: shrinkage in part of forest stand; up to 90% damage to crowns of remaining trees; hardly any growth in 1987 (approximately 200 ha)
20-50	5-10	Moderate damage to pine forests. Damage to needles in lower part of crown. No growth sites formed in 1986. Growth processes in 1987 associated with awakening of lateral dormant buds; restricted growth; changes in shape and dimensions of needles and shoots. Complete suppression of reproductive capability; evidence of genetic damage
10-20	3-5	Light damage to pines: reproductive capability inhibited during 1987; increased frequency of gene mutations

Table 5: Factors determining the consequences of radioactive contamination of forests in the Kyshtym and Chernobyl accident zones

FACTORS	KYSHTYM	CHERNOBYL
Quantity of radionuclides released by accident	7 · 10 <sup>16</sup> Bq	2 · 10 <sup>18</sup> Bq
Seasonal conditions	Autumn	Spring
Factor of retardation of radioactive aerosols by pine crowns	0.7 - 0.9	0.6 - 0.9
Time for accumulation by woody plants of 80 % of total radiation dose	6 - 8 months	1 month
Lethal dose for pine: -needles -apical meristem	≈ 50 Gy 15 - 25 Gy	100 Gy ≥ 25 Gy
Area of contamination level lethal for pine forest	20 km <sup>2</sup>	50 km <sup>2</sup>
Changes in herbaceous communities affected by radiation	Changes in species composition	Changes are insignificant

Fig. 2: Dynamics of Sr-90 in structural parts of pine (A) and birch (B)

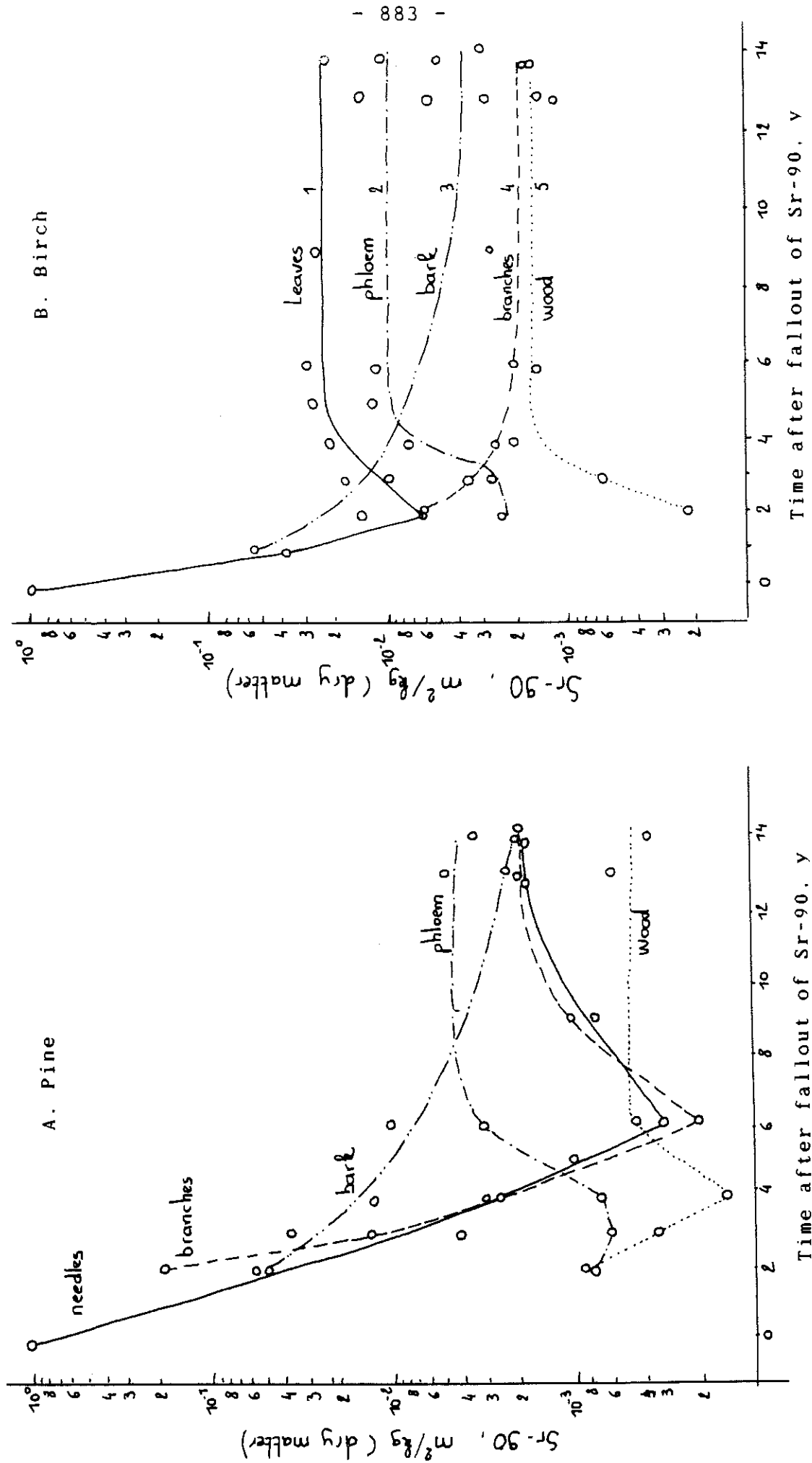


Fig.3: Flow diagram for Cs-137 migration in the forest ecosystem (1989)

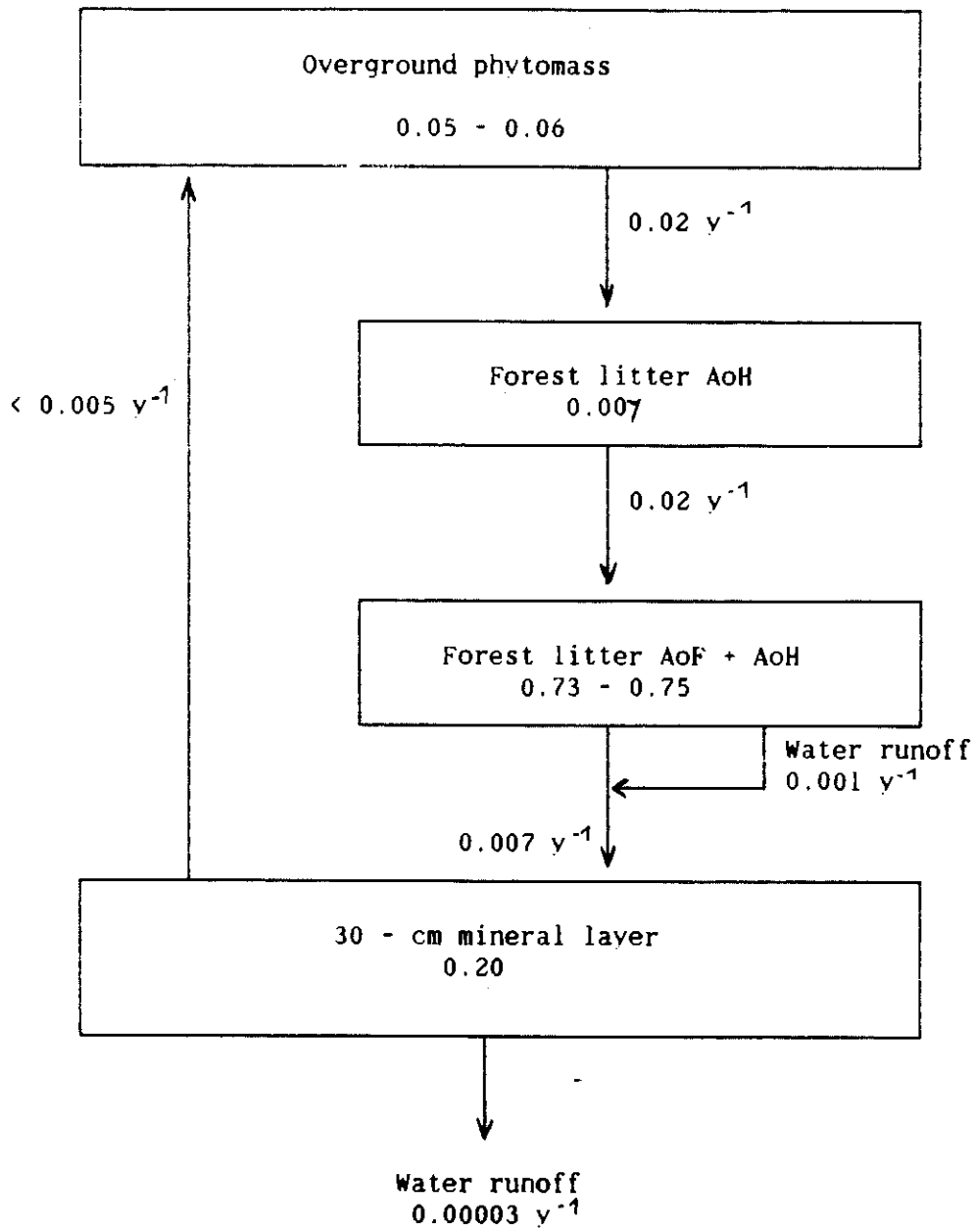


Fig.4: Dynamics of absorbed dose rate from  $\beta$ - and  $\gamma$  radiation in pine needles, Chernobyl zone

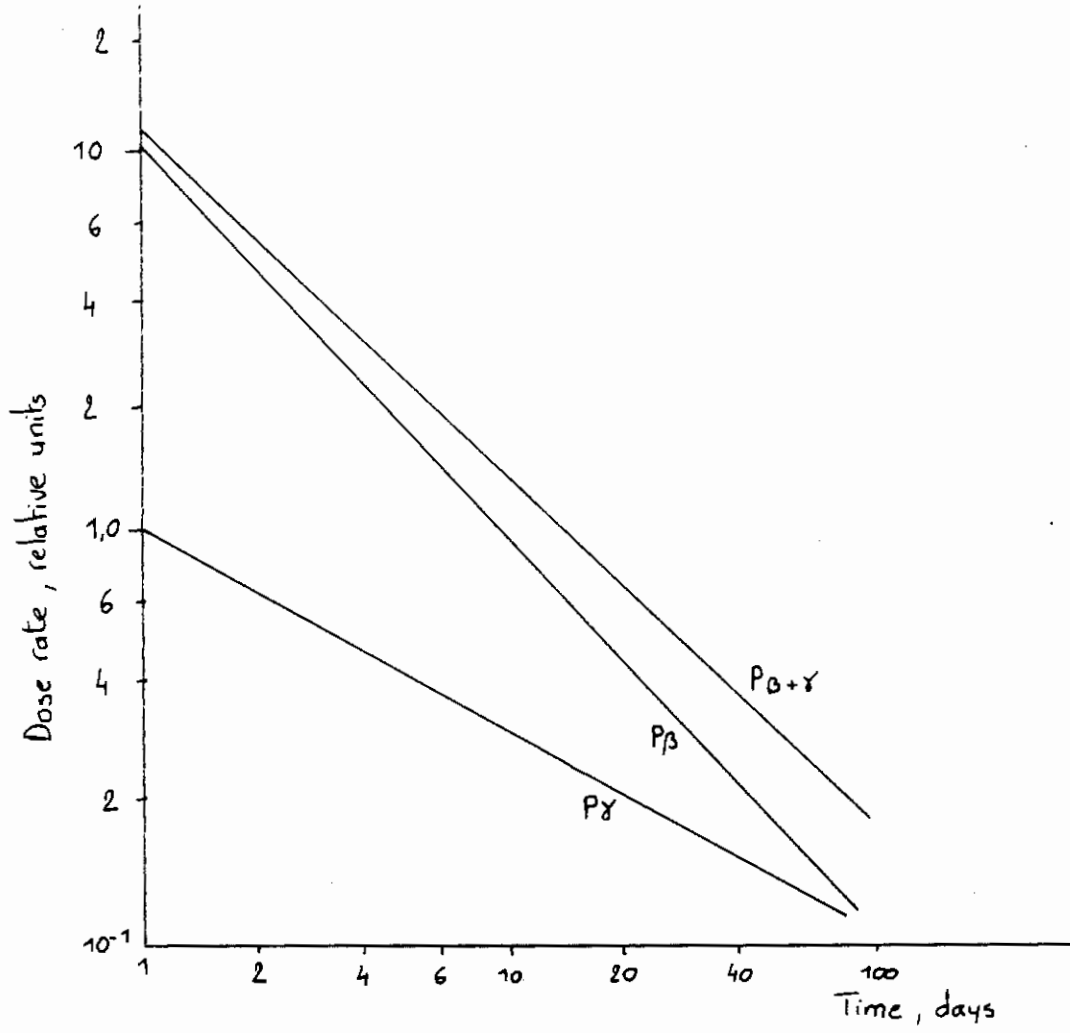


Fig.5: Dynamics of cryptophytes and hemicryptophytes in herbaceous plant community  
Contamination level - 150 MBq Sr-90/m<sup>2</sup> (Kyshtym)

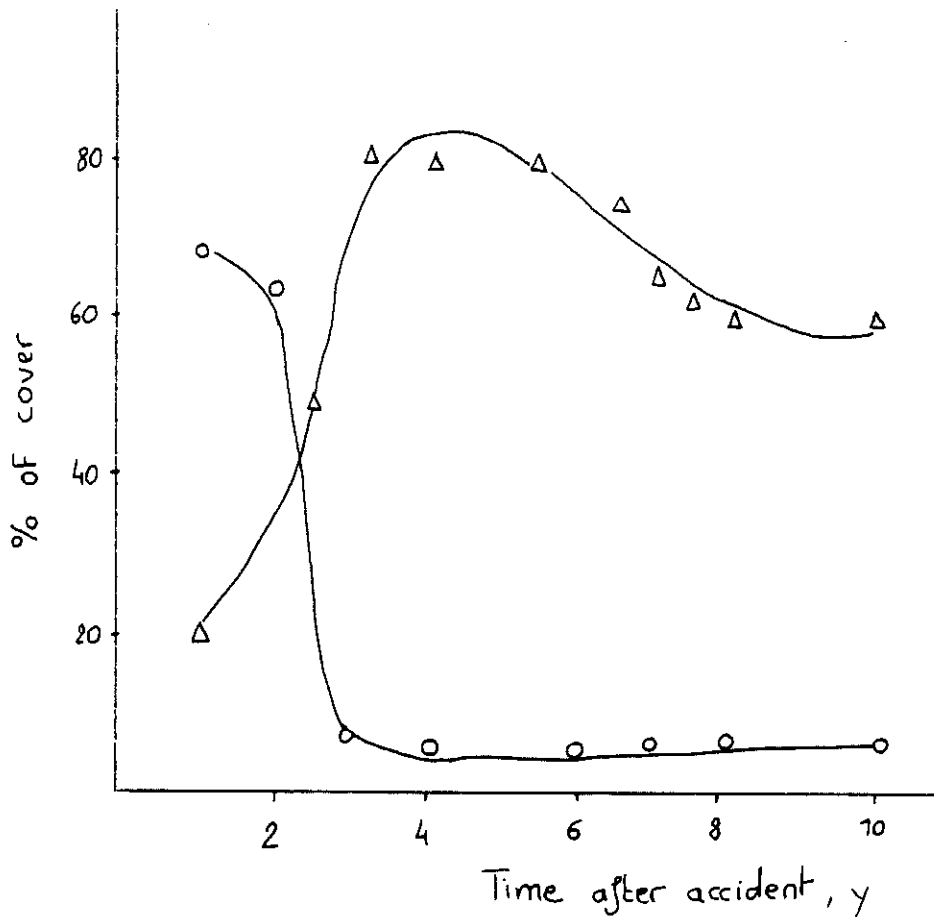
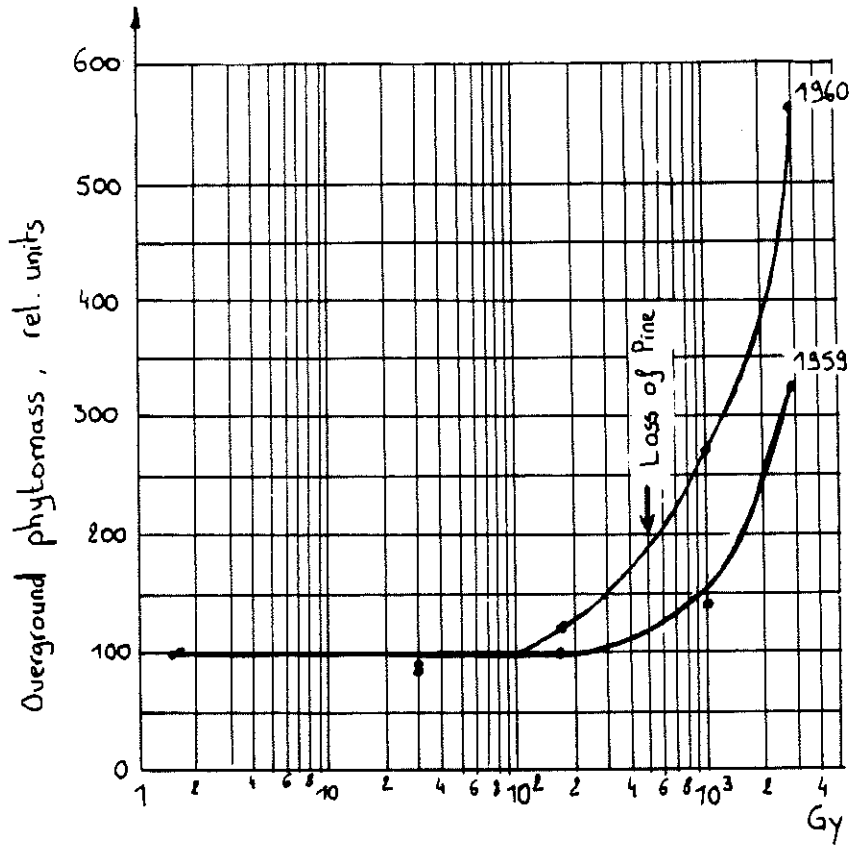


Fig.6: Change in overground phytomass of herbaceous plants in pine forest depending upon the radiation dose accumulated in pine needles (Kyshtym)







# **Non-classical Phenomena in Contemporary Radioecology**

**Y. KUTLAKHMEDOV**

The accident at the fourth unit of the Chernobyl NPP triggered a radioecological abnormality unique in character, dose burdens and scope. Powerful radiation dose fields caused the acute death of conifers ("red forest"). In an area in the Novoshepelichi forest the death of young fir trees was observed in 1986. "Chimera formations" - i.e. morphological changes such as pine-like shoots, multi-branching shoots, and other anomalies - were observed in 1987-1989.

The formation of powerful and - above all - heterogeneous dose fields in the 30 km zone around the Chernobyl NPP was typical.

We carried out a series of measurements using thermoluminescent detectors (TLDs) which showed that the dose distribution over a 100 m<sup>2</sup> plot ranged from 17 to 90 rad for the overground parts of plants, and from 3 to 24 rad for plant root systems (6-8 fold scatter with mean dose rate of 2-3 mR per hour), because of the considerable heterogeneity of the radioactive contamination.

There was a wide scatter (30-fold) in Ce-144 content in conjunction with a tail in the distribution due to the presence of "hot" particles. Ru-106 varied in a similar fashion and also had a "hot" tail.

Substantial vertical distribution of radionuclides was observed in the soil. Initially (1986-1987) the distribution followed an exponential pattern, while in 1988 "creep" through the soil profile was observed. This is a heterogeneous, non-linear radioecological process resulting from high dose burdens.

Plant biomass growth stimulation was observed in nature and in experiments. This is linked to suppression of the generative function accompanied by a compensatory increase in the growth of vegetative biomass.

The high dose burdens (uncharacteristic of other radioecological situations) led to an increase in plant biomass production, and - what is very important - high doses engender an increase in radionuclide removal. It appears that the irradiation dose promotes an additional afflux of radionuclides and plant biomass. We observed this phenomenon in a field experiment in which plants grown from irradiated seeds removed 1.5 to 3 times more radionuclides than the controls did.

High concentrations of radionuclides in soil solutions have significant radioecological effects. Thus, our experiments to estimate the relative speed of maize growth - (a) on washout from contaminated soils and (b) after acute gamma irradiation - showed that the relative biological effectiveness of chronic irradiation (the dose being assessed by calculation and with the aid of TLDs) was considerably higher (by a factor of 4 to 10) than that of acute irradiation.

A key feature of the radioecological situation after the accident was the significant increase in radionuclide removal over time. Removal with biomass constituted around 1%-3% in 1986-87 and 3-7% in 1988, while in 1989 under optimal experimental conditions we obtained 20-30% radionuclide removal from the soil. This points to a sharp increase in the mobility and solubility of the radionuclides over time.

Theoretical radioecology still lags behind the theory of radiobiological phenomena. The problems are indeed legion. We need to promote the development of non-linear radiobiology and radioecology.

The main problem is that dosimetry is difficult because of the heterogeneous topology of the dose fields. This in turn is due to the radionuclide mix and to radionuclide accumulation coefficients of changing dynamics in complex landscapes.

To facilitate prediction work, we consulted the study by Agre and Korogodin, which sets out the idea, method and concept of radiocapacity. We applied this approach to the Dnieper Cascade reservoir system.

The notion of a radiocapacity factor was advanced in connection with assessment of the radiocapacity of an isolated body of water:

$$F = kh / H + kh$$

where:

$k-k.H$  (translator's note: unclear characters photocopied from original)

h: thickness of the bed sediment layer sorbing the radionuclides

H: depth of body of water

The factor indicates which proportion of the radionuclides is contained in bed sediment and which in water (1-F).

An analysis of the situation after the accident showed that this method of assessing radiocapacity could be applied to the Dnieper Cascade. The summation formula for estimating the Cascade's radiocapacity is:

$$F_k = 1 - \prod_{i=1}^n (1 - F_i)$$

The estimate showed that while the radiocapacity of the individual reservoirs ranges from 0.6 (Kanev) to 0.8 (Kremenchug), the total radiocapacity of the entire system is 0.996 for Cs-137 and 0.6 for Sr-90.

Actual measurements of caesium in the bed sediment of the Kiev reservoir even now show activity in the region of  $10^{-5}$  to  $10^{-6}$  Ci/kg, which undoubtedly involves substantial dose burdens for benthon and zoo- and phytoplankton.

We created a radiocapacity model based on the Ukraine's agroecocenosis. The model includes: biomass yields, accumulation coefficients, the influence of irradiation on the removal of radionuclides, etc. This model takes into consideration irrigation using water-body waters and its impact on root removal of radionuclides and - in particular - the influence of the radioactively contaminated water on the removal of radionuclides from plant biomass in the areas under irrigation.

This means of assessing radiocapacity involves long and painstaking calculations. By applying this model to the assessment of dose flows to the population of the Ukraine we were able to calculate the basic contributions to the collective doses of various elements in the diet.

To this end we made use of:

1. Estimates of the dose from foodstuffs in regions with different levels of soil contamination, ranging from 10 to 0.02 Ci/km<sup>2</sup>.

2. An estimate and forecast of contamination of the Dnieper reservoirs under different regimens and different flood intensities (Institute of Cybernetics of the Ukrainian Academy of Sciences).

3. Crop harvests in different regions, the degree of irrigation and, particularly, the proportion of agricultural products in the Dnieper Cascade irrigation zone, which for certain oblasts amounts to 27-30%.

With the aid of the dose values for different radionuclides we calculated the expected collective doses from food intake for one year of life in real and predicted conditions.

The estimates predicted a total initial collective dose for the Ukraine through food intake alone of about 2 million man-rem. Given the actual behaviour of the Cascade in 1986 the dose through food intake alone was up by 14%, while extreme flooding would lead to a 26% increase in the dose. The mild flooding of 1989 increased the initial collective dose by 2.3%.

Additional radionuclide runoff from the River Pripjat floodplain, which is highly contaminated with Sr-90, could add 12% to the initial collective dose. The construction of cutoff dykes could lower this to 8%.

A further extreme ejection of radionuclides from the fourth unit could lead to a 32% increase in the expected annual collective dose through food intake.

In the presence of high collective doses for a population of 30-35 million the problem of variability in individual radiosensitivity assumes major importance in quantitative radioecology. Radiobiological data show that individuals vary greatly in their response to radiation. Thus, in drosophila individual radiosensitivity varies by a factor of 10 or more, the distribution having an assymetric log-normal character. In Switzerland an investigated human population also showed a ten-fold variability in sensitivity (depending on sex, age, living conditions, etc.).

A close examination of the quantiles in this assymetric distribution shows that given a mean dose of 1 rad over a population of 1 million, the collective dose amounts to approximately 3.2 million man-rem. This is a serious problem: how do we pinpoint and account for populations with high individual sensitivity? For such populations even low doses of 1 rad may equal 10 rem or more, and this may lead to somatic and genetic effects in these groups (all factors considered the collective dose probably lies between 2 and 12 million man-rem).

I have sketched just some of these new problems of quantitative radioecology that need to be described and solved, viz.:

1. Detailed dosimetry of dose flows among biological objects, plants, animals and man in the radioecologically anomalous zone (paying special attention to hot particles). It is clear that the dose to aquatic and terrestrial flora greatly exceeds the dose to man.
2. Estimation of radionuclide migration processes in the environment.
3. Investigation of individual radiosensitivity of the population in the zone affected by the accident.

This is something one might call "judicious radioecology" - to use Timofeyev-Resovsky's phrase - in the circumstances arising after the Chernobyl accident.



**Chromosome Mutagenesis in  
Populations of Aquatic Biota in  
the Black Sea, Aegean Sea,  
and Danube and Dnieper  
Rivers, 1986-1989**

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## ABSTRACT

We studied the level of structural mutagenesis in the reproductive and somatic cells of aquatic biota of various taxa from natural populations of neustic and benthic communities in the Black and Aegean Seas and the Dnieper and Danube rivers between 1986 and 1989. The cytogenetic research covered embryos, larvae and adult worms of Nereidae, Naididae, Tubificidae and Turbellaria, adult *Sagitta setosa*, young *Bivalvia* molluscs, embryos of Mysidacea, and growing roe of *Engraulis encrasicolus*, *Sprattus sprattus*, *Diplodus annularis*, *Mullus barbatus*, *Trachurus trachurus*, *Scophthalmus maeoticus*, *Abramis brama*, *Blicca bjoerkna*, *Rutilus rutilus* and *Stizostedion lucioperca*.

It was established that aquatic biota in the open waters of the Black and Aegean Seas had a lower level of chromosome mutagenesis than representatives of the fluvial communities. The intensity of mutagenesis was compared with the data published in the literature on radioactive contamination/chemical pollution of the aqueous medium in these areas.

The paper sets out statistical regularities in chromosome mutagenesis (inter-individual variability in the chromosome aberration rate and distribution of chromosome damage in cells), noting different patterns of chromosome aberration distribution among cells. On the basis of a large quantity of our own data from field and experimental cytogenetic studies involving aquatic biota, the paper considers the possibility of using - for the purposes of radiochemical-ecological monitoring - chromosome damage distribution in cells as an indicator of whether mutagens are radiation-related or not.

From 1986 to 1990 a study was made of the level of chromosome mutagenesis in the cells of 37 species of aquatic biota belonging to various taxa (worms, molluscs, echinoderms, chaetognaths, crustaceans, fish) in the natural populations of the neustonic and benthic communities of the Black Sea, the Aegean Sea and the Danube and Dnieper Rivers. The organisms in the neustonic and benthic communities are inhabitants of contour biotopes, which are regarded as the main "ecological targets" in aquatic ecosystems (Zaytsev, 1988).

The somatic tissues of aquatic biota at various stages of ontogenesis (embryos, larvae, fry) together with the generative tissues of sexually mature individuals served as the material for our research. Chromosome aberrations in cells at the anaphase-telophase stages of mitosis and the anaphase-telophase stages I and II of meiosis were analysed. A total of 43 samples were studied.

Fig. 1 shows generalized data on the level of mutagenesis in the cells of aquatic biota of various taxa. It can be seen from this that aquatic biota in the open waters of the Black Sea and Aegean Sea had a lower level of chromosome mutagenesis than representatives of the fluvial communities.

Some 60% of the samples from the sea had a mean level of chromosome mutagenesis of up to 2%, whereas in rivers the corresponding figure was approximately 20% of samples.

Data on the mean level of chromosome mutagenesis do not give a complete picture of the intensity of the mutation process in populations. It is therefore important to assess the heterogeneity of natural populations with respect to the rate of chromosome anomalies, which might testify a) to the stochasticity of interaction between mutagens in the medium and living organisms, and b) to the individual sensitivity of aquatic biota stemming from their latent genetic mutability. We checked the normality of the distribution of the aquatic biota samples with respect to rate of chromosome aberrations.

In the majority of marine samples there is substantial deviation from normal distribution, whereas in rivers a large degree of concordance with normal distribution is observed.

Analysis of the material available shows that the marine populations of aquatic biota generally have a low mean rate of chromosome aberrations, high dispersion and variation coefficient values, and asymmetrical distribution of individuals in terms of chromosome mutagenesis level. In rivers, in addition to an elevated mean level of chromosome rearrangements, we find low variation coefficient values because of the more homogeneous response of individuals to contamination, and their distribution is near normal.

As is well known, an analysis of the distribution of chromosome rearrangements among the cells of aquatic biota can yield substantial information on the way mutagens in the medium interact with hereditary structures.

According to the target theory (Li, 1963), under the action of infrequently ionising external radiation (gamma and X-rays) primary damage to chromosomes should be randomly distributed among cells in accordance with Poisson's law. A number of works on radiation-induced mutagenesis in a culture of human lymphocytes and in the cells of

plants do, in fact, contain data showing good correspondence between the cell distribution of chromosome aberrations and the theoretical Poisson distribution (Li, 1963; Luchnik, Bochkov, Sevankayev, 1969; Ganassi, Lyamin, Aptikayeva, Eyduş, 1971). However, other research involving the cells of *Crepis*, *Tradescantia* and peas (Catchside, Le, Thodey, 1946; Luchnik, 1968; Mitrofanov, Olimpienko, 1980) found that the distribution of chromosome rearrangements deviated substantially from random distribution. Some authors (Luchnik, 1968; Mitrofanov, Olimpienko, 1980) attribute this phenomenon to the processes involved in the incidence of, and recovery from, primary damage.

The literature does not contain much data on cell distribution of aberrations in the case of chemical mutagenesis. The rare data which are available mainly concern the distribution of chromosome breaks in cells when human lymphocytes are subjected to the action of antineoplastic preparations (phosphamide, thiophosphamide, mitomycin, dipin and photrin) (Bochkov et al., 1972; Yakovenko, Azhayev, Bochkov, 1974; Bochkov, Chebotarev, 1989). These show a consistent deviation from the random law in the cell distribution of chromosome breaks. It is shown that in the case of phosphamide, thiophosphamide and mitomycin the cell distribution of chromosome breaks conforms very well to geometrical distribution, whereas in the case of dipin and photrin it conforms to inverse binomial distribution.

In experiments specially set up by us to induce chemical mutagenesis in aquatic biota, we analysed the nature of the distribution of chromosome aberrations among cells under the action of mutagens such as DDT and chlophene widely found in the aquatic environment. The experiments involved brin embryos and the amphipoda *Gammarus olivii*, the data obtained being shown in Table 1. It can be seen that the empirical distributions deviate substantially from the Poisson distribution but concord well with the geometrical distribution.

Our data show, on the other hand, that in the case of aquatic biota the empirical distributions of chromosome aberrations almost always correspond to the theoretical Poisson distribution (Table 1) under the action of both external and incorporated irradiation (apart from a few individual cases).

We also set out to see how the distribution of chromosome aberrations among cells of aquatic biota from marine and fluvial populations corresponded to the theoretical Poisson and geometrical distributions. The results obtained are shown in Table 2. Greater concordance with the Poisson distribution was observed in only three species of fish from the Dnieper and the Dnieper-Bug estuary (or liman). In the other species of aquatic biota the distribution corresponded more to the geometrical distribution.

It is useful to compare the data obtained with the level of radioactive contamination and chemical pollution in the areas inhabited by the populations studied. According to S. A. Patin (1979), concentrations of organochlorine compounds, mercury and lead in inland seas already exceeded the minimum toxic and threshold concentrations for the most sensitive species of aquatic biota as much as 20 years ago. And in rivers the contamination gradient is even higher. Studies carried out during the First International Danube Expedition - when our cytogenetic data were obtained - revealed high concentrations of mercury, chlorinated hydrocarbons and oil in the water, and more so in the bed sediments (Polikarpov et al., 1989; Tarasova et al., 1989). After the Chernobyl accident, according to G. G. Polikarpov et al. (Polikarpov,

Kulebakina, Timoshchuk, 1989), in 1986 the concentrations of strontium-90 in surface waters of the northwestern part of the Black Sea ranged from 29.99 to 129.63  $\text{mBq}\cdot\text{l}^{-1}$ , while those of caesium-137 ranged from 12.59 to 185.18  $\text{mBq}\cdot\text{l}^{-1}$ . In 1987 the concentrations of strontium-90 and caesium-137 in the surface waters had fallen, as a result of migration processes, to between 18.52 and 62.96  $\text{mBq}\cdot\text{l}^{-1}$  and between 33.33 and 137.04  $\text{mBq}\cdot\text{l}^{-1}$  respectively. The highest concentrations of strontium-90 (333 and 571.3  $\text{mBq}\cdot\text{l}^{-1}$ ) were noted by these authors at two stations in the western part of the Black Sea.

In the Aegean Sea the mean concentration of strontium-90 in the water was 19.6  $\text{mBq}\cdot\text{l}^{-1}$  (Polikarpov, Kulebakina, Timoshchuk, 1989).

In spring and summer 1986 the concentration of strontium-90 in the Kiev Reservoir was equal to 1 800-5 180  $\text{mBq}\cdot\text{l}^{-1}$  (Izrael et al., 1987). In spring 1988 it had fallen to 666  $\text{mBq}\cdot\text{l}^{-1}$ . The caesium-137 concentration was 185  $\text{mBq}\cdot\text{l}^{-1}$  (Polikarpov, Kulebakina, Timoshchuk, 1989).

In spring 1988 the mean concentrations of strontium-90 and caesium-137 in the Danube waters were 11.85 and 14.88  $\text{mBq}\cdot\text{l}^{-1}$  respectively. The maximum concentrations of strontium-90 (16-20  $\text{mBq}\cdot\text{l}^{-1}$ ) were discovered along the lower reach of the Danube (20-255 km), while those of caesium-137 were found along the river's upper reach and amounted to 29.25-37.04  $\text{mBq}\cdot\text{l}^{-1}$  (Kulebakina, Polikarpov, 1989). The highest concentrations of caesium-137 and caesium-134 in the bed sediments of this river were found in the upper reach (in the Gabchikovo area), and amounted to 950 and 3 000  $\text{Bq/kg}$  dry mass (Rank et al., 1989).

These levels of radioactive contamination are far below those capable of inducing chromosome aberrations in aquatic biota, including the most sensitive among them - the embryos of sea fish (Tsytsugina, Risk, Lazorenko, 1973; Shekhanova, 1983).

It is therefore clear that in the overwhelming majority of cases damage to the chromosomes of aquatic biota in situ may have been caused by chemical pollution of the medium, with - as has been shown - the distribution of chromosome aberrations corresponding more to the geometrical distribution. Exceptions to this were the roe, located on the bed, of two species of fish in the Kiev Reservoir and one species of fish in the Dnieper-Bug estuary. These cases, particularly the latter, concurred more with the Poisson distribution, which - as has been shown experimentally - occurs under the impact of radiation. It should be noted here that a hot particle of ruthenium-103 was found in the bed sediment of the Dnieper-Bug estuary at the very spot where roe of Rutilus rutilus heckeli with a very high level of chromosome mutagenesis (30.5%) were collected (Tsytsugina, Lazorenko, Radchenko, Skotnikova, 1990). It is possible that in this case radiation from hot particles could have induced chromosome aberrations in the fish embryos.

Our analysis of the data found in the literature and of our own experimental and field data leads us to deduce that the nature of the cell distribution of chromosome damage may serve, for the purposes of radiochemical-ecological monitoring, as an indicator of whether mutagens are of a radiation-related nature or not.

In conclusion, I wish to express my thanks to V. N. Zhukinsky and his team for collecting the samples and establishing the species of the fish from the Kiev Reservoir.

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Table 1: Distribution of chromosome aberrations in the cells of aquatic biota (experimental data)

Species	Mean numb.of aberrations per cell	X <sup>2</sup> correspondence of Theoretical distribution		Mutagen
		poisson	geometrical	
Scophthalmus macoticus	0.123	20.19	3.95	DDT
Gammarus olivii	0.048	6.19	0.93	PCB
Scorpaena porcus	0.138	6.13	17.17	Y-91
"	0.188	5.27	10.04	C-14
"	0.207	0.74	5.26	C-14
G. olivii	0.065	0.08	1.27	Sr-90
"	0.120	5.11	20.26	Sr-90
"	0.146	3.01	17.96	Sr-90
"	0.156	2.24	12.11	Sr-90
Idotea baltica	0.103	0.10	1.55	Sr-90
G. olivii	0.089	1.09	3.81	-irrad.
"	0.164	0.42	5.45	-irrad.
Scorpaena porcus	0.145	1.84	7.53	-irrad.
"	0.151	2.56	17.50	-irrad.

Table 2: Distribution of chromosome aberrations in the cells of aquatic biota (field data)

Species	Mean numb. of aberrations per cell	X <sup>2</sup> correspondence of Theoretical distribution		contaminants discovered
		poisson	geometrical	
BLACK SEA				
Diplodus annularis	0.020	0.022	0.0006	
Scophthalmus macoticus	0.028	0.011	0.001	(Hg. PCB?)
Mullus barbatus	0.019	13.00	5.14	
Trachurus trachurus	0.035	21.60	7.46	
DANUBE				
Sphaerium corneum	0.052	0.019	0.05	
Nais sretscheri	0.077	0.18	0.039	
Naididae	0.062	0.070	0.007	DDT. oil (Tarasova et al. 1989)
DNEIPER				
Blicca bjoerkna	0.049	9.89	1.22	
Stizostedium lucioperca	0.066	0.23	0.67	
Rutilus rutilus	0.041	0.11	1.08	
Abramis brama	0.028	6.87	1.22	
Rutilus rutilus heckeli	0.319	1.308	50.467	Ru-103 (Tsytsuqina et al. 1990)
Monodacna caspia	0.092	0.453	0.003	





# **Doses of Irradiation to the Ukrainian Population as a Result of the Chernobyl Accident**

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In the middle of 1990 the whole problem of the assessment of exposure doses to the population living near the Chernobyl plant can be divided into several separate problems:

- storage, handling, organization and primary analysis of direct measurement results of external and internal doses to the people;

- usage of the available information for the models of retrospective reconstruction of the past exposure doses and for prediction of possible future doses;

- verification and validation of mentioned models by means of constant comparison of calculation results received on their basis to the results of current instrumental monitoring.

Three problems mentioned above are solving in the laboratory of mathematical models and radiation prediction in dosimetry and radiation hygiene department of AUSCRM USSR AMS.

The given report presents the description of the first results of this work.

#### 1. THE SYSTEMATIZATION OF DOSIMETRIC AND RADIOECOLOGICAL DATA

The Central Dosimetric Register (CDR) created at All-Union Scientific Center of the USSR Academy of Medical Sciences is the informational base for the estimation of individual and collective doses of public exposure as well as for radiation dosimetric assessment of ecological situation on the territories contaminated as a result of the Chernobyl accident.

The CDR structure is shown in Fig.1.

The primary storage of the information is provided in the so called Local Data Bases (LDB), which are formed in

accordance with main types of data. The second level is generalization of LDB data and creation of the radiation dosimetric territorial data base.

The main coordinating part of CDR is analytical section which consists of specially created software and provided input, assessment, systematizing and generalization of the primary information as well as development of mathematic models for calculation of doses.

Currently CDR contains 9 LDB of radioecologic and dosimetric types :

Radioecologic LDBs :

RED - information on level and temporal variations of exposure dose in the territory;

SOIL - data on radionuclide composition of soil depositions;

FOOD - data on radionuclide composition of foodstuffs contamination ( experimental gamma-spectrometric data);

SR - information of radiostrontium content in the environment.

The volume of radioecological LDBs is currently 46 000 records.

LDBs of individual dosimetry . CDR consists of 2 LDBs with information on individual doses of internal and external exposure from radiocesium:

LDB TLD keeps information on results of direct measurements of individual external exposure doses made with use of termoluminescent (TL) dosimeters .

LDB WBC includes data on results of direct measurements of radiocesium content in the body made with use of whole-body counters (WBC) and on values of individual internal irradiation doses calculated on the base of these measurements.

Each LDB includes the data on the man under examination.

LDB TLD includes currently detailed information on results of 25 000 measurements of external irradiation doses made in UkSSR in 1987 - 1990. LDB WBC stores information on measurement results and annual doses calculated on their base for 175 000 people living mostly on Ukrainian territories. 100 000 of them was

made in 1986.

Other LDEs shown on fig.1 contains the following radiation dosimetric information:

-J includes the 151 000 records for the results of measurements of thyroid activity and calculated individual thyroid doses (May - June 1986).

- "Pripjat-Chernobyl" includes data on 63 000 people evacuated from Pripjat and Chernobyl and approximately 40 000 results of retrospective dosimetry. It's task is to personificated retrospective restore doses of external exposure received before the evacuation (during 36 hours after the accident) on the base of mass requesting and selfinterviews;

-"Background" is a special data base. It's main task is to process information and to calculate radiation doses from natural sources which are not associated with Chernobyl accident.

Ecological and dosimetric territorial data base . Each information units (IU) of the CDR second level contains certain types of radioecologic and dosimetric information on a given settlement. Information is combined in 10 large IU (fig.1).

IU 1 - 3 include reference information:

1 - geographical IU contains detailed administrative description of a settlement and its geographical coordinates;

2 -population IU includes data on children and adults population (according to Central Statistic Office (CSO) UkSSR);

3 - IU "Protective actions" keeps information on monitoring level in a given settlement and takes account of limitation and protective actions;

IU 4 - 6 store generalized information on environmental contamination level in a given settlement.

4. RED includes generalized information on level and temporal variations of exposure dose rate in a given settlement and in its neighborhoods.

5. Contamin. density concerns information on surface radiocesium contamination density of the territory.

6. Contamin. of foods represents generalized information on specific radionuclide contamination of different components of

ration (milk, milk products, meat, vegetables, fruits, mushrooms, and so on).

IU 7 - 9 are intended for the generalized information on public doses in a given settlement in 1986-1990.

7. External exposure keeps information on quantity and results of direct measurements of individual external exposure doses made with use of TL dosimeters as well as mean values and parameters of dose distribution in different occupation, age and sex groups. Besides results of direct dosimetry this IU contains calculated values of external exposure doses made on the base of REM values measured in a settlement in different periods of time.

8. Internal exposure is intended for the information on the value and parameters of individual internal exposure dose distributions in different occupation, sex and age groups, receive with help of direct measurement (WBCs) and calculation methods.

9. Total exposure is created for the information on value and character of total doses distribution (doses of external and internal irradiation and their relationships are included).

Information of 4-10 IU is the result of the treatment of the CDR lower LDB information level.

10. Prediction is of peculiar importance (see 10 in fig.1). It contains the results of predicted doses of external and internal irradiation calculations for the coming 70 years for the people living in a given settlement. The assessments of expected dose are made with help of methods providing different level of conservativeness.

Ecological and dosimetric territorial data base keeps currently the information on 62 000 settlements situated mostly on the Ukrainian territory and on the russian and bjelorussian territories suffered from the accident.

It goes without saying, that the degree of information quantity for each region, district and settlement is different. First of all, it depends on the degree the accidental influence on a given settlement.

Further we show the main laws of individual external and internal exposure doses formation received as a result of analysis and handling of information accumulated in CDR during 1986-1990. They considered as a bases for the assessment of doses to population living in a given settlement.

## 2. DOSES OF EXTERNAL IRRADIATION

The most adequate source of information on external exposure doses to population are the results of direct measurements with use of TL-dosimeters. However, accounting for the fact that it is impossible to make such measurement to the whole population and that in Ukraine they have been made only since 1987, the information on temporal variations of external exposure source (gamma-irradiation from contaminated surface (and from radioactive cloud which was passing in May 1986)) is of extreme importance.

LDB RED (fig.1) contain information on radionuclide composition of irradiating surface and on measured levels of exposure dose rate (1 m above the soil surface) made in 10 000 different points on the territory of Kiev and Zhitomir regions during 1987-1989.

The bulk of points corresponding to the value of monthly dose of external exposure (open area) on the unit of radiocesium contamination density,  $D_{\gamma/\sigma}$ , calculated with use of these data is given in fig.2. The character of  $D_{\gamma/\sigma}$  change in time is essentially different in the first 15 months after the accident and in later periods. As the statistic analysis of data gave shown, this dependence can analytically be presented in the following appearance:

$$D_{\gamma/\sigma}(t) = \begin{cases} 61,1 * t^{-0,70} \frac{10^{-5}, \text{cSv/mon}}{\text{Ci/km}^2} & ; 1 \text{mon.} \leq t \leq 15 \text{mon.} \\ 9,27 - 0,006 * t, \frac{10^{-5}, \text{cSv/mon}}{\text{Ci/km}^2} & ; 15 \text{mon.} < t \leq 42 \text{mon.} \end{cases} \quad (1)$$

where  $t$  is time (in months) after the accident.

It must be stressed that empirical dependence (1) is based on the results of direct measurements RED. It's means that  $D_{\gamma/\sigma}$  include the natural background as the constant dose- component. If the average means of natural background change from 10,6 to 14,3  $\mu\text{R}/\text{hour}$  (or  $6,1 \cdot 10^{-5} - 8,2 \cdot 10^{-5}$   $\text{cSv}/\text{month}$ ) the effective half- time decreasing of external dose rate which is induced by radioactive track -  $T_{\text{ef}}$  change from 30 to 10 years. That means of  $T_{\text{ef}}$  is connected with the Chernobyl accident. Although that estimation is too rough it shows that from the 15 months after accident the decreasing of RED is determined by the physical half-time of Cs-137.

Mean individual dose of external exposure in a certain settlement,  $D_{\text{ext}}(T)$ , for the time  $t=T$  is determined in the following way:

$$D_{\text{ext}}(T) = K_{\text{tld}/\gamma} * D_{\gamma/\sigma}(T) * \sigma, \quad (2)$$

where  $D_{\gamma/\sigma}(T)$  is described by relationship (1);  $\sigma$  is contamination density with Cs-137 in a given settlement;  $K_{\text{tld}/\gamma}$  is a shielding factor which determines the relation of individual external dose measured with TL-dosimeter to the dose ( $D_{\gamma/\sigma}(T) * \sigma$ ) in the open area for the same time interval T.

The value  $K_{\text{tld}/\gamma}$  and its distribution are calculated with use of data on direct measurements of individual external exposure doses made on the Ukrainian territory which are kept in LDB TLD (fig.1)

Fig.3 shows the distribution of  $K_{\text{tld}/\gamma}$  values for adult population in 1987, 1988, 1989 as well as information generalized for 3 years. The analogous results for children are given in figure 4. The value of  $K_{\text{tld}/\gamma}$  is changing in a rather wide interval: from 0,01 to 0,4 for children and from 0,01 to 1 for adults. It reflects the individual character of children and adult behavior (time spent in- and out- of door, type of room, etc.) as well as occupational peculiarities of adult population.

The generalized information, i. e. total statistics and results of log-normal approximation of parameter distribution



$K_{\text{intd}/\gamma}$  is shown in fig. 5.

It must be stressed that information given in fig.3,4 and in the table of fig.5 is related to the territories with contamination density more than 5 Ci/km<sup>2</sup>. It is due to the fact that on the territories with lower levels of radiocesium contamination the contribution of internal exposure natural gamma irradiation which is not associated with accident fall-outs (natural gamma background, irradiation from building materials) highly increases.

For assessment of external exposure dose in a given settlement the procedure of making of random figures generator simulating experimental distribution  $K_{\text{intd}/\gamma}$  (fig.3,4) is used for N inhabitants, living in a given settlement. The value of 90% - quantile of simulated distribution is used in (2) as a value of  $K_{\text{intd}/\gamma}$ .

### 3. INTERNAL EXPOSURE

The main sources of information on doses of internal exposure to population living on the contaminated territories are the measurements of radiocesium content in the human body made with use of WBCs. These data are kept in LDB WBC (fig.1)

Fig.6 shows the dependence of individual annual dose of internal exposure (averaged value over settlement) registered in 1986 from radiocesium contamination density of the territory.

The analog information for children and adults measured with WBCs in 1988 and 1989 is presented in fig.7 and 8. Since 1987 the limitations on consumption of local foods were entered in a number of Ukrainian settlements in order to reduce doses of internal exposure. So the information on the settlements "under strict radiation control" and on the rest of the settlements where the limitations were not officially entered is given separately in fig.6 and 7. (Though limitations on local foods consumption were not officially entered, the self limitations took place).

The dependence of individual annual dose of internal exposure  $D_{\text{int}}$  from  $\sigma$  approximated by the function of the following appearance:

$D_{int} = a + b * \sigma$  , where a and b are the parameters.

The values of parameters a and b determined in each postaccidental year for children and adult population living in the settlements with different level of radiation control are given in table 2.

Coefficient b ( $10^{-5}$ , cSv/Ci/km<sup>2</sup>) given in fig.9, characterizes additional dose on the unit of contamination while coefficient a ( $10^{-5}$ , cSv) characterizes some dose of internal exposure which is common for all territories ("controlled" or "uncontrolled") and for different groups of population (children, adults). This dose is not dependent on contamination density in a given settlement. It reflects the existence of some source of internal exposure which is common for all territories and creates a common "dose pedestal". It can be explained by the process of distribution and mixing of foodstuffs delivered from the territories contaminated with radiocesium.

As can be seen from fig.6-8 and 9, the essential dependence of internal exposure dose from contamination density was observed only in 1986 and was practically absent in 1987-1989. It is characteristic that the mean value of individual annual dose remains practically unchanged (for "controlled" and "uncontrolled" territories) in 1987-1989.

The absence of temporal variations of internal exposure dose value, especially for the settlements "under strict radiation control", is an evidence of relatively high efficiency of protective actions.

Information presented in the table of fig.9 is the main for determination of mean internal exposure dose value in the settlements situated on the territories suffered as a result of the Chernobyl accident.

#### 4. LIFETIME DOSE

If the main information sources for assessment of dosimetric situation in a given settlement in the first four years are the results of direct dosimetry (TLD and WBC), then the only way to estimate dose which could be received in future ("lifetime dose" for 70 years) is the usage of some

model which describes the possible development of radioecological and dosimetric situation in future.

Models of two types are used for this purpose, i.e. conservative model with deterministic values of parameters ( $D_{dmp}$  is a predicted dose estimated by this model) and simulation stochastic demographic model ( $D_{sam}$  is a dose estimated by this model). Conservative model was accepted by working group of USSR NCRP (Barkhudarov R.M., Lichtarjov I.A., Savkin M.N. et al.) in 1988 in Kiev and was officially used for the assessment of predicted doses to population.

The main sources of conservativeness in the assessment of some dose  $D_{dmp}$  are :

- the usage of maximum high deterministic values of metabolic parameters which are not age-dependent;
- the usage of constant and age-independending "start" value of external exposure dose on the unit of radiocesium soil contamination density;
- the common "start" value of radiocesium concentration in milk produced in a given settlement which is the mean value multiplied to 1.7 (this coefficient determines 90% - quantile of distribution) also suggestion about independence of radiocesium contamination of vegetable part of diet from radiocesium soil contamination density;
- a common deterministic daily intake of milk and vegetables which is not depending on age;
- usage of maximum value of the parameter that characterizes the period of half purification of foodstuffs from radiocesium which is equal to 14 years;
- the assumption that all member of critical group (children born in 1986) live on the contaminated territory for 70 years.

Certainly, the values of  $D_{dmp}$  can be used to receive some primary "upper" approximate estimates of expected dose. However, the estimates received in such a way overestimate the values of risk. This value can not be used in the procedure of "cost-benefit" analysis to estimate the acceptability of protective actions because each "unit of conservativeness" unjustified included in

the model could highly increase the costs.

Simulation stochastic demographic model for the estimation of lifetime dose developed by the others at them receive possible distribution of individual "lifetime doses" for an arbitrary group of people.

Here the following factors must be taken into account :

$K_1$  , is the real statistic distribution (stochastic character) of model parameters characterizing the source of exposure (contamination of foodstuffs, their daily intake, behavioral regime of people, etc), age-dependence and variability of metabolic parameters;

$K_2$  a real sex and age structure and dynamics of population (subpopulation), living on contaminated territory;

$K_3$  is more real value of ecological half life (of foodstuffs) of radiocesium.

The relationship of estimates received with use of 2 models is determined as:  $D_{osm} = D_{dmp} / K$  , where  $K = K_1 * K_2 * K_3$  .

The application of simulation model for the assessment of "lifetime dose" in the critical group has given the following values of the factors:  $K_1 = 1.7$ ;  $K_2 = 1.1$ ;  $K_3 = 2.1$  which corresponds to the value of the conservative factor  $K = 4.1$  . For the estimation of  $K$  the values of 95%-quantile of individual dose were used. The refusal from milk (contaminated more than 10 nCi/l) assures the value of  $K$  equal to 6.2 .

It's important to note that all values of common and partial conservativeness coefficients mentioned above must be considered only as illustrative estimations. They means only one of variants of models realization in the volume of simulating population equal to 8 000 people. That's why now investigation of model for receiving of statistical distributions  $K_i$  depending on size of simulating population and different relationships between magnitude of external and internal exposure is conducting.

In conclusion it must be stressed that in this report we considered only some sources of conservativeness (conservative coefficients  $K_i$ )  $D_{dmp}$  . We could not estimate for instance conservative coefficients which are cased age - dependence and stochastic characters of human behavior in ununique field of

external exposure or dependence of internal dose from un milk part of diet. Preliminary estimations showed that this factors can still decrease the significance of prediction dose per life.

# CENTRAL DOSIMETRIC REGISTER STRUCTURE

<b>3</b>	<b>Hygienic IU</b>
	Evacuation data Counter measures Level of control

<b>2</b>	<b>Populational IU</b>
	Population and its structure 1986 1987 1988 1988 1998

<b>1</b>	<b>Admin. geographical IU</b>
	* AC republic region district type place latitude longitude destination from Chernobyl

**SETTLEMENT 6200**

<b>E C O L O G Y</b>	
<b>4</b>	<b>IU RED</b>
	**1986 **1987 **1988 **1989 **1998
<b>5</b>	<b>IU contamin. density</b>
	**1986 **1987 **1988 **1989 **1998
<b>6</b>	<b>IU contamin. of foods</b>
	**1986 **1987 **1988 **1989 **1998

<b>D O S E S</b>	
<b>7</b>	<b>IU EXTERNAL EXPOSURE</b>
	calcul. T L D **1986 **1987 **1988 **1989 **1986-1998
<b>8</b>	<b>IU INTERNAL EXPOSURE</b>
	calcul. M B C **1986 **1987 **1988 **1989 **1986-1998
<b>9</b>	<b>IU TOTAL EXPOSURE</b>
	**1986 **1987 **1988 **1989 **1986-1998
<b>10</b>	<b>IU PREDICTION 1990-2056</b>
	** dose of internal exposure ** dose of external exposure ** dose of total exp.

**ANALYTICAL UNIT**

<b>Radioecology</b>			
<b>Sr 1000</b>	<b>RED 20000</b>	<b>Soil 10000</b>	<b>Foods 15000</b>
<b>TLD 25000</b>	<b>WBC 175000</b>	<b>J 151000</b>	<b>PRIPJAT CHERNOBYL 63000</b>
<b>BACK-GROUND 10000</b>			
<b>LOCAL DATA BASES</b>			

FIG. 1.

Dependence of external dose per unit of contamination density on time

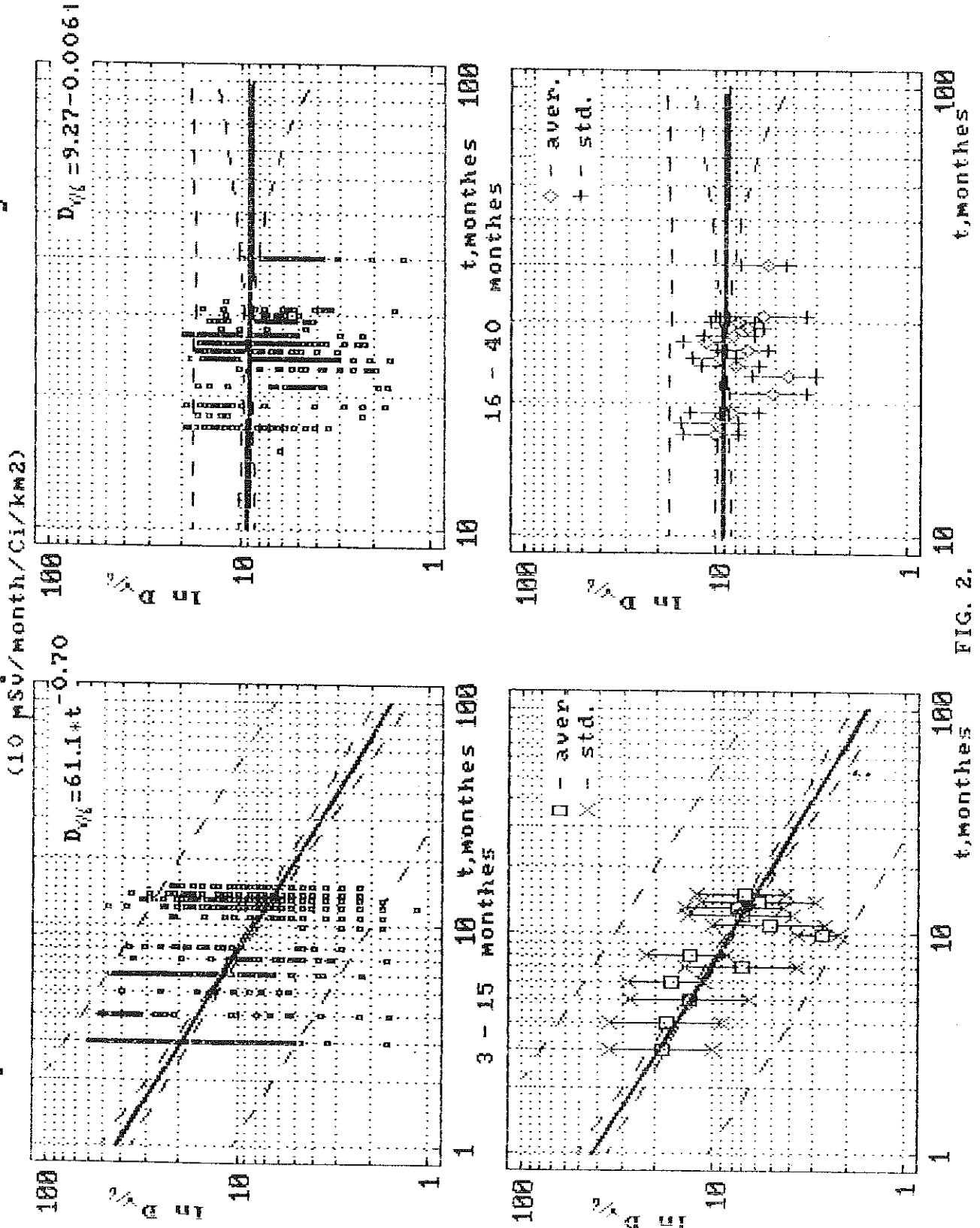


FIG. 2.

# ADULTS

$K_{t1d}/\gamma = D_{t1d}/(D_{\gamma_6} \cdot 6), G > 5 \text{ Ci/km}^2$   
 (lognormal approximation,  $f(n, \delta)$ )

$\hat{K}_{t1d}/\gamma$  - geom. aver.  
 H - men

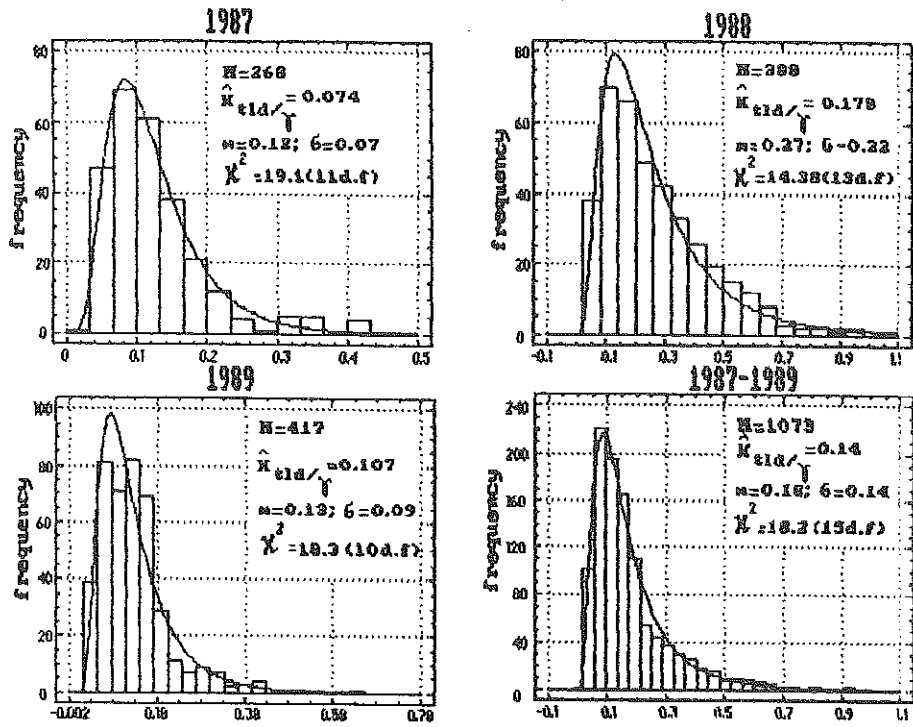


FIG. 3.

# CHILDREN

$K_{t1d}/\gamma = D_{t1d}/(D_{\gamma_6} \cdot 6), G > 5 \text{ Ci/km}^2$   
 (lognormal approximation,  $f(n, \delta)$ )

$\hat{K}_{t1d}/\gamma$  - geom. aver.  
 H - men

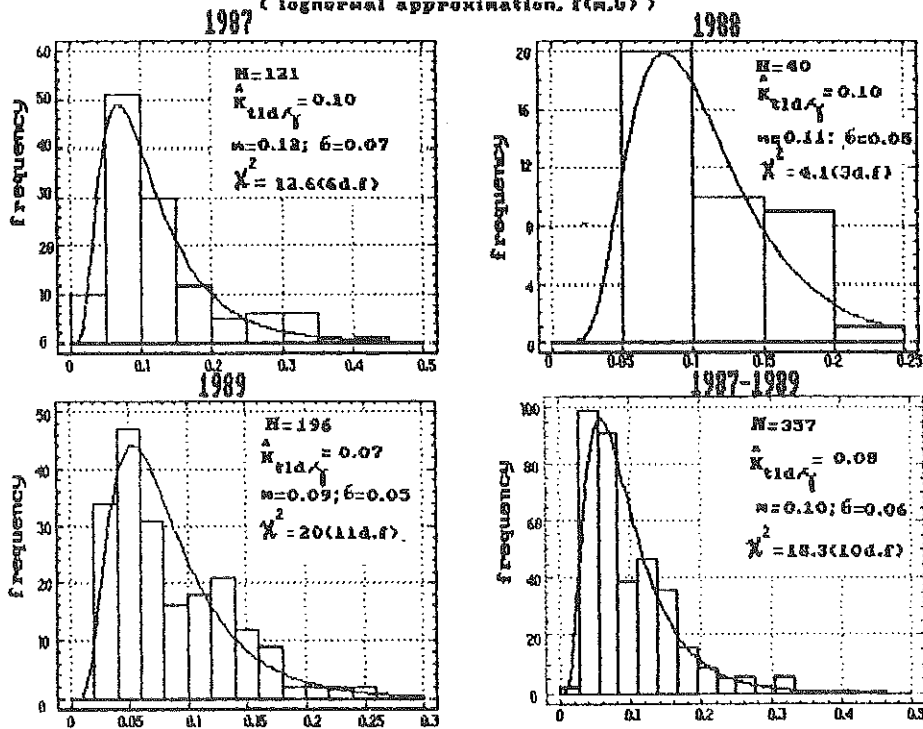


FIG. 4.



$$K_{tld/\gamma} = D_{tld} / (D_{\gamma/6}^{*} \cdot 6)$$

(General data 1987-1989,  $\bar{D} > 5 \text{ Ci/km}^2$ )

	N (men)	Summary statistic $K_{tld/\gamma}$		
		aver	std	Geom. aver.
Children	357	0,10	0,06	0,08
Adults	1073	0,18	0,14	0,14

# WBC MEASUREMENTS

N - settlements  
n - men

Adults

1986

Children

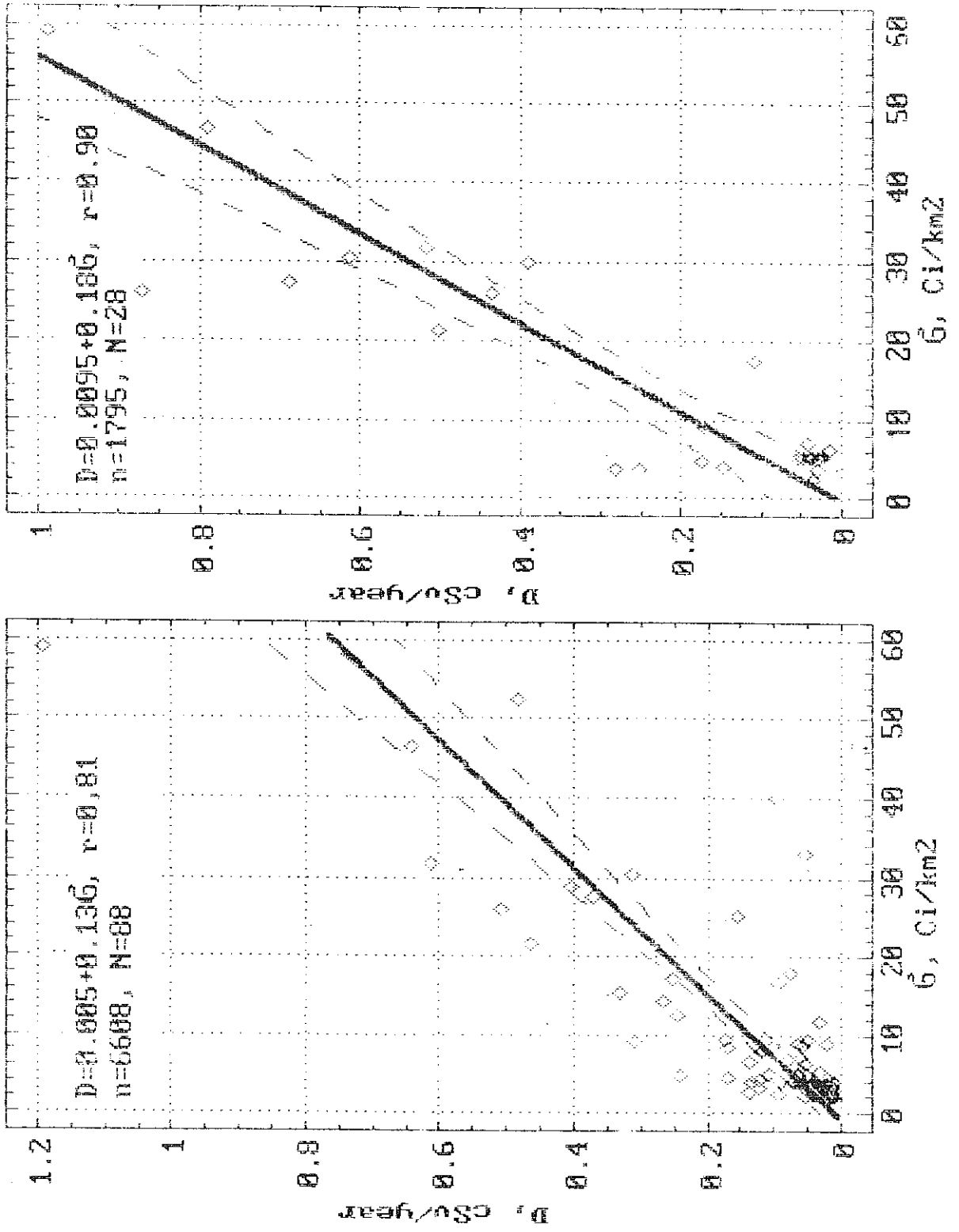


FIG. 6.

### WBC MEASUREMENTS

H - settlements  
n - men

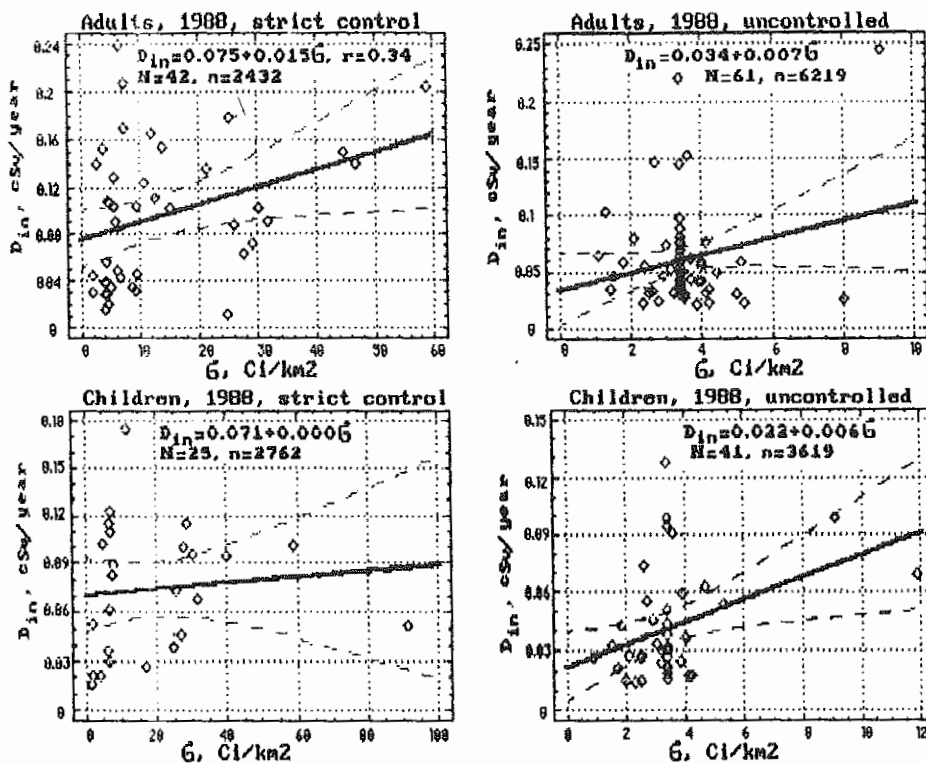


FIG. 7.

### WBC MEASUREMENTS

H - settlements  
n - men

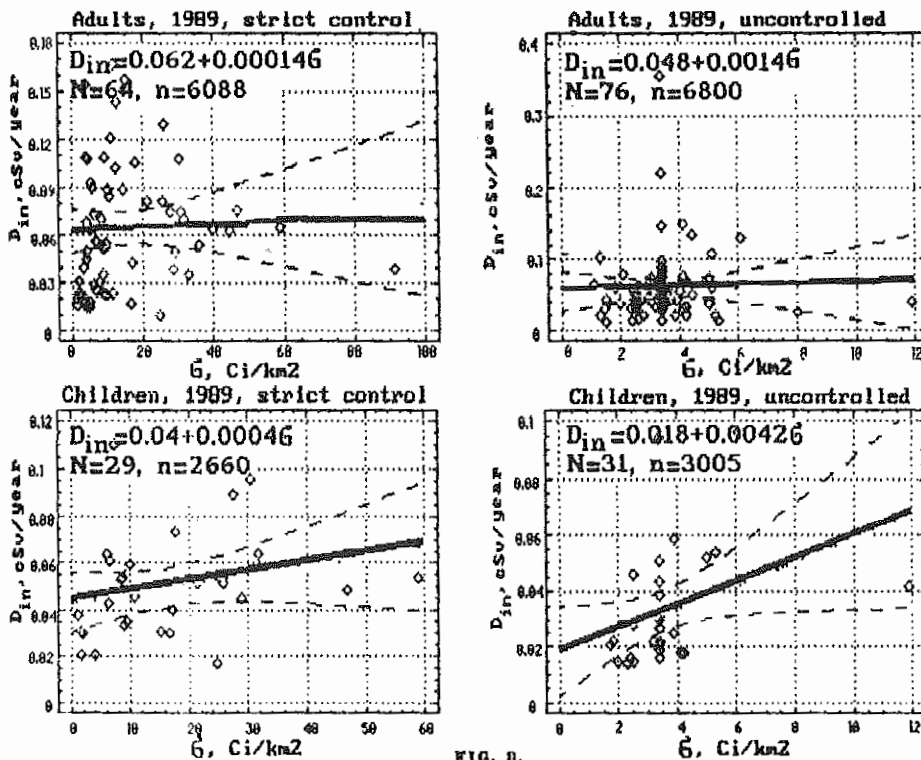
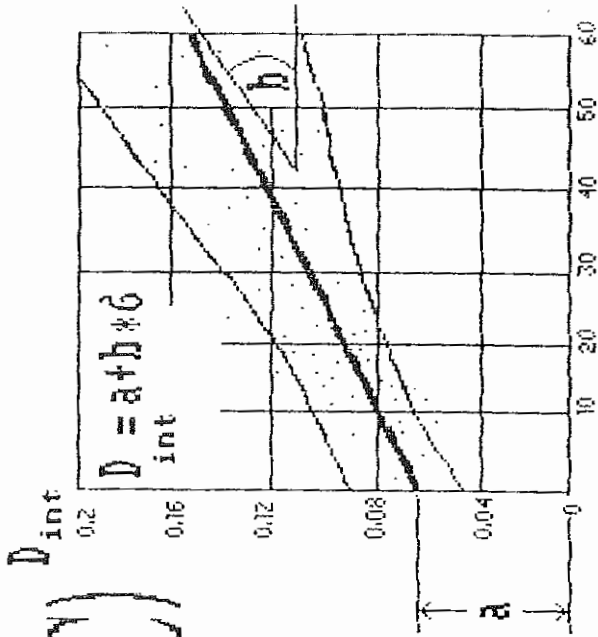


FIG. 8.

# INTERNAL DOSES (WBC)

$$D_{int} = a + b * \bar{C}$$

( $D_{int} = 10^{-5}$  Sv/year;  $\bar{C} - Ci/km^2$ )



	1986		1987		1988		1989	
	a	b	a	b	a	b	a	b
Children, s/c*	—	—	60 ± 15	1 ± 0	71 ± 11	0	43 ± 6	0 ± 0
Children, n/c**	5 ± 15	13 ± 1 <sup>m</sup>	20 ± 13	6 ± 3	22 ± 9	6 ± 2	18 ± 8	4 ± 2
Adults, s/c	—	—	71 ± 10	2 ± 1	75 ± 12	2 ± 1	62 ± 7	0
Adults, n/c	10 ± 37	18 ± 2	54 ± 16	0	34 ± 16	7 ± 4	55 ± 14	0

\*s/c - settlement under "strict control"

\*\*n/c - the remaining settlements

FIG. 9.



## Session VII

# **COUNTERMEASURES**



**The Experience of the Use of  
the Field and Stationary  
Methods of the Direct Control  
over Radioactive  
Contamination of the  
Environment by the Chernobyl  
Accident Products**

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The methods and means of direct continuous gamma-spectrometric measurements were used side by side with traditional methods of radiation control of environmental objects (sampling of soil-plant cover and water for further analysis) in the works related to the mitigation of the accident consequences at the Chernobyl NPS.

Being installed on different vehicles (helicopters, river research vessels, cars) or used in stationary conditions they allowed data on the dose rate and its anomalies in the different space-time scales to be obtained with high efficiency and proximate manner, to determine the different radionuclides contents in soil or their concentration in water "in situ" to control the quality of decontamination of the settlements etc.

Based on data from the continuous measurements (except the operative control) it was also possible to discover the space-time structure of radioactive contamination, to correct and refine the maps of radioactivity distribution plotted on the basis of interpolation of sample measurement results, to take representative samples according to gamma-survey results etc.

An advantage of the direct methods is also the possibility of direct data input into computer units to process, file systematize and store the data.

Gamma-quantum detection is mainly used for "in situ" continuous measurements of environmental parameters since beta- and alfa-radiation has a low penetrability even in air. Taking into account the occurrence of different scales of contamination it is obvious that the gamma-radiation field is more uniform as compared to beta-radiation since the non-uniformities of the local distribution are averaged over a considerably larger volume or area as a result of the larger free path of gamma-quanta. This also results in that, in contrast to the sampling method which is a differential method, the "in situ" measurements are able to yield at once the averaged estimates i.e. they are an integral method.

The quantum nature of gamma-radiation enables (after carrying out calibration actions for the used measurement geometry) the radionuclide contaminants structure to be assessed using the spectral composition of the gamma-field. As to such radionuclides as beta-emitting strontium-90 or alfa-active plutonium, their concentration with known initial ratio can be estimated from radionuclide activity of gamma-emitters /1/.

Nevertheless direct methods for control over soil-plant cover contamination with alfa-activity are also under development which is a subject discussed in the present work.

In this study we attempted to generalize five years of experience on the use of methods and means of direct continuous control of radioactive contaminants in the environment after the accident.

As an example of the use of these methods in stationary conditions we are considering here a stationary control of the radioactivity of the Pripiat' river water (1986-1989) and the use of direct methods of control of the water treatment cycle of the Dnieper water supply station (Kiev) in conditions of water input with increased content of technogenic radionuclides into the treatment facilities (1987-1988).

The use of direct continuous gamma-spectrometric measurements on the mobile control means is considered on the basis of experience in the use of a car complex.

#### A. Stationary use

##### 1. The continuous gamma-spectrometric control over the Pripiat' river water

The main purpose of the control consists in solving two problems :

- specific activity determination of gamma-emitters in water (in the first place iodine-131 and caesium-137)
- assessment of radionuclide deposition in water of the Kiev reservoir, a main water supply source for Kiev.

##### 1.1 Gamma-radiation detection

As a result of the accident and contamination of the water catchment area, twenty radionuclides-gamma-emitters were input into the Pripiat' river water. In these conditions it would be optimal to use immersible semi-conductors (HPSD), for example, "SHP-1" type manufactured by "ORTEC". However it is obvious that control must be concentrated on the most radioactive nuclides (iodine, caesium, strontium, plutonium). On the other hand the working conditions did not allow the detection equipment to be placed nearer than 200-300 m but in this case the use of HPSD is technically difficult. Also it is difficult to provide liquid nitrogen supply and production in field conditions.

To obtain source gamma-spectrometry information detection units based on sodium iodide crystals with different volume having high efficiency of gamma-quantum detection and acceptable energy resolution were used.

Just note two features of the direct measurement of the radioactivity level of natural waters :

- measurements are carried out in the emitting-dispersing medium which is water;
- measurements are generally carried out at small depths of natural pools (2-5 m).

The first feature leads to sharply increasing Compton's part of spectrum of any radionuclide. If there are some radionuclides with approximately equal specific activity in the water medium this results in sharply increasing total Compton's part of spectrum in its "soft-part" ( $\leq 500$  KeV) which deteriorates the "effects-to-background" ratio for this range where in particular the photopeak of iodine-131 occurs.

Water influence is also reflected on deterioration of energy resolution. This is shown in Fig. 1 by means of the example of measuring point and distribute sources of zinc-65.

The second feature consists in that the pulse quantity  $N_f$  measured with the detector at a certain depths is the sum of some components :

$$N_f = N_m + N_c + N_b + N_n,$$

- $N_m$  - actual radioactivity of water;
- $N_c$  - cosmos component distribution on the detector's own background ( $h=\text{const}$ ,  $N_c=\text{const}$ );
- $N_b$  - own background of the nature;
- $N_n$  - contribution of bottom natural and technogenic radioactivity.

The bottom radioactivity is a serious problem in measurements made in shallow water. In /2/ it is shown that when measuring with water-distributed cobalt-60 sources 98% of the gamma-quantum flux on to detector is created by radiation from a spherical source 65

cm in radius. Thus the presence of a water column of 2-2,5 m can be considered as an acceptable screen against the bottom natural and technogenic radioactivity.

In the initial period (just after localization of accident ejection) water activity was high enough (up to  $3 \cdot 10^{-8}$  Ci.l $^{-1}$ ) and  $N_f \approx N_m$ , however, these components begin to play an essential role, when  $N_m \ll N_c + N_b + N_n$ . Just this limits the use of the direct methods.

This limitation, as it is known, is quantitatively expressed by the sensitivity limit of the detection method which is determined as the minimum activity that can be measured in present conditions :

$$A_{\min} = \frac{\sqrt{\delta} \cdot t_{\alpha} \cdot G_{\delta}}{\delta \cdot S \cdot T}$$

- $\delta$  - basic error,
- $t_{\alpha}$  - Student's test,
- $G_{\delta}$  - standard deviation of background measurement,
- $S$  - measurement method sensitivity,
- $T$  - measurement time.

For each radionuclide the value  $\delta$  for the detector sensitivity is defined for this radionuclide in the considered measurement geometry (source uniformly distributed in water) and the value  $G_{\delta} = \sqrt{N_{\delta}}$  is measured in a radiation-free reservoir at the same depth.

These values were obtained by us when carrying out calibration actions on the basis of radiochemical analysis of water samples. When calculating total specific activity a value obtained for the observed set of radionuclides is defined as  $S_{\Sigma}$ .

It is clear that as the composition of radionuclides changes,  $S_{\Sigma}$  needs correction.

In Tables 1 and Fig. 2 are given results of the determination of the value  $S_{\Sigma}$  for a sodium iodide 80x80mm detector in the energy range of 0,15-1,8 MeV.

Table 1

<u>Date</u>	$S_z, \frac{\text{imp. sec}^{-1}}{\text{Ci. l}^{-1}}$
28.05.86	1.6 .10 <sup>11</sup>
22.07.86	1.3 .10 <sup>11</sup>
27.03.87	0.86.10 <sup>11</sup>
27.09.87	0.57.10 <sup>11</sup>

Decreasing gamma-activity of the Pripiat' river water resulted in need of using detectors with large values S. However it should be realized that increasing S leads to growth of mass of detector i.e. to growth of  $N_b$ . Our experience has shown that detectors of square section show better perspectives in this term as compared with the ones of rectangular section.

Taking into account the mean effective energy of fission products gamma-radiation amounted to 0,7-0,9 MeV for the initial period and then decreased to 0,6-0,8 MeV, these detectors are more "efficient" in increasing sensivity (S/m, m-detector mass) as compared with detectors of rectangular shape.

Note also that detectors were placed into hermetically sealed capsules from radiation-free stainless steel. In order to reduce radionuclide sorbtion on the capsule surface removable PVC covers were put on them.

## 1.2 Control, measuring and data processing equipment

The detector impulses were transferred through cable with low wave resistance\*) to the input of the continuous measurement equipment and multi-channel analyser.

Continuous measurement equipment consisted of the preliminary processing and documentation unit as well as a personal computer.

The preliminary processing and documentation unit comprised four ports to each of which one detector can be connected.

Each port included an input controlled spectrometric amplifier, four working differential discriminators as well as two-digit counters with liquid crystal indicators which can be connected to any discriminator.

The unit had a device for setting astronomical time, exposure time; comparator unit for setting the level "ALARM" as well as a numeric printer enabling to output actual information (in form of impulse amount for exposure time) with reference to astronomical time onto paper(tape) medium. Every path had also analogue data output from one of the discriminators (see Fig. 3).

Equipment operates in cycle mode preset with exposure time. Impulses from all the differential discriminators go into computer in parallel.

Its available software allows data to be processed in real time with any averaging base. It also allows to convert automatically information into specific activity units, to command for triggering of multi-channel analyzer (when exceeding level "ALARM" or by cycles).

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\*) single-circuit scheme of low voltage supply to detector and pulse transfer was used

Thus taking into consideration the specific conditions of operation the continuous control equipment could operate both with computer and without it (in case of failure)\*).

Energy "windows" of discriminators can be readjusted according to the changing radiation situation. Fig.4 gives an actual gamma-spectrum of the Pripiat' river water on 25.05.86 and indicates the fixed energy bands of detection : "iodine", "caesium", "integral".

Exposure time was varied depending on the radiation situation. So  $T=60$  sec. used in the initial period allowed to control an "overshoot" of activity with value of 0,04 Ci. Later on as activity dropped the exposure time was increased up to 600 sec.

Placing the detectors on anchored buoys allowed the measurements to be conducted in winter too (under ice).

Threshold sensitivity of the method using sodium iodide crystals of 80x80mm in size at the depth of 2 m amounted to  $0,7 \cdot 10^{-11} \text{Ci.l}^{-1}$  with exposure of 60 sec. and  $0,25 \cdot 10^{-11} \text{Ci.l}^{-1}$  with exposure of 600 sec.

The use of crystals with size of 80x400 mm allowed this value to be reduced twice.

The use of the direct continuous control equipment enabled activity of iodine-131 to be determined down to  $3 \cdot 10^{-10} \text{Ci.l}^{-1}$  with a basic error of  $\pm 30\%$ , total radiocaesium - down to  $5,8 \cdot 10^{-11} \text{Ci.l}^{-1}$  with basic error or  $\pm 25\%$ .

This equipment without any essential modifications was placed on the river vessels used for the mobile monitoring.

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\* ) in this case part of operations are carried out by hand.

2. Control over operation of treatment facilities of the Dnieper water supply station

The accident at the Chernobyl NPS resulted in radioactive contamination of the Kiev water reservoir and Dnieper river, the main sources of drinking water supply for Kiev. Here in the initial period radioactive contamination of the Kiev water reservoir took place both by way of activity fall-out on its surface and by way of input of contaminated waters of the Pripiat river, Uzh river a.o.. Subsequently only the second source contributed.

According to data /3/ in May 1986 the maximum activity of iodine-131 in water of the reservoir amounted to  $3 \cdot 10^{-8} \text{Ci.l}^{-1}$  (1100 Bq.l<sup>-1</sup>), in June 1986 it reduced up to  $4,3 \cdot 10^{-10} \text{Ci.l}^{-1}$ . In June 1986 activity of caesium-137 amounted to  $4,9 \cdot 10^{-10} \text{Ci.l}^{-1}$ , strontium-90 -  $3,5 \cdot 10^{-11} \text{Ci.l}^{-1}$ .

Continuous control equipment similar to the one used on the Pripiat river was installed on February 1987 at the treatment facilities of the Dnieper water supply station for the following purposes :

- control over gamma-activity level of drinking water,
- effectiveness assessment of treatment system operation,
- assessment of dynamics of radionuclide accumulation on the used sorbents.

Detector units (sodium iodide, 80x80mm) were placed so that all the basic stages of water treatment could be controlled :

- in receiving bowl - control over input water,
- in settler of fine filter - control over filter-cycle,
- in fine filter material - control over dynamics of radionuclide accumulation if filter,
- in pure water tank - control of output water.

From the standpoint of interpretation of the measurement results a difference from the use on Pripiat' river consisted in experimentally measuring the value S for the detector located in the filter material (zeolite).

To extend caesium-137 accumulation in zeolite the filter detector was substituted by a detector of smaller size (40x40mm). At the end of 1987 the output water activity was at the sensitivity limit of the method ( $3 \cdot 10^{-11} \text{Ci.l}^{-1}$ ).

B. Use of continuous control methods in cars

Besides what was mentioned, the use of direct methods of gamma-spectrometric control in cars has a number of further advantages discovered during operation in contamination areas :

- possibility of operation both in mobile and in stationary conditions by quickly changing control means location depending on situation;
- intercalibration of stationary means;
- aerogamma-survey results correction;
- operative assessment of horizontal migration of radionuclides on the control routes;
- possibility of detailed inspection of settlements and assessment of decontamination effectiveness.

### 3.1 Car complex for control and study on radionuclide composition of environmental contaminants

A block-diagram of apparatus part of complex is given in Fig.4. The technical means of complex enabled :

- to carry out the continuous measurements of gamma-field intensity and spectrum composition with two collimated scintillation detectors placed on the car top (on the two gates at height of 2m);
- to carry out continuous measurements of gamma field intensity with non-collimated scintillation detector-discover placed on the car top;
- to obtain data on radionuclide contents in soil by direct gamma-spectrometry on the foot by using an outboard collimated scintillation or semi-conductor detector;
- to determine the basic composition of radionuclide or total specific gamma-activity of surface waters by means of the immersible scintillation detector.

All the detectors and equipment are completely unified with the ones used in the stationary conditions in order to ensure measurement unification.\*)

Surveying procedure did not considerably differ from that accepted in the surveying nuclear geophysics /4/.

The procedure of the carried out measurements of gamma-field characteristics consisted in recording, processing and topographic referencing of its intensity and spectrum composition data transferred from the collimated detectors and in recording of counting rate from the detector-discover as well.

Interpretation of gamma-field intensity data has some features when obtaining them in motion.

As it is known that essential characteristics of the mobile continuous control means are a "linear resolution" value \*\*) of

\*) In the car there was used multi-channel (pulse-height) analyzer LP4900B (AFORA, Finland), which has shown high reliability when operating in the conditions of the strong environmental and mechanical effects.

\*\*) for aeroplanes - "area resolution".



measurements process; this value is determined by a constant of information collection time (using of  $\Omega$  or RC-integrators) and a car motion speed (V), on the one hand, and by the minimum required change detection of dose rate on the route areas from which the information is integrated, on the other hand. Properly speaking, just this characteristic determines a possibility of surveying with a certain extent of detail and control means efficiency as well. The use of the RC-integrators requires the readings of intensity I and coverage of anomaly  $\Delta$  (see Fig.5) to be regenerated, to get rid of inertial ability  $\sqrt{t}$  (where  $\tau$  - integration time). We used RC-integrators with a smaller quantization time of 10 sec. This enabled the intensities of anomalies with coverage of 40m and 20m to be detected with an accuracy of 5% and 25% respectively when moving at a speed of 4 km/h.

Estimates of "inertial" length  $\Delta X$  showed that it doesn't exceed 50m at a speed of 40 km/h and 5m at a speed of 4 km/h.

### 3.1.1 Measuring with the collimated gate (site) detectors

These detectors are used for carrying out measurements of gamma-field intensity and qualitative assessments of the spectrum composition.

The measurement scheme and basic measured and preset values are given in Fig.6.

Dose rate in point A as it is known is calculated from the ratio :

$$P_A = K_{\gamma} \cdot g \cdot \ln \left[ 1 + \left( \frac{r}{H} \right)^2 \right]$$

$K_{\gamma}$  - gamma-constant of radionuclide;

$g$  - radionuclide contents.

For the case of the uniform gamma-quantum flux from all the sides a collimator thickness is determined from the condition  $\Omega / 4\pi F \ll 1$ , where  $F$ =attenuation coefficient. Taking into account the mean effective energy of fission products of 0,7-0,9 MeV, the lead thickness of 3cm enabled this relation to be met.

The collimator was a lead cup with length exceeding the dimensions of scintillation crystal placed inside a length equal to the crystal size. This provided 10% of gamma-quantum falling into the collimator surface to reach the crystal the open cavity.

In the side wall of the collimator there was a slot coinciding with the detector crystal position and ensuring detection within solid angle  $\Omega = \pi/3$ , which enables to "refine" measurement geometry.

The collimator mounting on the top enabled to fix the collimator slot at angles  $\beta = 30^\circ$ ;  $45^\circ$  and  $60^\circ$  with the vertical. In this case (with  $\beta = \text{const}$ ) the detector readings will be related to contaminant contents with simple correlation and will not depend on distance from soil surface :

$$\Delta N = A \cdot g \cdot \frac{\Omega}{\cos \beta}$$

A- calibration coefficient depending on detector arrangement and spectrum composition.

Thus when using averaged spectrum it is possible to estimate  $\int_0^t$  of the individual radionuclides, in the first place the long-lived radionuclides of caesium-134, 137. In the course of continuous gamma-survey at a motion speed of 5-40 km/h a set of gamma-spectra provided data on the spectrum composition of the gamma-field averaged over a section of 1,5-10 km (in linear representation) to be obtained within  $10^3$  sec.

The need to obtain the spectral information was related to the existence of three types of contamination differing in caesium-134, 137 contribution in the total contaminant contents.

On that ground the coefficient of transfer from counting rate to exposure rate was different for the different contamination types. The use of a unified coefficient resulted in 50% error. This problem is considered in more detail in the section concerning calibration of the technical means of the complex.

Another difficulty in the interpretation of the measurement results was that it failed to find an acceptable linear regression equation between counting rate and dose rate for the whole range of exposure rate of 0,04-20 m R.h<sup>-1</sup> existing during the first year after accident; it is generally characteristic of the use of sodium iodide for dose rate measurement. For the individual subranges (0,04-0,4; 0,7-5mR.h<sup>-1</sup> etc.) such linear equations were obtained and used in calculations.

### 3.1.2 Measuring with outboard detector

When the data obtained in motion show a significant change\*) in dose rate or spectrum composition of the gamma-field the quantitative determination of the radionuclide contents was carried out standing still using the outboard scintillation detector with collimator (sodium iodide, 40x40mm or 63x63mm). The collimator is made in form of lead cylinder with open face part and wall thickness of 5 cm.

The measurement are carried out while protective means and detector are installed on the fixing support so that gamma-quantum "collection" takes place from an area of 1 m<sup>2</sup> which facilitates the interpretation of results in the units Bq.m<sup>-2</sup>.

\*) "Thresholds" for such decision depended on the problem to be solved.

In the early stage of the accident situation when a large radionuclide nomenclature was found a HP Ge-detector with volume of 70 cm<sup>3</sup> in the same geometry was used.

The measurements are occasionally followed by sampling of soil in the layer of 0-10 cm from measurement site for the further determination of strontium-90 and plutonium content.

The measurements made with immersible detector did not differ from the ones for the stationary location and were discussed above.

### 3.1.3 Calibration measures

The development of the calibration procedure for the complex means and their regular testing were very important for the proper interpretation of the results.

The calibration was carried out on specially chosen and preliminary studied sites being representative in landscape-geochemical respect. There were three sites of such kind in the area of the fuel-type contaminants (with different contents), three sites - in the areas of the contaminants with sharply fractionated nature (with different contents as well) and two sites - in the areas of contaminants with intermediate nature. The calibration was carried out stationary using both sampling from the collimator "lightened" surface of site (not less than 5) and tested means for measuring of dose rate used as the controls.

Also the value of the conversion coefficient between surface activity of caesium-137 and dose rate created with caesium-134, 137 for the direct measuring means was estimated.

The dose rate change caused by shifting source center into dee soil was also estimated.

Measuring "linear resolution" and minimum required change of dose rate is carried out during motion of the car in the different detection conditions either when driving over the bridge above the water surface where dose rate is decreased or when driving through the cross-roads where activity on the asphalt is less than on the sideways.

In 1990 the joint calibration of technical means of complex and the contamination direct measurement equipment represented by the Japanese specialists (sponsor of works is company of NHK, Japan) was conducted on the sites of sharply fractionated type contaminants. The works are conducted under the above mentioned scheme and also included a comparison of radiochemical methods for determination of strontium-90 and plutonium in the samples. As a whole, the direct measurement equipment showed a good coincidence of results. On completion of results processing those will be published in form of a joint report.

The carried out metrological measures have shown that a basic error of caesium-137 determination in the range of 0,05-50 Ci/km<sup>2</sup> by means of the scintillation detector changes from 50% for the lower limit up to 15% for the upper limit (for HPSD- 30% and 5% respectively).

An error of caesium-134 determination in the range of 0,3-15 Ci.km<sup>2</sup> is also close to these values.

Time of gamma-spectrum measurement here amounted to 10<sup>3</sup> sec.\*. The error of Ru-106 determination changing within the range of 1-4 Ci/km<sup>2</sup> is close to 100% for the scintillation detector and amounts to 30% for HPSD.

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\* Large value results in decreasing control efficiency.

Cerium-144 determination with a basic error of 30% is possible only when the value exceeds  $10 \text{ Ci/km}^2$ , provided the caesium contamination does not exceed  $5 \text{ Ci/km}^2$ .

As an example Fig. 7 shows the determination of the dose rate and contents of some radionuclides along the control route and the results of the continuous measurements of dose rate. More detail for assessing the horizontal migration on the same route is given in Fig. 8.

### 3.2. Car complex for radiation surveying and control over environment

The experience of using the car complex discussed in 2.1 indicated that resolving the practical problem of quick collection of information on the exposure rate with simultaneous referencing to the basic radionuclide composition of fall-out (in order to obtain assessments of the radiation impact on the inhabitants) is often difficult because of the used measurement geometry. In the contaminated areas there is a complex spatial structure of activity "spots" of different size (up to meters) some of which have radionuclide contents exceeding tens and hundreds of times a mean level and not getting into the field of vision of the collimator in the process of gamma-survey.

On the other hand so called "unapproved" radiation sources which have reached environment for different reasons (radioactive waste, sources of radioisotope diagnostics etc.) are found in the many settlements of the USSR.

Their search requires means with high efficiency and high spatial selectivity.

Solving these problems with use of the above discussed complex is a difficult matter.

It is clear that these difficulties are mainly related both to the assessment of dose rates from volume-area extended sources when using the collimated detector and to the impossibility to discover in motion a direction of maximum radiation rate indicating the source having a main contribution to dose in the given place.

The car complex which was developed to solve these problems combined the process of continuous measurements of gamma-field intensity and use of the gamma-location principle (crude estimate of space part direction from where maximum gamma-quantum flux falls down on to detector) and gamma-bearing principle (estimate of maximum radiation source direction).

The sensitive part of the gamma-locator comprised 6 sodium iodide crystals with hexahedral section ( $63 \times 250 \text{ mm}$ ) arranged close to each other on the generating line of a cylinder with diameter of 8 cm (Fig.9) made of tungsten.

Between adjacent faces of crystals there was a lead "shady" screen device of triangular section. Such scheme created a certain angular sensitivity for each detector. This part of the locator was placed on the car top at a height of 1,5 m.

Pulses from each detector were transferred independently to input of board computer, analyzed according to conversion of the counting rate into dose rate and represented in analogous and digital form to the operator on the display screen (Fig.10).

For the operator's convenience a car mimic panel with vector indicated a direction of maximum radiation source (space part) in the corner of screen.

For an accurate estimate of the source direction it was possible to use one of two alternatives :

- simultaneous connection of any of three adjacent detectors into operation mode of computer according to program of "Peleng" (bearing);
- use of four detectors (sodium iodide, 63x63 mm) with collimator in form of semi-cylinder placed on the car bumper at height of 1 m with base of 1,5 m.

The especially developed computer program of "Peleng" enabled also (after proper calibration against a point source) to estimate the distance to the maximum radiation source. To correct a car motion according to results of gamma-location a set of equipment included also a small-size telecamera with monitor. The video-image of the locality also displays the result of data processing under program of "Peleng" in analogous form (stroke with height proportional to dose rate positioned at the location of maximum radiation source) (Fig.9a).

During continuous surveys information on dose rate is recorded on magnetic floppy-disk and video-information - on the video tape recorder.

Composition of radionuclide contents is analyzed by means of "in situ" portable HP Ge-detector with collimator.

In case of need this detector can be placed on the car top for continuous gamma-spectrometric survey.

#### 4. Direct measurement of $\alpha$ - activity soil cover

The labor involved in the radiochemical determination of radionuclide contents has stimulated works for the creation of means for the direct assessment of the total activities.

Difficulties in the development of such techniques are well known and the existing means don't enable  $\alpha$ - particle flux to be determined remotely.

In conditions of soil covered with dense vegetation the use of the use of the contact means is practically impossible.

In the first phase of development of such means the task was undertaken to achieve remote direct determination of total activities for an  $\alpha$ -particles flux of  $\geq 0,5 \frac{\text{part}}{\text{cm}^2 \text{ sec}}$ \*

The physics of direct measurement process was based on the principle of detecting not  $\alpha$ -particles themselves which have too short paths in air but "clusters" (or "tracks") originating in air during its ionization by  $\alpha$ -particles and retained there for considerable time (until 15 min.). The problem consists in "pulling-in" the "cluster" aeroions by means of magnetic field or air eddy flow into a measuring counter.

In our model\*\*) we have realized the second principle.

The detection unit in the working position is located at distance of 30 cm to soil surface. Circulating flow transfers aeroions from areas of 0,26 m<sup>2</sup> to a detection unit of original development which is a gas-discharge counter to which aeroions are transferred by means of special transporting and ion-focusing electrodes. The detector is opened to atmosphere which enables to reduce the sensitivity threshold of the detector and to achieve energy of detected  $\alpha$ -particles of 100 KeV.

\*) accepted threshold for boundary of settle off zone (0,1 Ci/km<sup>2</sup> with plutonium)

\*\*) jointly with Moscow Engineering-Physical Institute

Fig.11 indicates a dependence of aeroions counting rate and detection effectiveness of aeroions from  $\alpha$ -particles of plutonium-239 on the distance to the source.. One can see that the detector perceives the aeroions from a source with an activity of 250 Bq at a distance up to 65 cm which shows the perspectives offered by the solution which we have chosen.

The accident at the Chernobyl NPS has stimulated works for development of field means for radiation monitoring for different purposes (5,6) in many countries.

Our experience in the development and use of such means as well as the experience of other researchers /7/ has shown that one of the main problems remained improving the accuracy of obtained results and the unification of calibration procedures on the special testing grounds.



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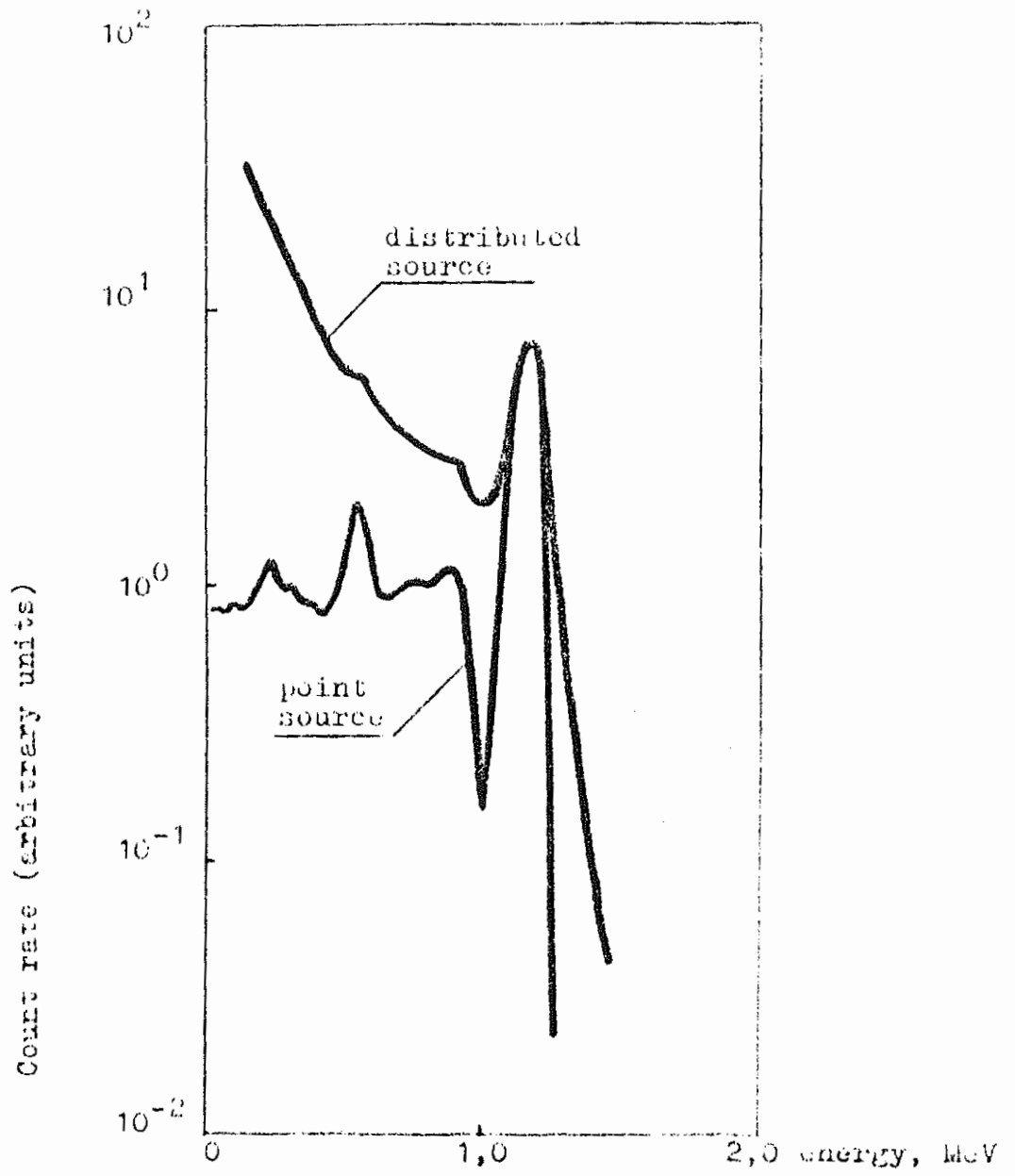


Fig.1. Gamma spectra of Zink-65 from a point and a distributed source.

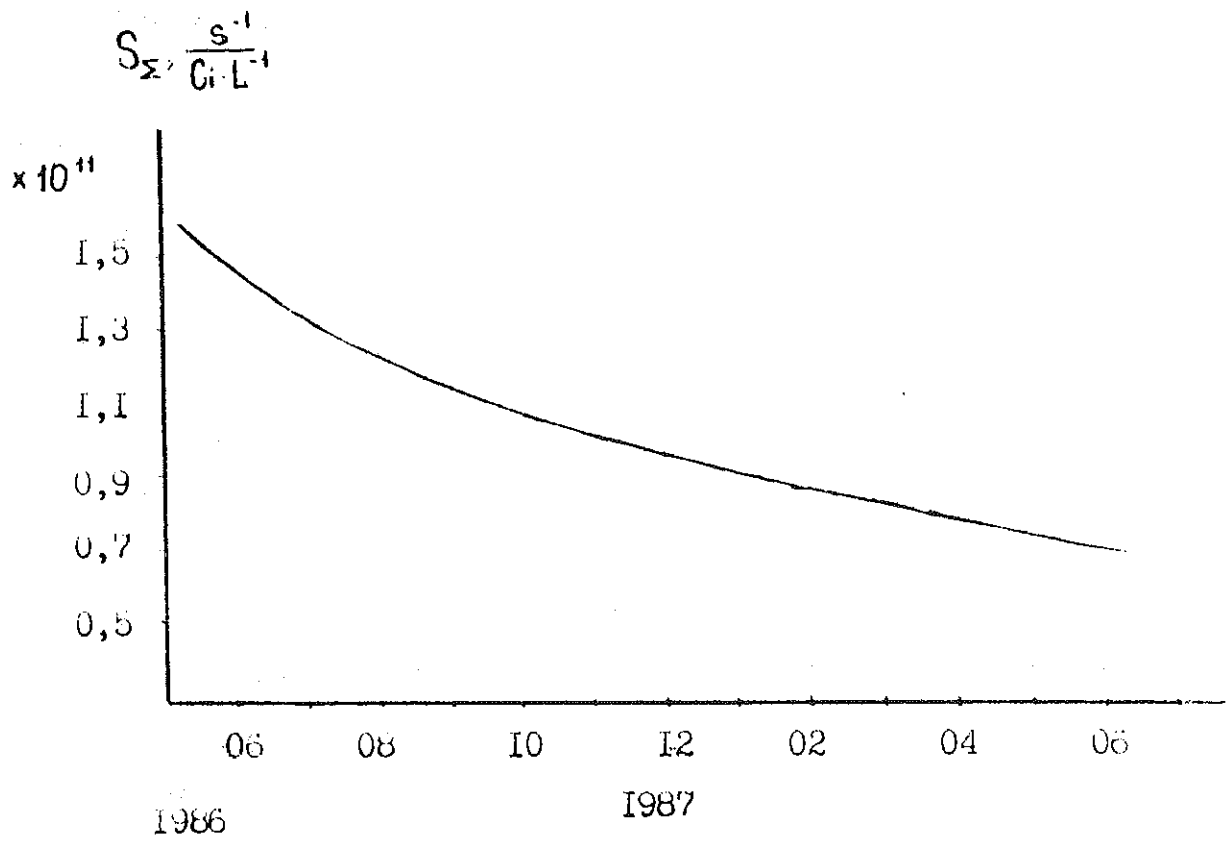


Fig.2: Sensitivity of measurements in the course of time in the river Pripiat.

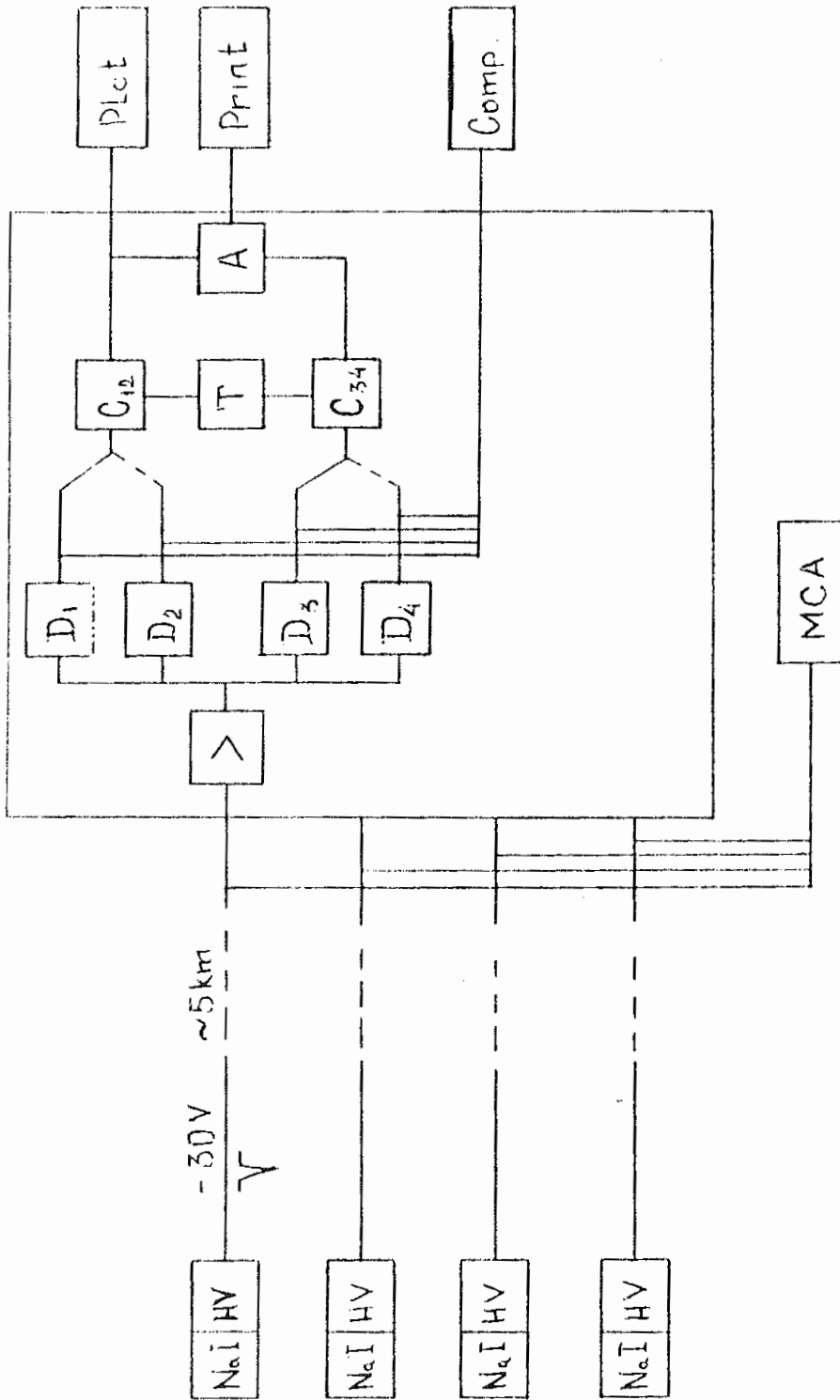


Fig.3 Circuit diagram of the equipment

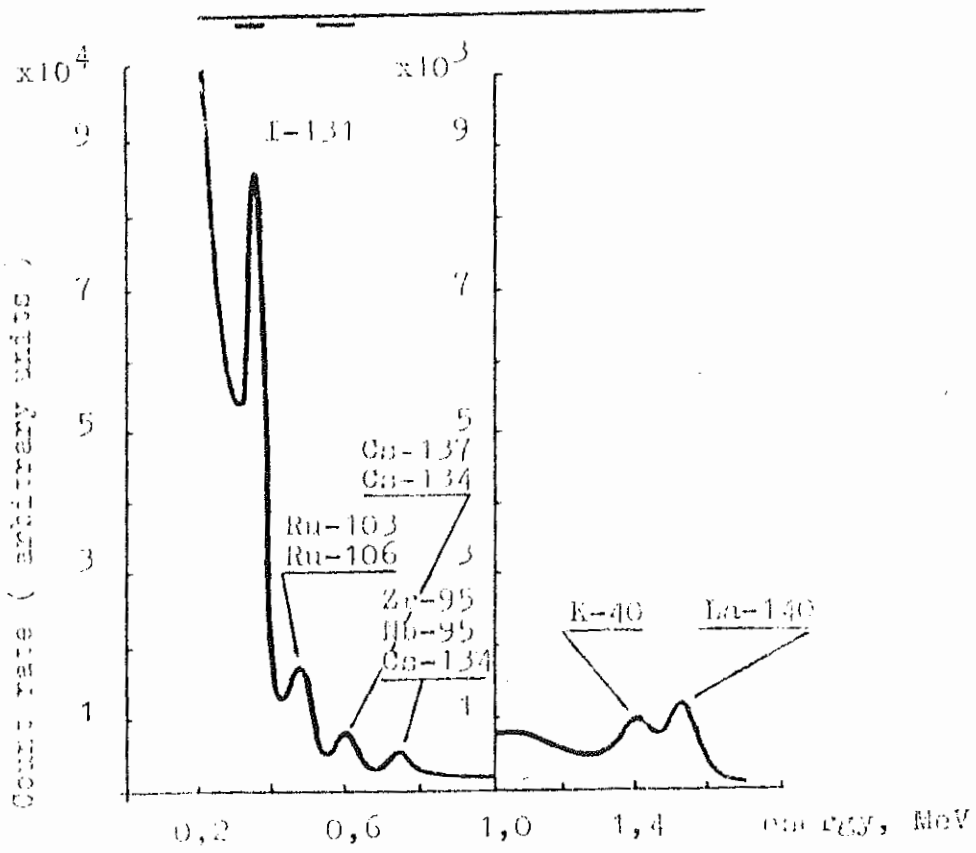
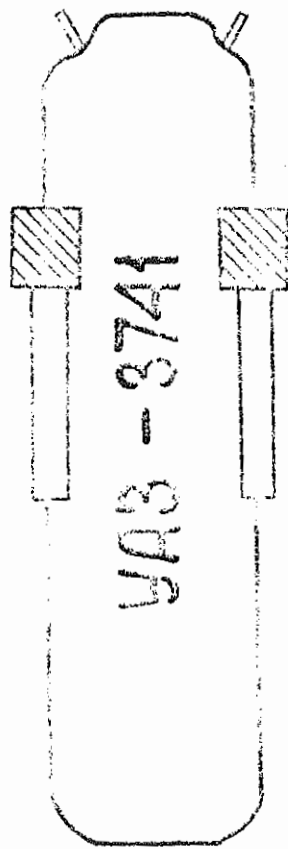
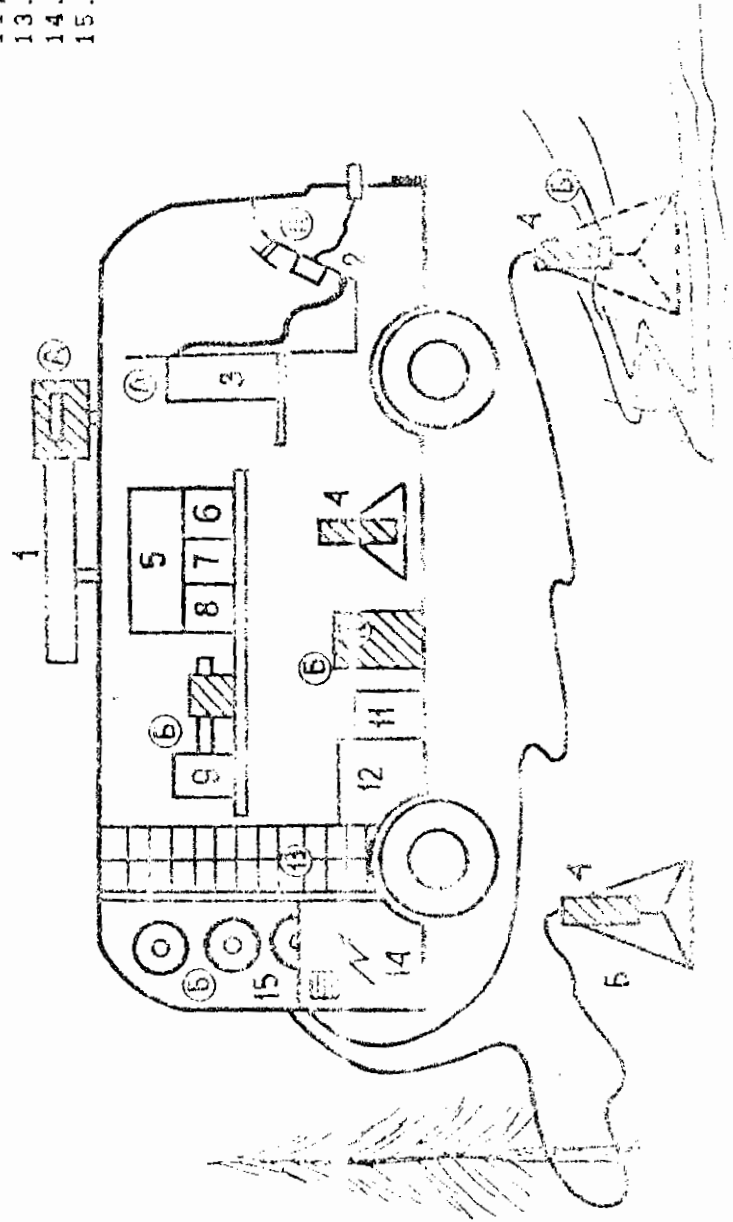


Fig.4: Gamma spectrum of water in the Kiev reservoir.  
(May 30, 1986)

Figure 3. Schematic of the equipment



1. Collimated detector
2. Intensitometer
3. MCA monitor
4. Outboard collimated detector
5. MCA
6. Tape-recorder
7. Floppy-disk unit
8. Printer
9. Low-level gamma-radiometer
10. Low-level beta-radiometer
- 11,12. Expedition equipment
13. Container for soil samples
14. Generator
15. Cable



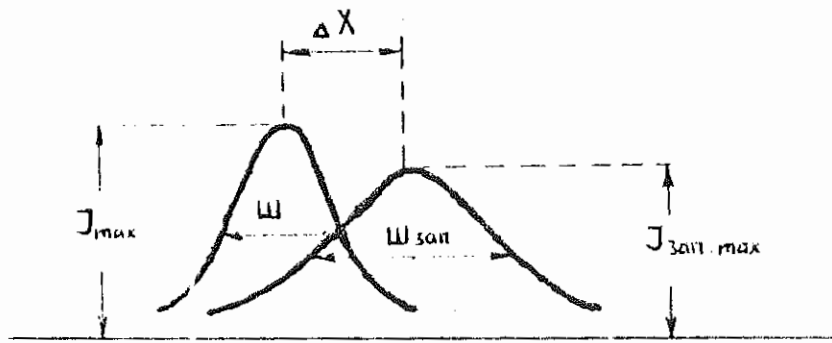


Fig.5 Record of anomaly ( $\Omega_{3ant}$ ) and in actuality ( $\Omega$ )

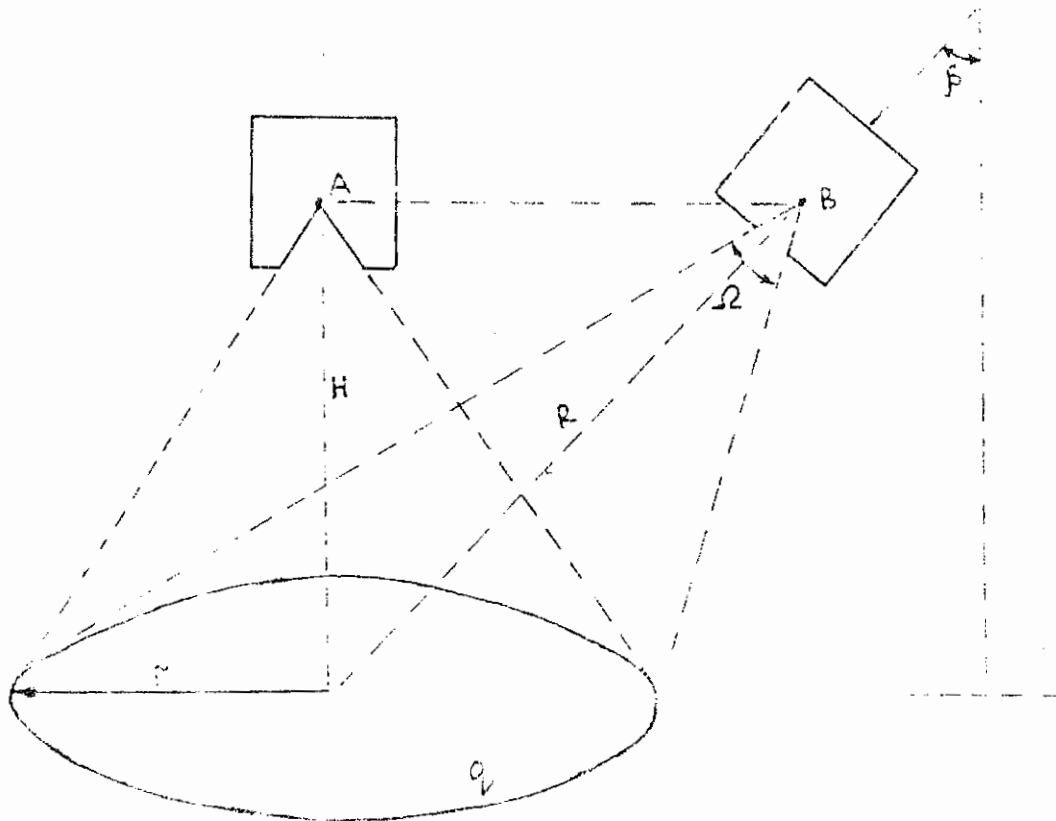


Fig.6 Measurement scheme

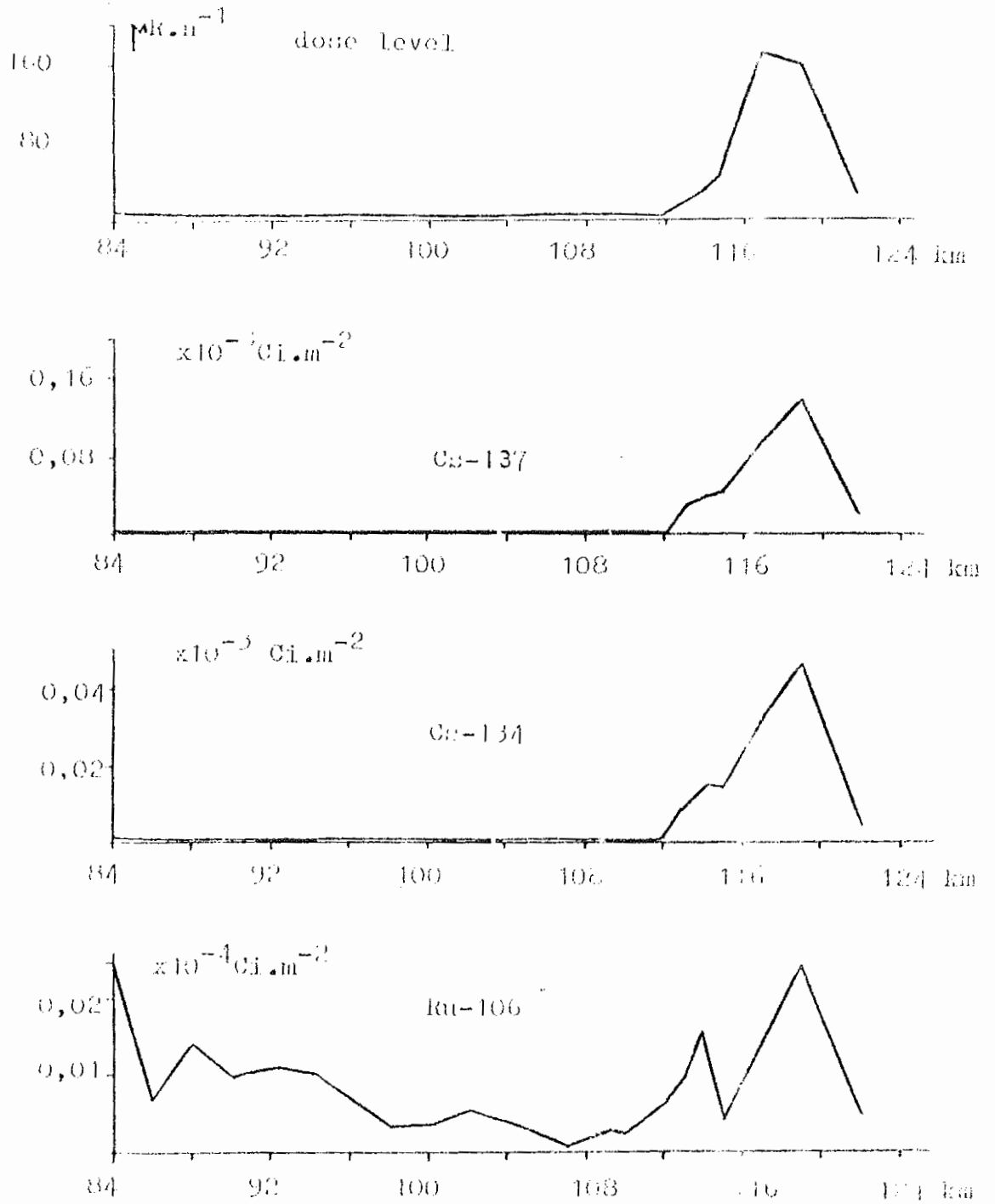


Fig. 7 Results of the continuous autoradiography on one of the routes in the 30-km zone (August, 1987)



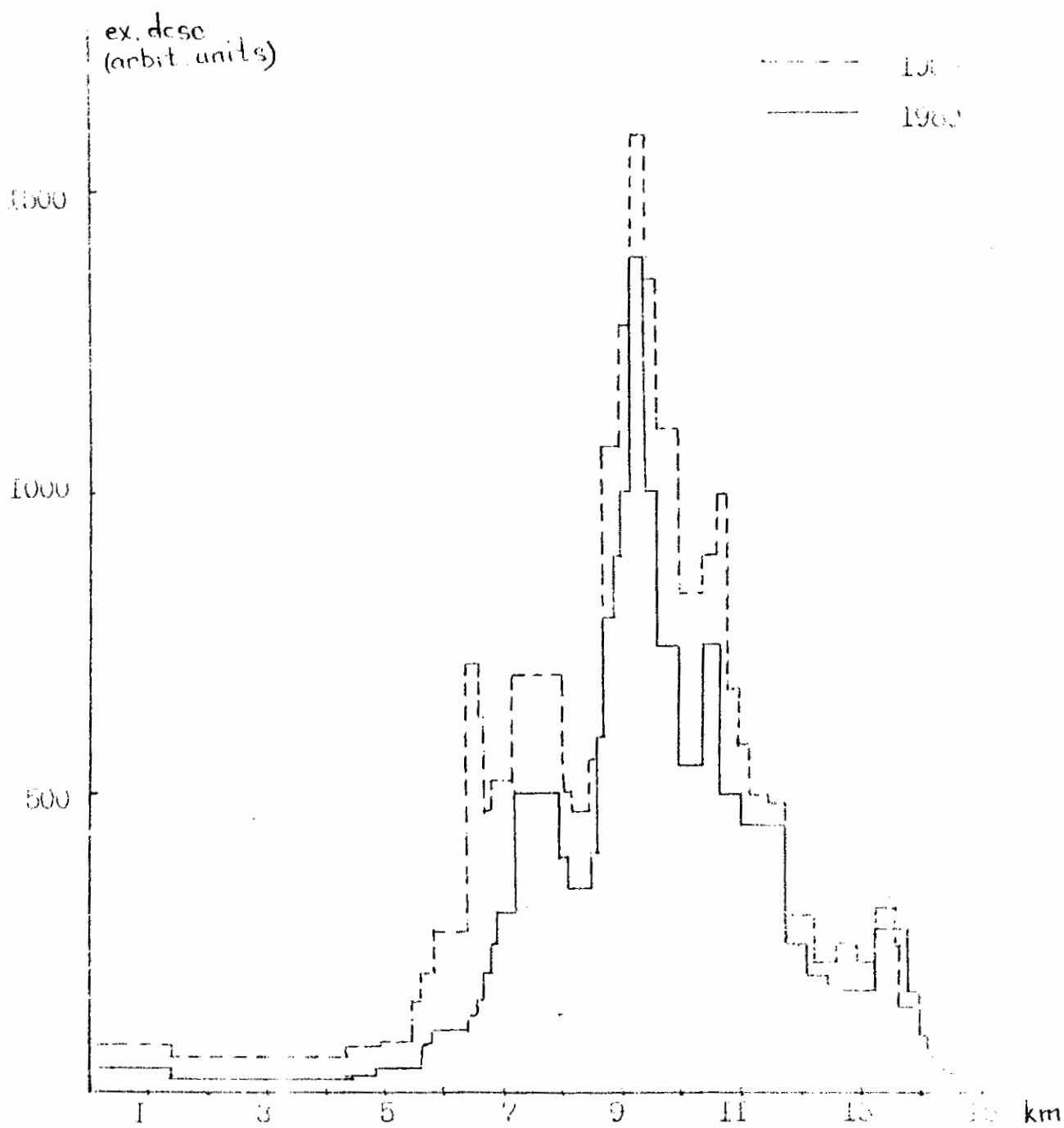


Fig.8 Results of the continuous measurements of doze rate with more details

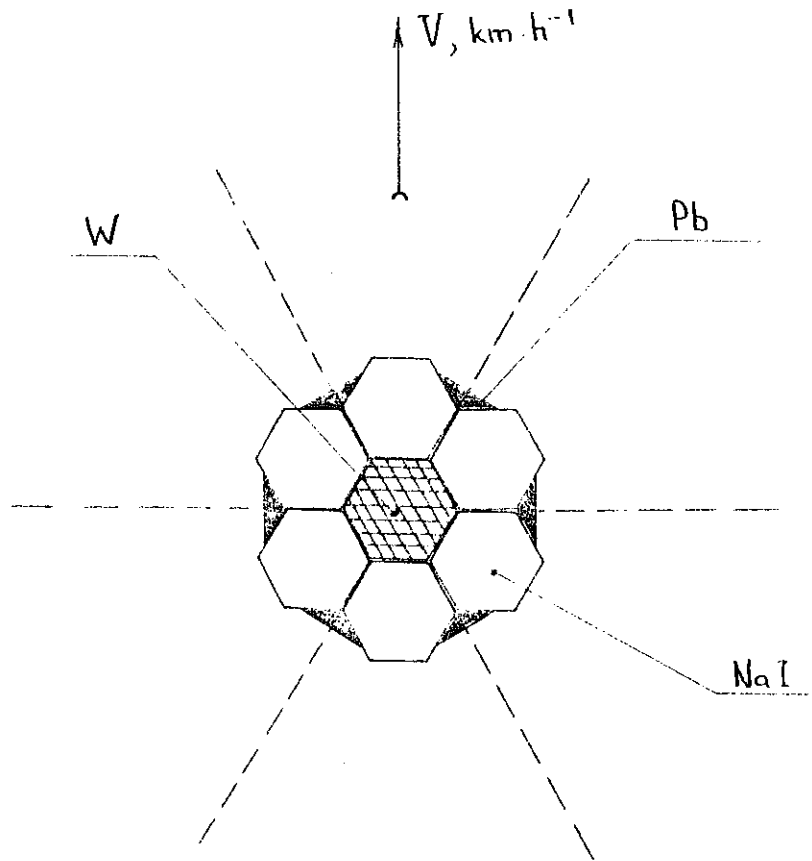


Fig.9 Scheme of the sensitive part of gamma-locator

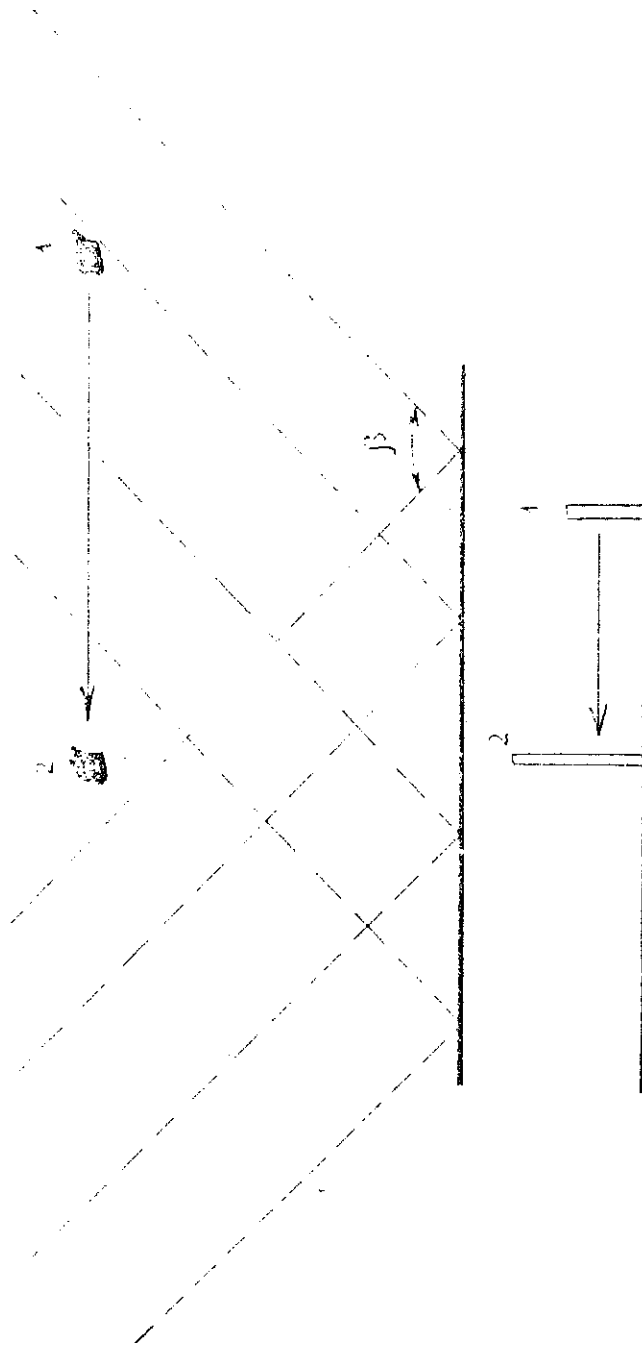
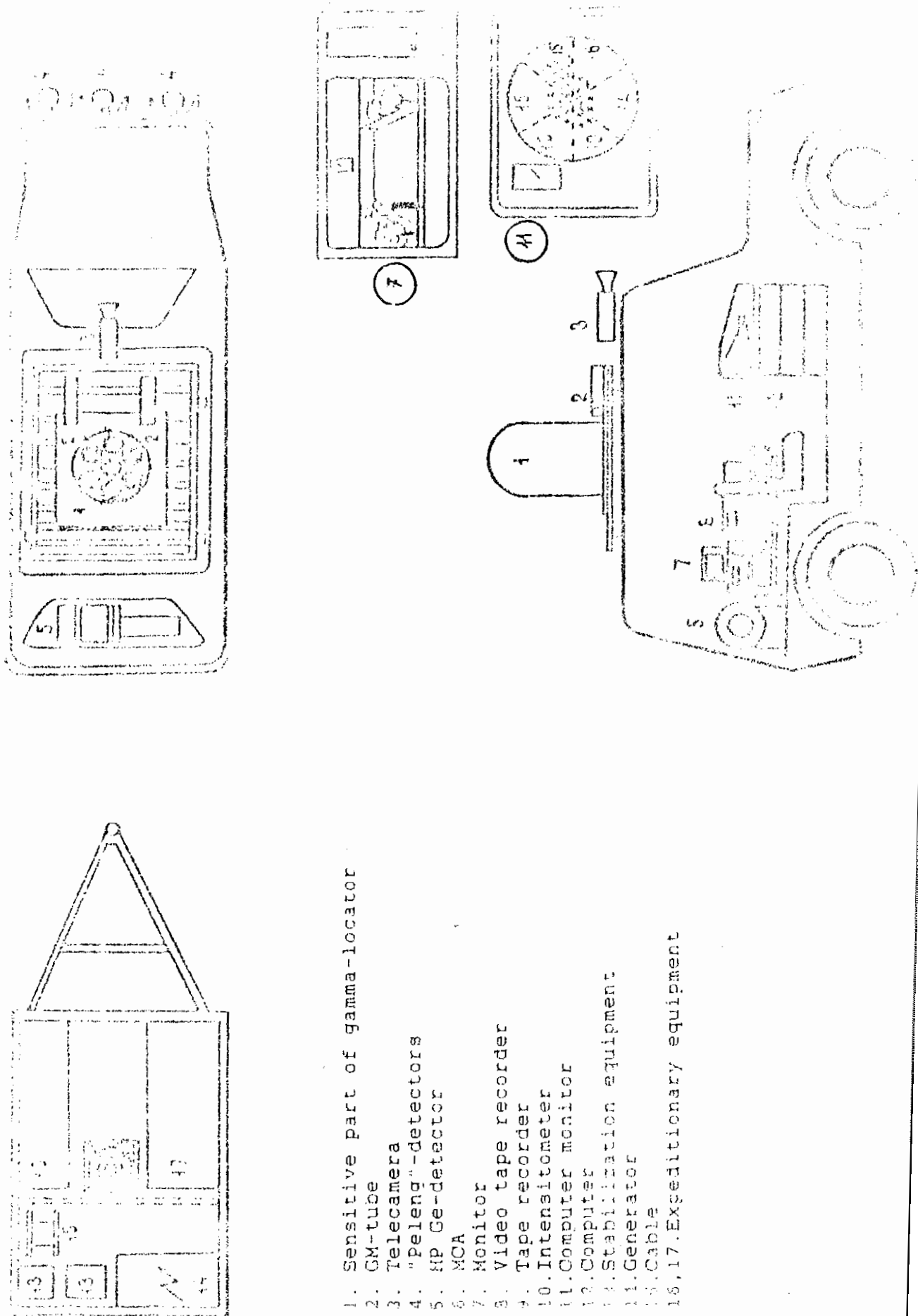


Fig.9a: Scheme of the videoimage of locality and results of data processing under programm of "Peleng"

Fig.10 Scheme of the equipment



1. Sensitive part of gamma-locator
2. GM-tube
3. Telecamera
4. "Peleng"-detectors
5. HP Ge-detector
6. MCA
7. Monitor
8. Video tape recorder
9. Tape recorder
10. Intensitometer
11. Computer monitor
12. Computer
13. Stabilization equipment
14. Generator
15. Cable
- 16, 17. Expeditionary equipment

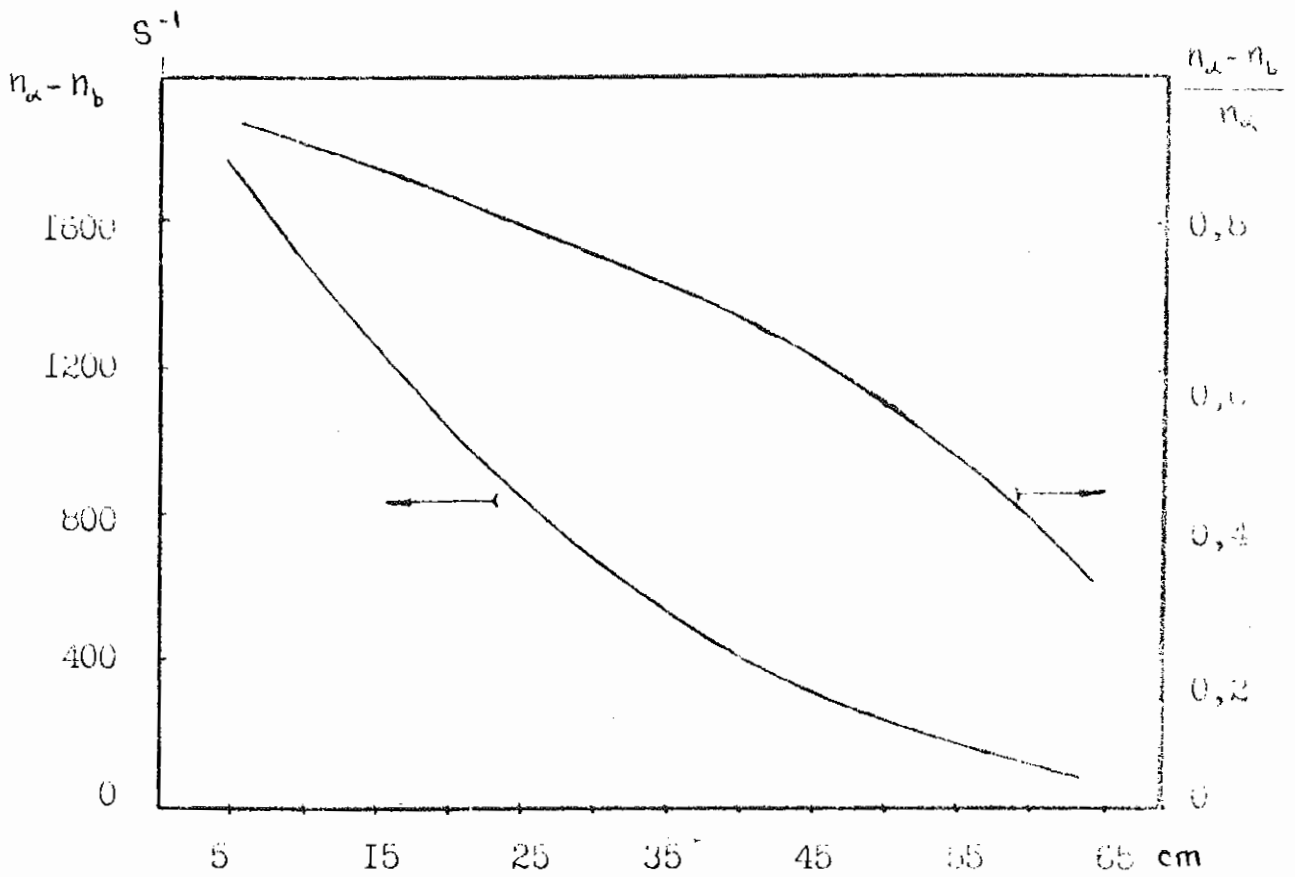


Fig.11: Dependence of aeroions counting rate and detection effectiveness of aeroions from  $\alpha$ -particles of plutonium-239 on the distance to the source.

**Comparative Analysis of the  
Effectiveness of Measures to  
Protect the Public from  
Radiation Following the  
Kyshtym Accident**

**G.N. ROMANOV, L.A. BULDAKOV, V.L. SHVEDOV**

## ABSTRACT

The formation of the Eastern Urals radioactive trail meant it was necessary to take a series of urgent and planned (or longer-term) measures to protect the public from radiation. The urgent protective measures included evacuation in the first 7-10 days of 1 100 people from the four villages nearest to the plant, since their external irradiation dose might have exceeded 100 rem in the first month. This evacuation, which later proved to be resettlement, prevented 91% of the potential 30-year dose. Parallel to this, the population was subjected to a number of hygiene measures, and dosimetric monitoring was introduced. Given that  $^{90}\text{Sr}$  played the leading radiological role, a  $^{90}\text{Sr}$  contamination limit of  $2 \text{ Ci/km}^2$  was introduced for the area, and higher levels required implementation of longer-term measures to protect the populace from radiation. Such longer-term measures included monitoring radioactive contamination of produce and agricultural products (where contamination was below  $2 \text{ Ci } ^{90}\text{Sr}$  per  $\text{km}^2$ ), their rejection and replacement by non-contaminated produce, restricting public access, banning economic activity, and decontamination of rural populated areas/arable land. Rejection and replacement of produce came late, were difficult to implement and had little effect on reducing  $^{90}\text{Sr}$  intake via food. For this reason, the longer-term measures included subsequent, additional resettlement of 9 600 people from a further 19 villages some 250-670 days after the accident. This measure prevented 20-38% of the potential 30-year dose, and from a modern radiological point of view was not wholly justified. Protection of the non-evacuated population exposed to contamination below  $2 \text{ Ci } ^{90}\text{Sr}$  per  $\text{km}^2$  mainly involved reducing the  $^{90}\text{Sr}$  concentration in the milk of cows belonging to the rural populace, this being achieved by banning the use of natural pastures and hay meadows. This measure reduced by a factor of 2 the  $^{90}\text{Sr}$  intake into humans. Agricultural production was restored in areas with contamination levels ranging from 2 to  $100 \text{ Ci } ^{90}\text{Sr}$  per  $\text{km}^2$ ; this involved setting up state farms specialising in meat production (beef, pork), which turned out produce containing 2.5-5 times less  $^{90}\text{Sr}$  than in surrounding farms.

The deposition in the Eastern Urals of a trail of radioactive fallout made it necessary to implement a series of measures to protect the public from radiation, which - according to their aims and periods of execution - can be divided into a) urgent measures, and b) planned or long-term measures. The urgent measures were taken to prevent large doses of external whole-body irradiation and of internal irradiation of the gastro-intestinal tract of the inhabitants in the populated rural areas closest to the nuclear installation. As for the public living further afield and in areas of low-level radioactive contamination, the planned protection measures were intended primarily to reduce the intake of  $^{90}\text{Sr}$  into the human body via locally produced food, and to prevent late stochastic (oncological) effects. Many of the long-term measures were combined with the restoration of agricultural production as a means of rehabilitating the contaminated territory.

The effectiveness of these steps - measured either in terms of a reduction in irradiation dose to the public or of a decrease in  $^{90}\text{Sr}$  concentration in the produce obtained - varied, and we can now select the most efficacious measures and reject the other, less effective ones.

Of the urgent public radiation protection measures, the most effective in lowering the public irradiation dose was evacuation during the first 7-10 days (Table 1). This prevented 87% of the potential 30-year dose of external irradiation and 91% of the effective dose equivalent. As the evacuation periods increased, the effectiveness of this measure, which was now planned evacuation, was reduced, and - from a modern radiological point of view - evacuation carried out 330-670 days after an accident is a crude, superfluous measure which does not warrant the socio-economic and psychological drawbacks involved.

Table 1

Irradiation doses prevented by evacuation,  
% of potential 30-year dose

Irradiation pathway	Evacuation times			
	urgent evacuation 7-10 days, 1 100 pers.	250 days 2 250 pers.	330 days 4 200 pers.	670 days 3 100 pers.
External irradiation	87	18	17	17
Irradiation of gastro-intestinal tract	84	23	23	9
Irradiation of bone and red bone marrow	99	88	49	40
Effective dose equivalent	91	38	28	20



Other urgent measures (Table 2) were directed at reducing external irradiation of the evacuated population through hygiene measures, and also at preventing further irradiation of non-evacuated members of the public in the event of their uncontrolled presence in highly contaminated areas or their use of produce and goods in populated areas abandoned after evacuation. The effectiveness of limiting public access to contaminated areas and banning the use of produce and goods does not lend itself to analysis, since these measures are of a preventive nature.

Table 2

Additional urgent measures to protect the public from radiation

Measures	Purpose of introducing measure	Time of introduction and scope of measure
1. Hygiene measures involving evacuated population	To remove radioactive contamination from surface of body and clothing	1957; 1 100 pers.
2. Ban on bringing out property and produce from the evacuated populated areas	To prevent further spread of radioactive substances and use of contaminated produce	1957; 4 populated areas
3. Limiting access to contaminated area	To prevent further irradiation of non-evacuated population	1957; approx. 200 km <sub>2</sub>

The planned or long-term measures, which began to be introduced in 1958, covered both the contaminated areas of the Eastern Urals in which the population continued to live, as well as the territory from which the population had been rapidly evacuated. In the zone where people stayed on (where, in 1958, maximum contamination was 100 Ci/km<sub>2</sub> <sup>90</sup>Sr and the minimum 1 Ci/km<sub>2</sub> <sup>90</sup>Sr), monitoring of the radioactive contamination levels in food and fodder was made compulsory (Table 3)

Table 3

Long-term measures in the area where the public stayed on

Measures	Time of introduction and scope of measures	Effectiveness of measures
Radiation monitoring of food and agricultural produce; rejection of produce	1958; 1 000 km <sub>2</sub> , 3 years; 100 000 analyses; rejection of some 10 000 t of produce	10-20% reduction in dose to public
Provision of fodder base for privately owned livestock	1961	<sup>90</sup> Sr concentration in milk reduced by a factor of 2

in most of the populated areas (covering a total area of some 1 000 km<sub>2</sub>), and accompanied by rejection and substitution of produce in cases where established permissible limits were exceeded.

The need to create eight special monitoring laboratories, provide technical equipment and train staff for them, led to substantial delay in initiating mass radiation monitoring everywhere, and in actual fact monitoring did not get under way fully until summer/autumn 1958 when the bulk of the most contaminated produce from the 1957 harvest had been consumed by the population or farm animals. Neither was it possible in practice to monitor all produce or replace all the rejected produce. This measure proved insufficiently effective and may be assessed as having reduced the dose to the public by 10-20%. The low efficacy of this measure led to further evacuation of the population, even as late as 670 days after the accident.

Whether the non-evacuated population was able to stay on for a prolonged period depended on the dose accumulated in bones and red bone marrow as a result of ingesting <sup>90</sup>Sr in food. Intake of <sup>90</sup>Sr with cows' milk plays a leading role in formation of the <sup>90</sup>Sr dose. Some 70-80% of the daily <sup>90</sup>Sr intake via food comes from milk. This predetermined one of the measures, namely the need to change the composition of the fodder fed to privately owned cattle by changing the fodder base. This involved excluding to the maximum the use of natural pastures and hay meadows, the produce of which contains 5-10 times more <sup>90</sup>Sr than that from cultivated land. Changing the composition of the fodder base in a number of populated areas reduced <sup>90</sup>Sr concentration in the milk of privately owned cows by a factor of 2.

A health protection zone, in which both residence and agricultural exploitation were forbidden, was set up in the part of the contaminated area from which the population had been evacuated by 1959 (Table 4). After the decision was taken to re-establish agricultural production on 80% of the previously abandoned territory, the health protection zone was reduced in area from 700 km<sup>2</sup> to 200 km<sup>2</sup>. Besides the creation of the health protection zone, the abandoned farmlands were ploughed as usual over a two-year period. This reduced the concentration of radioactive substances in the air above such land by several dozen percent.

Table 4

Long-term measures in the evacuation zone

Measures	Time of introduction and scope of measures	Effectiveness of measures
Creation of health protection zone (2 Ci <sup>90</sup> Sr/km <sub>2</sub> )	1958 - 700 km <sub>2</sub> 1962 - 200 km <sub>2</sub>	Prevention of further irradiation of non-evacuated population
Decontamination of agricultural areas	1958-1959; 20 000 ha arable land	Decrease in resuspension of radioactivity
Organization of specialized state farms and forestry undertakings	1961; 9 state farms, 3 forestry undertakings	Concentration of <sup>90</sup> Sr in meat and dairy produce from specialized state farms 2.5-5 times lower than that in produce from other state farms and private holdings

As shown above, the planned radiation protection measures in the evacuation zone were combined with agricultural rehabilitation of this territory. The main task, namely to get agricultural production going again and obtain produce with a <sup>90</sup>Sr concentration not exceeding established permissible levels, was achieved, and depended both on local levels of radioactive contamination and on the specific characteristics of <sup>90</sup>Sr transfer into the various kinds of agricultural produce. The lowest <sup>90</sup>Sr concentration is typically found in poultry, pork and beef, while the greatest is found in natural grasses, straw, silage crops and root crops. The following contamination levels, in curies of strontium-90 per square kilometre, were taken as permissible: food grains 2, beef 20, pork 100. Nine specialized state farms were set up in 1961 geared to meat production, and they are currently exploiting 85% of the land previously taken out of use, where the contamination is 2-100 Ci/km<sub>2</sub> of <sup>90</sup>Sr. This land makes up 10-15% of the area exploited by state farms. Having the state farms concentrate on meat production, while avoiding use of natural land as far as possible and adopting new methods, has resulted in produce 2-5 times "cleaner" than that from "unregulated" farming.

Several methods of reducing <sup>90</sup>Sr accumulation in produce were tried out. The most effective involved optimized land use based on classifying and using areas in line with their radioactive contamination levels, on the basis of the correlations between the established permissible levels of <sup>90</sup>Sr concentration in specific kinds of produce and the observed actual <sup>90</sup>Sr concentrations in such produce measured in terms of a common contamination unit (Table 5). Through skillfully allotting food and fodder crops to areas with varying contamination levels it is possible to reduce <sup>90</sup>Sr concentration in produce by almost 200 times as compared with unregulated allocation. Of the other measures, decontaminating soil by removing the contaminated top layer is also effective, reducing <sup>90</sup>Sr accumulation in produce 5 to 15-fold, but due to the high cost of this method it is best restricted to small areas, e.g. those under

Table 5

Influence of agrotechnical measures on <sup>90</sup>Sr concentration in agricultural produce

Method	Reduction of <sup>90</sup> Sr concentration as compared with customary practice
Optimization of land use (classifying farmlands according to their radioactive contamination level, the permissible <sup>90</sup> Sr concentration in a given type of produce, and the observed coefficient of <sup>90</sup> Sr transfer into produce)	max. 200-fold
Decontamination of farmlands by removing the top (5-10 cm) layer	5 to 15-fold
Deep ploughing and burying the top layer below the tillable horizon	2 to 7-fold
Physico-chemical improvement of soils by means of immobilizing substances	max. 2 to 4-fold
Increasing crop yields	max. 1% for each percentage point increase in yield
Selection of crop varieties	max. 4-fold
Removal of surface contamination from biomass during cultivation and harvesting	$n \times 1^0 - n \times 10^1\%$

vegetables. Deep ploughing and burying the contaminated top layer at a depth of more than 50 cm reduces  $^{90}\text{Sr}$  transfer into produce several-fold. All the other methods are less effective; for example, physico-chemical improvement of soils by adding substances which immobilize  $^{90}\text{Sr}$  uptake into the roots - in particular lime, sulphates, phosphates and silicates - results in an approximately two-fold reduction in  $^{90}\text{Sr}$  accumulation in produce. Agrotechnical methods, most of which reduce  $^{90}\text{Sr}$  uptake 2 to 4-fold, are suitable where actual produce contamination levels are close to the permissible levels, but will have no substantial impact in cases where the permissible levels are significantly exceeded.

In livestock farming (Table 6) the most sensible measure is to alter the composition of the fodder ration, which should be optimized both from the angle of its nutritional value and of including feed with a very low  $^{90}\text{Sr}$  concentration. In this connection the ration must not contain any fodder from natural land, since this is the most contaminated. In practice this measure reduces  $^{90}\text{Sr}$  transfer to milk and meat by several dozen percent. Another recommended measure is to maintain meat cattle on clean fodder for several months before slaughter. This reduces the  $^{90}\text{Sr}$  concentration in produce by a factor of 2 or more.

Table 6

Influence of zootechnical measures on  $^{90}\text{Sr}$  concentration in livestock produce

Method	Reduction of $^{90}\text{Sr}$ concentration as compared with customary practice
Selecting optimum composition of fodder ration	$n \times 10^1\%$
Increasing the yield of dairy cattle	$n \times 10^0 - n \times 10^1\%$
Keeping fattening livestock on clean fodder before slaughter	$n \times 10^1\%$

In conclusion, it can be said that, of the above-mentioned measures and methods to reduce irradiation of the public in the event of radiation accidents (the latter phase of which may be marked by what is known as the "strontium hazard"), only a few have a really marked effect. The most radical measure is evacuation of the population, provided this is done in the very early phase of the accident. As for ensuring that people can continue to live safely in areas contaminated with  $^{90}\text{Sr}$ , there are hardly any measures with an effectiveness factor of more than 2-4 which could be applied to private holdings. In state and other large farms optimization of land use and decontamination of farmland can be effective measures.

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# **Ways of Reducing the Build-up of Sr-90 in Crops in the Kyshtym Accident Zone**

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## ABSTRACT

In the zone of the Kyshtym radiation accident, with its leached clayey loam chernozem soil (pH 5.6, exchangeable  $\text{Ca}^{++}$  31.6 mg-equiv./100 g), the lowest  $^{90}\text{Sr}$  concentration - expressed in terms of contamination level (in units of Bq/kg per  $\text{Bq/m}^2$ ) - was observed in the grain of millet, maize and wheat and in potato tubers, varying for this group of crops from 0.051 to  $0.234 \times 10^{-3} \text{ m}^2/\text{kg}$ . The largest build-up was found in oil-bearing crops (flax, mustard) and root crops (carrots, beet, swede, turnip) -  $(0.73-1.37) \times 10^{-3} \text{ m}^2/\text{kg}$  and  $(1.23-4.15) \times 10^{-3} \text{ m}^2/\text{kg}$  respectively. Leguminous crops (peas, beans, French beans) occupied the middle section of this scale -  $(0.165-0.35) \times 10^{-3} \text{ m}^2/\text{kg}$ . Values for individual crops varied by a factor of 2-4 in different years in line with seasonal agro-meteorological conditions.

A 37-40% reduction in  $^{90}\text{Sr}$  build-up in the grain and stalks of vetch and oats was observed after deep ploughing to a depth of 50 cm. Transfer to the grain and vegetative organs fell by a factor of 2-4 in comparison with the control when mineral fertilizers (NPK) were added - in doses of  $9 \text{ g/m}^2$  of each element - to the upper soil layer after deep ploughing. The addition of  $\text{Na}_3\text{PO}_4$  to the surface of soil containing  $^{90}\text{Sr}$  in quantities equivalent to 100, 200 and 300% of exchangeable  $\text{Ca}^{++}$  content, and then ploughing this layer under to a depth of 50 cm, reduced transfer to wheat grain by a factor of 2.5-12. The addition of  $\text{Na}_2\text{SiO}_3$  in corresponding quantities proved less effective. Adding colophony or furfural-analine to this soil layer at the rate of 2% of layer mass, followed by ploughing to a depth of 50 cm, reduced the transfer to wheat grain and peas by a factor of 2-5, and by a factor of 4-8 when higher doses of colophony were applied. Adding colophony to the soil led to a decrease in  $^{90}\text{Sr}$  build-up in the following year's crops as well.

The Kyshtym accident in 1957 radioactively contaminated a considerable area, including forest tracts and cultivated land. There was an urgent need to study the behavioural processes of radionuclides in various parts of the biosphere, including the system of soils and plants, and to draw up practical measures for limiting uptake into biological and food chains. Given that a considerable part of the deposited radioactive mixture comprised  $^{90}\text{Sr}$ , the researchers focused their attention on this biologically dangerous radionuclide.

There was also great scientific and practical interest in determining the  $^{90}\text{Sr}$  accumulation levels in various species and varieties of plants in order to show which agricultural crops had the lowest accumulation levels when harvested.

Given the above, and because of the acuteness and importance of the problem at the time, our research had to cover the following points: 1) the uptake of  $^{90}\text{Sr}$  into crops and the accumulation level in the harvest after tilling (in various ways) and applying mineral fertilizers in field experiments; 2) the effect of applying sodium silicate, sodium phosphate, a furfural-aniline mixture and colophony to the soil on  $^{90}\text{Sr}$  accumulation in harvested crops in small-scale field experiments; 3) the differences in  $^{90}\text{Sr}$  accumulation in various species and varieties of plants harvested.

Large and small-scale field experiments were carried out between 1959 and 1966 on leached clayey loam chernozem soil (pH 5.6; exchangeable  $\text{Ca}^{++}$  31.6 mg-equiv./100 g). The  $^{90}\text{Sr}$  contamination density of areas used for field experiments varied between 0.63 and 2.4 MBq/m<sup>2</sup>, the maximum value for the small-scale field experiments being 150 MBq/m<sup>2</sup>. A wide range of agricultural plants was studied in the research project. NPK fertilizers were applied to the upper soil layer at the rate of 90 kg/ha of each element.

Experiments involving ploughing the soil at various depths and applying NPK mineral fertilizers to the upper soil layer led to a considerable reduction in  $^{90}\text{Sr}$  accumulation only in the grain and straw of vetch and in the grain of oats (Table 1). NPK had a less marked effect on wheat (17-40%) and virtually no effect on barley and maize.

When the soil contaminated with  $^{90}\text{Sr}$  was ploughed to varying depths and no fertilizers were applied, the uptake of  $^{90}\text{Sr}$  from the soil into crops varied within a relatively narrow range. In comparison with 'control' ploughing to a depth of 20 cm, deep ploughing (50 cm) reduced the concentration of the radionuclide only in the grain and straw of vetch and the straw of oats (37-40%). Some variants showed a trend towards an increase in the  $^{90}\text{Sr}$  content per unit mass of matter.

However, applying mineral fertilizers to leached chernozem soil produced substantial harvests of cereals and other crops without an increase in the  $^{90}\text{Sr}$  concentration, which is an extremely important practical consideration when growing crops on land contaminated by  $^{90}\text{Sr}$ .

In addition to tilling the soil in various ways and applying mineral fertilizers, various chemical substances (sodium phosphate and silicate, a furfural-aniline mixture and colophony) were used to improve the soils contaminated with  $^{90}\text{Sr}$ . The research was carried out in small-scale field experiments (using bottomless boxes measuring 50 x 50 x 50 cm placed in the soil).

It was established that application of tribasic sodium phosphate to leached chernozem soil in quantities equivalent to 100, 200 and 300% of the content of exchangeable calcium, combined with ploughing-under of the treated soil layer to a depth of 50 cm, considerably reduced the level of  $^{90}\text{Sr}$  in the wheat crop (Table 2). An increase in the phosphate dose resulted in a greater reduction. Application of sodium silicate in the same quantities was less effective than sodium phosphate (Table 3).

Overall, we concluded that addition of the above-mentioned reagents is reasonably effective in reducing the transfer of  $^{90}\text{Sr}$  from soil to plants without a significant reduction in the harvest or any deterioration in the chemical properties of the soil.

The idea behind application of the furfural-aniline mixture and colophony to the leached chernozem soil was to create a closely bound soil layer in which the  $^{90}\text{Sr}$  was in a form which plants could not assimilate easily. Moreover, the fact that these reagents have hydrophobic properties and are toxic for plant root systems may, in addition, limit the spread of roots in a soil layer treated with this reagent, and consequently reduce  $^{90}\text{Sr}$  uptake into plants.

The results of the studies (Table 4) showed that treating the soil layer contaminated by  $^{90}\text{Sr}$  (5.5 kg) with the furfural-aniline mixture (in the ratio of 1:2) or with colophony (at the rate of 2% of soil mass), followed by ploughing-under to a depth of 0.5 m, considerably reduced (by 50-60%)  $^{90}\text{Sr}$  accumulation in the wheat and pea crops as against the control. Application of colophony proved to be more effective than the furfural-aniline mixture in reducing the  $^{90}\text{Sr}$  concentration in the crop. Adding colophony in quantities corresponding to 2% of soil mass reduced the  $^{90}\text{Sr}$  concentration in wheat and pea crops by 2-5 times, and by 4-8 times when higher doses were used. Treating the soil with colophony significantly reduced  $^{90}\text{Sr}$  content in plants in the second year as well.

The results show that treating the soil with chemical compounds able to fix  $^{90}\text{Sr}$  in poorly accessible forms is generally a reasonably effective way of reducing  $^{90}\text{Sr}$  uptake into crops. Although it is not possible to recommend the use of furfural, aniline and colophony in agricultural production because of their scarcity and relatively high cost, the results show that appropriate substitutes with similar properties should be sought. At the same time, when evaluating similar reagents, it is above all essential to take into account the effects of the additives on the crops, how long they remain effective in reducing  $^{90}\text{Sr}$  uptake into plants and the outlay needed for soil improvement measures of this nature.

An important factor determining  $^{90}\text{Sr}$  uptake into agricultural and other plants is their biological characteristics (the structure of their root system, how deep their roots spread in the soil profile, and their absorption of macroelements and microelements, in particular of calcium, which is the chemical analogue of strontium). For scientific

and practical reasons, it was therefore extremely important to study  $^{90}\text{Sr}$  uptake into various taxonomic groups of plants, which would indicate which crops had the lowest  $^{90}\text{Sr}$  accumulation levels. To this end, multiannual field experiments were set up (from 1959 to 1966) on leached chernozem soil with a wide variety of agricultural plants. The contamination density of the area was  $2.4 \text{ MBq/m}^2$  of  $^{90}\text{Sr}$ .

Of all the plants studied (Table 5), the lowest  $^{90}\text{Sr}$  concentration in the marketable part of the crop (grain, roots, tubers) was observed in the grain of cereals (millet, maize and wheat), sunflowers and potato tubers. The highest concentration was found in oil-bearing crops (oil-bearing flax, false flax and mustard) and in root crops. The  $^{90}\text{Sr}$  concentration in leguminous crops lies between the values for these two groups. The  $^{90}\text{Sr}$  content per unit mass of matter was much higher in vegetative organs than in grain.

The results lead us to the conclusion that ploughing to varying depths (between 20 and 40 cm) with standard equipment was ineffective in lowering the  $^{90}\text{Sr}$  contamination of agricultural crops. Applying mineral fertilizers (NPK) - at the rate of 90 kg/ha of each element - to the upper soil layer, together with ploughing at various depths, especially deep ploughing, reduced the concentration of  $^{90}\text{Sr}$  in the grain and vegetative organs of agricultural crops by 2 to 4 times.

Chemical substances used to improve soils contaminated with  $^{90}\text{Sr}$  varied in their effect on  $^{90}\text{Sr}$  uptake into various agricultural crops.

Applying sodium phosphate and colophony to soil contaminated by  $^{90}\text{Sr}$ , with subsequent ploughing-under of the contaminated layer to a depth of 50 cm, was more effective in reducing the concentration of  $^{90}\text{Sr}$  in the crop than adding furfural-analine and sodium silicate.

Study of a broad selection of crops revealed considerable differences between varieties and species as regards radionuclide accumulation in the harvest.

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Table 1

The effect of NPK and the depth of ploughing on <sup>90</sup>Sr accumulation in the harvest of vetch and oats

Depth of ploughing, cm	Experiment variant	Oats			Vetch		
		grain	straw	grain	straw	grain	straw
		<sup>90</sup> Sr, % as against "control" ploughing	<sup>90</sup> Sr, % as against "control" ploughing	<sup>90</sup> Sr, % as against "control" ploughing	<sup>90</sup> Sr, % as against "control" ploughing	<sup>90</sup> Sr, % as against "control" ploughing	<sup>90</sup> Sr, % as against "control" ploughing
20*	Control NPK	100 56.9	100 65.3	100 46.1	100 41.9	100	100
30	Without fertilizers NPK	129 89.1	77.2 70.2	125 50	114 48.1	114	114
40	Without fertilizers NPK	197 51.6	71.2 60.3	115 59.6	106 62.9	106	106
50 (Deep Ploughing)	Without fertilizers NPK	83.8 24.7	60.3 52.4	63.4 38.4	61.7 39.5	61.7	61.7

\* The control involved ploughing to a depth of 20 cm with no application of fertilizers

Table 2

Effect and after-effect of tribasic sodium phosphate on  $^{90}\text{Sr}$  accumulation in the wheat crop

Variant	Part of the plant	First year		Second year	
		$^{90}\text{Sr}$ , % as against the control		$^{90}\text{Sr}$ , % as against the control	
Soil + $^{90}\text{Sr}$ (control)	Grain	100	100	100	100
	Straw	100	100	100	100
Soil + $^{90}\text{Sr}$ + $\text{Na}_3\text{PO}_4$ (100% exchangeable Ca)	Grain	44	44	126	126
	Straw	43	43	115	115
Soil + $^{90}\text{Sr}$ + $\text{Na}_3\text{PO}_4$ (200% exchangeable Ca)	Grain	<8	<8	72	72
	Straw	9	9	79	79
Soil + $^{90}\text{Sr}$ + $\text{Na}_3\text{PO}_4$ (300% exchangeable Ca)	Grain	<8	<8	44	44
	Straw	2	2	49	49

Table 3

Effect and after-effect of sodium silicate on  $^{90}\text{Sr}$  accumulation in the wheat crop

Variant	Part of the plant	First year		Second year	
		$^{90}\text{Sr}$ , % as against the control		$^{90}\text{Sr}$ , % as against the control	
Soil + $^{90}\text{Sr}$ (control)	Grain	100	100	100	100
	Straw	100	100	100	100
Soil + $^{90}\text{Sr}$ + $\text{Na}_2\text{SiO}_3$ (100% exchangeable Ca)	Grain	163	163	102	102
	Straw	159	159	93	93
Soil + $^{90}\text{Sr}$ + $\text{Na}_2\text{SiO}_3$ (200% exchangeable Ca)	Grain	35.0	35.0	90	90
	Straw	39.0	39.0	91	91
Soil + $^{90}\text{Sr}$ + $\text{Na}_2\text{SiO}_3$ (300% exchangeable Ca)	Grain	7.0	7.0	63	63
	Straw	14.0	14.0	56	56

Table 4

Effect and after-effect of applying colophony and furfural-analine on <sup>90</sup>Sr accumulation in pea and wheat crops, % as against the control

Variant	Part of the plants	Wheat				Peas	
		First year		Second year		First year	Second year
		Grain	Straw	Grain	Straw		
Control (Soil + <sup>90</sup> Sr)	Grain Straw	100 100	100 100	100 100	100 100	100 100	100 100
<sup>90</sup> Sr + furfural-analine (2% by weight of the treated soil)	Grain	51	70	48	125		
	Straw	41	100	41	129		
<sup>90</sup> Sr + colophony (2% by weight of the treated soil)	Grain	41	73	30	77		
	Straw	34	61	22	73		
<sup>90</sup> Sr + colophony (5% by weight of the treated soil)	Grain	24	49	17	60		
	Straw	20	45	13	52		



Table 5

Standardized concentrations of <sup>90</sup>Sr in the agricultural crops harvested in field experiments, 10<sup>-3</sup> m<sup>2</sup>/kg (average for 1959-1966)

Crop	<sup>90</sup> Sr	
	Grain	Straw
Lytstsens-62 spring wheat	0.174	1.72
Winer barley	0.142	1.94
Golden rain oats	0.187	2.26
Kazan millet	0.051	4.40
M-2 green bristle grass	0.171	3.23
Vyatka winter rye	0.095	1.56
Anakhoyskaya spring rye	0.110	2.01
Argayashkaya buckwheat	0.323	9.53
Voronezh-76 maize	0.108	3.23
Moscow-3 maize	0.054	3.64
Kapital pea	0.266	13.1
Nemchinovski-51 pea	0.171	8.01
Broad bean	0.354	-
Horse bean	0.155	-
Green bean	0.184	-
Oil-bearing flax	0.731	6.30
Penzenskaya mustard	1.37	3.78
False flax	0.972	5.73
Saratovski-169 sunflower	0.149	-
Fodder sunflower	0.111	-
Lorkh potato	0.234	0.101
Sugar beet	0.127	4.15
Nantskaya carrot	1.23	8.32
Osterzundomski turnip	4.15	13.2
Krasnoselskaya swede	2.43	10.9

**Measures (and their  
Effectiveness) to Improve the  
Radioecological Situation  
Given the Particular Features of  
the Contamination Caused by  
the Kyshtym and Chernobyl  
Accidents**

**N.P. ARKHIPOV, G.S. MESHALKIN, A.N. ARKHIPOV, N.I. BUROV,  
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There are more fundamental differences than similarities between the world's two major radiation accidents which occurred in the Soviet Union.

The Kyshtym accident was caused by a cooling system failure which led to the thermal explosion of a concrete tank containing high-level radioactive waste (comprising a soluble mixture of nuclear fission products low in caesium isotopes). The explosion led to the instantaneous release and subsequent dispersion and deposition of radionuclides over parts of the Chelyabinsk, Sverdlovsk and Tyumen oblasts (regions) of the RSFSR.

A classic cigar-shape trail of radioactive fallout formed; it had a very high contamination intensity gradient cross-wise but a relatively small gradient length-wise (in the wind direction), particularly far away from the centre of the explosion (Fig. 1).

In the Chernobyl accident, the reactor cooling system failure led not only to thermal explosions, but also to subsequent prolonged release of dispersed spent nuclear fuel, graphite and constructional materials. The initial release contained isotopes of highly volatile elements primarily in steam and gas phase form, while those of refractory metals were in fuel matrix composition form (uranium dioxide).

Prolonged dispersion of the radioactive clouds and jets emanating from the unit was accompanied - in line with the thermodynamics of this very complex process - by sorption and co-condensation of volatile radionuclides in the steam-gas phase onto the surfaces of solid aerosol particles, and by subsequent diffusion of these radionuclides into the particles; all this led to an extremely complex, but fairly regular, pattern of radioactive contamination forming over an enormous area in various directions from the Chernobyl NPP's stricken Unit IV (Fig. 2).

The complexity of the contamination pattern is not just due to its consisting of superimposed layers stemming from the different types of release, but also to the fact that individual sections of the contamination zone may differ substantially from one another in terms of overall fallout density, size of deposited particles, radionuclide composition, radionuclide ratios, and even of the biological availability of radionuclides (particularly strontium-90 and caesium-137) apparently changing over time.

The Chernobyl accident differs from that at Kyshtym not only because the release and contamination area involved were vastly greater, but also because the release included more or less all the radionuclides found in the spent fuel (uranium-235,238 plus the products of nuclear fission and neutron activation) and, what is more important, plutonium and other transuranic elements (Table 1).

TABLE 1  
General characteristics of the major Soviet  
radiation accidents

Characteristic	Kyshtym 29.09.57 (autumn)	Chernobyl 26.04.86 (spring)
Release of radionuclides into environment, MCi	2.1	50
Area where Sr-90 contamination over 2 Ci/km <sub>2</sub>	1 000	20 000
Radionuclide composition	Mixture of radioactive fission products low in Cs-137 (one year old)	U-235,238, radioactive products of fission and neutron activation
Soil-climate conditions	Forest-steppe, leached chernozem, evaporative mechanism	Byelorussian-Ukrainian Polesye, soddy-podzolic soils, eluvial mechanism
Time over which trail formed	11 hours	35-40 days
Main dose-forming radionuclides	Sr-90	I-131, Sr-90, Cs-137, Pu-238,240, Am-241, etc.
Form of radionuclide occurrence in release composition	Soluble nitrate-acetate salts	Combination of soluble (condensed) and insoluble (fuel matrix composition) forms
Distribution of radionuclides over area	Steady decrease length-wise along trail, high gradient cross-wise	Highly irregular throughout area
Total population in contaminated area, million persons	0.27	27, including 2.5 children
Population evacuated, thous. persons: immediately in subsequent years	10 -	116 50-70

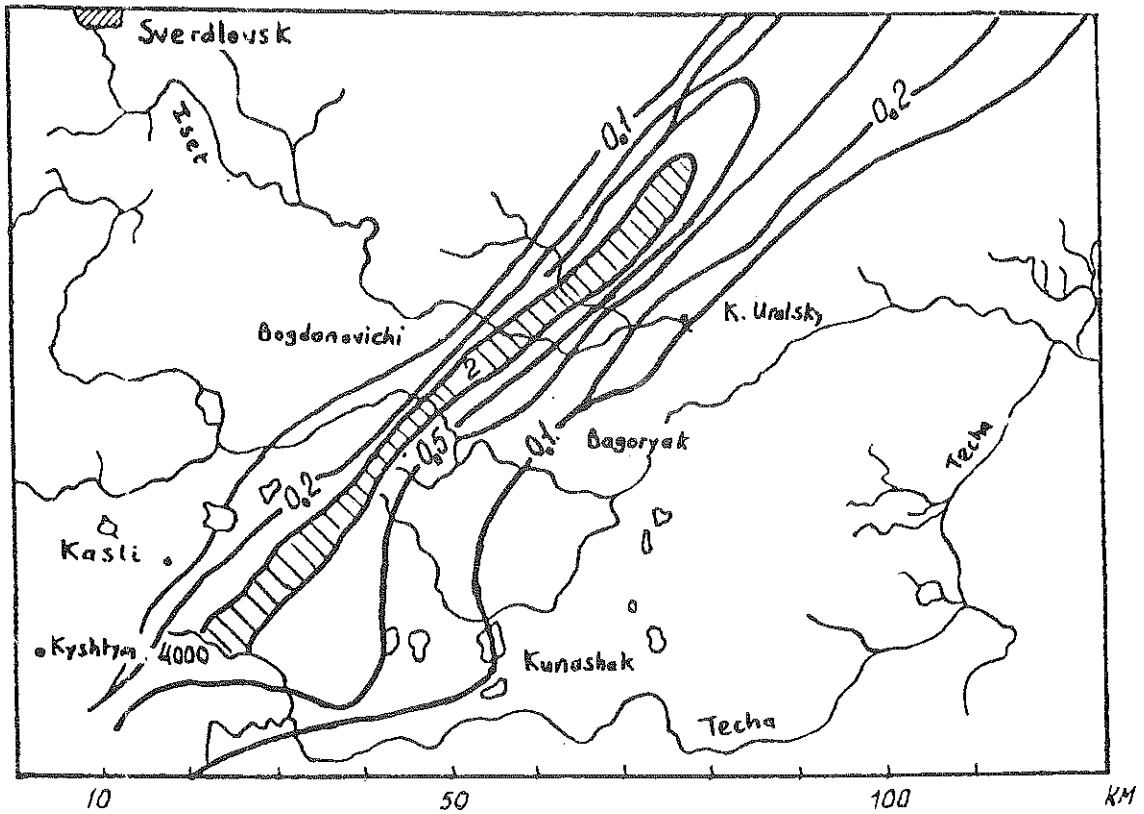


Figure 1: Sr-90 contamination (Ci/km²) at Kyshtym

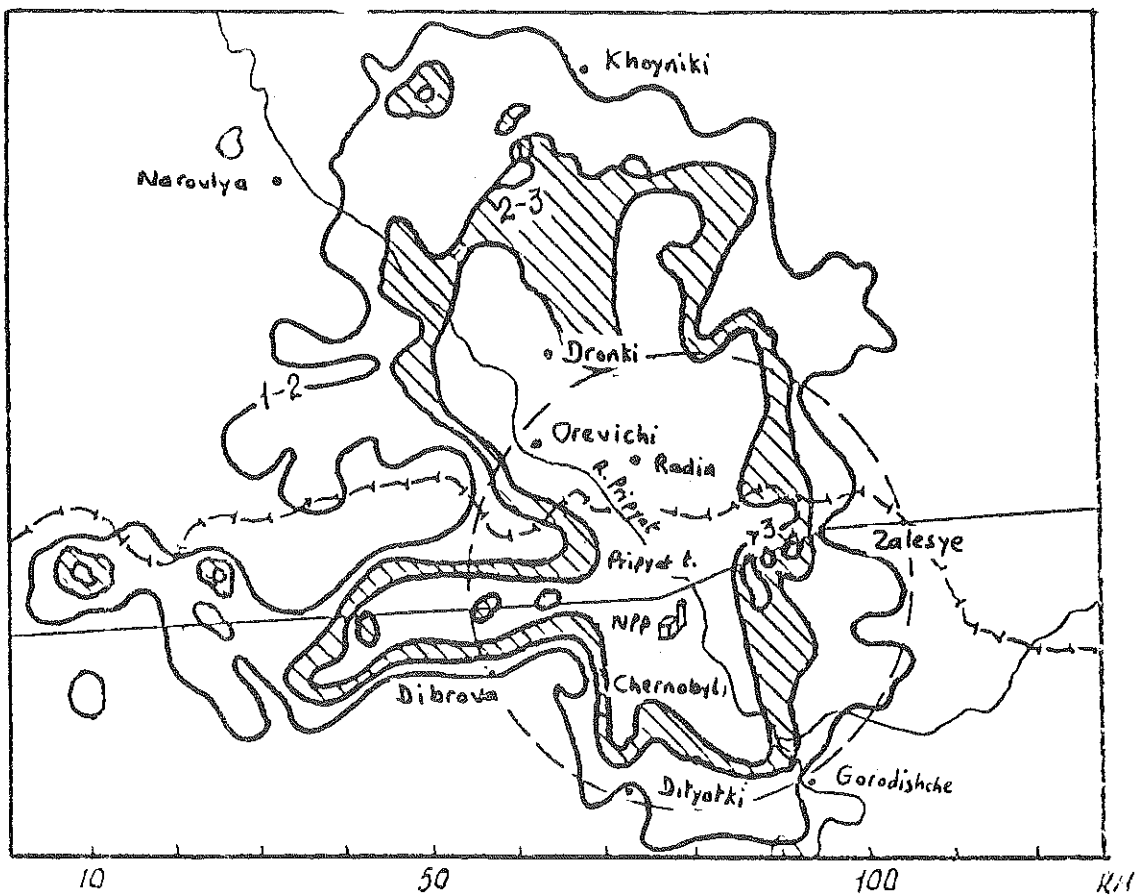


Figure 2: Sr-90 contamination (Ci/km²) at Chernobyl

The essential difference between the two accidents is not just that they occurred at different times of the year, but also that the soil-climate conditions in the two accident zones differ.

The Kyshtym accident region is in a forest-steppe zone with fertile grey forest soils and leached chernozems with an evaporative soil formation mechanism.

The Chernobyl NPP is located in the Byelorussian-Ukrainian Polesye (Pripyat Marshes), where infertile soddy-podzolic, mainly sandy, soils predominate with a low exchange capacity and an eluvial soil formation mechanism.

As far back as the early sixties, which was a period of intense global radioactive fallout, it was established that the Polesye region was the most vulnerable, radioecologically speaking, compared to other parts of the country. Suffice it to say that, even at that time, the concentrations of strontium-90 and caesium-137 in agricultural products quite often exceeded the permissible limits. Thus, the complexity of the Chernobyl accident was aggravated by the fact that it occurred in a region where the biological availability of the most dangerous dose-forming nuclides - strontium-90 and caesium-137 - is greatly enhanced.

The most effective emergency measures taken to protect the population against acute over-exposure to radiation after both the Kyshtym and Chernobyl accidents were the slaughter of privately owned livestock, and the evacuation of the inhabitants of the worst-contaminated places to uncontaminated regions where they were given housing and work. This prevented acute radiation sickness among the population.

Given the specific radionuclide features of the Kyshtym contamination, there was no official evacuation of the populace, and soon afterwards abandoned dwellings and other buildings were destroyed.

In the Chernobyl case, after the short-lived nuclides had decayed and the dose rate had fallen to safe levels, it was decided to allow some of the evacuees to go back to a number of settlements. The radiation conditions obtaining in the remaining (guarded) settlements temporarily abandoned by the inhabitants continue to prevent a decision being taken on allowing them to return, although we know of cases in which people have gone back to their homes without official permission (people of advanced age usually).

Over 1 000 such unofficial returnees are already again living just in the 30-km zone alone (in its least-contaminated sectors).

Following the Chernobyl accident there was a need - in contrast to Kyshtym - to pay serious attention to reducing the concentration of radionuclides in the air (given the presence in the contamination of a substantial quantity of plutonium isotopes to which strict norms apply). A large fleet of road-spraying vehicles and even helicopters were deployed for this purpose, thus helping to reduce dust formation on roads and in places where there were people.

However, dust suppression often involved applying a whole series of polymer-forming agents - such as sulphite-alcohol mix, latex, oil slurry, preparation MM-1, etc. - whose use was not justified at all. As a result, quite a few road accidents occurred, and use of such agents in areas of increased dust formation (quite often far away from places where there were people) - on a sandy plateau, for example - had basically no effect or was even detrimental, given that resources,

labour and materials were expended senselessly.

The truth is that the engineering and technical possibilities of rapidly improving the radiation situation over large areas are incomparably minuscule compared with natural processes (washout by precipitation, blowoff by wind, vertical migration in the soil and binding by soil and plants). The benefit of using engineering measures was therefore more psychological than real in improving the situation on agricultural land and in forests.

A similar conclusion can be drawn from the measures to decontaminate the town of Pripyat by removing soil from lawns and washing down asphalted streets and concrete-covered areas with special agents.

Fig. 3 shows the change in the gamma background on two sites in one part of the town, soil having been removed from one site, while no measures were taken at the other.

In addition, as our studies have shown, the presence of radionuclides in particles of fuel matrix composition of increased density (over 9 g/cm<sup>3</sup>) resulted in a very low value for the coefficient of redeposition (cr) even in the initial period after the accident (in June 1986 the cr for the town of Chernobyl was 2.5 x 10<sup>-8</sup>).

$$cr = \frac{\text{Concentration of radionuclide in air, Bq/m}_3}{\text{Level of soil contamination by the same radionuclide, Bq/m}_2}$$

Following this, as fallout particles penetrated deeper into the soil the cr value continued to fall rapidly, and by mid-1988 had stabilised at between  $n \times 10^{-9}$  and  $n \times 10^{-10}$  (Table 2).

TABLE 2

Coefficient of redeposition (5-10 km zone)

Before accident	1.4 x 10 <sup>-7</sup>	L. A. Buldakov et al. 1981
After accident Initial period	1.0 x 10 <sup>-4</sup>	G. N. Shinn, D. U. Wilson 1974
June 1986	2.5 x 10 <sup>-8</sup>	Experimental data
1987	3.4 x 10 <sup>-9</sup>	
1988	1.8 x 10 <sup>-10</sup>	
1989	1.1 x 10 <sup>-10</sup>	
1989	1.1 x 10 <sup>-10</sup>	Calculated data
1990	1.0 x 10 <sup>-10</sup>	

Individual values vary within a range of two orders of magnitude.

The pre-accident cr value in the Chernobyl NPP region was 1.4 x 10<sup>-7</sup> (different type of fallout). This value (or an even higher one) was used in official estimates of the importance of dust formation, and led to considerable over-estimation of the role played by the inhalation pathway in radionuclide intake by man when calculating the possible irradiation dose equivalent.

TABLE 3  
Livestock farming: various aspects involved following  
the two accidents

Kyshtym 29.09.57 (autumn)	Chernobyl 26.04.86 (spring)
1. End of grazing season	1. Start of grazing season
2. No "iodine hazard"	2. "Iodine hazard" almost everywhere
3. Decreasing biological impact (eventually to zero) with increasing distance from sources	3. Possible increase in degree of radiation damage with increasing distance from source
4. Slaughter and burial of animals in just three populated areas along the fallout trail	4. Slaughter of animals and scrapping of meat products at distances of up to 100 km from source
5. Absence of clinical forms of radiation pathology in animals in all populated areas except the three above	5. Signs of radiation damage in animals in many populated areas, with symptoms of athyroidism, hypothyroidism, leukaemia
6. Normal province in respect of microelement composition of soil	6. Endemic (depleted) province in respect of soil microelements



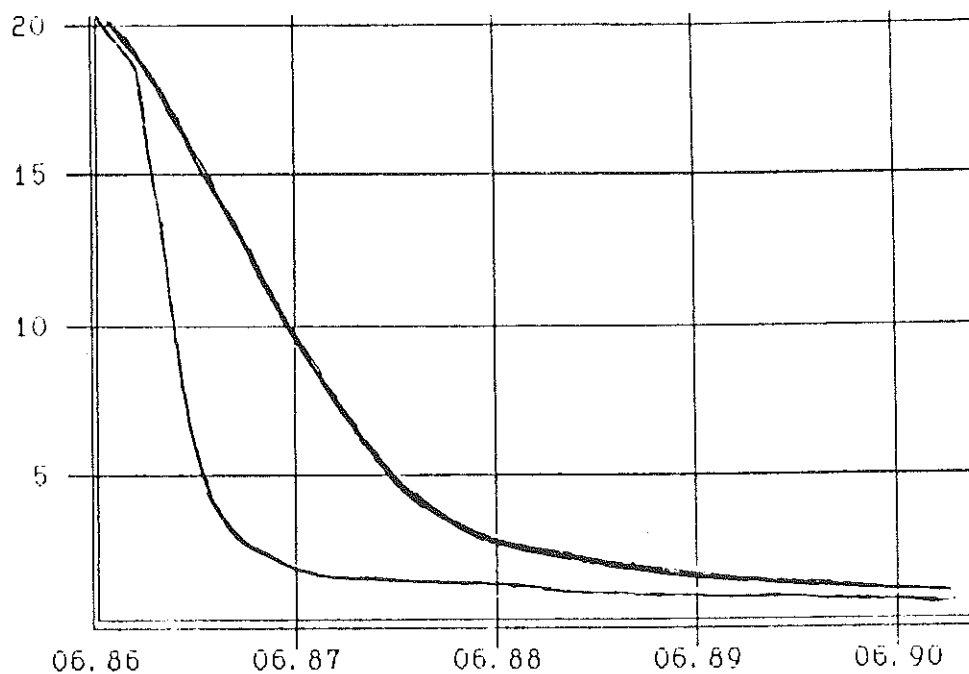


Fig. 3: Change in gamma background following decontamination, Pripjat town

The branch of farming most vulnerable to large-scale radiation accidents is livestock farming, for which Table 3 shows the various aspects involved following the Kyshtym and Chernobyl accidents.

The most effective protective measures in the livestock farming field after both the Kyshtym and Chernobyl accidents, was rapid removal of cattle from contaminated pastures and putting them into sheds or evacuating them elsewhere (if no stocks of fodder were available). Timely selection and culling (slaughter and burial) of animals with a high level of radiation damage helped reduce economic losses considerably (Table 4).

After both accidents the approach adopted was to keep animals not showing signs of radiation pathology on "relatively clean" fodder, which gives rise after several months to animal products within the provisionally permissible contamination levels.

Unfortunately, lack of fodder stocks severely limited application of this method following the Chernobyl accident. Furthermore, a considerable number of animals were slaughtered not just for radiological reasons but on organisational and economic grounds as well.

When the acute radiation hazard is over a start can be made - in the second stage of overcoming the accident consequences - on gradually restoring agricultural production as a whole on the basis of ploughing (including special types of ploughing) and recultivation of contaminated arable land.

A substantial proportion of the accident zone areas are covered by forest (Kyshtym 20%, Chernobyl 50-60%). Experience in both cases has shown that the forestry measures taken adequately ensured both effective ecological protection of forest stands and radiation-health protection of people who stayed on locally (Fig. 4).

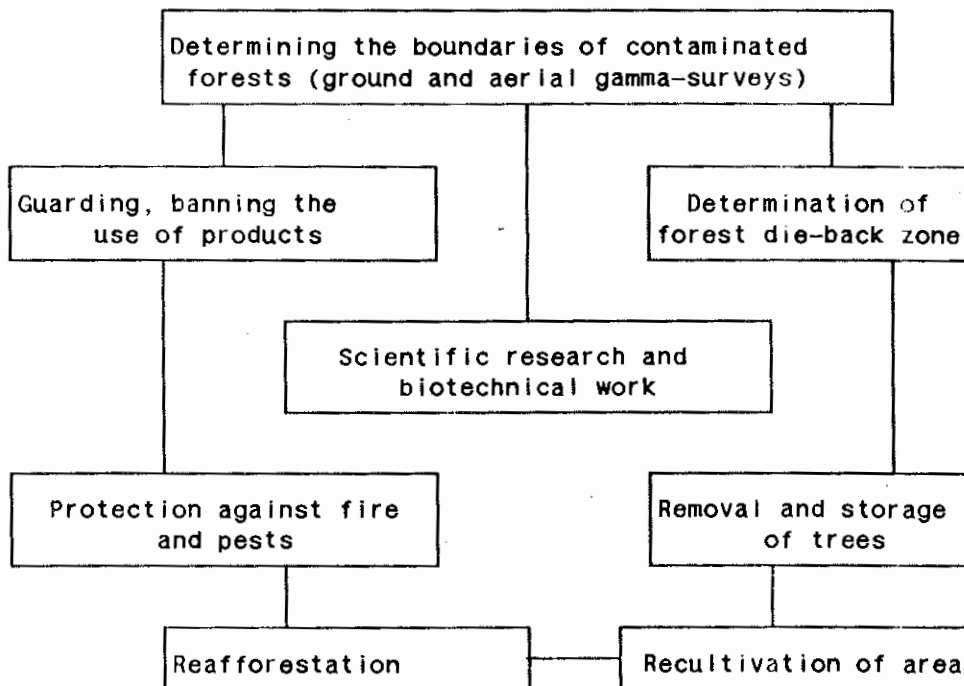


Figure 4: Measures in contaminated forests

TABLE 4  
Protective measures connected with agricultural  
production after the radiation accidents

Kyshtym 29.09.57 (autumn)	Chernobyl 26.04.86 (spring)
STAGE 1	
1. Grazing stopped, livestock kept inside	1. Grazing continued, no stocks of fodder
2. Evacuation of cattle from two populated areas only	2. Evacuation of cattle from hundreds of thousands of hectares in evacuation zone
3. Culling and burial of severely damaged and contaminated animals (about 200 head of cattle and 300 sheep)	3. Culling with subsequent compulsory slaughter of more than 300 thousand head of cattle/sheep/pigs, meat kept in refrigerated stores
4. Delaying slaughter of several thousand animals, slaughter, boning of meat and its sale (bones buried)	4. Animals maintained on "relatively clean" fodder, slaughter for meat, with subsequent processing before sale
STAGE 2	
1. Organisation of crop farming and fodder production on ploughed soils	1. Organisation of crop farming and fodder production after recultivation of land via special soil tilling methods, application of improvers and organic substances
2. Restoration of agricultural production on 80% of contaminated land	2. Partial restoration of agricultural production where contamination low

The radiation accidents led to complete die-back of pine stands in some sectors of the contamination zones, thus increasing the fire hazard and the likelihood of substantial dispersal of radionuclides in smoke over considerable distances.

At Kyshtym the dead forest (mostly located in a seldom-frequented part of the zone) was felled, stacked at a sufficient distance from the intact stands and covered with soil, after which it gradually decomposed more or less completely. Back in the clearing, a deposit formed on which thick herbage grew. Various crops were subsequently planted on part of the deposit, and these took very well.

At the Chernobyl NPP, where the dead forest was located right next to the three undamaged units readied for start-up, the dead stand and the top 10-20 centimetres of contaminated soil were - against the advice of radioecologists - buried in unlined trenches to improve the radiation situation.

These areas are called "sites for temporary localisation of radioactive waste" or STLRW, and the intention was to remove the waste from the trenches at a later date and re-bury it elsewhere. Adoption of this approach in the area around the Chernobyl NPP, which has a high water table (1-3 m) and sandy soils, meant that over 900 ha of such sites were created, with the result that the exposure dose rate fell to 10 mR/h (early 1988).

Whereas in cohesive soils it takes some 10 years before radionuclides in the buried waste start to escape from unlined trenches, in the above-mentioned case substantial concentrations of radionuclides, including Pu, were detected after only 2 to 3 years, especially in groundwater near the trenches. It became clear that, in the Chernobyl NPP context, the STLRW sites gave rise to the additional and technically complex problem of preventing long-lived radionuclides from migrating out of the trenches into the hydrographic network.

It is hoped that an optimum solution to this problem will emerge from current research into radionuclide migration rates, ways of creating artificial geochemical barriers along radionuclide migration routes, and also various methods of regulating the level of groundwaters, or even of cleaning them of radionuclides, as an alternative to the extremely expensive complete "exhumation" and re-burial of STLRW waste elsewhere.

Burying the "red" forest and the fertile layer of soil in the area around the Chernobyl NPP left a sandy wasteland facilitating dust formation and prone to deflation, where the residual exposure dose rate on the surface exceeded natural background radiation by up to three orders of magnitude. There was therefore a pressing need to reduce the amount of dust formed in the area.

At first, hopes were pinned on the use of polymer-forming agents, the main rivals here being sulphite-alcohol mix, latex and preparation MM-1. However, it became clear very quickly that when trying to reduce dust formation over a substantial area of land (one thousand ha. roughly), biological methods are second to none as regards effectiveness, reliability, economic expediency and ecological appropriateness.

The essence of these methods was to restore soil fertility by applying an improver (liming of acidic soils) and mineral and organic (peat)

fertilisers in accordance with agrochemical norms, and subsequent sowing of perennial grasses under a cover of winter and spring cereals (rye, oats).

Research by many science laboratories has shown that restoration of the vegetative cover is 4 to 8 times more economical and effective than using polymer-forming chemical reagents.

Furthermore, there are indications that polymerised latex intensifies the radionuclide redeposition process and prolongs air contamination because the film it forms soon breaks down. When this happens, air currents transport the fuel particles - now equipped with "wings" of their own - over very large distances.

A comparison of curves showing the decrease in the concentration of radioactive aerosols in the air at three sites at different distances from the stricken Chernobyl NPP unit (Pripyat 3 km, Chernobyl 16 km, Zorin 36 km) in 1987 shows them to be identical. This proves that decreases in concentration do not depend on the scale of the decontamination or technical engineering measures undertaken (these having been carried out at the first two locations, but not at the settlement of Zorin).

The first stage in recultivation of the industrial site area and part of the health protection zone (HPZ) - involving restoration of soil fertility and sowing of perennial grasses - meant that as early as autumn 1988 the hazard of increased dust formation and subsequent transport of radioactive dust in the Chernobyl NPP environs had been more or less eliminated, since almost all the above-mentioned area was covered by fairly thick grass (Fig. 5).

It should be noted that completion of the first stage in recultivation of land in the HPZ coincided with the start of the period in which the value of the coefficient of redeposition of radionuclides in the air-soil system within the 30-km zone stabilised at a fairly low level ( $2 \times 10^{-10}$ ), equivalent to a concentration of radioactive aerosols in the air over 1 000 times lower than the permissible limits.

This stabilisation in radionuclide concentration in the air was not due primarily to the decontamination and technical engineering measures employed, but to a number of natural factors, although it did coincide with completion of work to remove the "red" forest and subsequent recultivation of the HPZ area.

A further example of unsuccessful technical engineering measures in the Chernobyl NPP 30-km zone was the construction of over 130 protective dykes, aimed, according to those responsible, at preventing radionuclides from being washed out of the contaminated area and into the hydrographic network.

Given the eluvial nature of the local soils and the presence of radionuclides in matrix composition form, the most perceptible result of this measure was a rise in the water table and the demise of several thousand hectares of forest stands. These dykes had no substantial effect on the scale of surface run-off of radionuclides.

A comparison of the sequential nature of the measures undertaken to eliminate the consequences of the Kyshtym and Chernobyl accidents clearly reveals a common set of post-accident measures (Fig. 6).

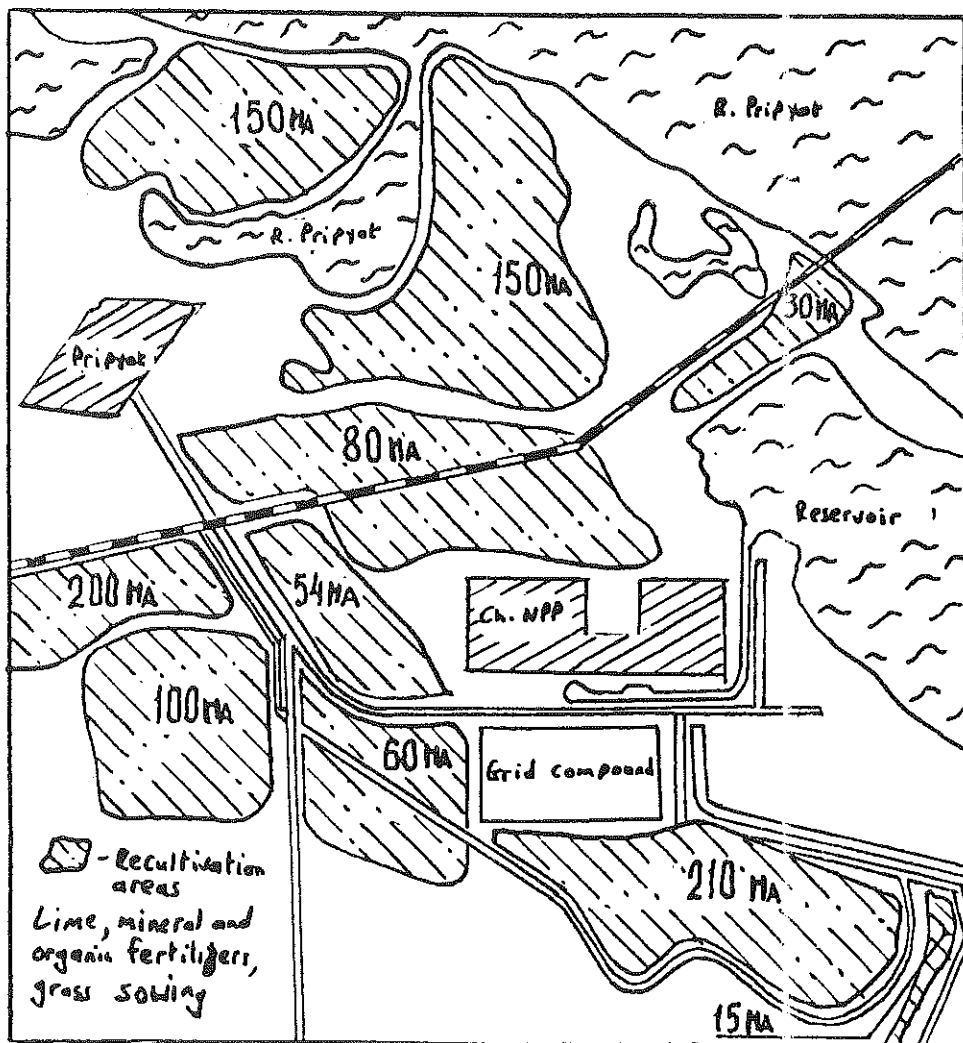


Figure 5: Plan of recultivation work in 1989

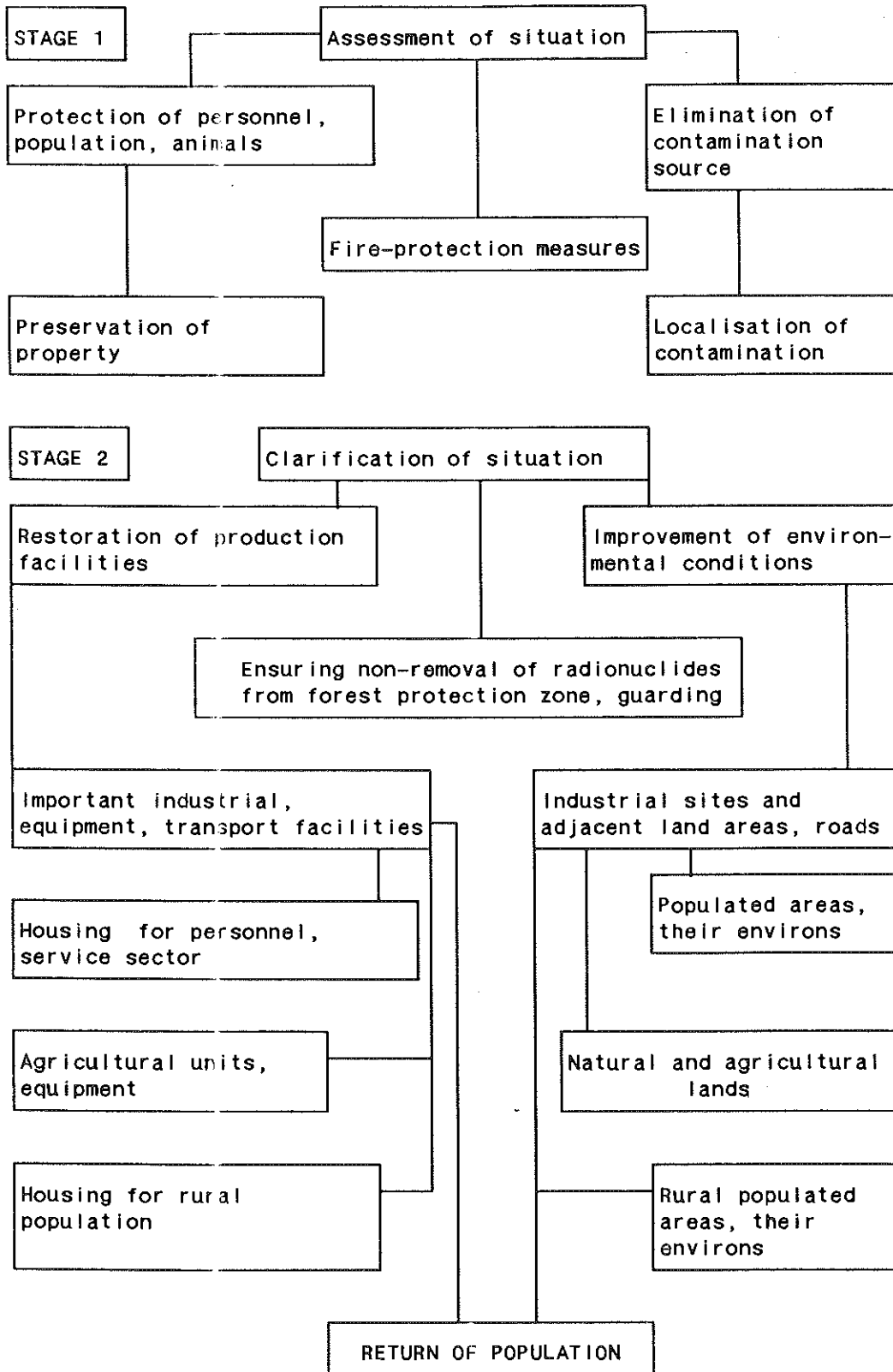


Figure 6: System of post-accident measures in evacuation zone

These cover two objective stages: Stage 1 ("hot" stage), when the aim is to prevent people and animals from dying and damage being done to property and to localise radioactive contamination, followed by Stage 2 (restorative stage).

Depending on the specific radiological and economic situation, the time interval between the said stages can vary, and both the unavoidable dose-related costs and the extent of the economic loss caused by the accident will depend on how long this interval is. Thus, correct assessment of the interval, taking account of social-psychological and other relevant factors, is of cardinal importance.

The time needed to stabilise radionuclide redeposition processes in the soil-air system can serve as an important guide for decision-makers.

It is quite natural that, in the second stage, the most important production units - whose shutdown increases the economic losses caused by the accident - should be the first to be revived (A), together with the communications, transport and housing facilities for the personnel required to get these units restarted and operating normally again (B).

The next phase in the restorative stage is the experimentally based economic "re-occupation" and exploitation of the least-contaminated agricultural and forest lands, followed by gradual expansion to areas with higher contamination levels (C).

The final stage here is the return of the evacuated people to populated areas previously readied for this purpose (D).

Practical implementation of these post-accident measures at Kyshtym and Chernobyl has revealed the following:

1. Full and rapid elimination of the consequences of major radiation accidents is impossible at present because the required facilities and methods are lacking. We can speak only of implementing a rational system of accident counter-measures to limit the negative consequences of an accident, and this in two stages: the "hot" stage (acute radiation hazard) and the restorative stage.
2. The "hot" stage includes the imperative tasks of saving the lives of personnel, population and animals and protecting them from radiation, and of guarding (and protecting from fire) the accident zone with all the property and natural resources it contains. This has to be seen to rapidly.
3. When implementing the restorative stage and improving the radioecological and radiation-health situation in vast areas, we must take into account not just radioactive decay but also the trends and dynamics of the potent processes of radionuclide redistribution (particularly in the soil-air and soil-groundwater systems) as governed by natural factors.
4. The main requirement for any protective post-accident measures in the environmental field is their ecological appropriateness, with priority being given to agro-biological measures over technical engineering solutions.
5. Restoration of economic activity in the Chernobyl NPP accident zone must be preceded by scientific and experimental checking of the planned methods and by pilot-scale trials of the same, since the



experience gained following the Kyshtym accident cannot be applied directly to the Chernobyl situation, in view of the physico-chemical features of the radionuclides in the Polesye region contamination.

6. When deciding to apply the devised methods on an industrial scale, it must be remembered that when used in actual conditions they will be substantially less efficient, due to the imperfections of the technical equipment involved and the virtual impossibility of achieving the same quality of results as in experiments.

**Obtaining 'Clean' Produce from  
Livestock Reared in Areas  
Contaminated by the  
Chernobyl Accident**

**R.G. ILYAZOV, S.K. FIRSAKOVA, A.F. KARPENKO**

## ABSTRACT

Considerable areas of Byelorussia devoted to intensive livestock farming were contaminated as a result of the accident at the Chernobyl NPP. The problem of furnishing livestock produce meeting Soviet Ministry of Health requirements (provisionally permissible levels) first arose immediately after the accident and is just as pressing nowadays.

The main aims of the studies were, in the long term, to devise and implement measures for the fodder/animal/livestock-produce food chain which would result in the maximum reduction of radionuclide levels in livestock produce.

It was established that the coefficients of radiocaesium transfer from the daily fodder ration varied from 0.004 to 0.016 for milk, from 0.03 to 0.06 for beef and from 0.10 to 0.11 for sheepmeat, which tallies with previous findings.

A procedure for producing meat from cattle reared in a contaminated environment was developed to provide "clean" produce from livestock farming. It was established that the rate of radiocaesium removal from muscle tissue is an exponential function with two periods of semi-elimination - 14 and 80 days. Scientific and experimental data were used to draw up practical recommendations for final fattening of cattle with "clean" fodder, thus allowing fodder stocks to be used rationally.

Ever since the accident occurred, milk with a high concentration of radiocaesium has been processed into butter. The standard method used to make cream butter reduces by a factor of 6 the radiocaesium content in the end product as compared to the original milk; and when melted, the butter contains hardly any traces of radiocaesium.

Considerable areas of Byelorussia devoted to intensive livestock farming were contaminated as a result of the accident at the Chernobyl NPP. The problem of furnishing livestock produce meeting Soviet Ministry of Health requirements (provisionally permissible levels) from contaminated holdings first arose immediately after the accident and is just as pressing nowadays. Currently livestock produce is contaminated primarily by the radionuclides of caesium ( $^{134}$ ,  $^{137}\text{Cs}$ ) which are biologically dangerous in the event of internal intake into the human body in diet.

The main aims of the studies were to devise and implement measures for the fodder/animal/livestock-produce food chain which would result in the maximum reduction of radionuclide levels in livestock produce. It is known that the ratio between the content of radiocaesium in the daily diet and 1 kg of animal muscle tissue (transfer coefficient) ranges from 0.04 to 0.06, which made it possible to plan animal diets and to establish limits for radiocaesium content in fodder.

A comparative analysis was carried out of the speed of removal of radiocaesium from the body of fattening livestock when summer feed and two types of winter feed were used. It was established that radiocaesium was removed most effectively by fresh mass grown on arable land and, of the winter food types, by silage-concentrate. When animals with an original muscle tissue contamination level of between 11 and 22 kBq/kg are transferred to "clean" feed (3.0-5.6 kBq/day) the speed of radiocaesium removal from muscle tissue is described by an exponential function with two periods of semi-elimination: 14 and 80 days (Fig. 1). Thus after 47 days, the original activity of muscle tissue had fallen by 4.6 times according to data from lifetime dosimetry.

Experimental data were used to work out practical recommendations for the final fattening of cattle, in the first and second stages of which the radiocaesium content in feed is not standardized and in the last stage of which animals are transferred to "clean" fodder (silage maize, concentrates).

If these recommendations are followed, rational use can be made of the fodder stocks with a high radiocaesium content.

A scientific experiment was carried out to obtain "clean" milk on farms with various levels of radioactive contamination. An outline of the experiment and the indicators of specific activity in milk are shown in Table 1, which shows that when cows are kept on "clean" fodder (11.0-26.0 kBq/day), within four or five days the radiocaesium content in milk falls by 3 - 4 times in comparison with the original level.

Ever since the accident occurred, milk with a high concentration of radiocaesium has been processed into butter. The standard method used to make cream butter reduces by a factor of 6 the radiocaesium content in the end product as compared to the original milk; and when melted, the butter contains hardly any traces of radiocaesium.

In addition to natural physical and environmental factors, the agricultural and livestock measures taken to reduce the transfer of radionuclides into livestock produce considerably reduced the amount of produce with contamination above permissible levels. For example, four years after the accident the proportion of "dirty" milk and meat produced on farms in the Gomel oblast (region) fell, in comparison with 1986, by 55 and 95 times respectively (Tables 2 and 3).

The multi-faceted programme to deal with the consequences of the Chernobyl accident is designed to obtain only clean livestock produce from agro-industrial production. Beef can be produced on natural grazing land with a contamination level of over  $15.0 \text{ Ci/km}^2$  by introducing specific breeds of beef cattle. Under this approach, beef production must be in two stages, as it is now: in the first stage using contaminated fodder from natural grazing land, and in the final stage using fodder grown on arable land or returning the herd to clean meadows. The development of migratory horse breeding for meat may also be a solution. If cattle with these levels of contamination are kept on pasture in the summer and the winter, it will be possible to avoid the production of "dirty" milk and at the same time to produce high quality meat. Branches of livestock breeding such as pig and poultry breeding should also be developed. In each contaminated rayon (district) it would be advisable to set up a study farm with highly qualified staff (livestock experts, vets, radiologists) who would be responsible for spreading scientifically based approaches to livestock farming appropriate to the given radiation conditions.

Table 1

Rates of reduction in radiocaesium concentration in cows' milk  
when they are transferred to "clean" fodder

Name of farm	Radiocaesium content in fodder $p \times 10^{-7} \text{Ci/day}$	Duration of experiment (days)	Specific activity of milk, $p \times 10^{-8} \text{Ci/l}$ Bq/kg		Degree of reduction (times)
			Before the experiment	After the experiment	
"Peremozhnik" collective farm, district of Bragin	6.0-7.0	4	1.5	0.4	3.7
"Strellichevo" state farm, district of Khoyniki	3.0-4.0	5	2.0	0.5	4.0
"Vysokoborsky" state farm, district of Vetka	5.0-6.0	5	2.0	0.5	4.0

Table 2

Coefficients of radiocaesium transfer  
Into livestock produce

TYPE OF PRODUCTION	
Milk	0.0043 - 0.0147
Beef	0.030 - 0.047
Mutton	0.10 - 0.11

Table 3

Production of milk on farms  
In the Gomel oblast after the accident

Year	Production (tonnes)	Production with a contamination level above the provisionally permissible level (tonnes)	Percentage of produce contaminated (%)
1986 (4th quarter)	450.1	299.3	66.5
1987	813.5	235.9	29.0
1988	860.5	146.3	17.0
1989	853.5	49.2	5.7
1990 (2nd quarter)	457.8	5.6	1.2

Table 4

Beef production on farms in the  
Gomel oblast after the accident

Year	Production (tonnes)	Production with a contamination level above the provisionally permissible level (tonnes)	Percentage of produce contaminated (%)
1986 (4th quarter)	341.0	64.8	19.0
1987	1 842.0	71.0	3.8
1988	2 055.0	24.0	1.2
1989	2 244.0	10.0	0.5
1990 (2nd quarter)	385.6	0.7	0.2





**Countermeasures at the  
Windscale Accident: The Origin  
of the Emergency Reference  
Level Concept and its  
Subsequent Evolution**

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## Introduction

The Windscale accident was the first large-scale accidental release of radioactive nuclides from a nuclear reactor and as might be expected it had a profound effect on thinking in relation to protection of the public in such circumstances.  $^{131}\text{I}$  was the most significant radionuclide released and it remains the case that for any operating or recently operating reactor from which there is an accidental release the isotopes of iodine will, at early times, be a significant, if not the most significant, hazard. It is therefore appropriate to consider the response to that accident, how it influenced future events in radiological protection and to ask whether there are still lessons to be learned.

## Historical perspective

The accident to pile No. 1 at Windscale on 10-11 October 1957 occurred at a time before intervention levels or counter-measures for accidents to nuclear reactors had been formalised. The circumstances of the accident have been extensively described (1) and the release of radionuclides estimated (2). The release of particulate material was small and just three nuclides dominated the release, namely  $^{131}\text{I}$ ,  $^{132}\text{Te}$  and  $^{133}\text{Xe}$ , no other isotope constituting much more than 10% of the  $^{131}\text{I}$ . Because the reactor had been shut down from normal operation on the 7th October and had only been operated at a very low power for two short periods (to induce nuclear heating to release Wigner energy), the greater part of the shorter-lived isotopes of iodine had decayed.

Neither  $^{132}\text{Te}$  or  $^{133}\text{Xe}$  are metabolically active thus, in the early stages of the release  $^{131}\text{I}$  presented the major hazard to the general public. This was recognised early on in the course of the accident, as was the fact that the major route of intake would be ingestion through drinking fresh milk. Thus an intervention limit of 3700 Bq/l. (0.1  $\mu\text{Ci}/\text{l}.$ ) was imposed in the vicinity of Windscale and between 11 and 13 October milk production in an area of 520  $\text{km}^2$  was banned for human consumption. This ban lasted up to 25 days when  $^{131}\text{I}$  levels had fallen below the intervention limit.

Crick and Linsley (3) estimate that this countermeasure saved  $3.5 \times 10^3$  man Sv to the thyroid glands of the population of Cumbria who received, as a result of the accident,  $3.2 \times 10^3$  man Sv to the thyroid. Estimates of thyroid doses made by Baverstock and Vennart (4) using the data of Clarke (2) on integrated activities of  $^{131}\text{I}$  in air are in good agreement with measured values for adults, suggesting that the milk ban was effective and that thyroid dose derived primarily from inhalation of  $^{131}\text{I}$ . However, it is noted (4) that for children the predicted doses are considerably smaller than those measured, suggesting that as well as inhalation the ingestion route was important and the milk ban not completely effective.

Largely in response to the Windscale accident, the concept of an Emergency Reference Level (ERL) was derived (MRC 5,6,7,8).

#### Emergency Reference Levels

The ERL of dose, as it was originally perceived in the U.K. (8), is a complex concept. It is not a limit but rather a minimum dose to individuals at which action to reduce exposure of the public should start to be considered. What action would be taken would be determined by a balance between the extent of dose reduction that could be achieved by particular countermeasures and the risks incurred in taking such countermeasures.

The rationale for such an approach lies at least partially in the particular circumstances that apply to accidents to the type of reactors used in the U.K., namely graphite cored, gas cooled reactors. Such reactors have a large core volume and thus a relatively low energy density in the core. Furthermore the pressure in the coolant circuits is relatively low. In the event of an accident leading to a release of radioactive materials, the situation tends to evolve over several hours rather than minutes as would be the case where energy densities and coolant pressures were higher. Thus it is reasonable, in the early stages of an accident to make sequential assessments of radioactivity in

the local environment to monitor how the situation is developing regarding doses to the public, while concentrating attention primarily on measures to limit the further release of radioactive material, thus optimising the chances of reducing the ultimate consequences of the accident, full attention having to be given to the public only when the ERL of dose is likely to be exceeded.

Thus, ERLs were specifically designed for application in the first few hours of the accident when resources at the accident site were likely to be scarce, and to apply to relatively small populations close to the source of the release. Table I summarises the ERLs of dose for the whole body and the thyroid recommended by the MRC (8) in 1975.

In 1977, responsibility for setting ERLs passed to the NRPB (9) who largely retained the principles but subsequently specified two levels of ERL applicable to three specific counter-measures, i.e. evacuation, sheltering and administration of iodine tablets (10). At the lower level action may be desirable but not essential, but at the upper level countermeasures should be introduced whatever the circumstances. In other words, the ERL became a dose limit in the true sense of the word and countermeasures were specified. This was a clear departure from the MRC philosophy which described the ERL of dose as "not a firm action level but as a dose where the responsible authorities should judge whether countermeasures should be introduced, full account being taken of the disadvantages and risks these countermeasures might create" (8). Thus the NRPB made the judgement on the disadvantages and risks implicit in the range of ERLs of dose specified. However, the transitory nature of ERLs was retained and indeed spelled out (10). The lower ERLs of dose adopted by the NRPB are either equal to or lower than those that were recommended by the MRC (1975); see Table 1.

In 1984 the ICRP published advice on the subject of unplanned releases (11). Up to that date the ICRP had taken the view that intervention levels were a matter for national authorities because individual circumstances in countries were so different that no universal guidance could be given

(12). The general principles for implementating counter-measures outlined in 1984 placed emphasis on dose to the individual and on the balance between risk saved and risk incurred. The ICRP divided the temporal evolution of the accident into three phases, early, intermediate and late.

In the early phase they proposed there was a need for procedures on the initiation of intervention to be included in emergency plans for any appropriate facility. These should anticipate the likely public exposure depending on the condition of the plant and the prevailing meteorological conditions. In an appendix the ICRP gives a table of dose equivalent levels for early phase countermeasures (see Table II). They give upper and lower limits for low risk countermeasures, sheltering and administration of stable iodine, and for higher risk countermeasures such as evacuation. For the low risk countermeasures, the lower level is made equal to the annual dose limit for the public, at that time 5 mSv, and the upper an order of magnitude greater. For the higher risk countermeasures, the values are 10 times higher.

For later phases the main body of the report assumes that the opportunity for consultation with an expert group will enable decisions to be made in the light of the prevailing circumstances. However, in the appendix a further table specifying dose equivalent levels for intermediate phase countermeasures is given. In this case the low risk countermeasures concern control of foodstuffs and the higher risk, relocation of population. The dose limits are numerically the same as the low and high risk countermeasures in the early phase.

In 1988 the NRPB initiated discussions on revising the ERLs for the U.K. (13). A number of modifications to the concept are proposed. Firstly, that dose limits for the public should not have any substantial influence on the setting of intervention limits. Secondly, primary consideration is given to the various countermeasures and then the time phases (early, intermediate and late) in which they might be appropriate. Table III summarises the proposed limits.

Commentary

In 1957 the Windscale accident highlighted the need for a means of decision-taking in the early stages of an accidental release of radionuclides to the environment and the concept of ERLs was subsequently born. Thirty or more years later the concept has evolved and its application broadened - for example ERLs were used to justify the absence of countermeasures to limit  $^{131}\text{I}$  intake through milk in the U.K. at the time of the Chernobyl accident (14).

In general, ERLs of dose have been reduced and, at the same time, the time scales over which they are applicable and the sizes of population to which they might apply have been increased. They have evolved from levels at which action should be considered, if dose can be saved, to formal dose limits. The revised concept leaves much less room for action determined by the individual circumstances of the accident. Are these changes to be welcomed?

In part the answer to this depends on whether it is a realistic assumption that accidental releases can be controlled at source. If they can, then the initial concentration of scarce resources to that purpose must be of value. The somewhat greater risk to a few is compensated by the benefit of a reduced overall impact of the accident. It should be noted here that the lower the values of ERL the less the opportunity for gaining this benefit is available. If the assumption is false then nothing is to be gained by not turning attention directly to the safety of the public and the risk-benefit equation concerns only the dose to be saved and the risk incurred by the countermeasures.

The use of the ERL concept as a dose limit without reference to dose saved is surely to be regretted, particularly where some risk might be attached to the introduction of a countermeasure.

We should also ask whether as rigid a regime of intervention limits, as has evolved, best serves our interests. The circumstances of an accident are highly variable and what can be done to minimise exposure varies from accident to accident, depending on numerous conditions that might apply in a specific location or at a specific time. The risks incurred by counter-measures also varies from place to place

and situation to situation.

Thus the value of treating each accident on its merits should not be under-estimated. To take just one example. In the U.K. no intervention measures were taken at the time of the Chernobyl accident to reduce the doses from  $^{131}\text{I}$ . In the Netherlands when  $^{131}\text{I}$  was detected in milk a ban on outdoor grazing of cattle for 4 to 5 days was introduced. This single measure is estimated to have reduced thyroid doses to the critical group by 50% (15). In none of the documents discussed above relating to ERLs is this countermeasure considered, yet for minimal cost and no risk whatsoever to the population, a significant dose saving was made. If the principles outlined by the ICRP are taken at face value, this is the kind of action we should expect.

Whether it was appropriate to use ERLs in the context of Chernobyl in European countries is questionable. There was no decision to be made between implementing countermeasures and mitigating the consequences of the accident. According to the main body of the ICRP report (11), a balance between saving dose to the individual and incurring risk from countermeasures should have been struck after the circumstances had been assessed by an expert group. If there are effective countermeasures (i.e. ones that significantly reduce doses to individuals) that incur no risk and are not inordinately expensive or disruptive without reference to the annual dose limits to the public. There will of course be doses too small for savings to be effective. It is thus to be regretted that the ICRP chose, in the appendix to the 1984 report (11), to give intervention levels for the intermediate phase of accidents; these, like the response of countries outside that in which the accident happened, should be a matter for action in the light of the prevailing circumstances.

The existence of a rigid regime of intervention levels does not preclude unanticipated action being taken if the circumstances suggest it would be beneficial. However, the existence of an apparently comprehensive system of regulation does not encourage actions beyond those that satisfy the regulations. It often seems to be the case that



recommendations by bodies like the ICRP are given biblical status and little thought is given to whether, in any particular circumstances, some modified or alternative response might not be more appropriate. It should be remembered that the ICRP's primary role is to make recommendations concerning the planning of exposure and not to provide a basis for assessing the actual consequences of a given exposure. It is therefore to be hoped that in future less rigidity is incorporated into the criteria for intervention and more emphasis is placed on developing strategies for dose reduction most appropriate in the circumstances of a particular incident.

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Table I

A comparison between ERLs recommended  
by the MRC<sup>a</sup> and NRPB<sup>b</sup>

Organ	MRC mSv	NRPB (dose equivalent mSv)		
		Evacuation	Sheltering	Iodine
Whole body	100	{ 500	5	-
		{ 100	25	-
Thyroid	300	{ 1500	250	250
		{ 300	50	50

a taken from reference 8

b taken from reference 10

Table 2

Dose-equivalent levels for early phase countermeasures

Countermeasure	Dose equivalent (mSv)	
	Organ Whole body	Lung*, thyroid and any single organ preferentially irradiated
Sheltering and stable iodine administration		
upper dose level	50	500
lower dose level	5	50
Evacuation		
upper dose level	500	5000
lower dose level	50	500

\* In the event of  $\alpha$ -irradiation of the lung, the numerical values apply to the product of RBE and absorbed dose in mGy. This RBE is expected to be substantially less than 10.

Taken from reference 11

Table III

Summary of proposed dose levels for the introduction of countermeasures to protect the public

Countermeasure	Organ	Dose Equivalent (mSv)	
		Lower level	Upper level
Sheltering	Whole body/ effective	3	30
	Thyroid	30	300
Evacuation	Whole body/ effective	30	300
	Thyroid	300	3000
Distribution of stable iodine tablets	Thyroid	30	300
Relocation	Whole body/ effective	3	30
Restrictions on food and drinking water	Whole body/ effective	1	10
	Thyroid	10	100

Taken from reference 13



**Reappraisal of Environmental  
Countermeasures to Protect  
Members of the Public  
Following the Windscale  
Nuclear Reactor Accident , 1957**

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## ABSTRACT

On 10 October 1957 one of the two air cooled reactors on the Windscale site, Cumbria UK, caught fire resulting in an uncontrolled release of activity to atmosphere, which lasted until the fire was quenched by water during the morning of 11 October 1957. Subsequent dispersion of activity was widespread. The most important nuclides, with respect to potential health problems to the general public, were  $^{131}\text{I}$  and  $^{210}\text{Po}$ . Significant quantities of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  were also released.

A decision was taken to implement a ban on the distribution of milk containing more than  $3700 \text{ Bq l}^{-1}$  of  $^{131}\text{I}$ . The consequent restrictions covered an area of  $518 \text{ km}^2$ ; extending from St Bees in the north to Barrow peninsula in the south. The ban lasted from 25 to 44 days within this area, during which time some 3 million litres of milk were disposed of. Monitoring of other foodstuffs and nuclides was undertaken, but no further countermeasures were considered necessary.

In the most heavily contaminated areas the maximum dose equivalent to a child's thyroid was estimated to be 160 mSv. The predicted dose, in the absence of any countermeasure, was 360 mSv.

Since that time, both the estimated inventory of activity released in the incident and the basis for limitation of radiation doses to members of the public (resulting from routine or accidental releases) have been reviewed several times. Based on current recommendations for off-site countermeasures required in the event of abnormal releases of activity to atmosphere, the actions undertaken following the 1957 Windscale Fire are reappraised.

It is concluded that the milk ban area would have been extended substantially. Limited action to restrict consumption of locally produced green vegetables and, probably, free range eggs would have been appropriate also. Greater emphasis on sampling of meat products, particularly from upland areas, would have been prudent. Notwithstanding this, the actions undertaken reduced both individual and collective doses arising and were consistent with the radiation protection standards then in place.

## INTRODUCTION

The accident at the Windscale works of the UK Atomic Energy Authority occurred on 10 and 11 October 1957. Some of the uranium fuel within one of the two air cooled reactors became white hot and there was an emission of fission products, especially those which are volatile or have volatile compounds. The course of events has been described in the Report of the Committee of Investigation [1]. The accident occurred during a controlled release of Wigner energy, which had become stored in the graphite moderator of the pile as a result of normal operations. The immediate cause of the accident was a second nuclear heating applied, before it was necessary and at too rapid a rate, because it was thought that the Wigner release was dying away and that parts of the graphite structure would therefore escape being annealed. The instrumentation of the pile was not sufficient in quantity or in distribution to enable a reliable judgement to be made and it is probable that this heating caused the failure of one or more uranium fuel cartridges.

The radioactivity was emitted to atmosphere from the 120 m stack of Pile No. 1, beginning some time before 14.00 (GMT) on 10 October and continuing to about 11.00 on 11 October 1957. Peaks in emission occurred about midnight on 10 October and again during the period 09.00 to 11.00 on 11 October when water was used to put out the fire. Water was actually turned on at 08.55 11 October and turned off at 15.10 on 12 October, by which time the pile was cold. During the earlier part of the release, the wind at the height of the stack-top was light and mainly from the SW. With the passage of a cold front over Windscale about 01.00 to 02.00 on 11 October, the wind veered to NW and became much stronger. An inversion existed some hundred metres or so above ground level and winds above this inversion differed quite markedly from the ground winds.

Shortly after midnight on 10 October, the Chief Constable of Cumberland was warned of the possibility of an emergency and men in the factory were instructed to stay indoors and wear face masks. No action was advised in the district.

Neither the total activity released nor the variation in rate of emission is known from measurements at source but estimates have been made, both at the time and later [eg 1, 2, 3, 4, 5]. The principal nuclides released are listed in Table 1. The physical and chemical nature of many of the emitted radionuclides remains uncertain. Some 5% of the total activity released may have been associated with graphite particules, up to 15  $\mu\text{m}$  in diameter, found in the neighbourhood of the pile after the accident [6].

During the course of the accident there was patchy rainfall throughout north-west England. The highest totals (8-10mm from 09.00 10 October to 09.00 11 October) were recorded in the Furness peninsula. From records of a rainfall recording gauge at Barrow it is known, however, that most of this fall occurred between 17.00 and 19.00 on 10 October, about 6 hours before the passage of the front and before the active cloud had reached the area. Traces of rain between 01.00 and 02.00 on 11 October, about the time of the passage of the front, were recorded at Silloth and it is likely that any rain associated with the front was heavier on the hills. The front became very weak as it passed south and there was no rain associated with it in the Midlands and southern England [7].

#### ENVIRONMENTAL SURVEYS AND COUNTERMEASURES UNDERTAKEN

Environmental surveys carried out after the accident have been reported by Chamberlain & Dunster [6] and Dunster et al. [8]. As soon as it was realized that there was a considerable release of activity to atmosphere, there were three possible hazards requiring immediate evaluation.

- i External radiation.
- ii Inhalation of activity.
- iii Ingestion of activity from contaminated food and water.

At about 15.00 on 10 October a survey van was sent out to the south of the factory, with a second van sent out about 17.00 to the north. These two vans maintained continuous patrols throughout the night and next day. During the period of release, and after, air activity measurements were made both on the site and in the district.

The highest radiation level measured was  $40 \mu\text{Sv hr}^{-1}$  (against a normal background of  $0.1 \mu\text{Sv hr}^{-1}$ ) at a point approximately 1.6 km downwind on the coast. Radiation levels were generally 1.5 to  $2 \mu\text{Sv hr}^{-1}$  in the area 3-5 km south of the factory and much lower in other directions. Exposures from radiation were assessed as small and insufficient to warrant protective action [8].

Levels of activity in air on site rose from a norm of 3 to  $5 \text{ Bq } \beta \text{ m}^{-3}$  to  $50 \text{ Bq } \beta \text{ m}^{-3}$  during 11.00 to 14.00 on 10 October, eventually rising to  $850 \text{ Bq } \beta \text{ m}^{-3}$ , with peaks up to  $1.5 \times 10^4 \text{ Bq } \beta \text{ m}^{-3}$  on the morning of 11 October. The average activity in air throughout the incident was around  $150 \text{ Bq } \beta \text{ m}^{-3}$ . In the district the highest value measured was  $1 \times 10^3 \text{ Bq } \beta \text{ m}^{-3}$ , during the night of 10 October, at a point about 3.2 km south-east of the factory. In general, values reported were much lower than this and from 12.00 on 11 October fell away rapidly. Air contamination was assessed to have risen to 'worrying' but not dangerous levels, and no action was taken [8].

Deposition of activity to ground was measured, or inferred, initially from three methods: gamma spectrometry of grass; gamma spectrometry of soil/root matt; and ground level gamma dose rate surveys [6, 8, 9]. The ground gamma measurements and estimated deposition of  $^{90}\text{Sr}$ ,  $^{131}\text{I}$  and  $^{137}\text{Cs}$  in north-west England are presented in Fig. 1. Peak concentrations were reported along the coastal and inland regions south east of the site to the Duddon estuary and Furness peninsula. In addition, enhanced deposition is apparent running towards the Wasdale, Eskdale and Duddon valleys. Deposition of activity close to Windscale is summarised in Table 2.

By noon on 12 October the first milk analyses were available (from milk collected during the evening of 10 October and the morning/evening of 11 October). Levels of  $^{131}\text{I}$  ranged from traces to  $1.8 \times 10^4 \text{ Bq l}^{-1}$ . The higher readings were exclusively for milk obtained on the evening of 11 October. Analysis of the Seascale morning milk of 12 October was completed at 15.00 and showed  $3 \times 10^4 \text{ Bq l}^{-1}$ . By about 21.00 on 12 October, arrangements were made to ban milk containing more than  $3.7 \times 10^3 \text{ Bq } ^{131}\text{I l}^{-1}$  and deliveries from suppliers within a 3.2 km radius of Windscale were withheld. As analyses were completed throughout 13 to 15 October, the restrictions on distribution were extended in stages to cover an area some

40 km long, 16 km wide at the south and 10 km broad at the north (Fig. 2); including the Barrow peninsula to -10 km north of Windscale. This amounted to 518 km<sup>2</sup>.

Samples were taken further afield, from the Lancashire coast, the north Wales coast, the Isle of Man, Yorkshire, Westmorland and the south of Scotland. By 19 October surveys had extended to Northern Ireland and the south of England. Milk restrictions were not extended into these areas.

The intensity of milk monitoring initially placed special emphasis on the fringe of the restricted zone. On 16 October, for instance, 58 milk samples were collected from outside the area and only 7 inside. By 22 October, when 260 samples were collected in one day, 230 were obtained from the restricted area with only 30 from outside, since by this time the area concerned had been defined clearly. A total of 3000 samples (2500 l) of milk were analysed in the period immediately following the accident.

Analytical results for milk (Table 3) confirmed that the main ingestion hazard resulted from <sup>131</sup>I. Other foodstuffs were also monitored, particularly vegetables, meat (including thyroids and bones), eggs and water. Peak concentrations recorded for iodine and strontium are presented in Table 4. No restrictions were placed on these. Soil, root matt, herbage and, occasionally, cattle dung were also monitored.

The rate of decrease of activity in milk and grass is illustrated in Fig 3. Milk restrictions were first relaxed in the eastern region (31 October), followed shortly afterwards by the south. By 4 November, the restriction was limited to a coastal strip extending 19 km south of Windscale, which persisted until 23 November 1957. The total period of restriction was thus about 8 weeks, latterly due to cautious assessment of the <sup>90</sup>Sr and <sup>137</sup>Cs levels rather than <sup>131</sup>I. During this time some 3 million litres of milk were disposed of through drains to sea [12].

## REAPPRAISAL OF COUNTERMEASURES REQUIRED

Countermeasures employed following the Windscale fire have been reappraised, based on emergency reference level "ERL2" values [14, 15] (which superseded the advice of the MRC [3, 16, 17, 18]) and the most recent advice from the NRPB [13]. Further guidance from the CEC is referenced [19] and the effects of the draft proposals of the ICRP [20] are considered.

### Atmospheric concentration of radioactivity

Data for air concentrations off-site are generally restricted to total  $\beta$  activity which suggest an upper limit of  $1000 \text{ Bq m}^{-3}$  in the district, with a lower limit of perhaps  $150 \text{ Bq m}^{-3}$  (based on peak district concentrations and average prevailing site concentrations respectively [8]). This information can be supplemented by appropriate modelling. The relative proportions of beta/gamma emitters in the release are well characterised as are the air samplers available at the time. It can thus be assumed that the total  $\beta$  measured in air was dominated largely by particulate activity such as  $^{99}\text{Mo}$ ,  $^{103}\text{Ru}$  and  $^{137}\text{Cs}$ . Both  $^{132}\text{Te}$  and  $^{131}\text{I}$  are retained less effectively by filter papers and a 20% retention factor may be appropriate [eg 34]. Retention of  $^3\text{H}$  was probably negligible. Strontium-90 was present at about 0.5% of the  $^{137}\text{Cs}$ . Concentrations of  $^{210}\text{Po}$  were around 20% of the  $^{137}\text{Cs}$  component. The integrated air concentration throughout the 24 hr release period can thus be estimated, as presented in Table 5.

As an independent model, the integrated concentration of each nuclide can be predicted from atmospheric dispersion models [21], based on best estimate releases and meteorological conditions prevailing. It is probably reasonable to assume Pasquill category D weather throughout the incident and all activity being blown into a sector of  $60^\circ$  width, to the south-east of the release source. The thermal buoyancy of the release introduces considerable uncertainty in determining an effective stack height. For simplicity, and probably somewhat conservatively, the actual stack height of 120m is used. Results obtained are presented in Table 5 and are in all cases encompassed by the range of air concentrations derived from total  $\beta$  activity. Such good agreement may be fortuitous, since the model is quite dependent on the assumed effective stack height, but does suggest

that results available are representative of the area of peak impact. Comparison with ERLs for sheltering the local populace, Table 5, shows that even under the most restrictive criteria (limited principally by  $^{131}\text{I}$  and  $^{210}\text{Po}$ ), and assuming the highest estimated airborne concentration, no immediate action was required. Issue of iodate tablets, to restrict the thyroid dose from  $^{131}\text{I}$ , would normally be recommended under the same criteria as sheltering the population and hence, again, this countermeasure was not required. Thus, even in retrospect, it can be concluded that the appraisal at the time of the incident was probably correct, that atmospheric concentrations rose to "worrying" but not "dangerous" levels.

#### External radiation levels

The peak recorded district radiation level following the Windscale fire was  $40 \mu\text{Sv hr}^{-1}$  which probably includes a component from cloud gamma dominated by noble gases (Xe mainly). Nonetheless, in isolation, this could have prompted a requirement to decontaminate some areas. However, such levels did not persist in the environment. Generally external radiation did not exceed  $2 \mu\text{Sv hr}^{-1}$  and decayed with a half-life of 8 to 9 days. No protective action was recommended, nor would such measures be taken on current criteria.

#### Activity in milk and other foodstuffs

Grass and soil samples collected from around Sellafield indicated significant deposition of  $^{131}\text{I}$ ,  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ ,  $^{103+106}\text{Ru}$  and  $^{95}\text{Zr}$  (eg Table 2 and Fig 2). Peak levels of activity recorded on grass are compared in Table 6 to modelled predictions. These are in reasonable agreement, apart from strontium where measured deposition at the site boundary was greatly in excess of modelled values (which predict a peak at 1.5 to 5 km from site). This can be explained by discharges of particulates from the Windscale piles prior to 1957 [35], and is consistent with data presented in Table 4 showing that concentrations of  $^{90}\text{Sr}$  were uniform through turnip tops and tubers from a local farm (indicating a substantial contribution from root uptake) whereas  $^{89}\text{Sr}$  was present in turnip tops at more than five times the level in the tubers (indicating foliar deposition).

Formal ground contamination DERLs for sheltering of the population have not been proposed, although it has been suggested [23] that sheltering may act to reduce significantly dose uptake from ground contamination. Appropriate DERLs can be scaled from published advice on the requirement to evacuate the population [24]. Again, however, it can be seen from Table 6 that there was no requirement for shelter based on ground contamination.

By contrast, deposition of  $^{131}\text{I}$  exceeded those levels established for the introduction of milk bans and, to a lesser extent, restrictions on green vegetable consumption. Modelled deposition levels suggest this milk ban would exceed 100 km (Table 7), although uncertainties in modelling preclude prediction beyond this distance. Advice to restrict green vegetable consumption may have extended about 20 km but would depend very much on local crops and availability for harvesting. The duration of any such restriction could be expected to mirror that for milk and would be dominated by physical decay and biological loss of  $^{131}\text{I}$  ( $t_{\text{eff}} \sim 4\text{d}$ ). Levels of  $^{137}\text{Cs}$  also exceeded the more limiting values at which a restriction may be imposed on milk and could have led to greater persistence of the ban in local areas.

Measured concentrations of  $^{131}\text{I}$ , recorded in milk on 13 October, confirm the greater extent of restrictions which would be advised currently (Fig 2), particularly if based on the recommendations of the CEC [19].

Information for other foodstuffs can also be compared to DERLs. The peak concentration of  $^{131}\text{I}$  reported in drinking water was 3.7 to 37  $\text{Bq l}^{-1}$ . This is well below current long term DERLs for substitution of freshwater supplies [eg 13, 19, 24]. At the time of the incident the maximum recommended concentration of  $^{131}\text{I}$  in drinking water was 220  $\text{Bq l}^{-1}$ , which is actually more restrictive than the 400  $\text{Bq l}^{-1}$  currently proposed by the CEC [19]. An  $^{131}\text{I}$  concentration of 1050  $\text{Bq l}^{-1}$  was recorded in the R. Calder immediately downstream of the Windscale drain but elsewhere the peak level was 85  $\text{Bq l}^{-1}$  in Linbeck (near Devoke water). Neither of these sources provide drinking water. Data relating to concentrations in fresh rainwater are not available.



Reported levels of  $^{90}\text{Sr}$  in vegetation, presented in Table 4, were well below current limits for the introduction of restrictions on consumption. Data for  $^{131}\text{I}$  are rather more extensive, particularly for leafy vegetables. The peak recorded concentration in cabbage outer leaves was  $\sim 23,000 \text{ Bq kg}^{-1}$ , from a farm near Holmrook on 17 October [33]. This implies an initial peak value of  $>30,000 \text{ Bq kg}^{-1}$ , which would be close to current values for withdrawing green vegetables ( $1.1 \times 10^5$ ,  $2.2 \times 10^4$  and  $3.2 \times 10^4 \text{ Bq kg}^{-1}$  respectively for ERL2, [13] and [13]mod). A decontamination factor of 10 to 50 was assumed at the time, for normal food preparation processes of washing and removing outer leaves [33]. This is in accordance with recent statements [24] and confirms that any advice to withdraw fresh vegetables would be limited in area and timescale.

Loss of activity from vegetables through October is presented in Fig 4. Generally, the highest concentrations were recorded in outer leaves of brassica (eg cabbage, lettuce, sprouts and kale). Lowest levels of  $^{131}\text{I}$  were recorded in edible parts of root crops and were not generally above  $400 \text{ Bq kg}^{-1}$ . The single highest value (of  $27,750 \text{ Bq kg}^{-1}$ ), was recorded on swede leaves from Barrow on 21 October. Samples to the north of the site showed very little activity.

Seaweed sampled from the local coast south of Windscale had initially high concentrations of  $^{131}\text{I}$  (Fig 4).

The peak concentration of  $^{131}\text{I}$  in locally produced eggs was  $54,000 \text{ Bq kg}^{-1}$  although generally values were not greater than  $25,000 \text{ Bq kg}^{-1}$  [33]. By 4 November values had declined to less than  $1,000 \text{ Bq kg}^{-1}$  (Fig 5). The CEC currently recommend restricting all minor foodstuffs containing more than  $30,000 \text{ Bq kg}^{-1}$ . For other foodstuffs a general limit of  $3,000 \text{ Bq kg}^{-1}$  is suggested [19]. Depending on the status assigned to egg consumption locally, this could imply a restriction being imposed for up to 15 days.

Fewer data are available for  $^{131}\text{I}$  in meat products, and all relate to lowland areas. Nonetheless, peak recorded values of  $85 \text{ Bq kg}^{-1}$  in trout,  $590 \text{ Bq kg}^{-1}$  in salmon roes (from the R. Calder and the Ravenglass estuary)  $740 \text{ Bq kg}^{-1}$  in mutton and  $3,367 \text{ Bq kg}^{-1}$  in beef (from around Seascale and Drigg), imply that there was no widespread uptake requiring restrictions.

Data for sheep thyroids (Fig 6), obtained from animals slaughtered at the Whitehaven abattoir, indicate a loss of  $^{131}\text{I}$  activity with a biological half-life of 3-4 days. This is in line with rates noted for local foodstuffs above and again suggests that peak concentrations recorded would not have persisted for long periods. As a prudent measure, however, greater emphasis would be applied currently to sampling upland areas.

#### Estimate of dose averted

Average radiation doses to the thyroid in local residents were 5-20 mSv in adults and 10-100 mSv in children; the maximum dose inferred from measured activity in the thyroid was 160 mSv to a child. For adults, the data suggest that the prime route for intake of  $^{131}\text{I}$  was inhalation (Table 8) but for children there is a discrepancy between assessed dose and estimated inhalation dose, indicating a contribution from ingestion. Without a milk ban, thyroid dose levels were expected to reach 70 mSv in adults and 360 mSv in children. The prompt introduction of restrictions on milk supplies thus reduced ingestion doses to the local population. Generally, some 75% of the estimated child ingestion dose was averted (Table 8), falling to around 55% for the most exposed child. For adults the proportion of dose averted was greater. These data are in accord with estimates that imposing a milk ban within 2-4 days of a release of activity should avert 65% to 85% of the dose which would otherwise be incurred via this pathway [29].

Doses incurred via other pathways (eg consumption of fresh vegetables) do not appear to contribute substantially to adult doses incurred locally. Uptake of other radionuclides was detectable (eg Table 9) but doses incurred were generally small by comparison to  $^{131}\text{I}$ .

Collective doses following the incident have been assessed elsewhere [5, 32]. Correcting for the age distribution of the population, the collective effective dose equivalent commitment from all pathways was about 150 manSv to Cumbria, 1900 manSv to the UK and 2000 manSv to Northern Europe. Of this about 23% in Cumbria was due to inhalation (mainly  $^{210}\text{Po}$  and  $^{131}\text{I}$ ) and 59% from milk consumption (mainly  $^{131}\text{I}$ ). For this region it was estimated that a further 108 manSv was averted by introduction of the milk ban. Elsewhere, only 30% of the dose derived from milk consumption (due to the much lower levels prevailing) and ~50% derived from inhalation.

## CONCLUSIONS

In the light of those data which are available, had protective measures following the Windscale pile fire been based on current criteria, actions other than the implementation of a milk ban would be limited to a restriction (probably short term) on local green vegetable consumption and probably eggs, based primarily on  $^{131}\text{I}$  levels. Under current criteria the area to which the milk restriction applied would be considerably greater than adopted at the time. Further emphasis would also have been placed on gathering data for fresh rainwater and upland meat products. Nonetheless, actions undertaken at the time reduced significantly both individual and collective dose uptakes and were consistent with the radiation protection standards then in place.

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TABLE 1. Estimate of principal nuclides released [3, 5, 26, 36]

Nuclide	Total release TBq	Nuclide	Total release TBq
<sup>3</sup> H	5000	<sup>129m</sup> Te	25
<sup>89</sup> Sr	5.1	<sup>129</sup> Te	25
<sup>90</sup> Sr	0.22	<sup>131</sup> I	600 1000 <sup>a</sup>
<sup>95</sup> Zr	7.5	<sup>132</sup> Te	600-1000
<sup>95</sup> Nb	7.5	<sup>134</sup> Cs	1 2
<sup>99</sup> Mo	36	<sup>137</sup> Cs	22 94 <sup>b</sup>
<sup>103</sup> Ru	40	<sup>144</sup> Ce	4 0
<sup>106</sup> Ru	5.9	<sup>147</sup> Pm	2 3
<sup>111</sup> Ag	0.52	<sup>210</sup> Po	8 8 <sup>c</sup>
<sup>115</sup> Cd	1.6	<sup>239</sup> Pu	0.0016

- a Crabtree [26] estimated that his figure (of 1000 TBq) could be in error by a factor of 1.5 either way. An equal release of <sup>132</sup>Te is assumed.
- b The upper end of this range is considered most probable [36].
- c Data from Stewart & Crooks [7] suggest a <sup>210</sup>Po/<sup>131</sup>I release ratio of 1:38.

TABLE 2 Activity Deposition to Grass

Distance (km)	Bearing from stack	Activity deposition (Bq m <sup>-2</sup> ) <sup>a</sup>						Total α
		<sup>137</sup> Cs	<sup>131</sup> I	<sup>103+106</sup> Ru	<sup>95</sup> Zr	<sup>90</sup> Sr	<sup>89</sup> Sr	
0.4	270°							592
1.0	320°	370		1110	740	36	278	
2.4	160°	2960		1110	3885	64	1417	276
3.2	140°	7215	370000	7030	10915	25	873	167
6.4	147°	9065	223850	7770	4070			
8.8	144°	5920	74000	4440	740			
16	150°	4625		6105	3700			
28	152°	185	19610	2960	1850			

- a Data from [8] and [33]; samples taken 18 October 1967.

TABLE 3. Radionuclides in Milk from two Farms near Windscale  
Activity Concentration (Bq l<sup>-1</sup>)

Date	Total $\alpha$	<sup>239</sup> Np	<sup>144</sup> Ce	<sup>137</sup> Cs	<sup>131</sup> I	<sup>106</sup> Ru	<sup>103</sup> Ru	<sup>95</sup> Zr	<sup>95</sup> Nb	<sup>90</sup> Sr	<sup>89</sup> Sr
Farm 1 <sup>a</sup>											
12.10.57	135	nd	74	26		<74	<74			<110	
13.10.57	135	nd	110	30		<74	<74			37	
28.10.57			<83	233		<67	25	50	120		
Farm 2 <sup>b</sup>											
25.10.57			<33	696	3700	<67	<25	<25	<2	0.85	13

a Farm 1.6 km NW of Windscale : nd, not detected. Data from [8] and [33].

b Farm 3.2km SE of Windscale : Data from [33].

TABLE 4 Strontium and Iodine in Foodstuffs Other than Milk

Material	Peak Activity Recorded (Bq kg <sup>-1</sup> ) <sup>a</sup>		
	<sup>131</sup> I	<sup>90</sup> Sr	<sup>89</sup> Sr
Turnips (tubers)	260	100	195
(tops)		100	1330
Potatoes	150	87	113
Cabbage (outer leaves)	22,940	27	
Kale	15,926	60	600
Rape		60	
Carrots	220		
Swede (tubers)	110		
(tops)	27,750		
Lettuce	3,890		
Sprouts (outer leaves)	9,075		
River Water	1,036		
Lake Water	85		
Drinking Water	37		
Eggs (free range)	53,700		
Meat (beef)	3,370		
(mutton)	740		
(fish)	590		

a Data from [8] and [33]; various locations and dates (13-25 October 1957)

TABLE 5 Integrated atmospheric concentrations of activity and ERLs requiring sheltering of the public

nuclide	From measured $\beta$ conc. <sup>a</sup>	Integrated Activity <sub>b</sub> (Bq s m <sup>-3</sup> )			
		Modelled peak [21]	ERL2 <sup>b</sup>	[13] <sup>c</sup>	[13] modified <sup>d</sup>
<sup>3</sup> H	1.6E8-1.0E9	7.1E8	5.4E11	3.2E11	3.2E11
<sup>89</sup> Sr	1.4E5-9.2E5	7.1E5	3.6E9	2.2E9	-
<sup>90</sup> Sr	5.9E3-4.0E4	3.1E4	2.8E8	2.0E8	2.5E8
<sup>95</sup> Nb/ <sup>95</sup> Zr	4.1E5-2.7E6	2.1E6	2.8E9	1.7E9	-
<sup>99</sup> Mo	9.7E5-6.5E6	5.0E6	3.9E9	2.3E9	-
<sup>103</sup> Ru	1.1E6-7.2E6	5.6E6	5.6E9	3.4E9	-
<sup>106</sup> Ru	1.6E5-1.1E6	8.3E5	1.1E8	6.6E7	-
<sup>131</sup> I	2.7E7-1.5E8	8.4E7-1.4E8	3.9E8	2.3E8	3.4E8
<sup>132</sup> Te	2.7E7-1.5E8	8.4E7-1.4E8	2.2E9	1.3E9	-
<sup>137</sup> Cs	2.4E6-1.7E7	3.1E6-1.3E7	1.7E9	1.0E9	1.0E9
<sup>210</sup> Po <sup>a</sup>	2.4E5-1.6E6	1.2E6	7.2E6	4.3E6	1.8E7

a Based on total beta activity in air measured on-site and in the district [8], and assuming a release of 800 TBq <sup>131</sup>I; 800 TBq <sup>132</sup>Te; 90 TBq <sup>137</sup>Cs.

b Lower level of intervention only; based on 5 mSv CEDE or 50 mSv single organ dose (including skin). Upper levels are 5x these values.

c Lower level of intervention only; based on 3 mSv CEDE or 30 mSv skin or thyroid dose. Upper levels are 10x these values.

d Modified using dose effectance from ICRP [20 25] and assuming intervention based on 3 mSv CE.

e Values for ERL are calculated assuming that <sup>210</sup>Po was present as submicron particles of oxide material, which can be modelled as inhalation class D [5, 22]. If inhalation class W is assumed the following intervention levels would apply:

	Bq s m <sup>-3</sup>
ERL2	6.9E6
[13]	4.1E6
[13] (mod)	5.1E6



TABLE 6. Activity deposition to ground and comparison to DERLs

nuclide	measured (peak)	modelled (peak) <sup>a</sup>	Activity deposition <sub>b</sub> (Bq m <sup>-2</sup> )				milk ban <sup>c</sup>		green veg restrictn.	
			ERL2	[13] shelter	[13] mod	[13] ERL2	[13] ERL2	[13] mod	[13] ERL2	[13] mod
<sup>89</sup> Sr	1.3E4 [11]	1.4E3								
<sup>90</sup> Sr	2.5E4 [11]	6.2E1								
<sup>95</sup> Sr	1.1E4 [8]	2.1E3								
<sup>103</sup> Zr	7.8E3 [8]	1.1E4	6.5E6	3.9E6						
<sup>106</sup> Ru		1.6E3	1.2E7	7.2E6						
<sup>131</sup> I		8.4E5-1.4E6	2.5E7	1.5E7						
<sup>132</sup> I	1.1E6 [2]	8.4E5-1.4E6	2.0E7	1.2E7	1.8E7	1.3E7	1.8E7	4.2E6	8.4E5	
<sup>137</sup> Te		8.4E5-1.4E6	6.5E6	3.9E6				3.6E5	7.2E4	8.2E4
<sup>137</sup> Cs	9.1E3 [8]	9.6E3-4.0E4	9.0E6	5.4E6	5.6E6	3.5E4	7.0E3	1.3E5	1.3E5	1.3E5

a A deposition velocity (V<sub>g</sub>) of 1E-2 m s<sup>-1</sup> is assigned for <sup>131</sup>I and <sup>132</sup>Te (cf 3E-3 m s<sup>-1</sup> determined at Preston [7]; 2.5E-2 m s<sup>-1</sup> experimental value [30,31]). For all other nuclides a value of 2E-3 m s<sup>-1</sup> is assumed (~ 15%-20% of the <sup>131</sup>I value [7,30,31]). These values are subject to considerable uncertainty.

b Scaled from advice on evacuation; based on inhalation dose from resuspended material.

c Based on 5 mSv CEDE or 50 mSv thyroid dose (ERL2); 1 mSv CEDE or 10 mSv thyroid dose [13]; 1mSv CE [13] mod.

TABLE 7 Predicted activity deposition with downwind distance

Nuclide	Distance (km)					
	0.5	0.7	1.5	5.0	10	100
<sup>89</sup> Sr	3.3E2	9.6E2	1.4E3	7.7E2	3.3E2	1.4E2
<sup>90</sup> Sr	1.5E1	4.4E1	6.4E1	3.5E1	1.5E1	6.4E0
<sup>131</sup> I <sup>a</sup>	2.6E3	8.0E4	1.1E6	6.4E5	2.6E5	1.2E4
<sup>137</sup> Cs <sup>b</sup>	6.1E1	1.9E3	2.6E4	1.4E4	5.9E3	2.6E2

a. Based on a mean release estimate of 800 TBq.

b. Based on a best release estimate of 90 TBq.

TABLE 8 Estimated doses to thyroid from <sup>131</sup>I following the Windscale accident, 1957

Place	Distance from Windscale (km)	<sup>131</sup> I in air (Bq s m <sup>-3</sup> )	Thyroid dose (mSv)			
			Estimated Inhalation [18]		Measured Thyroid activity	
			Adult	Child	Adult	Child
Seascale	3	3.0E8 [4,18]	18	36	5 [3]	8 [3]
Drigg	6	3.3E8 [4,18]	21	42	14 [3]	39 [3]
Holmrook	7	3.0E8 [4,18]	18	36	14 [3]	
Ravenglass	9	2.6E8 [4,18]	16	32	18 [3]	122 [3]
Bootle	18	1.5E8 [4,18]	9	18	14 [3]	60 [3]
Millom	31	7.4E7 [4,18]	5	10	4 [3]	
Ulverston	37	3.7E7 [4,18]	2	4	5 [3]	44 [3]
Barrow	38	3.7E7 [4,18]	2	4	3 [3]	
Leeds	150	1.1E6 [2,18]	0.07		1.5 [18,27]	
London	400	1.3E6 [3,26]	0.08		0.4 [28]	0.8 [28]

TABLE 9 Whole-body <sup>137</sup>Cs in subjects from the Windscale area [33]

Date	Number of subjects	Bq <sup>137</sup> Cs per g K
Mar - Sept 1957	4	3.6 ± 0.7
Jan 1958	3	11.8 ± 1.4
Mar 1958	1	16.6
Apr 1958	1	7.7
May 1958	1	6.8
July 1958	2	10.1

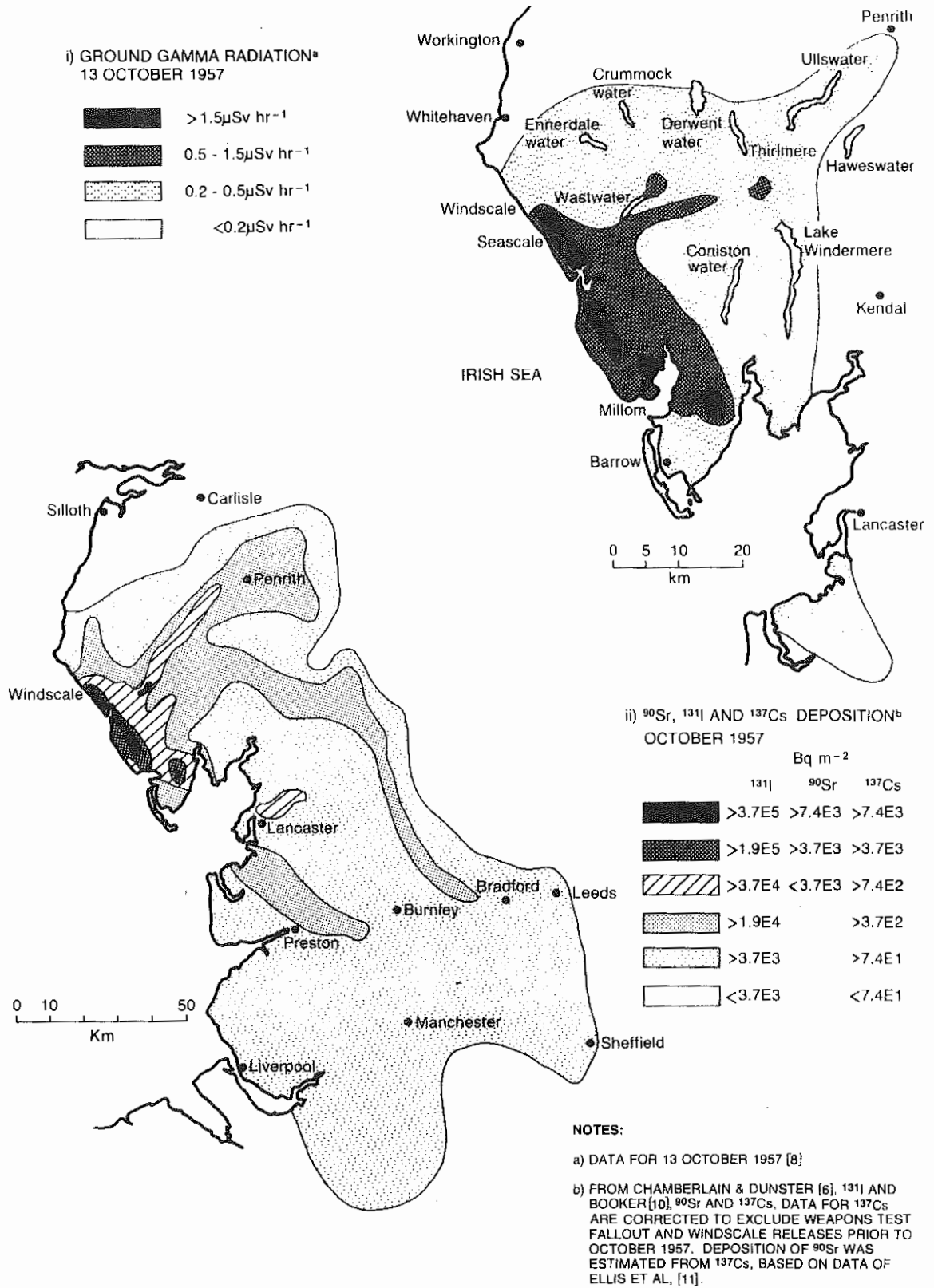
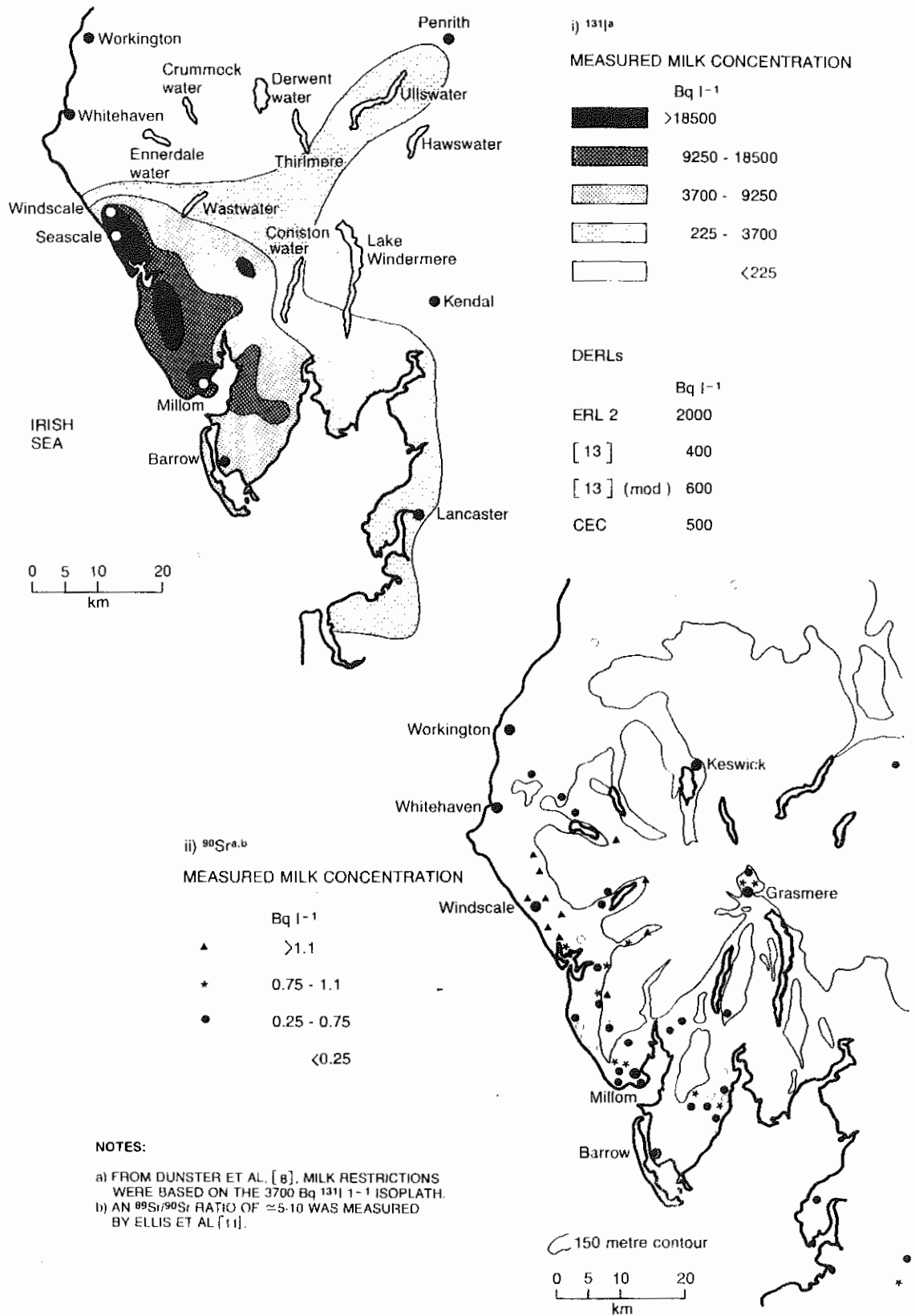


Figure 1 Ground Gamma Radiation and Activity Deposition in North West England following the Windscale Accident, 1957



**Figure 2 Maximum Activity Concentrations in Milk and Extent of Restrictions Instigated following the Windscale Accident, 1957**

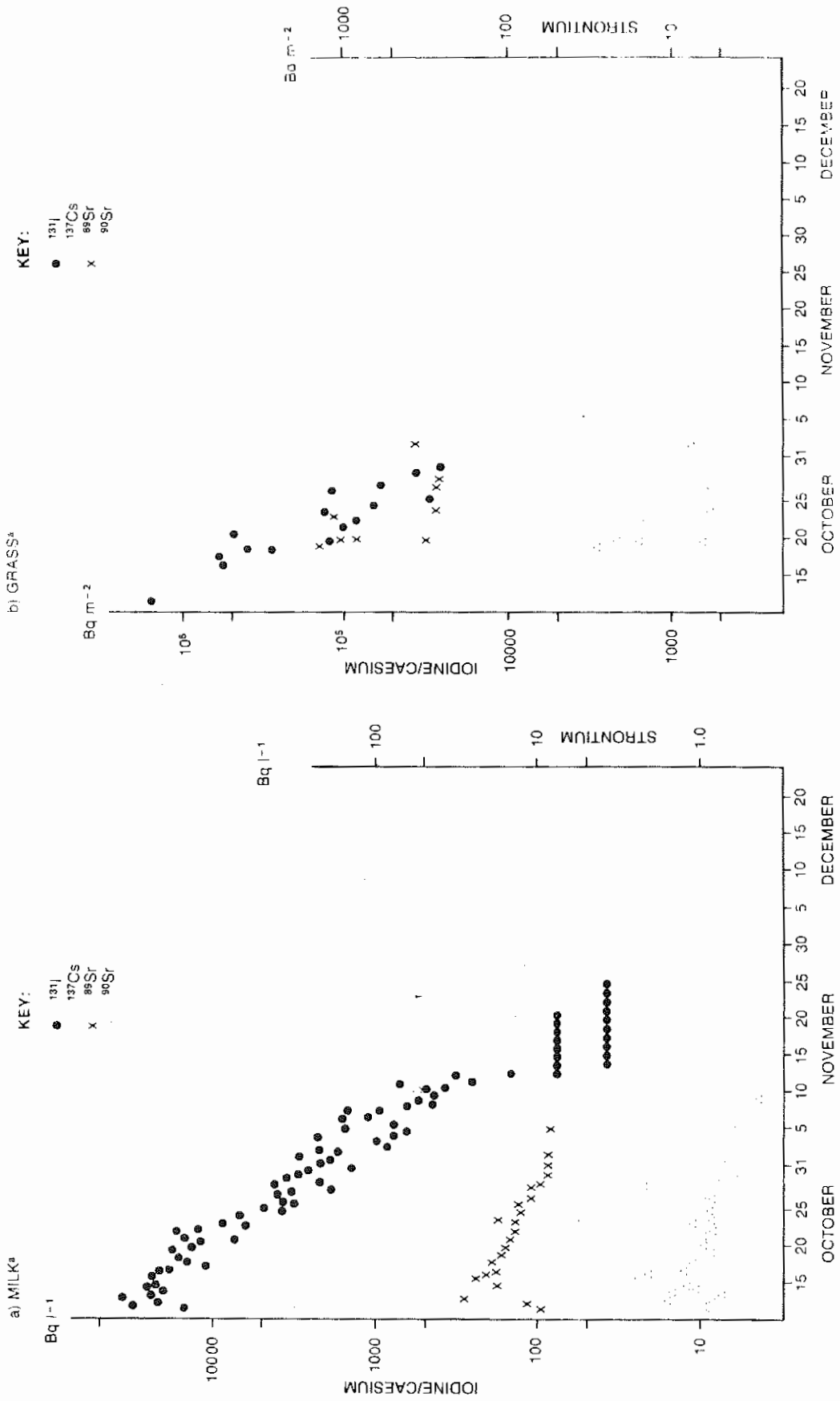
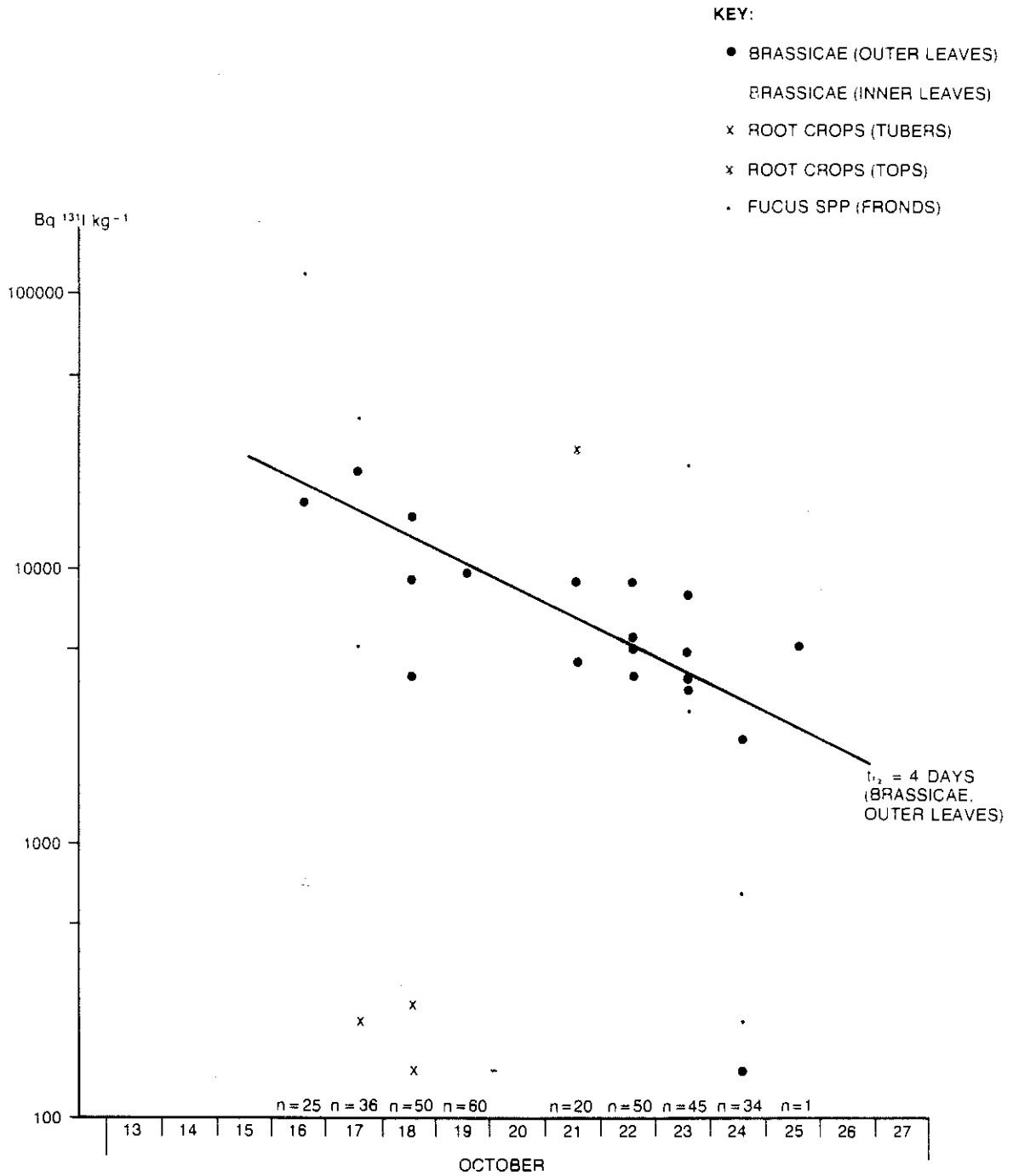
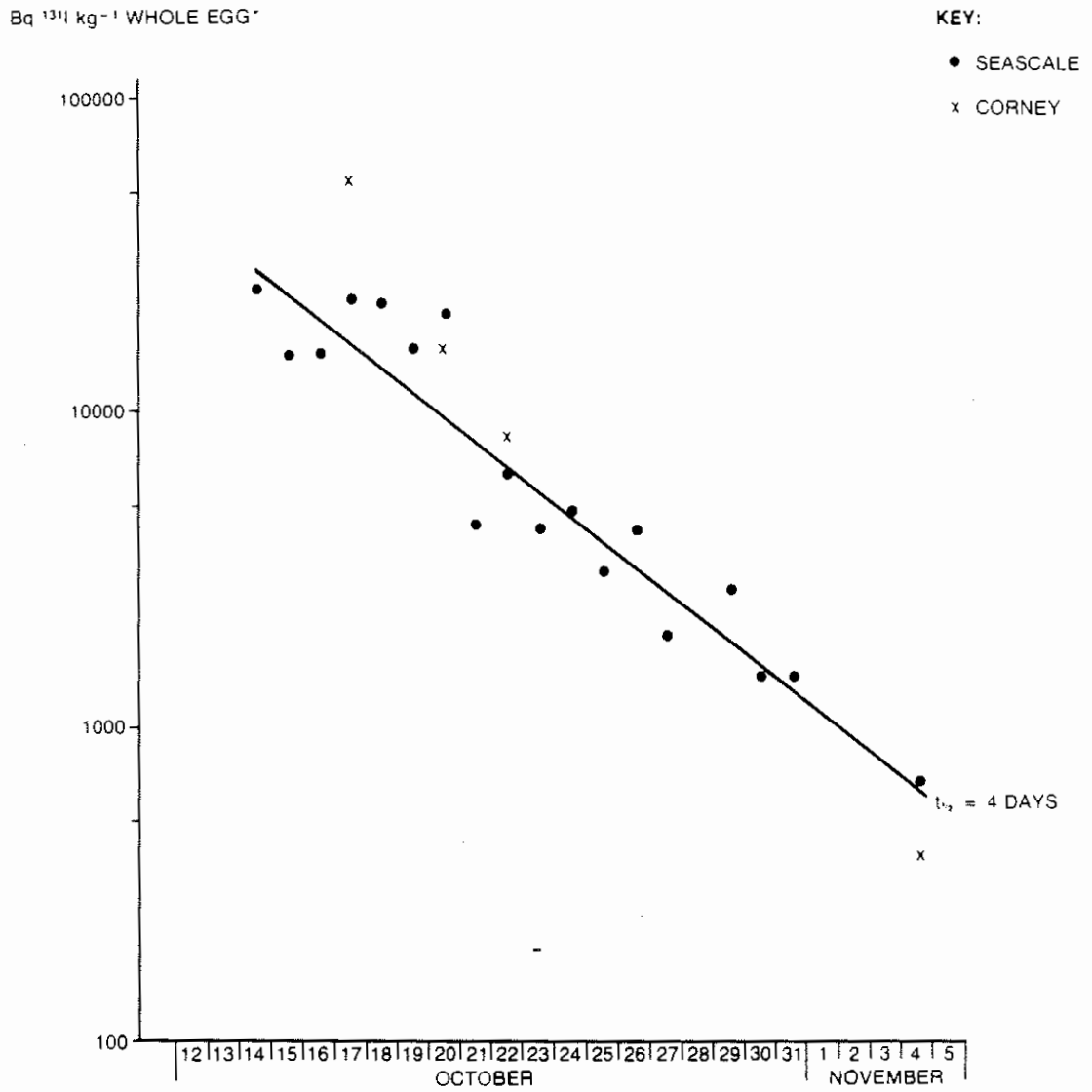


Figure 3 Loss of Activity from Milk and Grass October - December 1957



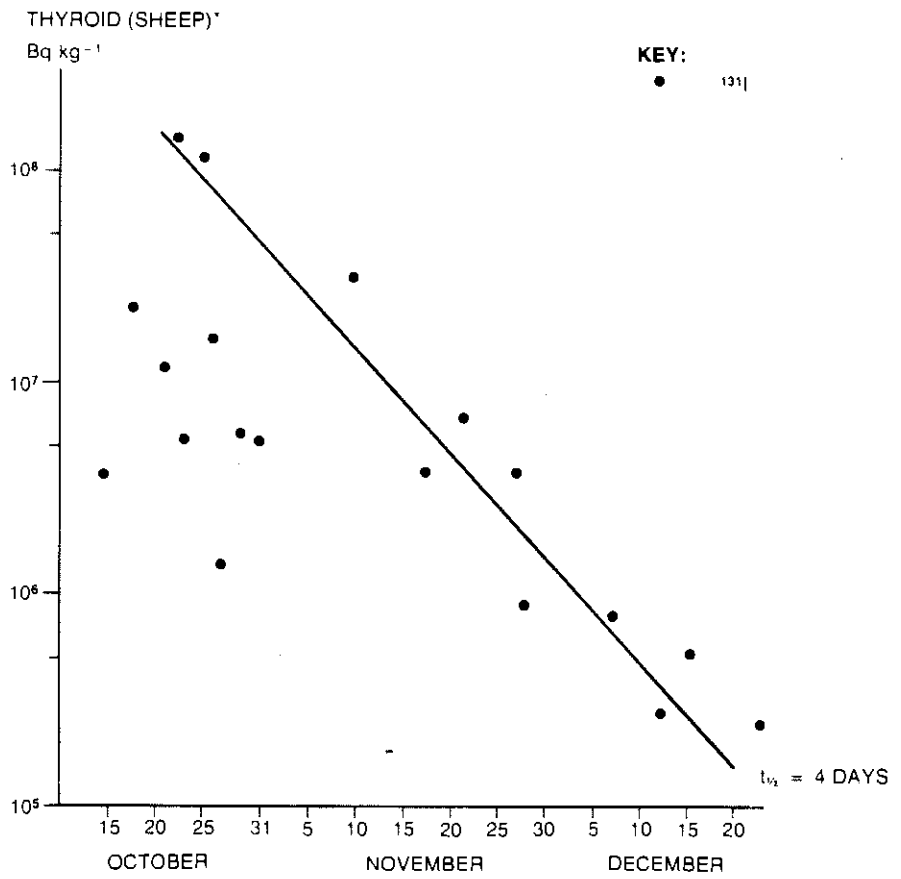
APPROXIMATE AREA OF VEGETABLE SAMPLING:  
 WINDSCALE TO MILLOM (BASED ON PEAK LEVELS)  
 SEAWEED FROM SEASCALE TO RAVENGLASS  
 n = TOTAL NUMBER OF SAMPLES FOR EACH DAY

**Figure 4 Upper Levels of  $^{131}\text{I}$  in Vegetables and Seaweed South and East of Windscale, October 1957**



\* DERIVED FROM ORIGINAL DATA [33] ASSUMING A STANDARD WHOLE EGG WEIGHT OF 60g.

**Figure 5 Concentration of  $^{131}\text{I}$  in Free Range Eggs from the South and East of Windscale, October-November 1957**



\* SHEEP FROM WHITEHAVEN ABATTOIR  
ERROR BARS OMITTED FOR CLARITY.  
EFFECTIVE LOSS RATE BASED  
ON INITIAL MAXIMA VALUES.

Figure 6 Loss of Iodine from Sheep Thyroids October - December 1957





**Windscale and Chernobyl:  
A Comparison of the  
Effectiveness of  
Countermeasures Taken in the  
UK**

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Abstract

The accidents at Windscale in 1957 and at Chernobyl in 1986 both gave rise to contamination levels in the UK for which countermeasures were considered necessary. Restrictions on the consumption of milk were imposed following the Windscale Fire, with the aim of reducing exposure to iodine-131. Following Chernobyl, members of the public were advised to avoid drinking rain water, and restrictions on the sale and distribution of sheep were imposed to reduce doses from caesium-137. The countermeasures which followed the Windscale Fire reduced the effective dose to the most exposed individuals by 80% and the corresponding reduction in the collective effective dose to the UK population was around 12%. The countermeasures imposed following Chernobyl resulted in a reduction in the maximum individual effective dose of 25% and the collective dose of around 3%. These differences are discussed. The countermeasures which followed each accident would have been more extensive had they been based on the radionuclide levels given in the present European Community regulations, but large additional dose savings would not have been achieved.

## Introduction

This paper provides a discussion of the different characteristics of the releases from the Windscale and Chernobyl accidents and their effect on the collective and individual doses received by the population of the UK. The countermeasures imposed as a result of these accidents are described and their effectiveness in reducing doses are assessed.

The effectiveness of these countermeasures in reducing doses is compared with measures based on current emergency intervention criteria and with each other. The aim was not to criticize what was done with the benefit of hindsight, but to determine whether these comparisons have implications for emergency planning in the future.

## A Description of Effects of the Accidents in the UK

### Windscale

The Windscale Fire resulted in a release of radioactive material into the environment, which was dispersed over parts of England, Wales and northern Europe. The main release continued from noon on 10th October until noon on 11th October 1957. The estimated quantities of radionuclides released<sup>(1,2)</sup> are presented in Table 1.

In the period directly following the accident, iodine-131 was identified as the radionuclide of immediate concern. A ban on the distribution of milk was imposed with the aim of reducing exposure to this radionuclide. Restrictions were introduced if the concentration of iodine-131 in milk exceeded  $3700 \text{ Bq l}^{-1}$  with effect from 11th October. The intervention level was based<sup>(3)</sup> on the decision to control the consumption of milk to avoid doses of greater than 200 mSv to a child's thyroid, which was a factor of ten below the level at which there was known to be evidence for the induction of thyroid cancer in children.

The restrictions on milk which followed the accident continued for a period of 25 days within most of the affected region, and up to 44 days in the area close to the site. The maximum area restricted was 520 square kilometres. The levels of iodine-131 in foods other than milk were also monitored<sup>(4)</sup>, and were estimated to give rise to doses below the criterion described above.

### Chernobyl

The Chernobyl accident occurred on 26th April 1986. The rate of the release of activity had reached relatively low levels by 6th May, by which time the main contaminated air mass had passed over southern Germany, Italy, Greece, eastern Europe and over the British Isles<sup>(5)</sup>. The release estimates are presented in Table 1.

Iodine-131 and caesium-137 were identified as the radionuclides of most importance. On 5th May 1986, three days after the plume from Chernobyl reached the South East coast of England, advice was issued to avoid drinking undiluted rain water, with the aim of reducing exposure to iodine-131. This advice was based on predetermined levels published by NRPB<sup>(6)</sup>. Later, restrictions were imposed to control exposure to caesium-137. In the middle of June, restrictions on the movement and slaughter of sheep containing total radiocaesium levels in excess of 1000 Bq kg<sup>-1</sup> were imposed in areas of Cumbria, North Wales and Scotland. These restrictions are still continuing, four years after the accident, although only 25% of the area originally affected continues to be restricted (August 1990). This represents a total of 647,000 sheep and 757 holdings<sup>(7)</sup>.

### The Assessment Methodology

The conclusions drawn in this paper are largely based on the radiological assessments of the two accidents which have been performed at the NRPB<sup>(2,5)</sup>. It is recognised that the models used in these assessments are likely to underestimate the contribution of livestock grazed on upland pasture to the collective effective dose. However, in general the individual doses from ingestion in

the areas of high deposition are likely to be overestimated because of the assumption that food is consumed locally. In the UK foods are distributed over wide areas and individuals in an area of high deposition are unlikely to consume only locally produced foods. However, these assessments still provide a useful basis for comparison and the relative effectiveness of each countermeasure strategy has been estimated in terms of the predicted individual and collective dose reduction.

The main aim of each of the assessments was to estimate the impact of the accidents on the population as a whole and, as a result, emphasis was placed on the calculation of the collective effective and thyroid dose equivalent commitments. Representative individual doses were also calculated. For adults, the dosimetric techniques used in both assessments were consistent with those recommended in ICRP Publication 30<sup>(8)</sup>; for infants and children, doses were calculated taking account of differences in body size and biokinetic parameters.

For the assessment of the Windscale Fire, a detailed spatial matrix of the distribution of activity was constructed. These data were applied to a radial grid, centred on Windscale, with distance bands increasing in size with increasing distance from the site. The population distribution was also arranged in this way and the calculations of collective dose were performed with this resolution. For the Chernobyl assessment, the UK was divided into seven regions which reflected the distribution of the deposition of activity, and which largely followed the pattern of rainfall during the passage of the plume. Collective doses were calculated on the basis of the population of the region, and activities in environmental materials which were assumed to be representative of the whole region.

#### The Individual Doses

Following the Windscale Fire, various estimates of the typical doses received by the local population were published<sup>(9,10,11)</sup>. Potential doses were predicted to be in the range of 5 to 20 mSv

for adults and 10 to 60 mSv for children. The maximum measured thyroid dose equivalent to a child was 160 mSv<sup>(2)</sup>; the corresponding maximum effective dose would have been 9 mSv taking account of all pathways and nuclides. The NRPB estimates of committed effective dose equivalent<sup>(2)</sup> are presented in Table 2. These results indicate that the restrictions on milk would have reduced the effective dose equivalents to the most exposed infants by around 80%, and to the most exposed adults by around 50%.

The effective doses to critical groups of adults and infants arising from the Chernobyl accident are also given in Table 2. The restrictions on the consumption of lamb are estimated to have reduced the dose to a critical adult by 25% and to an infant by about 10%. Drinking rain water had the potential of increasing the predicted dose to a critical individual by around 35%. However, since it is not certain whether this pathway would have affected any individual, this dose and the dose saving associated with the advice not to drink rain water, have not been included in Table 2 or the discussions of the effectiveness of countermeasures which follow.

#### The Collective Doses

The collective effective and collective thyroid dose equivalents to the UK population which arose from each accident are presented in Table 3. The effective dose from Chernobyl is 50% greater than that from Windscale, while the thyroid dose is under half of the Windscale value. This demonstrates the greater relative importance of the exposure to iodine-131 following the Windscale Fire.

The contributions of each pathway to the collective effective doses are given in Table 4. The inhalation pathway was dominant following the Windscale accident, while contributing only 3% to the collective effective dose from Chernobyl. This is a consequence of the the greater relative importance of iodine-131 and the release of polonium-210 during the Windscale Fire. It

also reflects the fact that the Windscale accident occurred in the UK and that the inhalation pathway would be expected to be significant immediately following the accident in the area surrounding the site. Following the Chernobyl accident, caesium-137 was relatively more important, as reflected in the importance of the external exposure pathway and foods other than milk to the collective effective dose equivalent.

#### A Comparison of the Effectiveness of Countermeasures

The restriction on the distribution of milk following the fire at Windscale was more effective in reducing both individual and collective doses than the countermeasures imposed following Chernobyl. This is largely a result of the differing nature of the accidents. Following Windscale, the effects of the accident were fairly localised. It was possible to implement countermeasures to reduce potentially high individual doses which also formed a significant fraction of the collective dose; around 12% of the total dose was estimated to have arisen within 50 km of the Windscale site<sup>(2)</sup>. Following Chernobyl, large areas of the UK were affected. The effects of the accident were widespread and were not restricted to a particular set of individuals, although higher levels of deposition occurred in parts of Cumbria, Scotland and North Wales.

There have been significant developments in emergency planning since the accident at Windscale occurred. Pre-defined intervention criteria have now been established. In the UK, the NRPB have derived emergency reference levels of dose<sup>(12,13)</sup> for use in the early stages of an accident. In addition, following the accident at Chernobyl, the Council of European Communities (Council) have issued a series of directives<sup>(14,15,16)</sup> on the maximum permissible levels of various radionuclides in foods. It is interesting to explore the dose reductions which could have been achieved if present criteria had been adopted.

The Council level for iodine-131 in milk is 500 Bq l<sup>-1</sup>. The use of this level following the Windscale accident would have



resulted in milk restrictions extending to a distance of almost twice that actually affected. It has not been possible to make precise estimates of collective dose reductions from the data included in reference 2. The use of Council intervention levels, would have had the effect of further reducing maximum individual doses by a factor of two. However, since this means that the total reduction in dose would be 90%, rather than the 80% achieved by the 1957 bans, this does not reflect a very significant change. In addition, the measurements of iodine-131 in green vegetables<sup>(4)</sup> exceeded Council criteria. However, the amount of vegetables grown during October 1957 is likely to have been limited, and this countermeasure is unlikely to have been very effective in reducing doses as a result.

Following the Windscale Fire, inhalation was the most important contributor to collective effective dose. However, individual inhalation doses are unlikely to have exceeded those at which current, and proposed, NRPB recommendations<sup>(12,13)</sup> would indicate that evacuation would be appropriate. The dose to the thyroid are likely to have been lower than the level at which the administration of stable iodine is likely to have been justified. However, since individual doses may have approached the lower ERL for sheltering, to be included in Reference 13, it is likely that if this accident had occurred under present conditions, precautionary sheltering and administration of iodine may have taken place. It is difficult to predict how much effect these measures would have had.

Following the Chernobyl accident the most important routes by which the population were exposed have been identified as the ingestion of milk and other foods and external irradiation from deposited activity. The level of caesium-137 at which sheep have been restricted is only slightly more restrictive than the level which would now be adopted and so the alternative criterion would have had little effect on either the individual or collective doses. The consumption of milk gave rise to a significant proportion of the collective effective dose. The peak concentration of iodine-131 in milk exceeded the Council levels

in the areas of maximum deposition<sup>(5)</sup>. It has not been possible to predict the reduction in collective dose which would have resulted from application of Council levels. However, restriction at these levels may have reduced the dose to the most exposed individuals by around 6%.

The countermeasures most appropriate to reduce doses from external irradiation are decontamination and the interdiction of land. Current ICRP advice<sup>(17)</sup> would suggest that relocation would be appropriate where the projected whole body doses in the first year were in the range of 50 to 500 mSv. The deposition levels in the UK did not give rise to levels in this range and these countermeasures would not have been appropriate.

### Conclusions

The restrictions on milk which followed the Windscale accident resulted in a reduction in the effective dose equivalent to the most exposed individual of around 80% and continued for a maximum period of 44 days. Following the Chernobyl accident, the application of restrictions on the consumption of lamb resulted in a maximum reduction in individual effective dose of the order of 25%. These restrictions are continuing four years after the accident. The estimated reductions in the collective effective dose to the population of the UK are 12% and 3% for the Windscale and Chernobyl accidents respectively. The application of current countermeasure criteria are likely to have had a limited effect on the levels of dose received.

The countermeasures imposed in the UK following the Windscale Fire were more effective in reducing individual and collective doses than those which followed the arrival of the plume from Chernobyl. They were also shorter in duration. The duration of the restrictions is a result of the different radionuclides of importance and the shorter half life of iodine-131 relative to caesium-137. The differences in dose reduction are largely the effect of the differing nature of the two accidents. The Chernobyl accident occurred overseas; its plume passed over large

areas of the UK and a large population were exposed to low doses. The Windscale Fire was more localised in its effect and action was taken to reduce potentially high doses to a small number of people. These conclusions suggest that it may be worthwhile to review the implications of applying countermeasures to reduce collective exposure, particularly for overseas accidents. Decision aiding tools, such as RADE-AID<sup>(18)</sup> would help in this process.

The continuation of restrictions on sheep following the Chernobyl accident result from difficulties in deciding on criteria for withdrawing countermeasures. Social and political judgements have an impact on these decisions and, as a result, a measure of the overall effectiveness of a countermeasure should include its cost and impact on society. An analysis of this sort is beyond the scope of this paper. However, it is worth noting that a system such as RADE-AID<sup>(18)</sup> may be used to analyse these factors, and could be applied to decisions on the withdrawal of countermeasures on due course.

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Table 1

Quantities of radionuclides released from the Windscale and  
Chernobyl accidents (TBq)<sup>(9)</sup>

Radionuclide	Windscale	Chernobyl
<sup>90</sup> Sr	0.07	8,000
<sup>106</sup> Ru	3	58,000
<sup>131</sup> I	740	260,000
<sup>132</sup> Te	440	48,000
<sup>133</sup> Xe	12,000	1,700,000
<sup>137</sup> Cs	22	38,000
<sup>210</sup> Po	8.8	-
<sup>239</sup> Pu	0.0016	25

Table 2

Individual committed effective dose equivalents following the Windscale and Chernobyl accidents (mSv)

	Windscale <sup>1</sup>		Chernobyl <sup>2,3</sup>	
	Adult	Infant	Adult	Infant
No food restrictions	8.2	36	1.1	0.9
With food restrictions	4.2	7.1	0.8	0.8

Notes:

1. Estimated at the point of maximum deposition density of iodine-131.
2. Representative of doses in the Cumbria and S.W. Scotland areas, where levels of deposition were highest.
3. These data do not include the drinking of rainwater pathway. This pathway had the potential of increasing the dose to critical individuals by as much as 0.3 mSv, a dose which would have been reduced by the advice to avoid drinking rain water.

Table 3

Collective doses to the UK population arising from the Windscale and Chernobyl accidents<sup>1</sup> (man Sv)

	Effective	Thyroid
Windscale	2 10 <sup>3</sup>	2.6 10 <sup>4</sup>
Chernobyl	3 10 <sup>3</sup>	1.2 10 <sup>4</sup>

Notes:

1. These estimates include an allowance for the countermeasures imposed. In the absence of countermeasures, the collective effective dose from Windscale would be about 12% higher and that from Chernobyl around 3% higher.

Table 4

Contributions of each pathway to the collective effective dose equivalent commitment (%)

	Windscale	Chernobyl
Inhalation	50	3
Milk	30	37
other foods	10	27
external	10	33





# **Comparison of the Countermeasures Taken for the Recovery of Rural Areas Contaminated by Radioactivity**

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## ABSTRACT

In 1989, an evaluation of the countermeasures taken for the recovery of rural areas was made. This evaluation was based on the analysis of the available information about eleven different scenarios where, in the past, an accident has taken place or a nuclear test has been done, and also on the information about laboratory and other field researches on the application of the countermeasures for land recovery.

In this evaluation, the Windscale (UK) and Chernobyl (USSR) accidents were considered too. At the end of this evaluation, important information was available about the accident that took place in Kyshtym (USSR) and, now, it is possible to extend this evaluation and to make an assessment of the countermeasures taken, especially in the three areas where a major accident has occurred (Windscale, Chernobyl and Kyshtym).

This work intends to compare the kind of countermeasures taken for the recovery of rural areas contaminated by radioactivity and their effectiveness. Considering the significance of the large volume of waste generated following the implementation of the countermeasures (i.e. removal of crops or other vegetation and of superficial soils), we have taken steps to gather all relevant information in order to include it in our evaluation and to indicate, also in this work, the effort required for the management of the generated waste in the three accidents mentioned above.

## INTRODUCTION

An evaluation of the countermeasures taken for the recovery of rural areas contaminated by radioactivity was made in 1989 which constitutes a part of the Spanish contribution to the CEC Post-Chernobyl Action [1,2].

This evaluation was based, on the one hand, on the analysis of the available information about eleven different scenarios where, in the past, a release of radioactivity took place impacting, with more or less strength, in some rural areas and, on the other hand, it was based on the information about laboratory and other field researches on the application of the countermeasures for the recovery of contaminated land. The scenarios reviewed were:

- Accidents: Windscale (UK, 1957), Rocky Flats Plant (USA, 1958) Palomares (Spain, 1966), Thule (Denmark, 1968) and Chernobyl (USSR, 1986).
- Nuclear tests: Bikini Atoll (1946-1958), Enewetak Atoll (1948-1958) and Nevada Test Site (1951-1975) (USA).
- Others: Hiroshima and Nagasaki (Japan, 1945) and Los Alamos (USA, 1942-1965).

The major accidents of Windscale and Chernobyl are included in the evaluation above mentioned. About the accident which took place in the southern Urals (Kyshtym, USSR), enough information was only available when the evaluation of those eleven scenarios had already been finished. Nowadays, it is possible to include, also, this last case and the present paper tries to compare the countermeasures taken in the three accidents (Kyshtym, Windscale and Chernobyl), considered with a major impact in the rural areas, taking into account the kind of contamination produced and the different countermeasures applied, stressing their advantages and disadvantages with respect to their efficiency and feasibility.

## THE CONTAMINATION

First of all, a brief description of the three accidents and the levels of contamination originated by the release of radionuclides in these cases are presented:

**Kyshtym:** In September 1957,  $7.4 \times 10^{16}$  Bq of radioactivity was released from a military facility, where nuclear weapons were produced. The accident was due to a break down in the refrigeration system used in the

concrete tanks which contained residue of highly active nitroacetates. Sr-90 was the main contaminant released and three provinces of the USSR were affected. 15000 Km<sup>2</sup> reached a level of contamination higher than  $3.7 \times 10^9$  Bq/Km<sup>2</sup>, 1000 Km<sup>2</sup> of them more than  $7.4 \times 10^{10}$  Bq/Km<sup>2</sup> and 120 Km<sup>2</sup> about  $3.7 \times 10^{12}$  Bq/Km<sup>2</sup> [3].

**Windscale:** In October 1957, one of the two natural uranium reactors, graphite moderated, was affected by fire during a deliberate release of stored Wigner energy. The fire was quenched by pumping water on to the core during 20 hours; during this period fission products activity (about  $12 \times 10^{14}$  Bq) was discharged to the atmosphere. The two major radioisotopes involved were I-131 ( $7.4 \times 10^{14}$  Bq) and Te-132 ( $4.4 \times 10^{14}$  Bq) and less amounts of Cs-137 ( $2.2 \times 10^{13}$  Bq), Sr-89 ( $3 \times 10^{12}$  Bq) and Sr-90 ( $7.4 \times 10^{10}$  Bq). The higher value of radiation level measured on the ground was 400 times superior to the normal background and an area of approximately 520 Km<sup>2</sup> was established for milk consumption restriction, during 6 weeks [4].

**Chernobyl:** In April 1986, an explosion happened during a test being carried out on a turbogenerator, at the time of a normal scheduled shutdown of the reactor (one of the four graphite moderated pressure tube type reactors). During 9 days radioactive products of fission (estimated up to  $2 \times 10^{18}$  Bq) were released from the fuel during the accident, not taking into account Xe and Kr. The main radionuclides were I-131 ( $2.6 \times 10^{17}$  Bq), Cs-137 ( $3.8 \times 10^{16}$  Bq) and Sr-90 ( $0.8 \times 10^{16}$  Bq). About half of this radioactivity remained in a radius of 30 km around the nuclear power plant, but this accident also affected a great area of the USSR, reaching some part of Europe too [5].

## THE COUNTERMEASURES

The main countermeasures applied in the three scenarios were:

### Kyshtym

- Ploughing of 20000 Ha in a traditional way.
- Deep ploughing (more than 50 cm in depth) in an area of 6200 Ha.
- 106000 Ha of agricultural production was blocked for a minimum of 4 years, giving way to it progressively afterwards.
- Setting up state farms for the control of the animal feedstuff contamination and the production of foods with low radioactivity.
- Restriction of food consumption like meat and milk.
- Limitation on the land use where the Sr-90 contamination was

superior to  $7.4 \times 10^{10}$  Bq/km<sup>2</sup>.

#### **Windscale**

- Restriction of milk consumption, where the contamination of I-131 exceeded  $3.7 \times 10^3$  Bq/l, since the third day after the accident until six weeks later.

#### **Chernobyl**

- Removal of vegetation (i.e. 400 Ha of forest around the nuclear plant was felled and buried).
- Removal of surface soil.
- Addition of a layer of non contaminated soil from far away areas.
- Deep ploughing and application of chemical products.
- Application of lime, zeolites, clay or other soil amendments and fertilizers and afterwards a normal ploughing.
- Application of fixatives (i.e. special foams).
- Use of contaminated products, after their storage, as food for the cattle.
- Use of contaminated products as raw material in industrial processes.
- Restriction in the consumption of contaminated food.
- Prohibition on the use of the land for agricultural production where Cs-137 contamination was higher than  $1.48 \times 10^{12}$  Bq/km<sup>2</sup>.

### **COMPARISON OF THE COUNTERMEASURES**

A comparison of the main countermeasures applied in the three scenarios is intended by the analysis of their advantages and disadvantages with respect to their efficiency and feasibility. The outstanding information about the three accidents is gathered in table 1.

#### **Removal of crops and vegetation**

Concrete information about the application of this countermeasure is referred only for the case of Chernobyl and in relation to natural ecosystems, composed mainly by coniferous forest. In fact, the decontamination works of the 400 Ha of "red forest", where trees and shrubs were felled and covered by soil, was obtained an average level of gamma-radiation 35-40 times lower than the initial one and, in some more contaminated spots, this rate became up to 1000 times lower [6].

It seems that devegetation work was made, also, after the accident of

Kyshtym, between 1959 and 1963, in a heavily contaminated zone of the Chelyabinsk region where pine, fir and other conifer species died from chronic internal and external radiation [7], but this information is not very clear.

However, in rural areas an average of 50% of contamination could be reduced by this action, but its effectiveness depends on the kind and density of vegetative cover, on the growing state of crops and on the kind of deposition of the contamination. Decontamination factors, also, depend on the period of time between contamination and cutting-taking away, that is why devegetation must start after deposition as soon as possible.

The main advantage is that the soil layer and its productivity are not disturbed, but the application of this countermeasure could create a big amount of waste, produce resuspension of the contaminants during the operations and involve an environmental impact.

In the case of forests, devegetation needs the use of special machinery and decades will be required for revegetation; nevertheless, it seems to be very efficacious.

#### **Removal of surface soil**

This decontamination method was applied in the near area of Chernobyl plant by mechanical means. The most effective procedure was scraping the contaminated soil by bulldozers and, also, by other building machinery (loaders, scrapers and graders) [8]. But clear information about the application of this technique is not available in relation to the agricultural lands, in areas far from Chernobyl or far from the location of the other two accidents considered.

However, the complete removal of soil layer (approximately 10 cm) seems nearly 100% effective, not excessively costly and can be done quickly. Its practicability depends on the type of soil and the topography of the area; that could impose conditions related to the kind of motorized equipment to be used.

The main disadvantages involve the use of special machinery, generate a loss of fertility, create a big amount of waste, produce resuspension and have a very important ecological impact (i.e. native animals would be lost although they would be replaced in time from adjoining areas).

After this remedial action was applied, fertilizers, organic matter or clean soil might be required.

The revegetation could be extremely difficult, but would become

easier if, during the soil removal, a part of the vegetation remains in situ.

In areas where the remaining of vegetation is interesting to be kept and the top soil is easily loosened, the use of vacuum-type cleaners could be practicable. In this case the total volume of the collected radioactive material can be reduced greatly in comparison with conventional earthmoving techniques.

#### **Normal and deep ploughing**

In Kyshtym, 20000 Ha were ploughed in a traditional way from 1958 to 1959 and during the two years later 6200 Ha were deep ploughed in depth superior to 50 cm. By deep ploughing (approximately 1 m) the gamma-dose was reduced by a factor of 10, it was an effective method for soil treatment [3].

In Chernobyl a range of agrotechnical measures was implemented in contaminated regions of the Ukrainian SSR, Byelorussian SSR and Russian SFSR in 1986 and 1987, including deep ploughing. By simultaneous use of agrochemical products in the first year following the implementation, these measures have reduced the levels of radioactivity in agricultural products by a factor of 1.5-3 [9]. However, information about the surface ploughed and the kind of plough used is not yet available.

The normal ploughing can be made by conventional farm machinery and procedures, and it is one of the cheapest methods. However, in the case of deep ploughing, special machinery is needed and the fertility of the soil can be altered.

The main disadvantage of ploughing is that the radionuclides, although being greatly diluted, are still available to be uptaken by the plants.

#### **Application of fertilizers and others amendments**

Large quantities of inorganic fertilizers were applied to hundreds of thousands Ha of contaminated land in Chernobyl. Steps were taken to reduce the transfer of radioactive substances from the soil to the crops by applying lime, phosphoric and potassic fertilizers and certain sorbents (zeolite) to the soil [9].

The reduction of radionuclides uptake could be achieved by providing cations competition. The chemical similarity between Sr and Ca means that the use of calcic amendments could reduce the radiostrontium uptake by the



crops. That could probably be the reason of the use of this kind of fertilizers in the case of the recovery activities in the state farms at Kyshtym [3].

The main advantage of this treatment is the good feasibility in comparison with other actions for contaminated land recovery. Also, it could allow a selective uptake reduction of radionuclides and corrects the fertility and acidity of the soil.

However, none of the soil amendments are highly effective at the same time for all the radionuclides. Their use depends on the type of soils, on the kind of radioactive contamination and on the availability in situ of large amount of the amendment to be applied.

#### **Application of fixatives**

After the Chernobyl accident, special polymers foams were applied to fixate the radionuclides onto the contaminated surfaces [8] and, possibly, onto the contaminated agricultural land. The use of this treatment was probably made, also, in Kyshtym, but clear information about the application of fixatives in rural areas is not available.

The application of foam, asphalt, road oils, etc. stops the resuspension and the spreading of contaminants, and could increase the effectiveness of the later operations which involve the removal of the contaminated surface, i.e. the application of 5 cm of polyurethane foam can absorb almost 85% of the activity. This method could be used as an initial step for further decontamination operations and recent investigations (CEA-France) have produced promising results.

When fixatives are just applied, without removal, no wastes are generated, it could last many years and their usage may be less expensive than other treatments. However, its main disadvantages, related to agricultural areas, are that the land is almost useless during the period of cover, water would not penetrate into the soils and some drainage is needed.

#### **Restrictions of land use and food consumption**

In 1958, 59000 Ha were taken out of the normal agricultural use in the Chelyabinsk region and 47000 Ha in Sverdlovsk region as consequence of the Kyshtym accident. At the beginning of 1961 these soils had been progressively rehabilitated for agricultural production by the creation of nine state farms and in 1989, 40000 Ha of the land restricted in

Chelyabinsk came back for normal agricultural uses [3].

The goal of these specialized farms was the control of the animal feedstuff contamination and the production of meat and milk with minimum concentration of Sr-90. The efficiency of this agricultural system, evaluated in the basis of the reduction of Sr-90 concentration into the products made in these farms, is expressed with reduction factors of 2-7 for meat and 3-4 for milk.

In the case of Windscale, at the time of the accident there was not prescribed emergency reference level applicable to milk contamination with radioiodine and an emergency value was derived, restricting the distribution of milk from those areas where the contamination of I-131 exceeded  $3.7 \times 10^3$  Bq per litre. The zone within which this milk ban was applied was extended in an area of  $520 \text{ Km}^2$ . During the period from the 3rd day to the 6th week after the accident, farmers whose milk was restricted from human consumption were paid compensation of a total amount of about 60000 pounds [10].

In relation to the Chernobyl accident, where Cs-137 contamination was higher than  $148 \times 10^{10}$  Bq/km<sup>2</sup>, a prohibition was imposed on the use of land for agricultural production. It has been recommended that such land should be transferred to the Soviet State Forest Administration for organization of a special forest preserve. In zones with a lower level of contamination, agricultural production is permitted with various limitations and recommended procedures, depending on the nature and level of contamination. These procedures include a change in crop structure and type of animal husbandry, and implementation of the agrotechnical and agrochemical measures above mentioned [11].

### **Food processing**

The information available about this countermeasure, related to the accident of Kyshtym, is only the creation of state farms where a controlled production of feedstuff, meat and milk was made [3] but, it is not clear if some activities in food processing were started at the same time.

There is no information about milk processing in the case of Windscale.

After the accident of Chernobyl, depending on the level of radioactive contamination of foodstuffs, changes were made in methods of storage, processing and ways of utilization [11].

Nowadays, the removal of radionuclides from contaminated products and crops is possible by special treatments in industrial processing. Ion-exchange treatment of milk, fruit juices, purées and vegetable could reduce its radionuclide content perhaps more effectively than other decontamination treatments. However, in this example, some essential nutrients are, also, removed.

#### GENERATED WASTE

In the case of the Windscale, a direct information, about the volume of milk poured into the Irish sea [12], is actually available by the British Milk Marketing Board, referred to 187 millions litres [13]. An estimation could also be possible on the basis of the total cost of the compensation paid to the farmers which was of the order of 60000 pounds (price referred in 1969 but it is not clear if it concerns the year of the accident) [10].

A volume of 4000 m<sup>3</sup> of trees was generated during the decontamination activities of the 400 Ha "red forest" near the west side of Chernobyl NPP. This forest in 1987 was surrounded by soil bank of 2-2.5 m high, trees and shrubs in it were felled and covered by soil layer of 1 m high. Layer of rich soil was filled up and grass was sowing [6].

In the case of soil removal a big amount of waste is produced because, removing only 5 cm of surface soil in 1 km<sup>2</sup>, a volume of 50000 m<sup>3</sup> of waste is created. About two millions m<sup>3</sup> of low-level radioactive waste would be generated for management, if it is assumed that only an area of 40 km<sup>2</sup> needs to have removed soil.

#### DISCUSSION

The real decontamination methods of rural areas are the removal of soil surface (5-10 cm) and the removal of vegetation with effectiveness of 95-100% and about 50% respectively. Both need special machinery, could produce a loss of fertility and productivity of the soil and an environmental impact.

The ploughing, normal or deep, produces a dilution of the contamination and could reduce the radionuclides uptake by the crops, the levels of external radiation and the resuspension. It can be made by traditional practices and machinery but the radionuclides are still

available to be uptaken by the plants. With ploughing it could be envisaged the simultaneous use of products to control the roots absorption and growth. Recent efforts try to place the contaminated soil layer (0-5 cm) deeper than the root layers (about 60 cm) to avoid the uptake of contaminants by the plants, without disturbance of the 5-60 cm profile. However, the correct application of this method requires the improvement of the equipment and machinery actually available.

The application of fertilizers is an easy method although none product can be considered highly effective for general application. However, by simultaneous application during ploughing could be efficacious.

The use of fixatives stabilizes the contaminants until decisions for land recovery are made. Some special foams could be sprayed and its later removal, in elastic solid form, could result real decontamination.

The temporal restriction of food consumption depends on the characteristics of the contamination and the agricultural products. If the main contaminant has a short half-life (as I-131 with no more than 8 days) and is susceptible to entry into important products of daily consumption (as milk), it seems reasonable, like in the case of Windscale, to apply this countermeasure.

The processing of the contaminated foodstuffs could contribute to reduce the loss of crops and other agricultural products affected by radioactivity. The efficiency on this reduction depends on the general acceptance to consume the processed products, that is why special education in this sense is needed.

There are some alternatives to mechanical or chemical treatments of rural areas as the change of the management of the contaminated land, including modification of the soil uses and the farming practices. In fact, in contaminated areas, it seems possible to grow some crops, with low transfer of the radionuclides through the food chain, or introduce in the normal practices, one of the countermeasures above mentioned.

Finally, an important point to bring out, closely related to the application of countermeasures, is that the most effective of them, which is the removal of vegetation and soil, generates the biggest amount of waste. The management of this waste must be considered a very important problem, so additional research is needed to be done in order to look for effective, practical and economical solutions within this field.

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**TABLE 1 EFFICIENCY OF COUNTERMEASURES APPLIED IN KYSHTYM, WINDSCALE AND CHERNOBYL  
(A FAIR TO GOOD FEASIBILITY IS ESTIMATED FOR ALL THE COUNTERMEASURES)**

SCENARIO	MAIN CONTAMINANT	COUNTERMEASURE APPLIED	EFFICIENCY OF COUNTERMEASURE
Kyshtym	Sr-90.	<ul style="list-style-type: none"> <li>* Ploughing (normal or deep).</li> <li>* Restriction of food consumption.</li> <li>* Limitation on land uses.</li> <li>* Creation of state farms.</li> </ul>	<ul style="list-style-type: none"> <li>* Reduction of <math>\gamma</math>-dose by a factor of 10 (deep pl.)</li> <li>* Reduct. of incorpor. radioactiv. by a factor &gt;10</li> <li>* Reduction factors of 2 to 7 for produced meat. 3 to 4 for produced milk.</li> </ul>
Windscale	I-131.	<ul style="list-style-type: none"> <li>* Restriction of milk consumption.</li> </ul>	<ul style="list-style-type: none"> <li>* Activity of consumed products <math>&lt; 3.7 \times 10^3</math> Bq/l.</li> </ul>
Chernobyl	Cs-137, Sr-90, I-131.	<ul style="list-style-type: none"> <li>* Removal of vegetation.</li> <li>* Removal of soil.</li> <li>* Deep ploughing.</li> <li>* Addition of non contamin. soil.</li> <li>* Application of soil amendments.</li> <li>* Restriction of food consumption.</li> <li>* Use of contaminated products as feedstuff and/or raw material.</li> <li>* Prohibition or change in the use of land.</li> </ul>	<ul style="list-style-type: none"> <li>* <math>\gamma</math>-radiation 45-50 times lower (forests).</li> <li>* Obtainable decontamination factors &gt; 20.</li> <li>* Reduction of the radioactivity in agricultural products by a factor of 1.3 to 3.</li> <li>* Reduction of internal irradiation dose by a "very substantial" factor.</li> </ul>



# POSTER SESSIONS





Session I

**Radioecological Studies and  
Modelling**

Part 1

Forest Ecosystems



# **Study of the Radiocaesium Cycle in Forest Ecosystems (Evergreen and Deciduous Trees)**

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## **ABSTRACT**

*Determinations made after the Chernobyl accident on rain water collected under spruces and oaks show that there is a good correlation between the deposition fluxes of both radiocesium and stable potassium, at least under spruces, suggesting that potassium could be used as an indicator of the behaviour of  $^{137}\text{Cs}$  in contaminated trees. Four years after the accident it seems that a short-term (1-3 yr) equilibrium has been realized in the contaminated trees and that radiocesium is now transferred by trees to the soil mainly via throughfall. Estimation of the global contamination of the different compartments of the forest is given.*

## INTRODUCTION

The accident of Chernobyl as a pulse injection provided an opportunity to study the behaviour of radionuclides in the environment and particularly in forest ecosystems which have been affected as trees are effective aerosol interceptors (1, 2, 3). In this particular ecosystem, radiocesium is of special interest because of its long radioactive half-life and because of its easy availability to biota and food chains.

Rain gauges have been installed in 1982 in a spruce forest (Eastern Belgium), in order to collect chemical pollutants in clearings and under canopies. The observations made in 1986 emphasized the role of canopies which are able to efficiently accumulate air pollutants during dry periods (4).

In the case of the radioactive aerosols from Chernobyl, deposition occurred mainly as a consequence of a rain episode which took place on May 4<sup>th</sup> and which was responsible for a solubilisation of radiocesium and for a direct transfer of this radionuclide into tree leaves. By comparison with the radioactivity collected in clearings, it was deduced that about 80% of radiocesium had been retained by the foliage while about 100% of the other detected radionuclides ( $^{103}\text{Ru}$ ,  $^{131}\text{I}$ ,  $^{132}\text{I}$ ,  $^{132}\text{Te}$  and  $^{140}\text{La}$ ) had been leached to the ground (5).

Determinations made on tree samples collected during the autumn 1986 showed that radiocesium was present in the leaves of all species and particularly in spruces needles although buds were not yet open at the time of the accident. In autumn 1986, new needles radiocontamination in  $^{137/134}\text{Cs}$  is about 50% of the levels observed in pre-1986 needles. This confirms that radiocesium had penetrated into existing older leaves after deposition (in May) and that a certain

translocation had already occurred into younger needles/leaves a few months thereafter.

Because of this very important transfer, we decided to follow two years after Chernobyl the behaviour of radiocesium in the same forest (under spruces and oaks).

## EXPERIMENTAL PART

Tree and throughfall water samples were collected initially in three different forests in southern Belgium. However, because of the too low levels observed in two of these forests, we decided to concentrate mainly on the most favourable case (the "Grand Bois de Vielsalm"). Vegetation and throughfall waters are collected at regular intervals and determined for radiocesium and stable element content. Sample preparation and analytical procedure may be describe as follow:

- rain water is sampled under trees and in clearings by means of 0.28 m<sup>2</sup> plastic collectors. Back to the laboratory, the 10 to 20 litres samples are first filtered (Schleicher & Schuell - folded - rapid - 385 mm paper filters) in order to eliminate plant debris and particles collected together with rain water. This fraction is oven-dried and counted for radiocesium content. It is referred to as "insoluble fraction". The filtrate (containing soluble cesium) is then extracted by stirring (1,5 hours) with a suspension of 2 g of copper hexacyanoferrate which is able to retain Cs very efficiently. The precipitate is collected on a filter and this solid matter is gently charred overnight at 450°C in order to reduce its volume to a fraction of cm<sup>3</sup>. This procedure allows a much better counting geometry during the radioactivity determination (80 cm<sup>3</sup> Ge-Li detector coupled with a 4096-channel

analyser Canberra Series 35 Plus). Soluble potassium was determined by ion chromatography (Dionex ion chromatograph 10) of filtered rain water aliquots (detection limits are of the order of 0.5 ppm).

- plant organs are isolated in the laboratory after collection and weighted as fresh matter and as dry matter. If necessary, plant samples are charred to provide reproducible counting geometries and reduce counting times.

## EXPERIMENTAL RESULTS

Radioactivity was measured in different tree samples and it was observed that:

- $^{137}\text{Cs}$  contamination is higher in spruce needles than in oak leaves of the same age: around 80 to 120 Bq/g of dry weight for needles and around 60 to 80 Bq/g of dry weight for oak leaves (if sampled in autumn).
- new shoots were more contaminated with about 210 and 120 Bq/g of dry weight for needles and oak leaves respectively in spring 1990. In the same time radioactive levels of older needles decrease to around 50 to 90 Bq/g dry weight. These results reflect radiocesium translocation phenomena. Highs levels observed during growing period are an artefact due to the higher water content of younger tissues ( $\geq 90\%$ ): if we consider data per g of fresh weight, levels of contamination decrease to 50 and 23 Bq/g fresh weight for needles and oak leaves respectively.
- $^{137}\text{Cs}$  contamination is of the same order in older needles, twigs wood and bark of spruces and oaks.



Contamination of wood itself is very low during growing season, if not undetectable. During winter 1990, in spruce trunk samples, we detect 13 Bq/g dry weight in alburn and 3 Bq/g dry weight in duramen.

On the other hand, in a spruce stand, needles constitute the main reservoir of potassium (44%), trunk (wood + bark) comes second (34%) and branch wood third (22%) (6). If the distributions of radiocesium and potassium follow the same pattern in trees, one has to conclude that, at the present time, leaves are the principal reservoir of  $^{137}\text{Cs}$  in trees contaminated in May 1986. Twigs are probably to be taken into account, but it is difficult to estimate their relative importance in the global contamination of the tree and in the contamination of leaching rain water.

Given needles and leaves are the main reservoir of leachable Cs (and K), they are the main source of transfer of radioactivity from trees to soil in a forest. After the radioactivity has been redistributed into the biologically active regions of the plant, rain operates as an external transfer agent: leaves are washed and radioactivity is progressively transferred from the leaves to the soil.

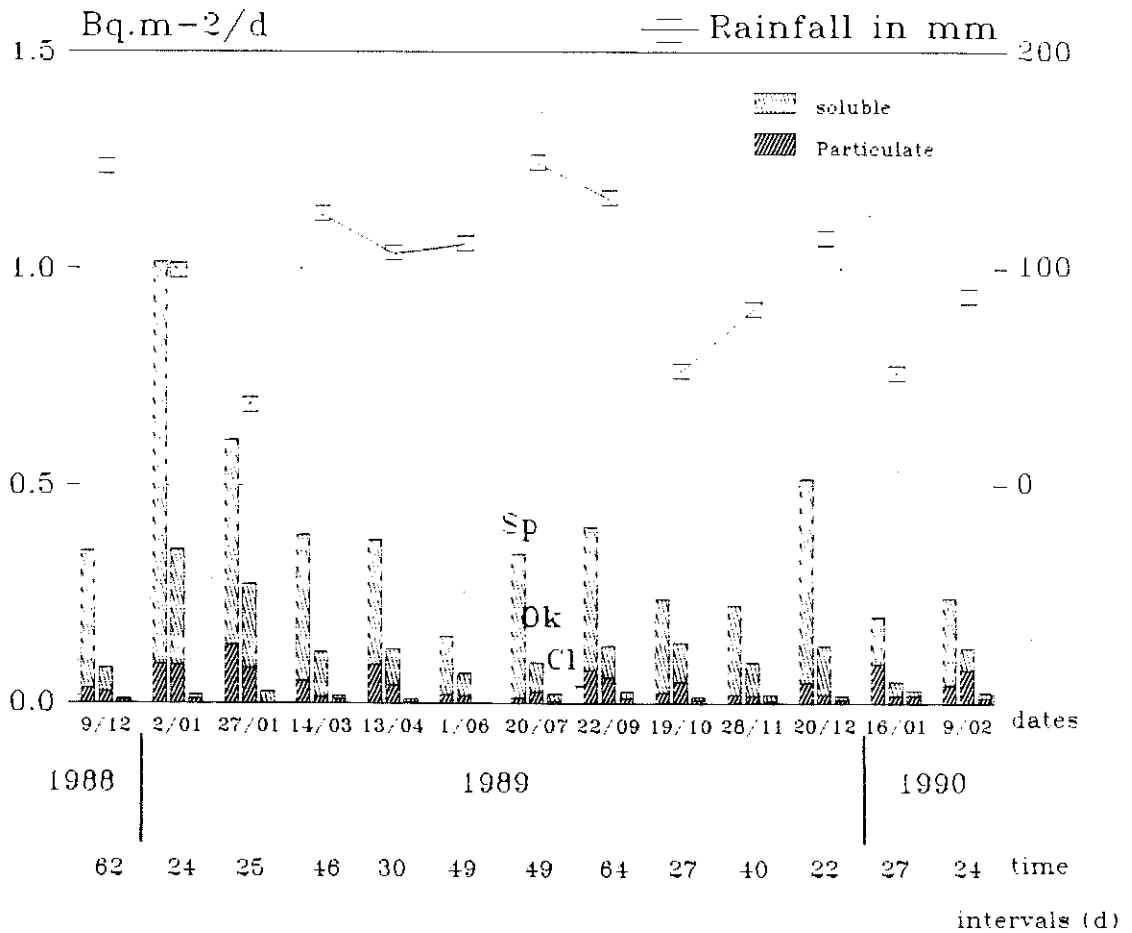
This is observed in throughfall waters collected under trees. Figure 1 shows the results of measurements made between the end of 1988 and the beginning of 1990 (deposition fluxes in  $\text{Bq}\cdot\text{m}^2$ ). Each bar on the graph represents the mean from 4 determinations under spruces and oaks (results are given for soluble and insoluble  $^{137}\text{Cs}$ ). For comparison, the graph shows the activity collected in clearings (due to resuspension) which is negligible. Rainfall is presented in mm of rain.

We observed that throughfall radioactive levels are variable in relation to time. Witkamp and Frank, 1964 (7),

on  $^{137}\text{Cs}$  tagged forest show that leaching is variable, this is partially attributed to varying Cs content of leaves.

FIGURE 1

$^{137}\text{Cs}$  activity of particulate and soluble phases in throughfall waters collected under spruces (Sp), oaks (Ok) and in clearings in the forest of Vielsalm (Belgium). Rainfall is expressed in mm. The abscissa gives the sampling dates and the time intervals (d) between two samplings.



We tested different correlations to explain the reasons for these variations of the washing rates. Table I presents the non-parametric correlation coefficients ( $r_s$ , Spearman coefficients) which have been calculated for different

pairs of variables ( $^{137}\text{Cs}$  and stable  $\text{K}^+$  fluxes vs rainfall). Over a whole year, very poor correlations are observed under both spruces and oaks.

TABLE I

Non parametric correlation coefficients  $R_s$ :  $^{137}\text{Cs}$  vs rainfall and stable  $\text{K}$  vs rainfall under spruces and oaks (whole observation period: December 1988 - February 1990). Values are given as:  $R_s$ , (level of significance %) and  $N$  of observations.

	Cs vs rainfall	K vs rainfall
Spruces	0.48 (90.2) 13	0.25 (59.5) 13
Oaks	0.13 (33.2) 13	-0.19 (47.0) 13

On the contrary, as shown in table II, there is a highly significant correlation (non-parametric) for spruces and a somewhat poorer for oaks, between radiocesium and its chemically related potassium especially during winter.

TABLE II

Non parametric correlation coefficients  $R_s$ :  $^{137}\text{Cs}$  vs stable  $\text{K}$  under spruces and oaks for whole observation period (December 1988 - February 1990) and as a function of the seasons. Values are given as  $R_s$ , (level of significance %) and  $N$  of observations.

	Spruces	Oaks
Whole period	0.92 (99.99) 51	0.48 (99.97) 52
Winter	0.97 (99.99) 20	0.83 (99.99) 20
Spring	0.92 (99.99) 12	0.85 (99.96) 12
Summer	0.96 (99.95) 7	0.35 (60.07) 8
Autumn	0.92 (99.99) 12	-0.17 (40.80) 12

This is presented in figure 2A and 2B where *linear regressions* are suggested, merely as a working tool. Correlations are evident for both species, but a much weaker slope is observed for oaks and this could be interpreted as follows:

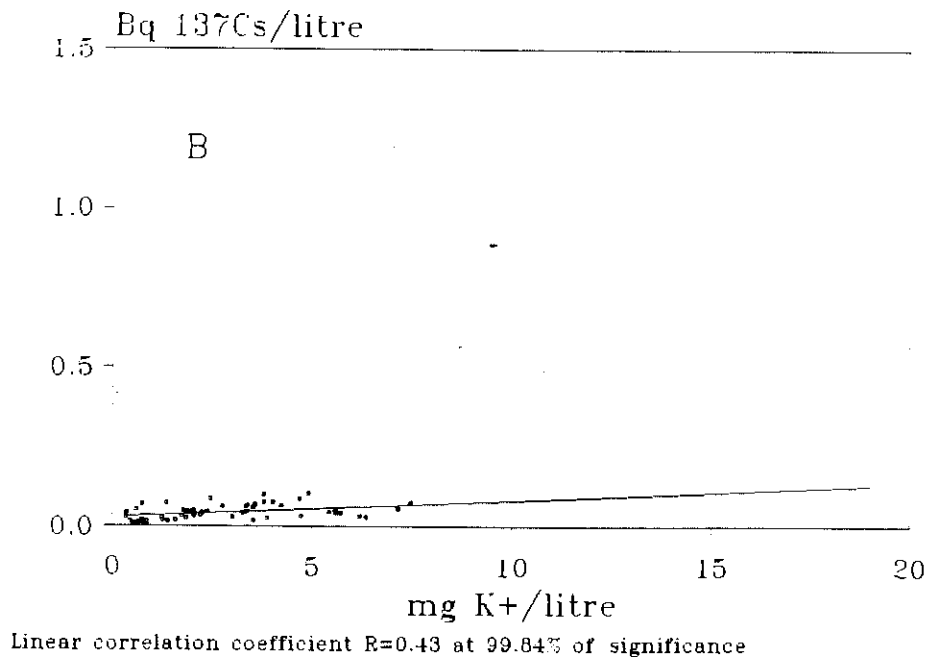
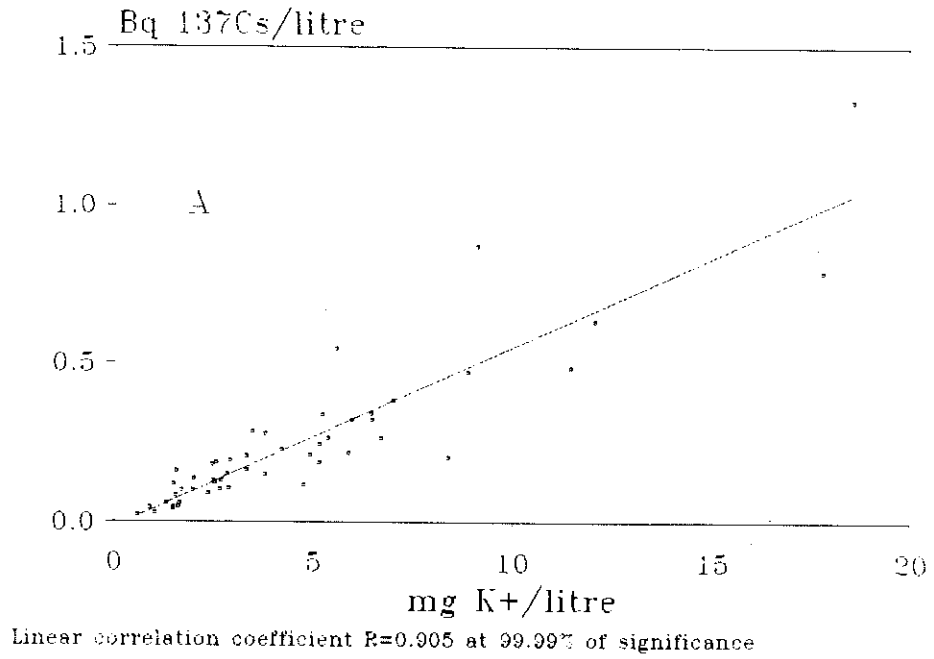
- four years after the accident, contamination by radiocesium could be less in oaks because, at the time of the Chernobyl event, oak buds were not yet open. By now, at the time of bud opening, contamination is about equal in oak leaves and in spruce needles, but with the advancement of the growing season, it significantly enhances in spruce needles.
- potassium content in oak leaves is greater: this was shown by measurements made in our laboratory (confirmed by Nys and al. (6) and by Cole and Rapp, (8)):  

oak leaves:	12 mg K/g DM
spruce needles:	6 mg K/g DM
- in general, potassium is washed at higher rates under deciduous trees (Cole and Rapp (8), measured  $21 \text{ kg K}^+ \text{ ha}^{-1} \text{ a}^{-1}$  under oaks against  $12 \text{ kg}$  under spruces).

Whatever the explanation for the differences between oaks and spruces, potassium seems to be a good indicator of the current washing rate of radiocesium from leaves. Seasonal variations of potassium in leaves are reported by several authors: Olsen, 1948 in Duvigneaud (9), show that the K pool increase in spring, stabilize in summer to decrease in autumn. This autumnal decrease of K level in leaves is attributed part to washing off and part to translocation to trunk, branches, roots ... (10). On the other hand, Ranger and al. (11), observe that 74% of the potassium necessary to the biomass production (new leaves, new age rings of tree) comes

FIGURE 2

Relationship between  $^{137}\text{Cs}$  and stable K in throughfall waters collected under spruces (A) and oaks (B) in the forest of Vielsalm (Belgium).



from older part of the tree (preexisting wood, needles). Opposite movement is observed at the end of summer and in autumn when foliar potassium is redistributed into woody part of the tree. During this period of dormancy (decrease of cell activity) cellular potassium leaves the cell (12).

The existence of a potassium cycle in trees and the correlations observed between radiocesium and potassium lead us to suspect that radiocesium and potassium movements are bound. Some experiments are now conducted in Vielsalm forest to quantify cycles of cesium and potassium in spruce needles and to determine the washing off availability of these two elements.

Our forest samples allow us to make a rough estimation of the distribution of radiocesium in different part of the spruce stands. Table III shows that the major part of the  $^{137}\text{Cs}$  (88.3%) is fixed in the soil. Tree vegetation retain about 3% of the total ecosystem activity.

TABLE III

Estimation of the distribution of  $^{137}\text{Cs}$  activity in contaminated forest of Vielsalm (Belgium).

	Biomass 10 <sup>3</sup> kg/ha	Bq/kg DM	<sup>137</sup> Cs 10 <sup>3</sup> kBq/ha	%
Needles	18	56	1.01	
Twigs	21	58	1.22	
Bark	10	60	0.60	3.03
Wood alburn	55	13	0.71	
duramen	55	3	0.16	
Roots	34	80	2.72	2.23
Litter	48	165	7.92	6.48
Soil O/OAh	180	270	48.60	39.80
Ah	910	55	50.00	40.94
B	1312	7	9.18	7.52

## CONCLUSIONS

$^{137}\text{Cs}$  is a long-term threat in contaminated forests and it is of prime importance to have a good knowledge of its behaviour in order to employ realistic counter-measures following an accident.

After deposition and washing of the leachable fraction of radiocesium deposited onto plant surfaces,  $^{137}\text{Cs}$  penetrates into leave tissues and translocation begins to operate since spring 1986. At the same time, transfer begins between tree foliage and soil.

Seasonal cycles are important in translocation phenomena and three years of measurements do not demonstrate any significant decrease of the contamination levels in tree tissues. Trees represent a non negligible reservoir (more than 5% including root system). This reservoir ensures, up to now, a rather constant supply of radiocesium. All these observations suggest that a mid-term steady state has been attained by now in the trees and that radiocesium is now washed away at a rate determined by the physiological activity of trees. This rate will slow down with time as radiocesium becomes depleted in trees at the expense of soil. This rate will also vary with time as root system uptake will become significant after radiocesium will be more or less trapped by organic matter and clay.

Long-term observations are still necessary to confirm these assertions as well as the similarity of behaviour between potassium and radiocesium. If confirmed, this similarity could greatly enhance our knowledge of contamination pathways in forest ecosystems.

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# **Study on the Bioavailability of Radiocaesium Following Contamination of the Forest Floor**

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## ABSTRACT

After the Chernobyl accident, the radioactive fallout have put forward the high capacity of forest ecosystems to catch radionuclides and particularly, the strong retention of radiocesium on soil surface.

Forest floor is characterized by the surperposition of purely organic (organic horizon: O) and readily mineral (hemiorganic (Ah) and mineral (B) horizons) materials. The migration capacity of radiocesium through such a profile as well as the influence of the different layers on the retention and remobilisation processes of the radionuclide is, by now, few understood. The importance of radiocesium bioavailability and the dynamic of its biological cycle depends from the transfer or storage possibilities of cesium into the different soil layers.

The migration possibilities of cesium in forest soil columns (undisturbed profile) following a surface contamination and also the redistribution of the radionuclide through the profile have been investigated. The radiocesium bioavailability in the different layers was estimated by activity measurements in the soil solution and from different extractions of the solid phase of the soil.

## INTRODUCTION

The reactor accident in Chernobyl and the following radioactive fallout have put forward the high interception rate of radionuclides by forest canopy and the long-term storage in soil surface after deposition (1). Cesium-137, with a half-life of 30 years, was an abundant radioactive constituent in Chernobyl fallout even if Cs was partly incorporated into undissolved forms (2).

During the last four decades, studies concerning the behaviour of radiocesium in forest soils have been nearly inexistent although investigations into agricultural soils and mineral clays have received much more attention. It was found that  $^{137}\text{Cs}$  partitioned largely with clays (3) and that, depending upon the type of soil, 40 to 80% was unexchangeable (4). Illite was the most effective sorbent by "fixation" in the wedge zone of clay minerals (5). The Cs availability was also influenced by the concentration of exchangeable potassium (6).

Results obtained at Oak Ridge National Laboratory on the behaviour of long-lived radioisotopes in three forest ecosystems (Eastern Tennessee) indicate that over 90% of the radiocesium is still in the soil 30 years after soil contamination (7). The percentage of  $^{137}\text{Cs}$  was found to be a linear function of organic matter content of the soil samples.

The recent post Chernobyl observations (8-9-10) have confirmed that the forest soils strongly retain radioactive Cs in the upper organic rich layers. The availability of radiocesium seems however to be low, particularly in deeper hemiorganic layers (11). The variability observed is in relation with the humus type and could be explained by some particular factor as potassium amounts. In the upper mineral soil, the  $^{137}\text{Cs}$  content was correlated to the % of the orga-

nic carbon which suggested that organic matter plays an important role in the retention of this radionuclide (12).

Unlike agricultural soils, forest floor is a superposition of purely organic (Of - Oh horizons) and readily mineral material (Ah - B horizons) with variable conditions of humus type. By now, only few data obtained "in situ" give information about the availability of radiocesium or of any dynamic process that may influence it through the various compartments of a forest soil profile. The interpretation of activity measures with soil depth is often difficult because of the numerous controlling factors (physical and chemical) and their high spatial heterogeneity.

Therefore, laboratory leaching experiments in soil columns were designed to better understand the physico-chemical aspects of the phenomena governing the vertical movement of radiocesium and its availability evolution through the profile.

The aim of this paper is to discuss several ways of expressing the radiocontamination of a forest soil profile and to compare the radiocesium behaviour with K in different phases of the soil with regard on their bioavailability.

## MATERIALS AND METHODS

Two soil cores (undisturbed profile) were sampled, both from a Brown Acid Soil developed under a Norway spruce (*Picea abies* (L.) Karst) in Belgian Ardennes. Some important properties of the various soil layers of the profile are shown in table 1.

to selectively displace the  $^{134}\text{Cs}$  linked to organic fraction. Extractions are made on air-dried soil at a 1/10 soil-solution ratio by shaking for 24hrs, followed by centrifugation at 10000 rpm for 10 minutes.

Before characterization, all extracts (except for NaOH extracts) were filtered at  $0.45\ \mu\text{m}$  (polysulfone membrane) in order to remove the colloidal material and microorganisms. Radiocesium activity in extracts was measured in a NaI detector (PACKARD-Minaxi Auto Gamma: 5000 series) and K concentrations by atomic absorption spectrophotometry.

## RESULTS AND DISCUSSION

The migration of  $^{134}\text{Cs}$  in the forest profile seems extremely slow in spite of the daily leaching during eight months. The effluents of both columns were collected every week but no radioactivity was detected.

The distribution of radiocesium in the various horizons (Fig. 1) shows that, as expected, 72% of the total activity remain in the top layers of the columns (0-6,3 cm), consisting mainly of raw humus (Of horizon) and soil rich in organic matter (OAh horizon).

As pointed out in the introduction, there are several reasons why discrepancies might occur when information about the contamination of a forest soil is expressed by weight (Bq/g soil). As the migration of radiocesium proceeds through very different soil layers, it is thus reasonable to assume that the volumic expression (Bq/cm<sup>3</sup> soil) gives a more realistic information. In our case (Fig. 2), the results expressed by weight overestimate by a factor 4 the contamination of the upper organic layers (Of-OAh horizons), comparatively to the other layers (Ah - B horizons).

Figure 1

Distribution of radiocesium as a function of the soil layers

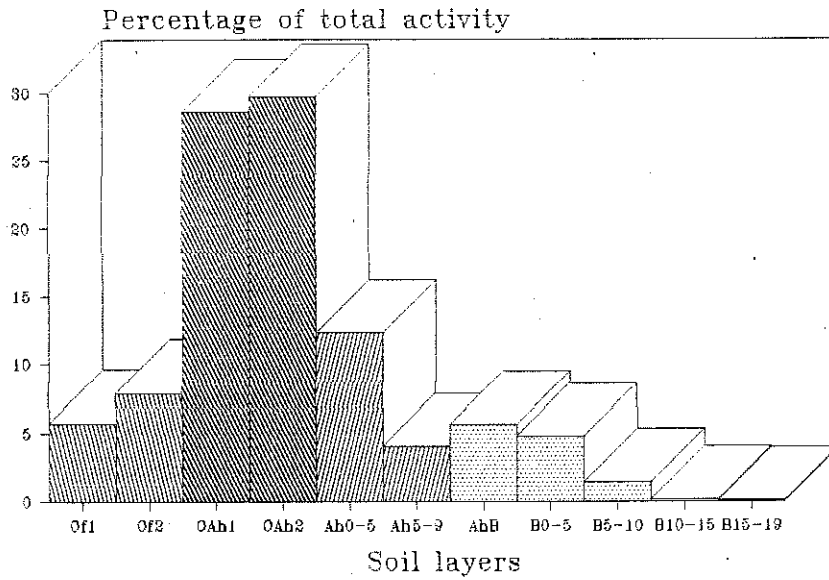
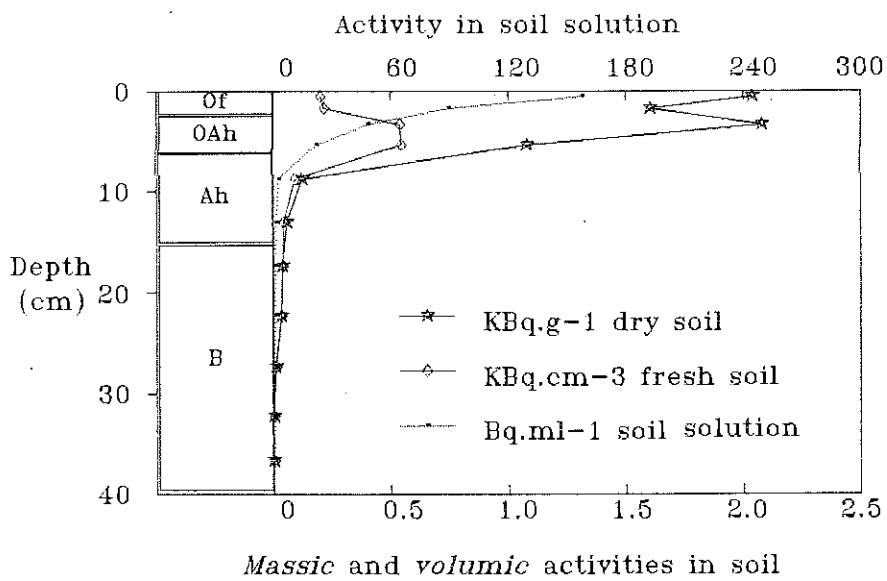


Figure 2

$^{134}\text{Cs}$  profiles in the spruce soil

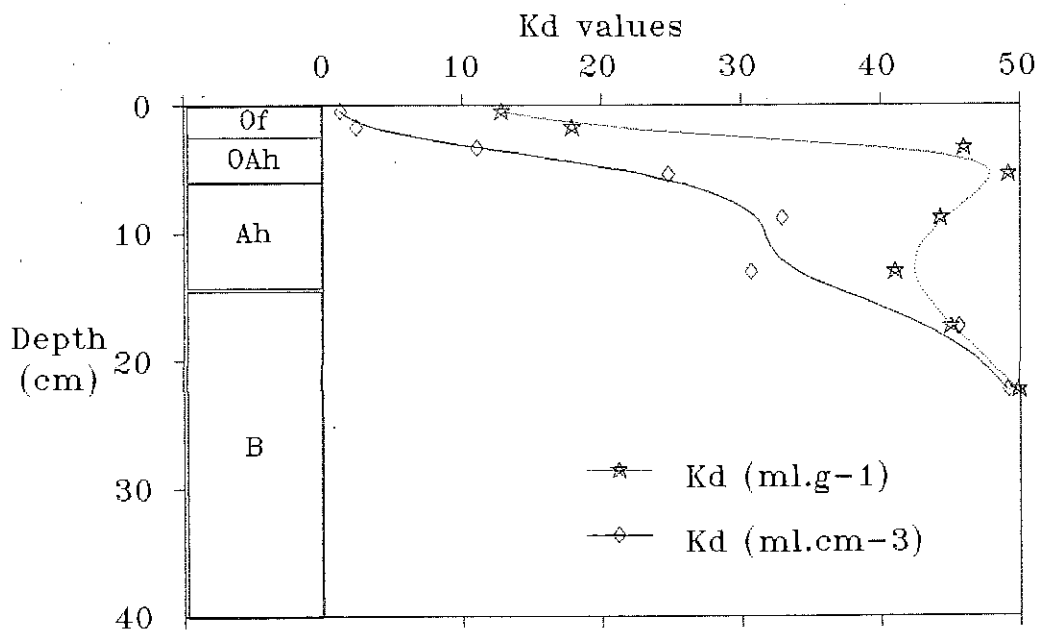


In these conditions, the use of the retention factor (Kd) for the prediction of the radionuclide movement or transfer can be misleading, especially from forest soil horizons, rich in organic matter, where plant root systems are particularly developed.

According to Adams (13), the soil liquid phase mediate between the plant root and the solid phase. The measure of the radioactivity in soil solution can thus be used to give a first indication of the mobility and of the bioavailability of radiocesium (Fig. 2). The Kd values (ratio of Cs-134 present in the solid phase and in the soil solution) were calculated using the soil activity expressed by weight or by volume. The coefficient distributions obtained in these ways are shown in Fig. 3.

Figure 3

Evolution of the Kd values in the soil profile





The comparison of these values yields the following results:

- + In the Of OAh horizons (humus layers), the Kd values obtained from massic activity of the soil are considerably higher (factor 2 to 10) than those obtained from volumic activity. In the hemiorganic horizon (Ah), the difference become, however, much smaller (at most a factor 1.5). In deeper mineral layers (B horizon), the two expressions yield essentially the same Kd values.
- + Kd values obtained for the upper organic layers (Of horizon) are much more smaller than the values obtained in deeper horizon but increase rapidly with depth.

The transfer from one horizon to the next one should thus result in a decrease in the rate of migration of the radiocesium or of its bioavailability. Our results confirm that the highest sorption occurs in the deeper mineral soil layers, especially in B horizon. However, Kd values suggest a more rapid evolution of the sorption process, already active in deeper organic layer (OAh: 2.4 - 6.3 cm) and then a relatively high bioavailability limited to raw humus layer (Of: 0 - 2.4cm).

As shown in table 2, the percentage of  $\text{NH}_4\text{Ac}$  extracted cesium in organic layers don't exceed 20% of total Cs content; the minimal value is found in OAh horizon where the total Cs content is, nevertheless, the most important. Relative highest values are found in deeper horizons. A comparable distribution of values is found for  $\text{CaCl}_2$  extraction but with a lower order of magnitude. These values were compared with measures of K concentration made from the same extracts.

Table 2

Proportions of the extractable  $^{134}\text{Cs}$  and K

horizon	% of NaOH, $\text{NH}_4\text{Ac}$ and $\text{CaCl}_2$ extracted radiocesium				$\mu\text{g/g}$ soil of $\text{NH}_4\text{Ac}$ and $\text{CaCl}_2$ extracted potassium		
	NaOH (1)	$\text{NH}_4\text{Ac}$ (2)	$\text{CaCl}_2$ (3)	(3)/(2)	$\text{NH}_4\text{Ac}$ (4)	$\text{CaCl}_2$ (5)	(5)/(4)
Of <sub>1</sub>	2.83	20.58	6.95	33.7	281	272	96.8
Of <sub>2</sub>	0.87	10.63	2.71	25.5	227	190	84.0
OAh <sub>1</sub>	0.27	1.89	0.21	11.0	172	171	99.3
OAh <sub>2</sub>	0.56	0.57	0.04	7.2	109	72	66.2
Ah <sub>0-5</sub>	0.88	1.92	0.08	4.1	72	45	63.4
Ah <sub>5-9</sub>	1.28	10.37	0.55	5.3	48	26	54.0
AhB	2.14	23.71	0.54	2.2	30	15	50.0
Bo-5	2.75	38.81	0.57	1.4	26	13	50.0
B <sub>5-10</sub>	2.41	44.21	-	-	27	12	44.4
B <sub>10-15</sub>	-	53.84	-	-	27	11	40.7

Quantities of K removed from both solvents are very similar in organic horizons (Of - OAh), thus subjecting that the great majority of the available K ion does not occupy specific sites on the substrate of the organic layers; to the opposite,  $\text{NH}_4^+$  remove more important quantities of exchangeable K in deeper mineral horizons (Ah - B). According to Bolt and al. (14), Ca ions are very ineffective in removing K from specific "edge-interlattice" sites on clay minerals. In contrast, poorly hydrated cation such as  $\text{NH}_4^+$  is highly competitive for the same ion-exchange sites.

As for exchangeable K in mineral layers, the difference in organic layers between the extractibility of Cs by  $\text{NH}_4\text{Ac}$  and by  $\text{CaCl}_2$  suggest a high selective sorption, indicating special reaction mechanisms other than non specific adsorption. If we take into account the existence of such specific exchange-sites for Cs on mineral clays (15), the discontinuous presence of a mineral fraction alone or combi-

ned with organic colloids (16) should explain partly the high sorption process at the surface soil of the column. Indeed, Cs extraction by dissolution of the organic fraction (NaOH extract) (table 2) don't show preferential retention on organic matter.

## CONCLUSIONS

Our study has put forward the risk to mislead the interpretation of activity measurements for forest soils if based on expression by weight.

In other way, the calculation of Kd ratio between the soil solution activity and the "volumic" soil activity gives a realistic indication for the evaluation of the bioavailability and of the mobility of radionuclides in a multilayered soil as a forest soil profile.

The results obtained for Cs speciation suggest that mineral components combined with the organic fraction in the upper organic horizons could play a role in the high Cs retention at the surface of the soil.

Part of this accumulation might also be due to microbial immobilization but this assumption was not tested in our study.

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# **A Systems Approach to Transport of Radionuclides in Forest Ecosystems**

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## ABSTRACT

Wide scale deposition of radionuclides over Europe from the Chernobyl accident has required detailed investigations of the effected regions. Our initial studies of generic forest ecosystems revealed a lack of understanding of the transfer and cycling of radionuclides and their resulting risks to man. To assess the radiation hazard from the forest ecosystem pathway we must determine if the forest acts as a source or sink for deposited radionuclides. This paper presents our progress on a systems approach used to evaluate pathways of radionuclides deposited on forest ecosystems. The approach considers relevant pathways and processes required in assessing the radiation dose risks to man.

## INTRODUCTION

Deposition of natural radionuclides, nutrient and trace elements is part of the hydrogeobiochemical cycle that redistributes materials on earth. Pollutant elements from industrial sources and artificial radionuclides from nuclear weapons tests and from the Chernobyl reactor accident in the USSR, has illustrated the importance of assessing transfer functions to determine appropriate population dose exposures. The wide scale deposition of radionuclides over natural and seminatural ecosystems of Western Europe has permitted the development of new models to evaluate some of the monitoring data taken to protect the population living in the path of the fallout. However, the monitoring data often has been confusing because of the lack of an overall approach to the chemical, biological and geological mechanisms responsible for the radiation dose delivered to man. We question the importance of the forest in the overall dose received by man. Is the forest a sink or a secondary source for radionuclides which, in time and under certain conditions, may contribute to the future radiation dose to man? We believe that it is important to synthesize and evaluate an overall or "global" picture of the factors that may influence the dose delivered. Only then can realistic models be constructed and validated.

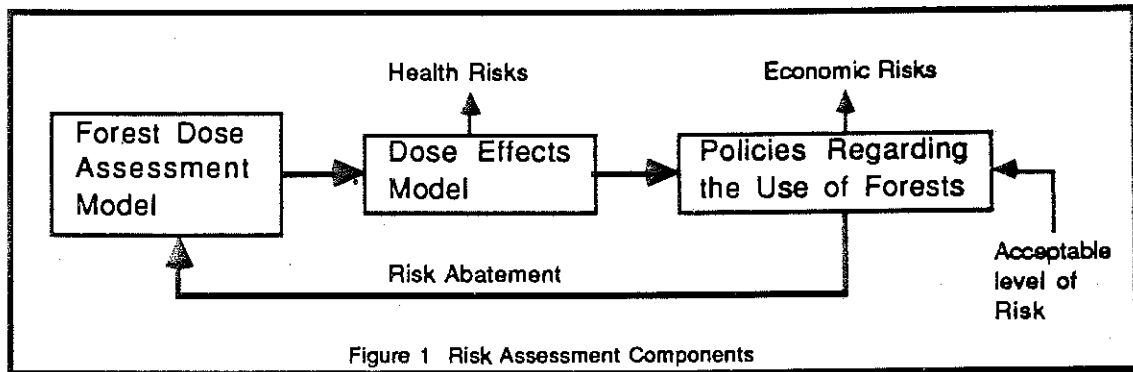
The goal of this study is to assess the risk to man from an accidental release of radionuclides into forest ecosystems. This is not only important from an environmental safety point of view but also from an economic one. The forest is a source of lumber, game, assorted vegetables, berries and herbs which if contaminated, may have a significant economic impact on the region affected. Given a set of policies on the use of contaminated forests, this study will address three main questions:

- ( 1 ) What are the important radionuclide pathways through the forest to man?
- ( 2 ) How long does the contaminated forest remain a risk to man?
- ( 3 ) What is the level of this risk over time?



## CONCEPTUAL RISK MODEL

This study is concerned with estimating the risk to man from an accidental deposition of radionuclides into forest ecosystems. The components necessary for this risk assessment are given in Figure 1 below:



The first component in Figure 1 is a forest dose assessment model. The goal of this assessment model is twofold: (1) to estimate the radiation dose contributed directly to man through the food chain and direct gamma radiation shine subsystems and (2) to estimate the flow of radionuclides out of the forest through the resuspension and aquatic pathways. In order to accomplish the above objectives, it is essential to understand the transfer and transport of radionuclides into and out of the forest ecosystem.

Once an assessment is made of the dose to man from the forest the health risks of such contamination needs to be estimated by a dose effects model. Thus, inhalation, gamma shine and ingestion doses obtained from the forest must be converted into increased incidents of cancer or other health effects. The dose to man is affected by policies regarding the use of a contaminated forest in the determination of an "acceptable" level of risk.

## SYSTEMS APPROACH

A systems approach has been used in both defining and formulating the relevant pathways of radionuclides to man through the forest. Figure 2 sets up the global perspective on the direct and indirect pathways to man through the four subsystems: Urban, rural, aquatic and marine. Using this global perspective, a schematic model has been formulated which delineates the pathways into and out of the forest ecosystem. The boxes in bold along with

the bold arrows in Figure 2 show the important pathways and subsystems interacting with the forest and its eventual effect on man. The focus is on those pathways entering or leaving the dotted box along with the interactions within this dotted box. The only radionuclide flow into the forest ecosystem is from the source deposition. The source deposition originates mainly from the initial accident fallout but also includes those radionuclides resuspended from both urban and rural systems. Radionuclides deposited on the forest may take two indirect pathways to man: (1) runoff to the aquatic subsystem or (2) resuspension. Precipitation in the form of rain or snow results in infiltration and overland flow of radionuclides into the aquatic subsystem which may reach man through drinking wells and the aquatic food chain. Resuspension by fires, wind and floral pollination also may reach man through deposition to both urban and rural systems from the source. Radionuclides deposited into the forest from the source may take two direct pathways to man via: (1) the consumption of contaminated food from the forest and (2) direct gamma radiation shine. This radiation dose pathway may occur through residence in and exposure to contaminated forests or lumber. Using a systems approach,

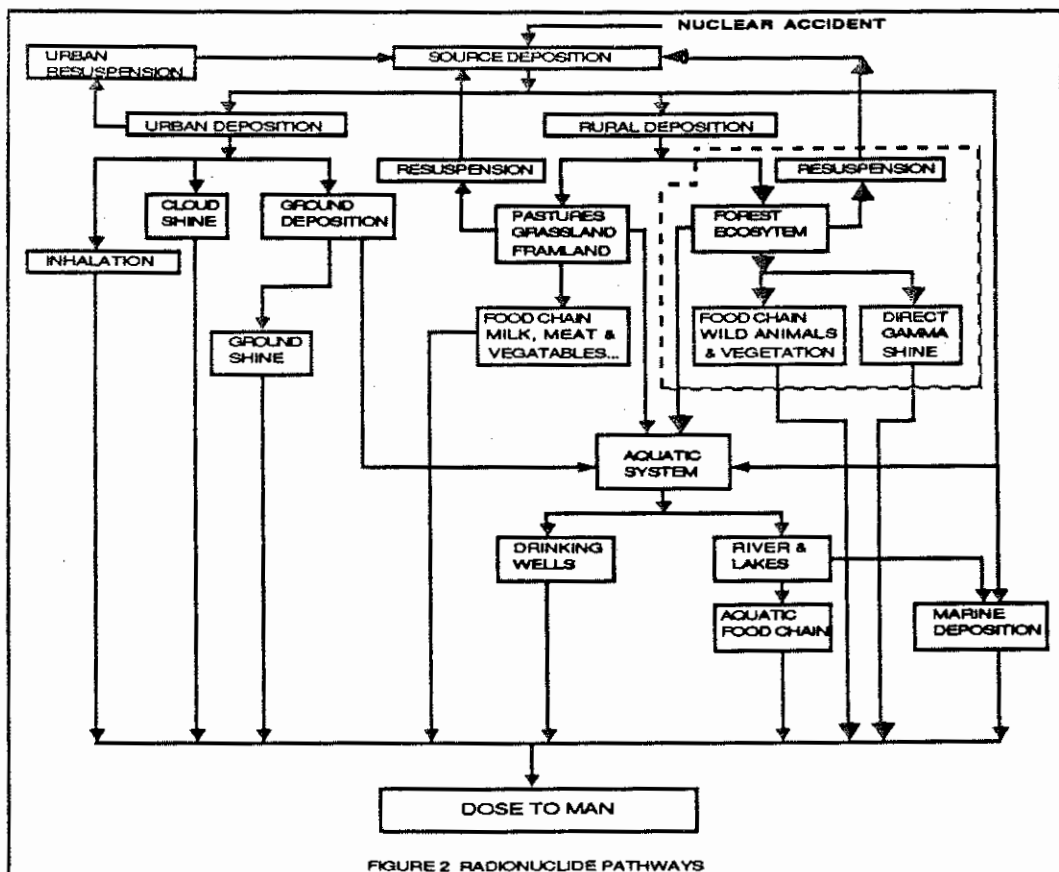


FIGURE 2 RADIONUCLIDE PATHWAYS



subsystems. For example, the ground component within the forest may contribute radionuclides to man through the aquatic subsystem by watershed runoff, to source deposition through the resuspension subsystem or directly to man through the direct gamma radiation shine subsystem.

### Source of Contamination

Radionuclides from nuclear accidents may be deposited into the forest by two means: (1) precipitation and (2) dry deposition. Precipitation in the form of rain or snow may carry airborne particles (aerosols) and deposit them on the forest ecosystem. Interception of these aerosols may be by the overstory, the understory, animals or the ground. Dry deposition may be intercepted by the same forest subsystems as wet deposition.

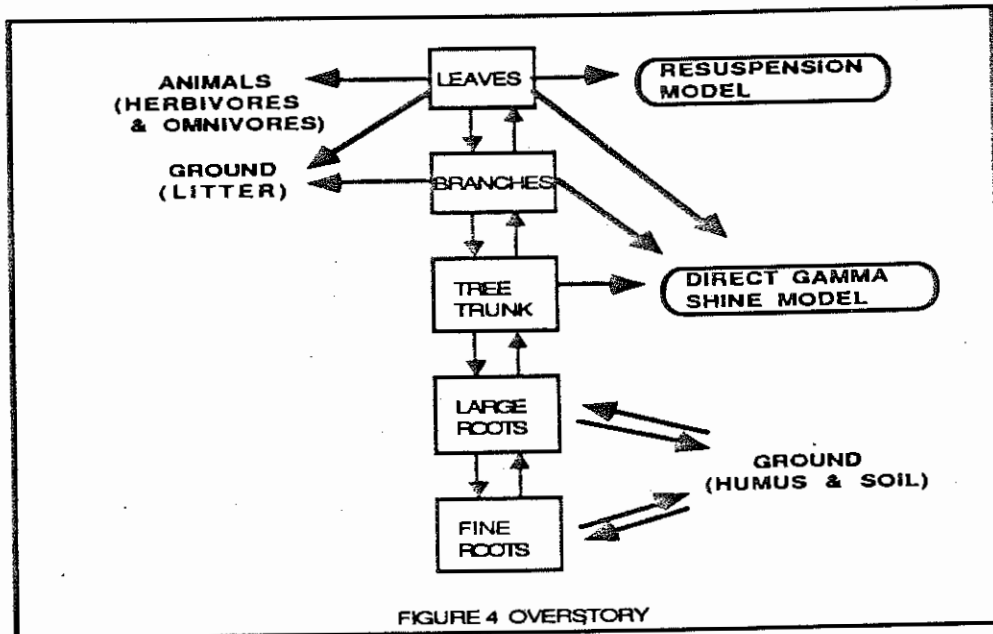
The aerosols constitute the major reservoir of pollutants in the atmosphere. The nature of radioactive aerosols produced from anthropogenic sources depends on the mechanism by which the radioactivity is released. In the Chernobyl reactor accident, which involved both the nuclear fuel melting and a graphite fire, aerosols were generated by the accident. The maximum plume altitude reached approximately 1500 meters for transport by the prevailing winds. The chemical form of the material released was variable and included graphite from the reactor core and boron carbide, lead, dolomite and clay which was dropped on the accident, which produced particles observed to range in size from less than one micrometer to tens of micrometers [1]. Following the formation of these radioactive aerosols, the fate of the radionuclides became the fate of the carrier aerosols [2]. Thus, emergency measures taken in an attempt to control the reactor, impact the transport, deposition and ultimate fate of released radionuclides.

Consequently, the formulation of realistic predictions of the impact of accidentally released radionuclides requires information on the characteristics of these aerosols and on their retention by foliage in relation to the species and in relation to the growth status of the plant and the climatological conditions. The following processes can be distinguished:

- ( 1 ) Wet deposition by rain or snow.
- ( 2 ) Impaction of aerosols.
- ( 3 ) Impaction of mist, fog, cloud droplets.
- ( 4 ) Absorption of gases.
- ( 5 ) Reemission.

### Overstory

The overstory pathway component consists of the tree portion of the forest, Figure 4. Radionuclides can be transported in either direction, from roots to foliage or foliage to roots through the Xylem and Phloem transport systems [3].

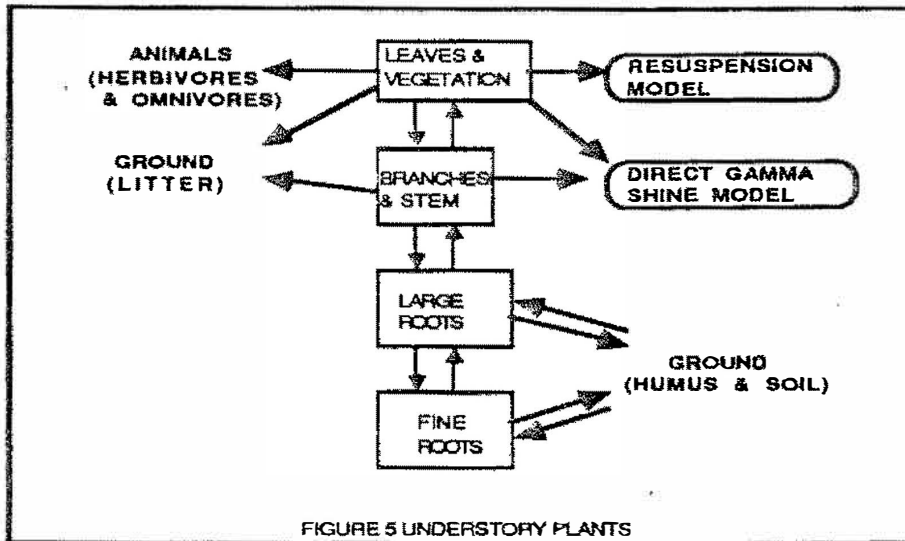


The bold arrows in Figure 4 signify the transport of radionuclides out of the overstory components to either other forest subsystems or to subsystems outside of the forest. For example, radionuclides may be transported within the forest by pathways from the leaves component to the ground by annual shedding of leaves or to animals by herbivore or omnivore consumption. Transport may proceed out of the forest subsystem by a pathway from the leaves component to the resuspension subsystem by aerial travel by wind or fire.

The most important characteristics of the overstory that influence the interception, transfer and transport of radionuclides within and out of the forest include: The type and behavior of overstory leaf component, the process of adsorption and leaching of radionuclides from the canopy, and the characteristics of the root system and its process of exchange of radionuclides with the ground component of the forest.

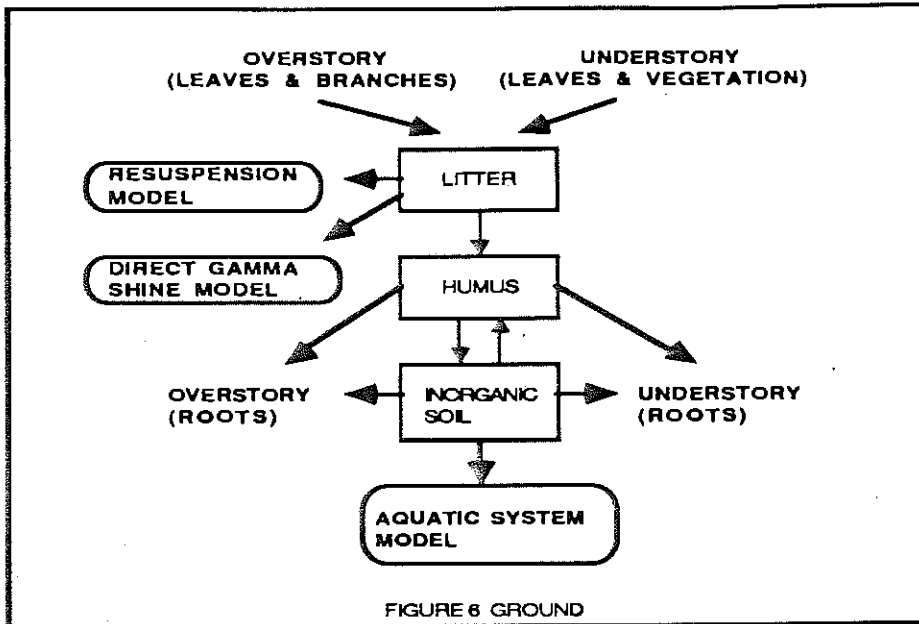
### Understory

The understory pathway consists of radionuclide interaction with the low lying bushes and plants as shown in Figure 5. In this Figure bold arrows signify transport out of the understory either to other forest subsystems or out of the forest. Similar to the overstory component radionuclides can be transported between the root system and the vegetation. Of particular importance are the food sources such as berries, herbs, and mushrooms.



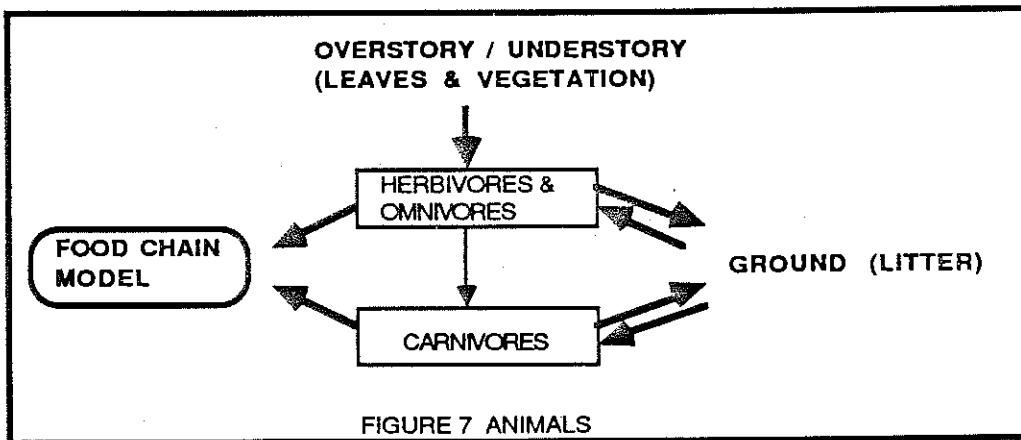
### Ground

The ground pathway consists of radionuclide interaction with the litter, humus and inorganic soil compartments as shown in Figure 6. This pathway includes the complex interactions of the root, soil and litter system where uptake and deposition proceed on an annual cycle in temperate zones. Some of the most important characteristics of the ground system relative to radionuclide transport include its chemical composition (i.e. clay content), its organic concentration, and its nutrient level.



### Animals

The animal pathway includes birds, animals and insects as shown in Figure 7. This is at the top of the food chain leading to man and could include a significant source of radiation dose through the dietary patterns of a critical group of a potentially exposed population.



### MODELING APPROACHES

The approach to modeling the forest system is to first define those factors that strongly influence the behavior of radionuclides in each forest subsystem. For example, the transfer of radionuclides within and out of the ground subsystem may be affected by such factors as clay or sand content of soil, the potassium and ammonium concentration, the pH,

the organic content of the litter, etc. Data for the inclusion of all the factors in the complex forest ecosystem may be unavailable and even may be unnecessary in our first approach model. Thus, the approach taken in formulating those "most important" factors influencing radionuclide behavior is the use of "expert opinion." Experts in such fields as plant physiology, soil chemistry and forestry will be given a questionnaire in which they will be asked to judge the importance of different factors on the radionuclide cycling within and transport out of the forest. By using the psychometric technique of paired comparisons it may be possible to arrive at a general consensus of which factors are deemed to be the most important. The result of such an analysis will reveal both the order of importance of each factor and its relative importance to an absolute zero weight.

Once descriptive models are constructed for the components of the forest ecosystem and the "most important" factors are chosen, a mathematical model will be developed for the behavior of radionuclide cycling within the forest over time and subsequent contribution to the dose to man (forest dose assessment model). Two other models will also be created, one for the affect of a given policy on both the associated economic and health risks (policy risk model) and another to relate radiation dose to health effects (dose effects model). The policy risk model will be developed based on a scenario approach for different policies effecting the usage of contaminated forests. Thus, different policy scenarios will be developed and presented to decision makers along with the associated health and economic risks. The mathematical model translating radiation dose to health effects will be taken from literature.

#### **Forest Dose Assessment Model**

The goal of a forest dose assessment model is to estimate over the long term the dose received by man given specified policies on the usage of contaminated forests. The model will include the following processes:

- ( 1 ) The transfer and transport of radionuclides within and out of the forest.
- ( 2 ) The regeneration, growth and death of forest biomass.
- ( 3 ) The interaction of man with the forest given selected usage policies.

The approach to mathematically assessing the transfer of radionuclides within and out of the forest ecosystem is based on: (1) the descriptive models of radionuclide transfer within and transport out of the forest and (2) the conservation of mass principal. Due to



the long term nature of several radionuclides, it is also necessary to consider the behavior of forest biomass over this time horizon. Several events may occur in forests over the long term (i.e. years) including fires, growth, death and human deforestation. The modeling of the processes of regeneration, growth and death of forest biomass (i.e. forest succession) over time has been studied by several authors [4]-[7]. In order to capture the interactions of man and wild animals with the forest, a spatial model of the forest will be constructed. Such a spatial model will consist of a three dimensional representation of the forest components: Overstory, understory, and ground. Semi-imposed at each point in space (where there exists a forest component) will be the mathematical model representing this component. Some of the goals of such a spatial model are to capture the spatial heterogeneity of the forest, to capture the lateral interactions and flows of radionuclides, and to create the ability to execute different scenarios of forest use by man.

#### **DATA REQUIREMENTS**

In order to calibrate the forest dose assessment model time dependent data regarding the level of contamination in different forest compartments is required. These compartments have been specified in our systems approach and include such processes as:

- ( 1 ) Atmospheric transport and deposition.
- ( 2 ) Interception by the forest canopy.
- ( 3 ) Transport and translocation in the canopy.
- ( 4 ) Forest litter interception - precipitation and throughfall.
- ( 5 ) Root uptake and discharge of radionuclides in soil.
- ( 6 ) Infiltration and evapotranspiration induced mobility.
- ( 7 ) Rates of transport through the root zone.
- ( 8 ) Rates of loss to the water table.

In order to obtain the required data we will rely on several sources. These sources of experimental measurements include published and unpublished studies on:

- ( 1 ) Fallout from nuclear weapons tests deposited on forests.
- ( 2 ) Releases at weapons production plants to nearby forests.
  - ( a ) Releases of nuclear waste to soil.
  - ( b ) Atmospheric releases from waste concentration operations.

- (c) Atmospheric releases from nuclear fuel production and power reactors.
  - (d) Subsurface releases to unconfined aquifers where re-emergence at springs occurs.
- (3) Resuspension to and from forests.

We believe that an additional source of information for the model application can be obtained from "expert opinion" using an appropriately designed Questionnaire. The data resulting from such a survey can establish ranges of values, the relative importance of different parameters and can guide us to critical data requirements.

## DISCUSSION

By using a systems approach we have defined the potential pathways for radiation contamination of man through forest ecosystems, and the connection of these pathways with particular forest compartments. In order to determine the level of risk to man over time it is now necessary to construct a mathematical model of the cycling of radionuclides within the forest and the transport of this contamination through the defined pathways to man. The models used to date for radionuclide cycling in forests have implicitly lumped the entire forest into several compartments (i.e. litter, leaves, and soil) without regard for spatial heterogeneity [8]-[17]. This heterogeneity of radionuclide distribution among forest elements along with its location in space may have a large effect on the dose to man. For example, the potential ranges of dose to man from direct gamma shine is certainly effected by the spatial location of radionuclides in relation to man and the concentration of these radionuclides at that particular emitting location. Lateral interaction among forest components may also play a role in the redistribution of radionuclides among forest components and possibly out of the forest. Such processes as leaching of the canopy, wind blown litter and ground dispersion may redistribute radionuclides laterally among the different elements of the forest. Using a spatial model these movements may be captured. Finally, by using a spatial model, different policies regarding forest use may be tested. For example, a possible estimation of the dose to man from a walk through the forest may be accomplished by randomly choosing walking routes and then applying the direct gamma shine model to calculate the subsequent dose. By randomly generating walking routes a probability distribution as a function of dose can be estimated. The same type of analysis may be accomplished in order to estimate the dose to wild animals by the consumption of contaminated vegetation.

To calibrate the proposed models it is necessary to obtain data collected over time from different forest compartments. Some of this type of data does exist from current and past studies [18]-[24] however quite possibly there will be gaps. In order to determine the affect of these gaps on our data requirements both sensitivity and uncertainty analysis will be employed. Sensitivity analysis allows one to estimate the sensitivity of the final answer (i.e. risk to man) with respect to a particular parameter estimate while uncertainty analysis allows for the description of how this estimate impacts the confidence placed in our final answer. This is important in defining areas in which a lack of data may be critical and more research and measurements must be done or in fact further resources to estimate the impact of this parameter may be wasted and unnecessary. The question of how critical each parameter is to the final estimate of risk and how much uncertainty this parameter introduces will be a useful tool in analyzing for "acceptable" data sets.

## CONCLUSIONS

Due to the Chernobyl nuclear reactor release in 1986 and subsequent radiation contamination of surrounding forest ecosystems, the question of what hazards this contamination poses to man is of current concern. At present there does not exist a framework useful for the assessment of risk from the release of radionuclides into forested ecosystems. Thus, our work will contribute to the development of such a framework, to include the structural elements and processes of the forest ecosystem, the factors affecting policy decisions and the interactions between deposited radionuclides in the forest, effect on man, and policies regarding forest usage. The development of such a framework will require a multidisciplinary approach integrating current knowledge and methodologies from such fields as Chemistry, Meteorology, Radiation Health, Forestry and Operations Research. The integration of such different expertise is essential and the authors welcome and appreciate constructive criticism and suggestions.

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# **Radioecological Studies and Modelling**

Part 2

Other Ecosystems, Food Transfer





# **A Comparative Assessment of the Impact of the Windscale and Chernobyl Accidents on Cs-137 Levels of Uplands Lamb in West Cumbria Using the 'RUINS' Model**

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#### ABSTRACT

Preliminary predictions of the  $^{137}\text{Cs}$  contamination of upland lambs in west Cumbria resulting from the Windscale and Chernobyl accident are presented. The predictions are made using the RUINS model and are tentative, particularly for the Windscale accident given the paucity of data available with which the model can be tested. For a typical scenario, whereby upland lambs are fattened on lowland pasture and slaughtered during February  $^{137}\text{Cs}$  activity concentrations in the first year after deposition of  $8000 \text{ Bq m}^{-2}$  are predicted to have been in the region of  $8.8 \pm 3.5 - 28.8 \pm 11.52 \text{ Bq kg}^{-1} \text{ FW}$  for the Windscale accident and  $1.1 \pm 0.5 - 2.8 \pm 1.2 \text{ Bq kg}^{-1} \text{ FW}$  for the Chernobyl accident depending upon grazing pressure. If an earlier slaughter date is assumed then the values increase for the Windscale accident to  $240 \pm 96.0 - 290 \pm 116.0 \text{ Bq kg}^{-1} \text{ FW}$ , but there is little effect for the Chernobyl accident.

For lambs fattened upon upland pasture, currently unusual, but which did occur in 1957: mean  $^{137}\text{Cs}$  activities in 1957/58 have been estimated to be in the range  $2320 \pm 928.0 - 2800 \pm 1120.0 \text{ Bq kg}^{-1} \text{ FW}$ .

In west Cumbria in the first year after each accident the Windscale fire is concluded to have resulted in higher  $^{137}\text{Cs}$  activity concentrations of upland sheep for human consumption than the Chernobyl accident. This was principally due to the timing of the

accidents, but could have been exacerbated significantly by differences in farming practice. However, deposition from the Chernobyl accident was more widespread therefore affecting a greater number of sheep.

## INTRODUCTION

West Cumbria in the United Kingdom is probably unique in having received similar pulses of  $^{137}\text{Cs}$  deposition from two separate nuclear accidents; the Windscale fire in 1957, and the Chernobyl explosion and fire in 1986. The object of this study is to compare the relative impact of these two incidents in terms of  $^{137}\text{Cs}$  contamination of lambs for human consumption.

Caesium-137 from the Windscale fire was principally dry deposited and the actual amount deposited in different areas was influenced by distance from the plant. The distribution and nature of the fallout is reported by Booker (1958) and deposition values, both for typical hill farming areas and lowland pastures, are quoted as  $8000 \text{ Bq m}^{-2}$ . Chernobyl  $^{137}\text{Cs}$  fallout in west Cumbria was wet deposited and exhibited a high degree of spatial variability (Beresford *et al* 1990). This is possibly due to variations in rainfall intensity and/or surface water movement, ie runoff/runon. To enable a direct comparison to be made we have taken a single deposition value of  $8000 \text{ Bq m}^{-2}$   $^{137}\text{Cs}$  for both incidents and pasture types.

Much of the area is upland where the major agricultural activity is sheep farming. Under normal practice lambs are born in late spring (typically May) and will graze on the open fell during the summer. In early to mid-autumn (eg October) all the sheep are generally moved onto improved upland pasture in the vicinity of the farm buildings. Those lambs selected for slaughter are then removed to lowland pasture for fattening over winter. Most will be slaughtered before late spring, typically in February.

There were differences in farming practice at the time of the Windscale fire. Some lambs were fattened at the upland farm rather than being removed to the lowlands. Furthermore the grazing density was generally lower than that currently used (Thompson & Kirby 1990). These two factors, together with the timing of the two accidents, will lead to differences in the contamination of lambs for human consumption.

#### The RUINS Model

In this paper the use of the RUINS model to calculate  $^{137}\text{Cs}$  activity concentrations in lambs' muscle following the two accidents is described. The development and testing of this model has been described elsewhere (Crout et al 1990, in press, pers comm, Galer et al 1990) and only particular points worth mentioning are presented here.

It should be emphasised that the vegetation sub-model is simplistic. In particular the effect of grazing is modelled simply by an increase in vegetation turnover, and thereby an increased rate of  $^{137}\text{Cs}$  loss. In particular the use of a simple rate constant, modified only by rooting density, to describe root uptake can only be justified by the lack of data to support a more sophisticated approach. Experimental work to develop this aspect of the model is currently underway.

The RUINS package can be used to generate crude confidence limits on its predictions based upon uncertainties in the input parameters. Previous experience suggests that uncertainties on the predictions here will be approximately  $\pm 40\%$  1 year after deposition rising to  $\pm 60\%$  4 years after deposition. Nevertheless, whilst there may be some uncertainty in the absolute values predicted, the overall trends and differences between scenarios are unlikely to be significantly affected.

## SCENARIOS

In order to compare the two accidents several farming practice scenarios were used and  $^{137}\text{Cs}$  activity concentration in lambs at slaughter were calculated in each case. Two times for slaughter were used, December and February. Slaughter in February would be normal and is representative for most lambs originating from the uplands. Comparatively few lambs would be slaughtered in December. However this slaughter month has been included to provide an example of an early slaughter time for upland lambs.

Two types of pasture were considered, a typical lowland pasture and a improved upland pasture. Parameter sets to enable the model to simulate these types of pasture have been developed previously from experimental work (Crout et al 1990, pers comm). In each case no consideration was given to the nature or contamination level of the lambs' previous pasture. This simplification was possible because the biological half life of  $^{137}\text{Cs}$  in lambs is relatively short at about 11 days (Howard et al 1987) and therefore any influence of earlier grazing will be negligible, even at the earlier slaughtering date of December.

Interception values assumed in the model for lowland pasture were 0.25 for the deposition from the Windscale accident (Simmonds et al 1979) which agrees with the figures given by Chamberlain (1970) for a herbage density of approximately  $100 \text{ g m}^{-2}$  (DW). For wet deposition from the Chernobyl accident a value of 0.12 was used for the lowland pasture (Fulker 1987). For the upland pasture 0.32 was used based on data from Livens et al (pers comm). Retention on vegetation was assumed to have a half life of 14 days (Simmonds et al 1979).

Two levels of grazing density were used, one nominally high, the other low. The values used differed for the upland and lowland pastures, reflecting the nature of the pasture and are given in Table 1.

Table 1. Grazing Densities (sheep ha<sup>-1</sup>)

Stocking rate	Upland	Lowland
High	10	16
Low	2	5

These scenarios are intended only to demonstrate the influence of various farming options on the contamination of lamb by <sup>137</sup>Cs as calculated by the model. They are not intended as a definitive statement of farming practice either currently or in 1957.

#### RESULTS

The calculated mean lamb muscle <sup>137</sup>Cs activity concentrations are presented in Tables 2(a) and 2(b) for the Windscale accident and Tables 3(a) and 3(b) for the Chernobyl accident. Results are presented for up to 4 years after each accident. For each scenario the calculated mean <sup>137</sup>Cs activity concentrations in lamb at the first slaughter after the Windscale fire are higher than those after the Chernobyl accident.

Lambs fattened on upland pasture are predicted to be consistently more contaminated than those grazed on lowland pasture. Whilst lambs fattened on lowland pasture are virtually uncontaminated one year after both accidents, lambs fattened on upland pasture still remain significantly contaminated 4 years later. There is also a consistent difference between the contamination of lamb from intensively and lightly grazed pasture. In general intensively grazed vegetation is predicted to be less contaminated resulting in lower lamb <sup>137</sup>Cs activities. There is, however, an exception to this in the December slaughter of lamb fattened on upland pasture in the first year after the Windscale fire.

Table 2 Predicted  $^{137}\text{Cs}$  activity concentration for lamb muscle after the Windscale accident, assuming deposition of  $8000 \text{ Bq m}^{-2}$ , based on different scenarios

a)  $^{137}\text{Cs}$  activity concentrations of lambs fattened on lowland pasture (mean  $\pm$  95% confidence limits  $\text{Bq kg}^{-1} \text{ FW}$ )

Year	Time of slaughter			
	DECEMBER stocking rate		FEBRUARY stocking rate	
	High	Low	High	Low
1957/58	240 $\pm$ 96.0	290 $\pm$ 116.0	8.8 $\pm$ 3.52	28.8 $\pm$ 11.52
1958/59	1.0 $\pm$ 0.47	2.0 $\pm$ 0.94	1.0 $\pm$ 0.47	2.0 $\pm$ 0.94
1959/60	1.0 $\pm$ 0.54	2.0 $\pm$ 1.18	1.0 $\pm$ 0.54	2.0 $\pm$ 1.18
1960/61	1.0 $\pm$ 0.60	2.0 $\pm$ 1.20	1.0 $\pm$ 0.60	2.0 $\pm$ 1.20

b)  $^{137}\text{Cs}$  activity concentrations of lambs fattened on upland pasture (mean  $\pm$  95% confidence limits  $\text{Bq kg}^{-1} \text{ FW}$ )

Year	Time of slaughter			
	DECEMBER stocking rate		FEBRUARY stocking rate	
	High	Low	High	Low
1957/58	2800 $\pm$ 1120	2320 $\pm$ 928	1860 $\pm$ 750	1900 $\pm$ 76
1958/59	390 $\pm$ 180	480 $\pm$ 2266	360 $\pm$ 171	440 $\pm$ 210
1959/60	330 $\pm$ 179	370 $\pm$ 199	330 $\pm$ 180	360 $\pm$ 194
1960/61	310 $\pm$ 188	340 $\pm$ 206	310 $\pm$ 186	340 $\pm$ 204



Table 3 Predicted  $^{137}\text{Cs}$  activity concentration for lamb muscle after the Chernobyl accident, assuming deposition of  $8000 \text{ Bq m}^{-2}$ , based on different scenarios

a)  $^{137}\text{Cs}$  activity concentrations of lambs fattened on lowland pasture (mean  $\pm$  95% confidence limits  $\text{Bq kg}^{-1}$  FW)

Year	Time of slaughter			
	DECEMBER stocking rate		FEBRUARY stocking rate	
	High	Low	High	Low
1986/87	1.2 $\pm$ 0.5	5.0 $\pm$ 2.2	1.1 $\pm$ 0.5	2.8 $\pm$ 1.2
1987/88	1.2 $\pm$ 0.6	1.9 $\pm$ 1.0	1.1 $\pm$ 0.6	1.9 $\pm$ 1.0
1988/89	1.2 $\pm$ 0.7	1.8 $\pm$ 1.1	1.1 $\pm$ 0.7	1.8 $\pm$ 1.0
1989/90	1.2 $\pm$ 0.7	1.8 $\pm$ 1.2	1.1 $\pm$ 0.7	1.8 $\pm$ 1.2

b)  $^{137}\text{Cs}$  activity concentrations of lambs fattened on upland pasture (mean  $\pm$  95% confidence limits  $\text{Bq kg}^{-1}$  FW)

Year	Time of slaughter			
	DECEMBER stocking rate		FEBRUARY stocking rate	
	High	Low	High	Low
1986/87	792 $\pm$ 356	1008 $\pm$ 453	557 $\pm$ 250	780 $\pm$ 327
1987/88	356 $\pm$ 188	406 $\pm$ 214	348 $\pm$ 184	492 $\pm$ 207
1988/89	331 $\pm$ 201	363 $\pm$ 220	327 $\pm$ 198	359 $\pm$ 218
1989/90	315 $\pm$ 213	343 $\pm$ 231	311 $\pm$ 210	339 $\pm$ 229

## DISCUSSION

We should emphasise the tentative nature of the results presented above. They are model predictions and no more reliable than the model itself. Whilst this has shown encouraging preliminary agreement with field data (Crout *et al* pers comm) it has not been fully tested in all respects.

One clear conclusion of this work is that in the December or February following the Windscale accident the  $^{137}\text{Cs}$  activity concentrations in lambs originating from the uplands were higher than in the corresponding months after the Chernobyl accident. The reason for this is clear. It is simply that more time elapsed between the deposition of Chernobyl  $^{137}\text{Cs}$  and slaughtering than was the case after the Windscale fire. Vegetation  $^{137}\text{Cs}$  activities therefore had longer to decline.

For Chernobyl even in the 'worst case' scenario for lowland pasture (of a low grazing pressure and lambs slaughtered in December) sheep  $^{137}\text{Cs}$  activity concentrations are predicted to be less than  $10 \text{ Bq kg}^{-1}$  FW (fresh weight), which is well below the UK action level of  $1000 \text{ Bq kg}^{-1}$  FW (total  $^{137}\text{Cs}$  activity). It can therefore be concluded that in the case of the Chernobyl accident there would be few problems with lowland pastures. Indeed no lowland farms in west Cumbria are currently restricted following the Chernobyl accident.

The situation in 57/58 was different. Animals slaughtered in December are predicted to have had mean  $^{137}\text{Cs}$  concentrations between  $240 - 290 \text{ Bq kg}^{-1}$  FW depending upon grazing pressure. By February this range would have declined to  $9 - 29 \text{ Bq kg}^{-1}$  FW. These values are all at least 6 times higher than for the Chernobyl situation. Had current regulations been in force in 1957 restrictions may have been placed upon lowland farms in the affected area. This is because the model calculates mean  $^{137}\text{Cs}$  activities. Current UK regulations require that no single sheep above  $1000 \text{ Bq kg}^{-1}$  FW should be slaughtered. Within a flock there can be great variability in  $^{137}\text{Cs}$  levels, some animals consistently maintaining a higher level than their counterparts.

Therefore a mean level of 240 - 290 Bq kg<sup>-1</sup> FW does not guarantee that no action will be required. One or two animals may be found at or above the 1000 Bq kg<sup>-1</sup> FW limit. In subsequent years contamination levels are predicted to have been negligible, being consistently below 10 Bq kg<sup>-1</sup> FW. This fall is due to the large fixation capacity of most lowland soils for caesium, rendering it unavailable for plant uptake.

The lack of data on sheep <sup>137</sup>Cs contamination following the Windscale fire makes it difficult to comment on the validity of the models predictions. However from data in Booker (1958) we have calculated the <sup>137</sup>Cs activity concentrations in lamb from <sup>137</sup>Cs levels in vegetation quoted in February for a pasture at Seascale. Taking Bookers' value of 78 Bq m<sup>-2</sup> and assuming a vegetation biomass of 100 g m<sup>-2</sup> (DW), a transfer coefficient for year old lamb muscle of 0.33 (Howard 1989) and a daily herbage intake of 1 kg DW, we calculate an equilibrium <sup>137</sup>Cs value for lamb of 260 Bq kg<sup>-1</sup> FW. This is higher than our predicted range of 9 - 29 Bq kg<sup>-1</sup> FW and may be due to differences in the soil type at Bookers' sample site compared with the soil used in our experiments to produce the model. Alternatively the underprediction of the model may be due to the use of interception and retention half-lives on vegetation which are too low. However, the high activity of vegetation found by Booker may partly be due to contamination of vegetation by soil. For pastures around Sellafield, soil has been shown to account for up to 50% of the dry weight of vegetation samples in winter months (Beresford & Howard pers comm).

The difference between the consequences of the two accidents is due largely to their relative timing. The Chernobyl accident occurred in late April, which is almost the ideal time for standard UK hill-farming practice. The Windscale accident occurred in October and was therefore much closer to the times of slaughtering. Even so its consequences were mild compared with the potential impact of a comparable accident occurring when fattened lamb is slaughtered, between December and April. However, our analysis has been confined to upland sheep and we should bear in mind that lowland lambs would be slaughtered earlier.

The scenario envisaged whereby lambs would be fattened on upland pasture is not a common agricultural practice nowadays and therefore the values given for the Chernobyl accident in this case are unlikely to be of much significance. However some lambs may have been fattened in this way during the period when the Windscale accident occurred and these will be relevant. In this case the high availability for uptake of  $^{137}\text{Cs}$  from these upland soils with a comparatively high organic content means that  $^{137}\text{Cs}$  levels in vegetation, and hence sheep, decline more slowly than in lowland pastures.

As the model is presently constructed it predicts that animals from intensively grazed pasture will be less contaminated than those from partially utilized pasture. This is because  $^{137}\text{Cs}$  activity in vegetation has two loss mechanisms in the model, removal by grazing and loss by tissue senescence and death. The fraction of vegetation loss via senescence falls as grazing increases. At full utilization the rate of vegetation removal is greater than at zero grazing and therefore the rate of  $^{137}\text{Cs}$  loss from the vegetation compartment increases with increasing grazing pressure. As the rate of uptake from soil solution is constant in the model increasing grazing pressure results in a lower  $^{137}\text{Cs}$  content in the vegetation compartment. This effect is partially masked when vegetation activity is expressed as a concentration. An intensively grazed pasture will have less standing biomass than an ungrazed pasture and this is accounted for in the model. Nevertheless the model suggests a general decrease in vegetation  $^{137}\text{Cs}$  activity concentrations for a given site as grazing intensity increases. This is illustrated in Fig 1 which shows vegetation activity concentration resulting from an arbitrary  $1 \text{ Bq m}^{-2}$  deposition event for a pasture with several levels of grazing. Whilst these results are somewhat surprising there is experimental field evidence to provide qualitative support (Salt & Mayes in press). However it MUST be emphasized that these aspects of the model are the least well developed and further work is in progress. At this stage of the model development these results should therefore be treated with caution.

The results in Table 2(b) show an exception to the general rule that high grazing pressure leads to lower lamb contamination in the scenario where lambs are fattened on upland pasture during the first year after the Windscale accident. This is because slaughtering is assumed to have occurred only shortly after the accident and therefore some initial deposit may still be present on vegetation surfaces. Such deposit may be less available for absorption by the sheep (Howard 1989). For a given amount of deposit any situation which results in a higher ratio of initial deposit to root uptake of  $^{137}\text{Cs}$  into vegetation should result in lower sheep  $^{137}\text{Cs}$  activity concentrations. In the period following deposition high grazing pressure will accelerate the removal of initial deposit increasing the rate at which  $^{137}\text{Cs}$  becomes available for root uptake, recycled via faeces and urine. This mechanism leads to the anomalies in Table 2(b).

#### CONCLUSIONS

These results must be considered as only tentative, but taking them at face value we can draw the following conclusions.

1. For a typical scenario whereby upland lambs are fattened on lowland pasture and slaughtered during February  $^{137}\text{Cs}$  activity concentrations in the first year after deposition are predicted to have been in the region of 9 - 29  $\text{Bq kg}^{-1}$  FW for the Windscale accident and 1.1 - 2.8  $\text{Bq kg}^{-1}$  FW for the Chernobyl accident depending upon grazing pressure. If an earlier slaughter date is assumed then the values increase for the Windscale accident to 240 - 290  $\text{Bq Kg}^{-1}$  FW, but there is little effect for the Chernobyl accident.
2. For lambs fattened upon upland pasture, currently unlikely, but which occurred in 1957: mean  $^{137}\text{Cs}$  activities in 1957/58 have been estimated to be in the range 2320 - 2800  $\text{Bq kg}^{-1}$  FW depending upon grazing pressure.
3. For both accidents the  $^{137}\text{Cs}$  levels remain higher for longer for the lambs fattened on upland pasture compared with those fattened on lowland pasture.

4. In west Cumbria in the first year after the accident the Windscale fire is concluded to have resulted in higher levels of contamination in sheep originating from the uplands than that due to the Chernobyl accident. This was principally due to the timing of the accident, but could have been exacerbated significantly by differences in farming practice. However, Chernobyl contamination was more widespread over the United Kingdom and therefore affected a greater number of sheep.

Whilst the RUINS model is still undergoing development it is already clear that mechanistic models of this type could play a very useful role in identifying agricultural management practices which can help reduce the radionuclide contamination levels in foodstuffs. For brevity we have concentrated here on upland sheep systems, but the model can be equally well applied to the case of lowland sheep, beef and milk production. It could also be readily extended for other animals, such as goats and deer which are important elements in the food chain in other countries.

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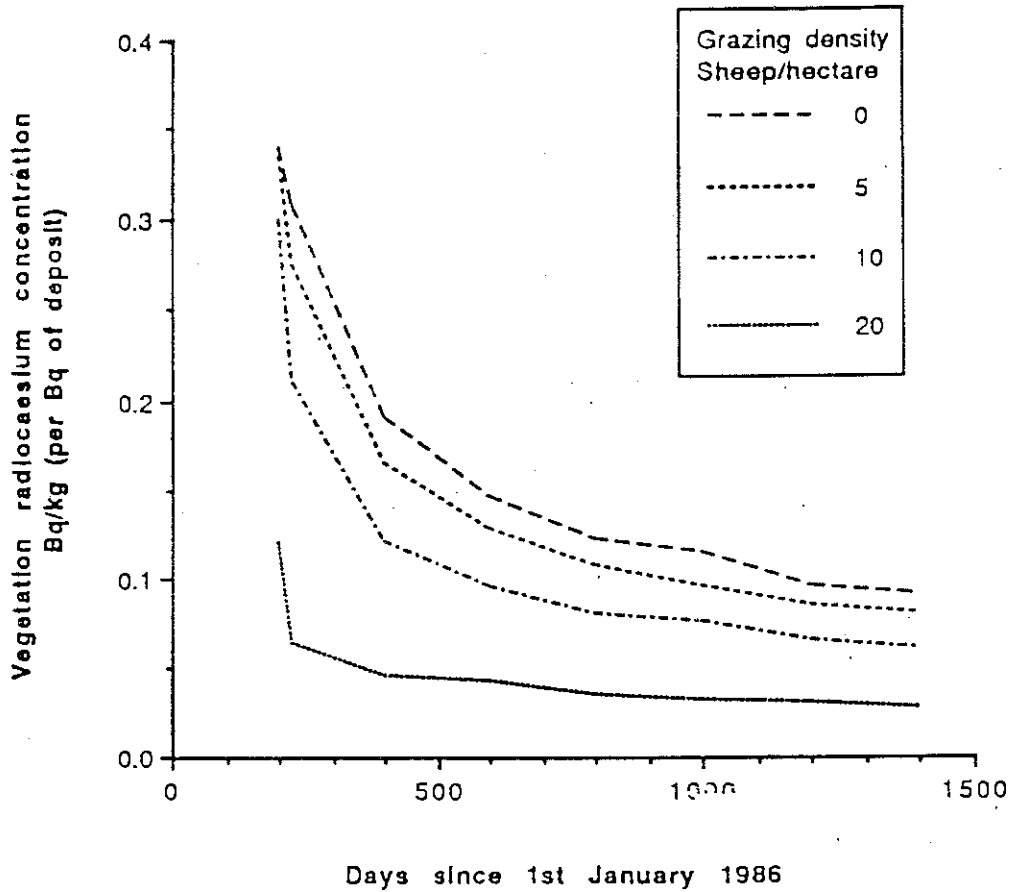


Figure 1 Predicted radiocaesium activity concentrations in vegetation resulting from an arbitrary 1 Bq/m<sup>2</sup> deposition event for a pasture with different grazing densities.

# **Concentration of Radiocaesium in Grain Following the Chernobyl Accident**

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## ABSTRACT

Radioactivity measurements and dose assessments in Hungary after the Chernobyl accident have shown that the consumption of baker's ware contributes significantly to the internal dose of man. Flour and bread have been contaminated mainly due to radiocaesium deposition onto the of cereals at the end of April and beginning of May, 1986.

Because of the different seasonal and growing conditions of biomass, the interception fraction of the standing winter wheat became higher than that of the summer wheat. Therefore, the contribution of grain to the internal dose was relatively high in Hungary where near to 90 per cent of flour and bread is produced from winter wheat in comparison with other countries.

The average concentration of  $^{137}\text{Cs}$  in winter grain harvested in summer 1986 was 32 Bq/kg with a range of 11-140 Bq/kg. The bran contained almost half of the total radiocaesium of the grain with about 20 per cent of the weight. The  $^{40}\text{K}$  concentration of grain was 149 Bq/kg. The  $^{137}\text{Cs}$  concentration in white bread commonly used in Hungary was 22 Bq/kg in average.

The concentrations found in bread samples from the whole country showed a high variation due to the uneven deposition of radioactive substances. The  $^{137}\text{Cs}$  concentration in winter grain was  $0.0075 \pm 0.0017$  Bq/kg normalized to 1 Bq/m<sup>2</sup> deposition density. The  $^{137}\text{Cs}$  concentration in grain harvested in 1987 or later became less than 1 Bq/kg. It suggests that the root uptake of radiocaesium by cereals must be very small.

The per caput committed effective dose equivalent due to consumption of baker's ware was estimated as 50 micro-Sv.

## INTRODUCTION

Following the accident at the Chernobyl nuclear power plant in April, 1986 the contamination of the vegetation was mainly due to the deposition of radioactive substances from the air to the surface [1,2]. Therefore, the plants with higher interception factor during the first post-accident days contained greater radioactive contamination than those with less interception. The difference between the interception ratio of the standing winter and that of the summer wheat has influenced mainly the radiocaesium activity in the grain and in the baker's ware, too. Other pathways for contamination of the grain may be the rainsplash and the root uptake. Both of them were negligible in comparison with the direct deposition onto the plants, in 1986. The human intake of the radiocaesium and the committed effective dose equivalent due to the consumption of baker's ware are determined on the basis of the radiocaesium concentration in the foodstuffs and the consumption rates.

## METHODS

Grain, flour and bran samples in the period of 1986-90 were taken from mill-works round the whole country partly by the National Institute of Food Hygiene and Nutrition and partly by the Trust of Grain Industry. The  $^{137}\text{Cs}$  concentrations of the samples were measured by semiconductor-detectors in Marinelli beakers. The total deposition of  $^{137}\text{Cs}$  on the ground surface was determined by in situ gamma-spectrometry and by concentration measurements in soil samples [3,4,5]. To estimate the per caput committed effective dose equivalent, the AGEDOS age-dependent methodology [6] was used. The consumption rates were taken from the Year-Books of the Hungarian Bureau of Statistics.

## RESULTS

Deposition of the radiocaesium was strongly inhomogeneous in Hungary, depending mainly on the local rainfall. Therefore, we have normalized the contamination of the winter wheat, flour and bran to the site specific deposition density of  $^{137}\text{Cs}$ . *Figure 1.* shows the  $^{137}\text{Cs}$  concentration in the grain in relation to the deposition density together with the confidence intervals (65 and 95 %-age). The slope of the simple regression line is  $6.50 \pm 1.46$ , the interception is  $10.5 \pm 7.1$  and the correlation coefficient is  $r=0.536$ . The slope differs from zero significantly with a probability level of  $p < 0.001$ , but the interception does not ( $p=0.15$ ).

The distribution of  $^{137}\text{Cs}$  contamination in different components of the grain is presented in *Table I.*

According to the mean values, it seems that the bran contains the largest part of the radiocaesium, similarly to the distribution of potassium, in spite of the fact that the weight of bran represents only about 20 % of the total weight. The regression analysis shows a weak correlation between the  $^{137}\text{Cs}$  concentration (normalized to the unit deposition) and the  $^{40}\text{K}$  concentration in the wheat ( $r=0.435$ ).

The concentration of radiocaesium in the rye (harvested in 1986) was similar to that in the presented winter wheat. The samples of oat and maize sown in spring contained  $^{137}\text{Cs}$  concentrations of 1-10 Bq/kg. All the grain samples harvested before the Chernobyl accident and after 1986 show concentrations of  $^{137}\text{Cs}$  less than 1 Bq/kg, while the soil in the root zone contains it in a range of 10-150 Bq/kg.

The committed effective dose equivalent was calculated based on the  $^{137}\text{Cs}$  concentration in bread and the consumption rate of bread. *Figure 2.* shows the dose contribution of the cereals, mainly from winter wheat, in comparison with other exposure pathways. The dose due to the radiocaesium fallout and assessed from activity concentrations in and consumption rates of foodstuffs has overpassed the dose calculated from the whole body  $^{137}\text{Cs}$  measurements, by a factor of near to 2.

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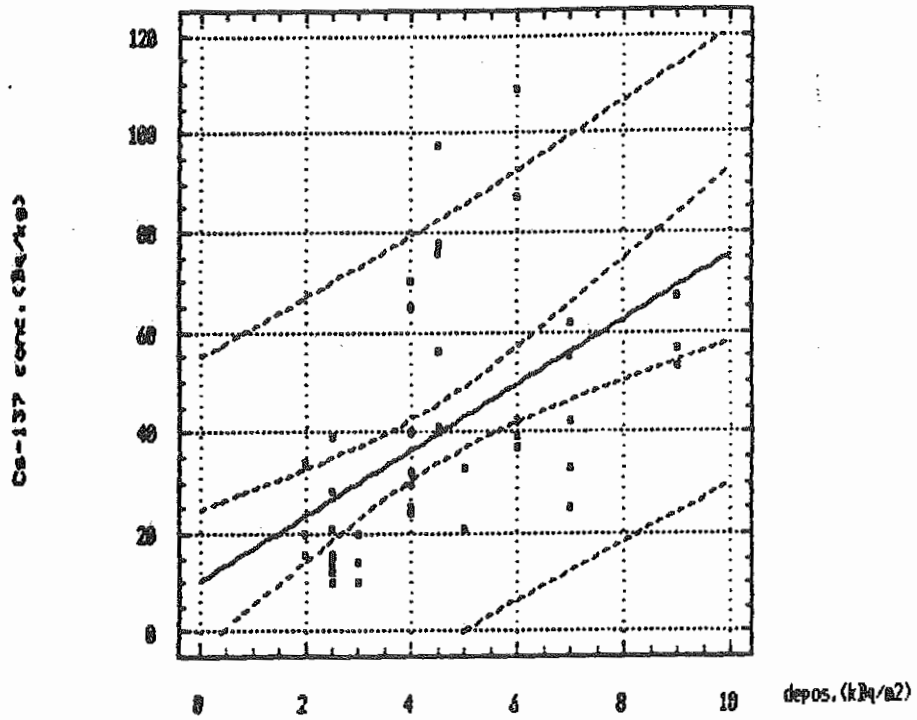


Figure 1. The  $^{137}\text{Cs}$  concentration in winter wheat (Bq/kg) depending on the  $^{137}\text{Cs}$  deposition ( $\text{kBq}/\text{m}^2$ ). Samples taken from twenty different regions of Hungary

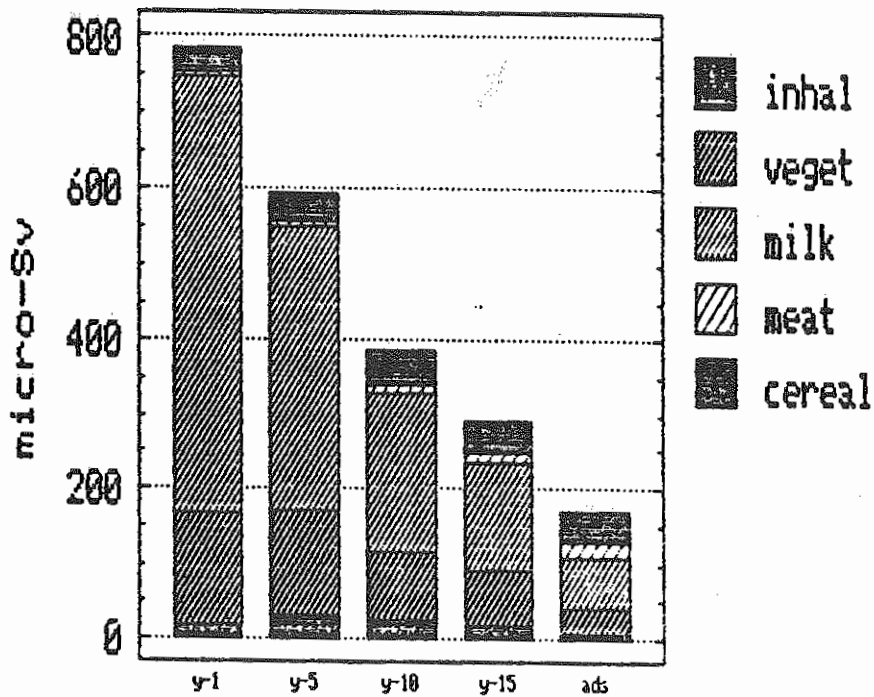


Figure 2. The average values of the committed effective dose equivalents ( $\mu\text{Sv}$ ) received on various intake pathways by different age groups (1-, 5-, 10-, 15-years old) of children and adults

Table I. The mean values and standard deviations of  $^{40}\text{K}$  and  $^{137}\text{Cs}$  concentrations in whole wheat, flour and bran. (In brackets the number of samples)

	wheat-grain	wheat-flour	wheat-bran
$^{40}\text{K}$ (Bq/kg)	149±42 (75)	45.8±16.6 (27)	397±17 (8)
$^{137}\text{Cs}$ (Bq/kg:kBq/m <sup>2</sup> )	9.04±4.77 (51)	4.41±1.83 (27)	26.3±10.4 (9)
ratio of mean values	16	10	15





# **Cs-134 Transfer from Water or Food to the Ciprinid Tinca Tinca Linnaeus: Uptake and Loss Kinetics**

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ABSTRACT

Experiments with  $^{134}\text{Cs}$  and the fish Tinca tinca Linnaeus (fam. Cyprinidae), as a part of a more extensive work, concerning a simplified freshwater trophic chain using water from Fratel dam, (at Tejo River), were undertaken. The whole work concerns the Contracts (CCE) nº BI6 - B - 198 - P and nº BI6 - 0245 - P.

Direct uptake from water, during a period of about 30 days, leads to a kinetics expressed by the power function:  $\text{CF}_{(t)} = 0.58 t^{0.781}$  (t in days), the concentration factor (CF) referred to wet weight.

Retention study, showed the existence of two biological half-lives,  $\text{Tb}_1 = 7$  days and  $\text{Tb}_2 = 87$  days, which might concern respectively, the  $^{134}\text{Cs}$  desorption from the transit organs and the loss of the assimilated isotope from the storage organs. The retention function (%) is:

$$R_{(t)} = 25 e^{-0.104t} + 75 e^{-0.008t} \quad (t \text{ in days}).$$

In the accumulation through the food chain, using planktonic crustacean Daphnia magna Straus (Cladocera) as prey, a transfer factor (TF) related to wet weight of both fish and prey, is estimated through the power function:  $\text{TF}_{(t)} = 0.022 t^{0.578}$  (t in days).

Finally, the retention study following the food pathway contamination, stresses the existence of one long term component, with hal-life  $\text{Tb} = 61$  days. Retention function (%) is expressed by one single exponential:

$$R_{(t)} = 87 e^{-0.0113t} \quad (t \text{ in days}).$$

The transfer factor kinetics seems to point out to a rather slow process, leading to lower  $^{134}\text{Cs}$  concentration values, than the contamination through the water.

The loss of the assimilated  $^{134}\text{Cs}$ , uptaken through both pathways, water or food, is a slow process. The longer biological hal-life is very important in Radiological Protection, once it may be attributed to the radionuclide loss from the muscular mass.

separating feces from the water and then filtering all the water through paper filter, retaining the non-ingested material where total  $^{134}\text{Cs}$  content was also determined.

At the end of experiment, retention factor, i.e., the ratio total  $^{134}\text{Cs}$  content in fish / total  $^{134}\text{Cs}$  ingested, was  $0.0364 \pm 0.0219$ . The transfer factor (TF) defined as the ratio  $^{134}\text{Cs}$  concentration in fish ( $\text{Bq g}^{-1}$ , w.w.) /  $^{134}\text{Cs}$  concentration in food ( $\text{Bq g}^{-1}$ , w.w.), shows a kinetics represented in Figure 4 according to power function:  
$$\text{TF}_{(t)} = 0.022 t^{0.578} \quad (t \text{ in days}).$$

In a bibliographic review concerning radioactive cesium transfer factors in freshwater fish [16], the following data are referred: Lebistes reticulatus / Daphnia magna - 0.67 (D.magna contaminated through the water); Lepomis macrochirus / D.magna - 0.06 and 0.44 (D.magna contaminated respectively through water and labelled food); Cyprinus carpio / D.magna - 0.29 and 0.001 (D.magna contaminated through water).

The transfer factor kinetics seems to point out to a rather slow process, leading to lower  $^{134}\text{Cs}$  concentration values, than the contamination through the water. On the other hand it is referred that the contamination through the trophic transfer is a more efficient process [6]. The reluctance that fish showed for the acceptance of dried daphnids as food, may have affected the transfer factor evaluation.

However, the evaluated retention factor is very close to  $0.034 \pm 0.004$ , obtained in similar experimental conditions, dried daphnids given to carps [7].

### 3.2.2 Retention

Retention and loss data refer only to three fishes, although the contamination period has been initiated with more specimens, some of them having died. The mean CF in fish was 0.026 at the end of a 13 days uptake period, through labelled dried daphnids, meaning a cesium concentration in fish of  $5.1 \text{ Bq g}^{-1}$ , for 9.7 g of total weight. The loss experiment lasted for 45 days.

Although no significant concentration of the radionuclide in the fishes was observed, it remained strongly attached and only a single half-life has been determined,  $T_b = 61$  days.

## 1. INTRODUCTION

The radioecological assessment studies made for the case of the normal operation of the nuclear power plants or for the case of an accident, which purpose is the evaluation of the effective dose equivalent to man, are based on the concentration and transfer factors in the terrestrial and aquatic ecosystems. The knowledge of the specific site factors is of the utmost importance. Our interest on the Fratel dam in Tejo River, only a few kilometers from the river entrance in Portugal, comes from the fact that in Spain there are three nuclear power plants at Tejo River banks or tributaries. Therefore, a CEC Contract (nºBI6-B-198-P) on the study of the Radioecology of Tejo River was settled. Besides this problem, under the scope of the CEC programme "Post-Chernobyl Actions", the contract (CEC) nº BI6-0245-P was established, whose objective was the study of the radiocesium transfer in that particular freshwater ecosystem.

$^{134}\text{Cs}$  was chosen, because of the importance it presented following the Chernobyl accident, and because it may also be present in the liquid effluents from the nuclear power plants equipped with pressurized water reactors.

An experimental study concerning the  $^{134}\text{Cs}$  uptake and loss in a simplified trophic chain (a microalgae, a small filter feeding crustacean and an omnivorous fish) in Fratel dam water was undertaken, a radioecological model being the final purpose.

This paper concerns the  $^{134}\text{Cs}$  transfer from water and from food (crustacean Daphnia magna Straus) to the fish Tinca tinca Linnaeus. Retention processes were studied and biological half-lives were evaluated.

The  $^{134}\text{Cs}$  transfer from water to the microalgae Selenastrum capricornutum Printz (Chlorophyceae), as well as the transfer from water and food (S. capricornutum) to the planktonic crustacean D. magna (Cladocera), have been subjected to former studies [1], [2].

## 2. METHODOLOGY

The fish concerned in the experimental food chain under study is Tinca tinca Linnaeus (Sub-class Teleostei, family Cyprinidae), an omnivorous species. The tenches used in the experiments were young fishes of about one year old.

The planktonic crustacean Daphnia magna Straus (Cladocera), a filter feeder that may reach 5 mm in length [3], was used as prey organism.

The water used in the experiments was collected from the Fratel dam and its chemical composition is presented in Table 1. The stable cesium content, determined by INAA, was  $6 \times 10^{-5}$  mg l<sup>-1</sup> [4]. For the experiments, water was filtered through ashless cristalyne retention paper.

<sup>134</sup>Cs was used under the form of cesium chloride, therefore at the ionic form Cs<sup>+</sup>.

The experimental basins were kept at the temperature of  $20 \pm 2^{\circ}\text{C}$ ; aeration through an air pump and a 15 hours day-light period, were provided.

In the uptake experiments through <sup>134</sup>Cs labelled water, the tenches were fed five days a week with 30 frozen daphnids per fish. Food was quickly eaten, therefore its contamination in the radioactive water was avoided.

The water radioactivity was kept stable by the addition of distilled water, when necessary, to compensate any evaporation. Fecces were regularly removed to avoid their ingestion as cesium labelled material.

The uptake experiments through the <sup>134</sup>Cs labelled food, D.magna, were carried out keeping fish into individual basins with 1 liter of inactive water and feeding them with four meals a week, consisting of <sup>134</sup>Cs labelled dried daphnids. Water was renewed each time a new meal was furnished and also before each weekend.

In what concerns the retention experiments, fishes followed previously a contamination period, through the water or the food, being afterwards transferred into basins with inactive water, which was renewed five times a week. They were regulary fed with non-contaminated daphnids.

At each sampling time, tenches were previously washed in inactive water, then they were anesthetized in a MS222 (SANDOZ) aqueous solution ( $0.5 \text{ g l}^{-1}$ ), and weighed. Specimens were put in individual plastic contain-

ers, filled with water to avoid asphyxia, for the radioactivity measurements. The fish radioactivity was evaluated in relation to a standard of similar geometry. Water radioactivity was determined in soluble and in particulate phases by filtration through 0.45  $\mu\text{m}$  Millipore membranes.

The detection system consisted on a 4"x 4" well type NaI(Tl) detector (the well is 1 1/4" x 2 1/2") connected to a multichannel analyser.

### 3. RESULTS AND DISCUSSION

#### 3.1 Contamination through the water

##### 3.1.1 Uptake

The experiment was carried out in two aquaria. In aquarium A 6 tenches were kept in 12 liters of water with an initial radioactivity of  $3.1 \times 10^4 \text{ Bq l}^{-1}$ ; 2.63 g was the average fish weight, resulting in  $1.32 \text{ g l}^{-1}$  of initial biomass. In aquarium B 8 tenches were kept in 8 liters of the same water, whose initial radioactivity was of  $4.0 \times 10^4 \text{ Bq l}^{-1}$ ; the average fish weight was 4.09 g and the initial biomass  $4.09 \text{ g l}^{-1}$ . The experiment runned for 32 and 34 days respectively.

Concentration factor (CF), defined as the ratio  $^{134}\text{Cs}$  concentration in fish ( $\text{Bq g}^{-1}$ , wet weight) /  $^{134}\text{Cs}$  concentration in water ( $\text{Bq ml}^{-1}$ ), was evaluated. At the end of the experiment CF equilibrium has not been reached in any of the two fish groups.

Figure 1 shows the CF kinetics for the aquarium A and each point represents the mean value from three fishes sampled at random, while for the aquarium B, Figure 2, all fishes were measured at each sampling time, each point being a mean of eight values. The individual differences are rather evident. A certain fluctuation in the water radioactivity was observed, which might have been due either to the adsorption on the basins surface or to the evaporation effect, besides the radioactive transfer into the fish.

The adjustment of data to a power function model [5], led to  $\text{CF}_{(t)} = 0.31 t^{0.695}$  and  $\text{CF}_{(t)} = 0.75 t^{0.707}$ , respectively for aquarium A and aquarium B, (t in days).

Due the similarity of the experimental conditions and results, an adjustment was done using the simultaneous values from both cases and a joint

analytical model was estimated (for  $t=31$  days):  $CF_{(t)}=0.58 t^{0.781}$ .

According to a bibliographic study [6] concerning the radioactive cesium transfer studies in freshwater fish, the concentration factor in the whole body ranges from 0.3 to 17, where the values we determined are well fitted.

The accumulation of the radioactive cesium is dependent on the potassium content either in water or in fish, because those elements are chemically similar, following the same metabolism. The  $K^+$  concentration in Fratel dam water (Table 1) is higher than other values in the literature, 1.1 to 2.1  $mg\ l^{-1}$  in Rhone River [7], and 1.7  $mg\ l^{-1}$  an average value for the European rivers [8]; the  $K^+$  concentration evaluated in Cyprinidae from Fratel dam presents a mean value of 1.97  $mg\ kg^{-1}$  (w.w.) [9], very similar to the values 2  $mg\ kg^{-1}$  for tenches and 1.9  $mg\ kg^{-1}$  for carps (both w.w.) given respectively in [7] and [10].

Therefore, very likely, the  $K^+$  content in water might act on a chemical competition with the  $^{134}Cs$ , decreasing its accumulation in the organism.

Another important physiological aspect is that the internal medium of freshwater fish is hypertonic in relation to the water, therefore they do not drink much water and the ionic absorption at gills and skin is very reduced [11].

### 3.1.2 Retention

This experience, with 4 specimens whose initial mean weight was 9 g (7.0 to 10.9 g), lasted for 92 days. In Figure 3 is shown the evolution of the retention, in percentage of the initial radioactivity concentration, each point representing the mean  $^{134}Cs$  retention of the four fishes.

The retention analysis, showed that data fit to an exponential model with two rate functions:  $R_{(t)}=25 e^{-0.104t}+75 e^{-0.008t}$  (t in days).

The compartment of low retention (25%), where  $^{134}Cs$  half-life is  $Tb_1=7$  days, may be interpreted as corresponding mainly to the transit organs (blood, digestive tract, kidneys, gills and liver) and also to the skin where the radionuclide is accumulated by a simple adsorption process. The compartment of high retention (75%),  $Tb_2=87$  days, might correspond to the storage organs, mainly the muscular tissue, where the metabolized radio-



nuclide was concentrated. This situation has already been recognized by several authors [7], [12], [13], [14], [15].

From a bibliographic study [6] in experiments where the contamination pathway was the water, the following range of values was compiled:  $Tb_1$  from 2 to 25 days and  $Tb_2$  from 20 to approximately 400 days, where the weight of the individuals and water temperature played an important role. The biological half-life increases with the age and with the decreasing temperature.

### 3.2 Contamination through the throphic transfer

#### 3.2.1 Uptake

A group of 10 tenches about 1 year old, with an initial mean weight of 2.5 g (0.7 to 3.7 g) was set up into individual containers for a continued ingestion of  $^{134}\text{Cs}$  labelled dried daphnids.

The contaminated daphnids were collected, dried, weighed and their radioactivity measured. A conversion factor of 1/13.89 was experimentally determined [2] to obtain  $^{134}\text{Cs}$  concentration in  $\text{Bq g}^{-1}$  wet weight, from the dry weight measurements. Portions were prepared to serve as labelled fish meals. Each portion was first suspended into 1 liter of water, for about 3 hours, and its radioactivity was measured before being supplied to a fish. With this previous operation, about 90% of the total cesium concentrated in the dried daphnids was lost to the water, the remaining 10 % being premanently retained. So, the water in the individual containers never became contaminated.

The ingestion of dried daphnids has shown to be accepted with reluctance. Cases have been where an individual meal stayed untouched for more than one day. Also, some times much of the material was rejected after having been taken into the fish mouth; as a result a general weight decrease, along the experiment, was observed. Wide individual differences concerning food intake. and cesium uptake were noticed.

The mean daily ingestion was of  $0.435 \pm 0.183$  g (wet weight), while the mean  $^{134}\text{Cs}$  concentration in food was of  $98.7 \pm 13.7$   $\text{Bq g}^{-1}$  (w.w.).

The cesium concentration in the fishes was regularly measured, along with the determination of the previously ingested material. This was made by

Scattering of experimental points is wide, which may be due to the relatively low radioactivity in fish. So, these values led to a rough adjustment ( $r=0.729$ ), Figure 5, which is expressed by the retention function:

$$R_{(t)} = 87 e^{-0.0113t} \quad (t \text{ in days}).$$

According to a literature review [6], regarding the throphic transfer of radioactive cesium in fish, some authors stated that a short-term component was practically inexistent, while others mentioned a quite evident multicompartmental system. The recognition of one single retention compartment has been mentioned [5] for Cyprinus carpio fed with  $^{137}\text{Cs}$  labeled dried daphnids, the biological half-lives in two parallel experiments being 25 and 45 days.

Nevertheless, the evidence of only one long term retention component may be admitted, because a redistribution of the radioactive cesium may happen: some organs, like the digestive tract, may very likely loose radioactivity, which may be concentrated in other organs mainly the muscles [17].

#### 4. CONCLUSIONS

It is evident from the present results that the direct  $^{134}\text{Cs}$  uptake from water, is more important than the food chain pathway, when dried daphnids are used as a prey. Actually this is what generally happens in short-term experiments.

The biological half-lives evaluated in this paper are well fitted within the bibliographic values.

The loss of the assimilated  $^{134}\text{Cs}$ , uptaken by both pathways, water or food, is a slow process, the evaluated longer half-lives being respectively 87 and 61 days.

Therefore it appears that the assimilation would be the more important  $^{134}\text{Cs}$  concentration mechanism.

The longer biological half-life is very important in Radiological Protection, once it may be attributed to loss of the radionuclide from the muscular mass, as it behaves like a strong retention compartment.

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Table 1  
Characteristics of the water from Fratel dam (1987-1989)

Chemical components	Concentration (mg l <sup>-1</sup> )		
	Geometric mean	Minimum - maximum	
Ca <sup>2+</sup>	44.0	25.0	- 67.0
Mg <sup>2+</sup>	12.0	7.9	- 22.0
Na <sup>+</sup>	20.3	9.3	- 27.0
K <sup>+</sup>	2.69	0.90	- 6.5
NH <sub>4</sub> <sup>+</sup>	0.299	0.034	- 0.772
Cl <sup>-</sup>	28.8	11.0	- 48.0
SO <sub>4</sub> <sup>2-</sup>	58.8	25.0	- 97.0
NO <sub>3</sub> <sup>-</sup>	1.05	0.272	- 2.46
NO <sub>2</sub> <sup>-</sup>	0.003	0.002	- 0.014
P <sub>2</sub> O <sub>4</sub> <sup>-</sup>	0.35	0.88	- 0.32
Total Fe	0.116	0.029	- 0.340
Total P	0.564	0.380	- 0.95
Total N	3.31	0.92	- 12.0
<hr/>			
Total Hardness (in mg l <sup>-1</sup> )	133.0	56.0	- 238.0
pH	7.8	6.9	- 8.9

Legends of figures

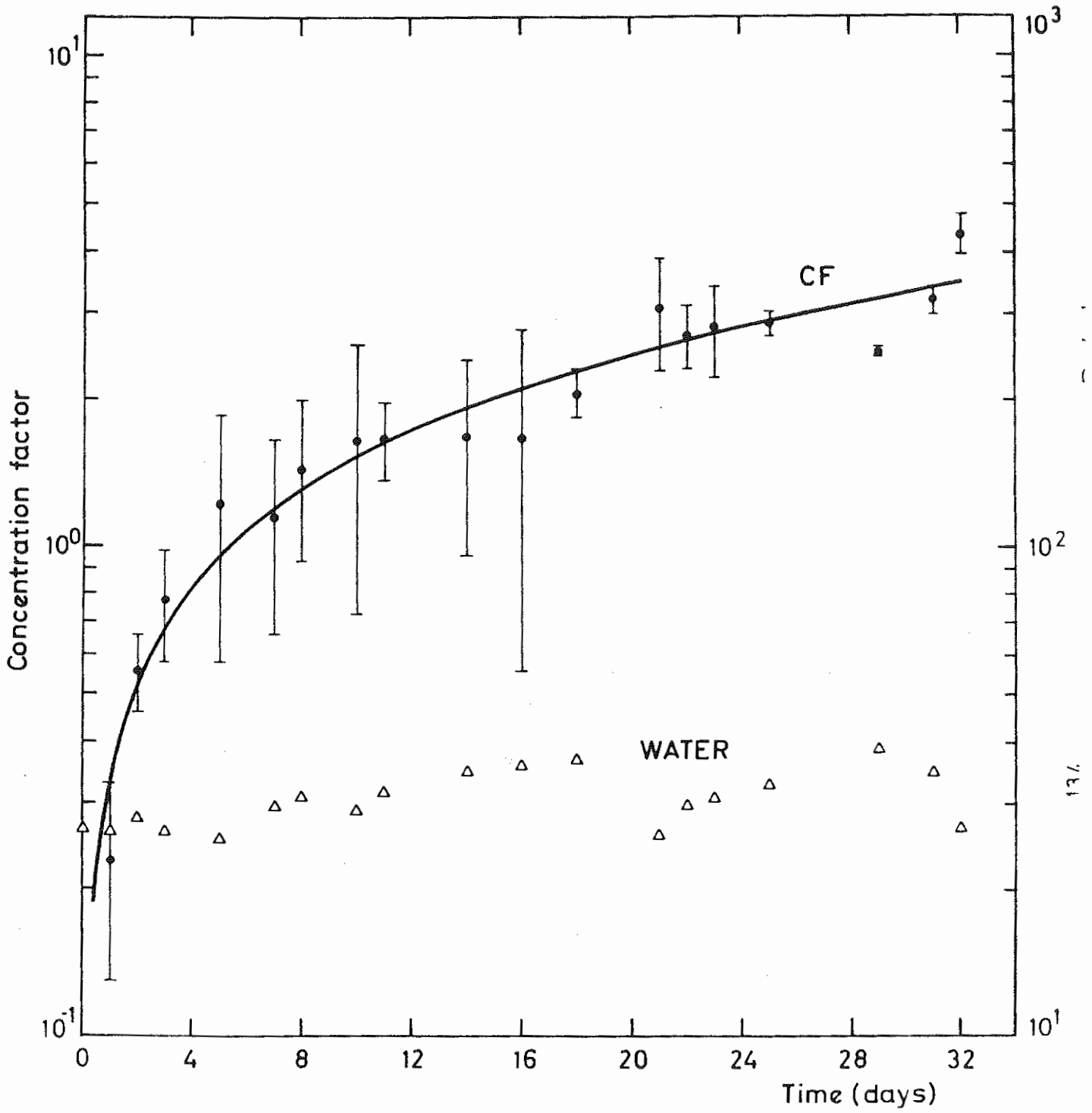
Figure 1 - Evolution of the  $^{137}\text{Cs}$  concentration factor (CF) in *T. tinca*, with  $1.32 \text{ g l}^{-1}$  of initial biomass, and of the water radioactivity (aquarium A).

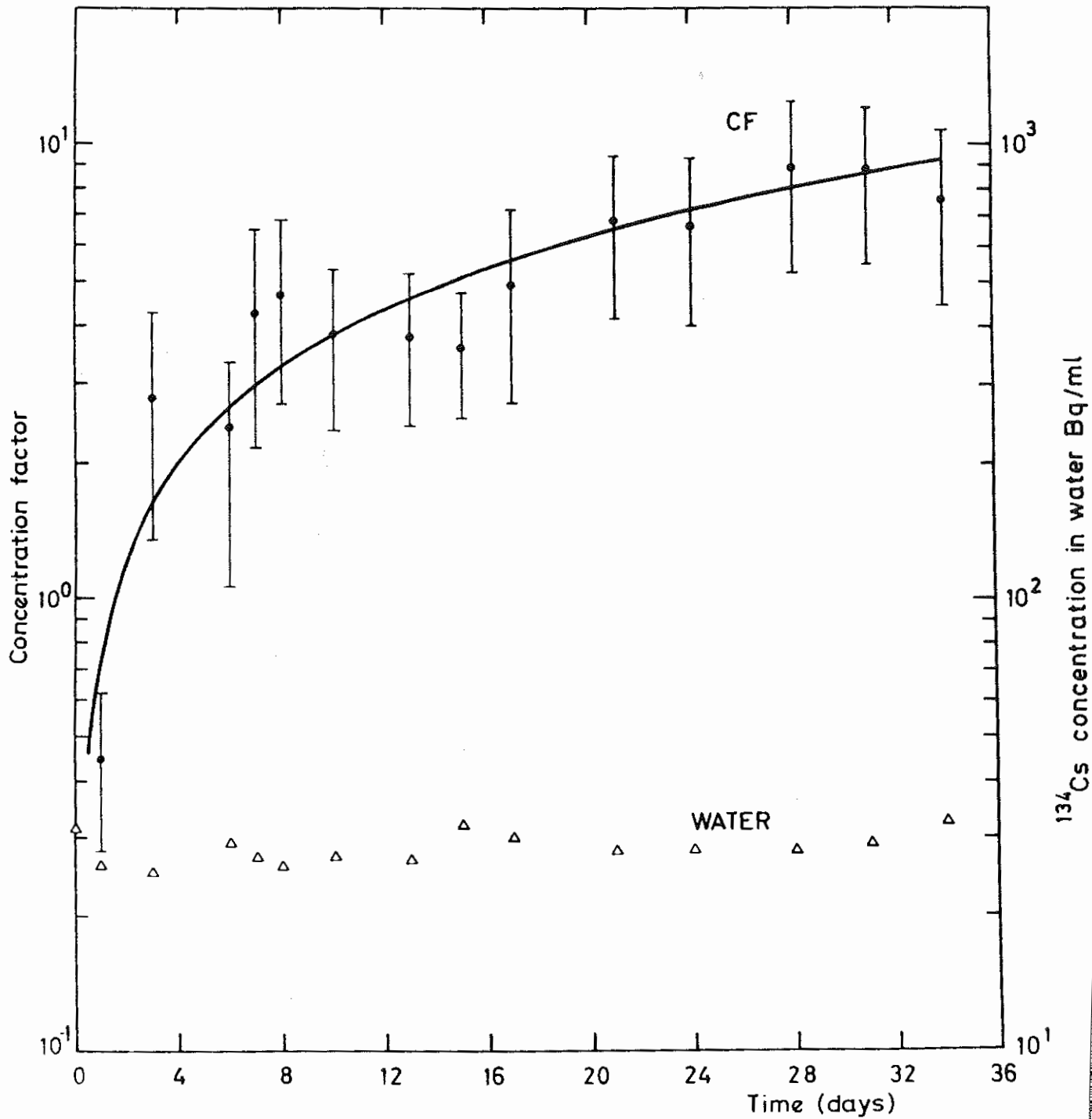
Figure 2 - Evolution of the  $^{137}\text{Cs}$  concentration factor (CF) in *T. tinca*, with  $4.09 \text{ g l}^{-1}$  of initial biomass, and of the water radioactivity (aquarium B).

Figure 3 -  $^{137}\text{Cs}$  retention in *T. tinca* contaminated through the water, showing two rate functions

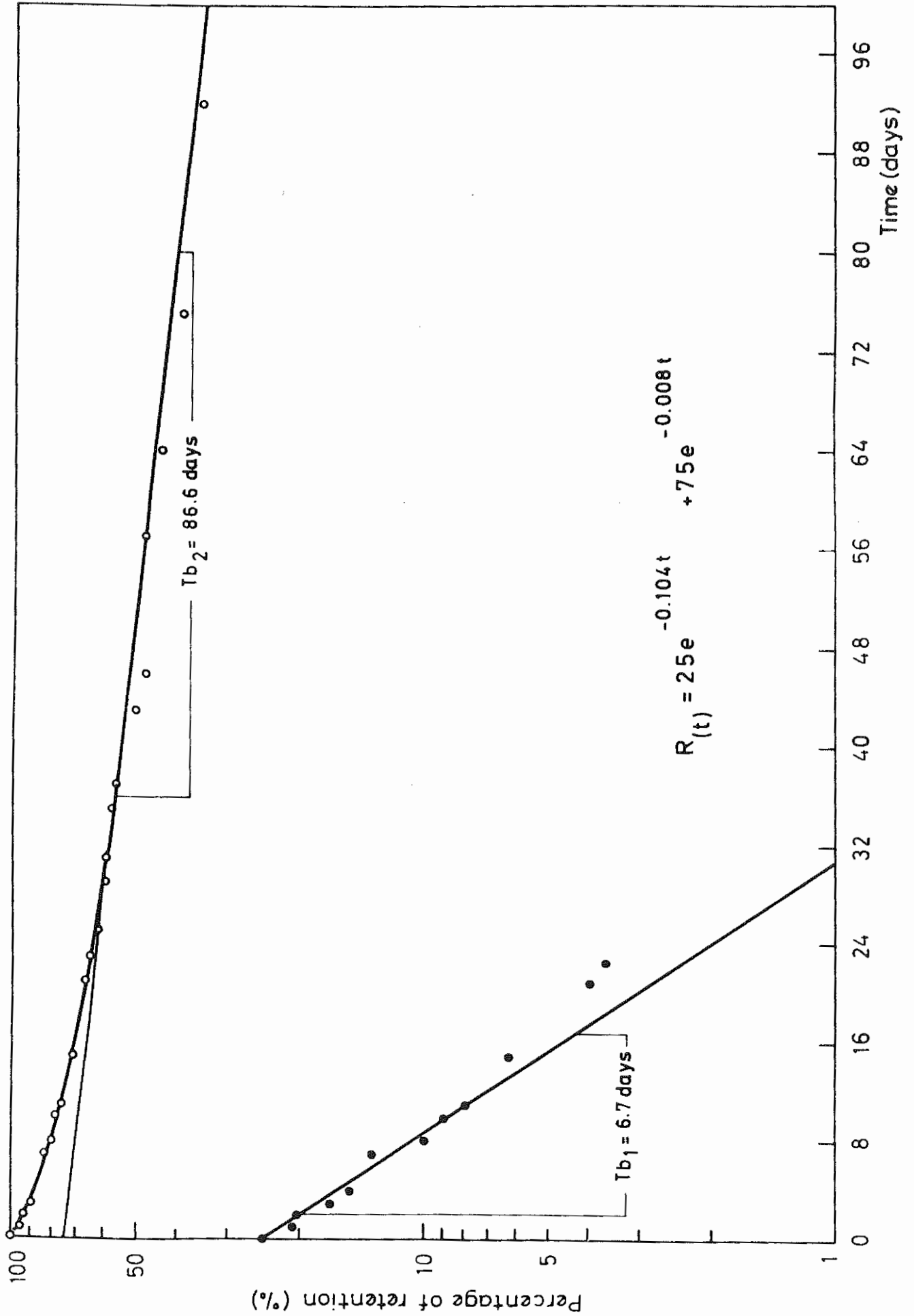
Figure 4 - Evolution of the  $^{137}\text{Cs}$  transfer factor (TF) in *T. tinca*, contaminated through labelled daphnids.

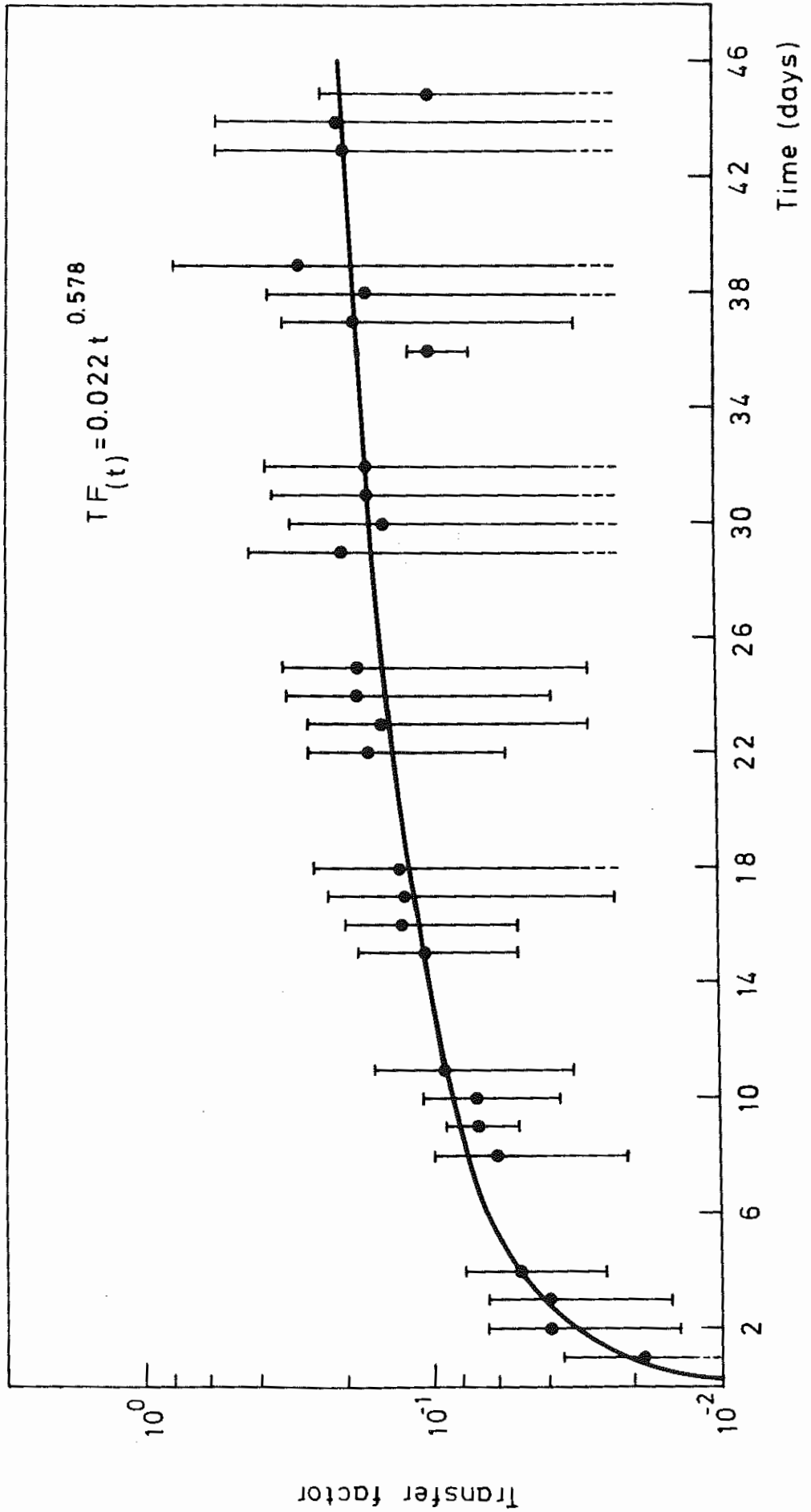
Figure 5 -  $^{137}\text{Cs}$  retention in *T. tinca*, contaminated through labelled daphnids, showing a single rate function.

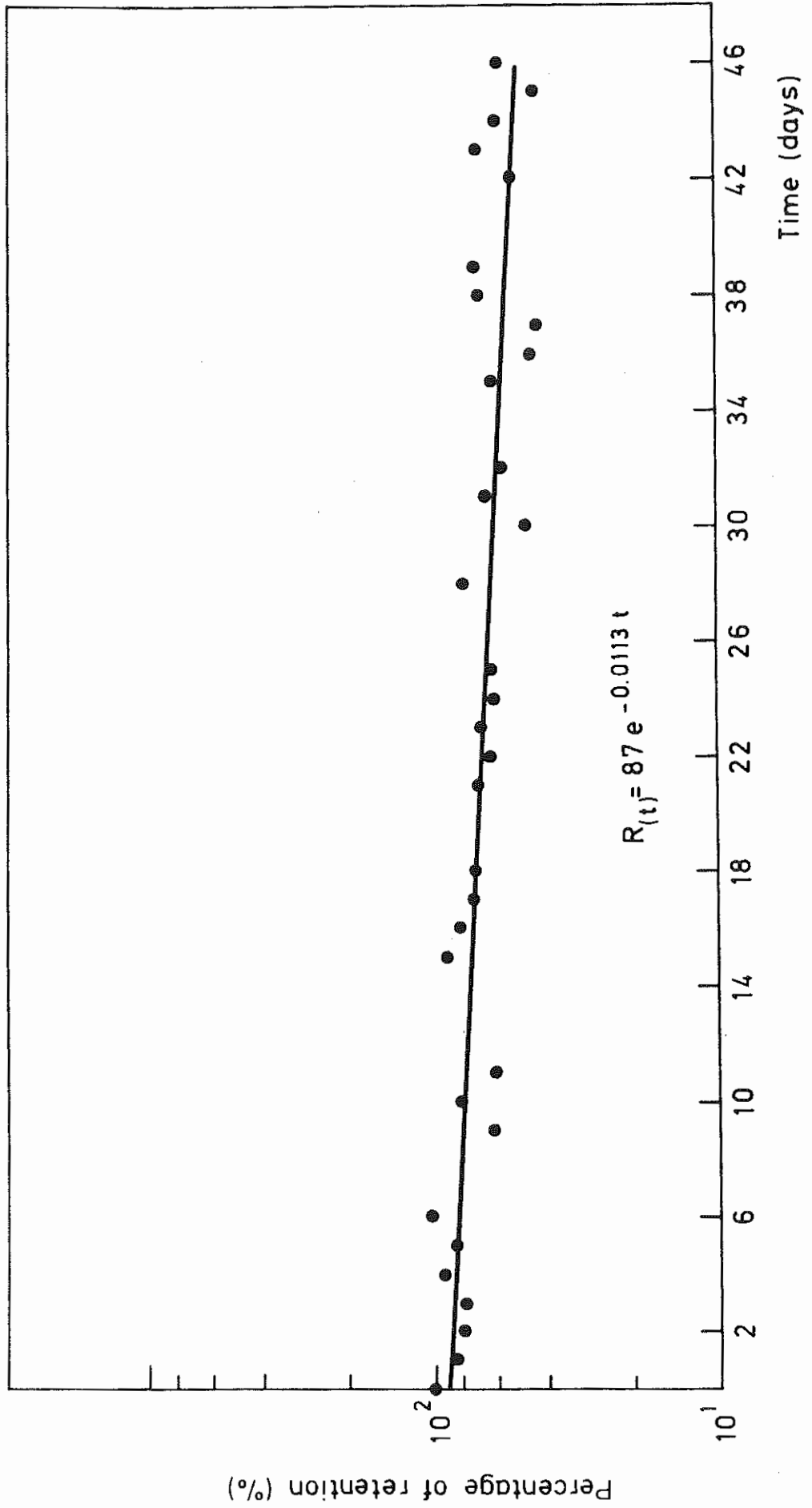












Session II

**Impact Assessments,  
Remedial Action**



# **Consequences of the Chernobyl Fallout for Wild Populations in South-Eastern Sweden**

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### ABSTRACT

An investigation was carried out in Sweden aimed at studying the impact of the Chernobyl fallout on the environment. Wild small mammals (orders Rodentia and Insectivora) were used as biological indicators of radioactive contamination of a region of south-eastern Sweden. The animals were captured in three differently contaminated areas, and for control, in an area with negligible fallout. The results obtained show that the activity in the captured animals was correlated with surface deposition. The differences between the species investigated and the influence of feeding habits on the contamination levels are discussed.

A mutagenicity test (bone marrow micronucleus test) was also performed on the rodents in order to verify the occurrence of a genetic damage. A positive correlation between the increase of genetic damage and both  $^{137}\text{Cs}$  content in muscle and surface contamination in trapping sites was found. The estimated doses absorbed by the investigated animals were too low to explain the obtained effects. An explanation of this discrepancy between dose and measured biological effect is not available, yet similar results have been repeatedly reported after the Chernobyl accident and should be matter of further discussions. It is possible that an increased frequency of chromosomal aberrations might occur already at minimal dose gradients.

## INTRODUCTION

The radioactive fallout following the Chernobyl accident in 1986 caused an extensive contamination of the environment in several European countries. The fallout levels recorded in the Swedish territory (among the highest in western Europe) involved mainly upland pastures and forests, with consequences for grazing ruminants and wildlife [1] and in particular small mammals, which are particularly exposed to ground-deposited fallout through direct contact with and ingestion of radioactive food.

For this purpose, a study was carried out aimed at examining the contamination in Sweden of wild rodents and insectivores two years after the Chernobyl fallout and the possible genetic damage present in rodents living in differently contaminated areas. Mutagenicity tests have been employed with success on wild populations to evaluate the biological effects of ionizing radiations [2, 3]. This paper reports the activity of  $^{137}\text{Cs}$  recorded in the captured animals and discusses the variations exhibited in the trapping sites with possible relationship to feeding habits. The results of a mutagenicity test (bone marrow micronucleus test) used to investigate the extent of low-dose damage, showed the existence of a correlation between environmental radioactivity and somatic cell damage.

## MATERIALS AND METHODS

The investigation was performed in undisturbed areas of south-eastern Sweden mainly covered by coniferous forests mixed with deciduous species. Three sites at different contamination levels and, for control, one with negligible ground deposition, were considered. The fallout contained several radionuclides, but this study focused on radiocesium which constituted by far one of the greatest sources of radioecological concern. Fig. 1 shows the location of the experimental sites, with the soil deposition as mapped by aircraft [4]. Since the fallout exhibited extreme local variations and the resolution of this aerial mapping was considered too poor, more accurate local measurements were performed. The ground deposition in the trapping sites was determined by measuring the contents of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  in homogenized soil cores (5.5 cm diameter, 10 cm depth) collected from surrounding areas. After subtraction of old fallout-caesium present in the deeper profile of the collected soil cores, the ground deposition of  $^{137}\text{Cs}$  (at the time of the investigation) resulted in 1800 Bq m<sup>-2</sup> at site 1 (control), 22000 Bq m<sup>-2</sup> at site 2, 90000 Bq m<sup>-2</sup> at site 3 and 145000 Bq m<sup>-2</sup> at site 4. The ratio  $^{134}\text{Cs}/^{137}\text{Cs}$  was constant throughout, 0.26.



The trapping sites were selected with great care far from roads and industrial or domestic settlements. In order to assess the eventual occurrence of heavy metals with known mutagenicity, the soil contents of As, Cd, Cr, Hg and Pb were determined with atomic absorption spectrometry (Perkin-Elmer 5000) after dissolution in  $\text{HNO}_3$ . The captures were performed in July-August 1988 with live traps using high-quality, uncontaminated baits. Altogether 104 wild rodents (*Clethrionomys glareolus* Schreb.) and 64 insectivores (*Sorex araneus* L.) were collected. Sex and somatic features were determined in each animal, whereas age was determined only in *C. glareolus*. After removal of skulls and digestive organs, the carcasses were weighed, homogenized and placed into plastic containers for activity determination.

The radiometric analyses were performed through gamma-spectrometry using a HPGe-detector (ORTEC, rel. eff. 37.3 %, FWHM 1.88 keV at 1.33 MeV), surrounded by a shielding consisting of 1 mm copper, 5 cm iron and 10 cm low-active lead in a special low-background laboratory. The output signal of the detector was fed through a linear amplifier (ORTEC 572) into a computerized 4096-channels analyzer for gamma-spectrometry (ORTEC Adcam).

For the bone marrow micronucleus test, both femurs were dissected and their marrow cells were flushed out with fetal bovine serum and pipetted several times. The cell suspension was centrifuged at 800 rpm for 5 minutes and the cell pellet was resuspended in a small amount of fetal bovine serum and smeared on a clean glass slide. The slides were air dried, fixed with methanol for 5 minutes, and stained with May-Grünwald and Giemsa for microscopic examination [5]. The samples were randomized and scored blind on codes, and the frequency of micronucleated polychromatic erythrocytes (MPCEs) in 2000 polychromatic erythrocytes (PCEs) per individual was counted by the same analyst.

## RESULTS AND DISCUSSION

The  $^{137}\text{Cs}$  contents in the collected animals are presented in Table 1. Wide fluctuations of  $^{137}\text{Cs}$  activity were observed, even in animals trapped at the same site. Nevertheless, Pearson correlation analysis showed that the  $^{137}\text{Cs}$  contents were fully correlated with ground deposition (*C. glareolus*  $r=0.78$ ,  $P<0.001$ ; *S. araneus*  $r=0.76$ ,  $P<0.001$ ). These results, which are clearly dependent on the diet of the captured animals, also reflect the habitat of the trapping sites. Several studies have described the diet of *C. glareolus* as composed of mixed vegetable food, such as mushrooms, shrubs, mosses, seeds and vegetative parts of the plants and of animal food, mainly invertebrates

[6, 7]. Mushrooms, in particular, constitute an important part of their diet [8, 9]. The ready uptake of radioactive substances by mushrooms and their accumulating properties is established [10, 11, 12, 13], and after the Chernobyl fallout the content of  $^{137}\text{Cs}$  in mushrooms has been found to be elevated both in Sweden [14] and in other countries [15, 16]. Even though the contribution of other contaminated food cannot be excluded, it is likely that mushrooms, as suggested for other animals [17], constituted the major component of the radioactive contamination of *C. glareolus*.

The total mean  $^{137}\text{Cs}$  concentration in *S. araneus* ( $2445 \pm 348$ ) was lower than in *C. glareolus* ( $3493 \pm 524$ ) and exhibited a more limited variability (cf. standard deviations). This reflects both the elevated metabolism and the diet of this animal, based on insects [18, 19] originating largely from deeper soil layers less contaminated by the Chernobyl fallout.

The results of the bone marrow micronucleus test performed on the individuals of *C. glareolus* are shown in table 2. The frequency of MPCEs/1000 PCEs differed significantly among sites (ANOVA,  $P=0.007$ ) and Pearson correlation analysis showed that the average frequency of MPCEs/1000 PCEs of each population was significantly correlated with both ground deposition ( $r=0.99$ ,  $P=0.012$ ) and body burden ( $r=0.97$ ,  $P=0.020$ ).

The soil contents of heavy metals in the trapping sites were rather low, with the following ranges: As 0.9-4.6 ppm, Cd <0.5 ppm, Cr 3.8-16 ppm, Hg 0.05-0.36 ppm and Pb 13-47 ppm. These values were far below the levels found in Swedish polluted areas [20, 21] and, also in this case, statistical analysis did not reveal any significant correlation between these results and the frequencies of MPCEs/1000 PCEs.

The whole-body dose rates due to the chronic exposure to  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$  and natural background received by the investigated rodents were also estimated. The external beta contribution, assumed to be self-absorbed, was not considered, while the external gamma contribution was determined using the data of Beck and De Planque [22] on the exposure rate for infinite plane sources (exponential distribution in the soil profile, relaxation length 3 cm and soil density  $1.6 \text{ g/cm}^3$ ). It was also assumed that the rodents were in contact with the contaminated surface layer, without taking into account the time spent in deeper soil locations, which throughout the year is limited to only 10-30 % of the time [23]. For the external contribution due to background exposure to naturally occurring radionuclides, it was used the value of  $4.0 \mu\text{Gy/d}$  [24].

The internal beta dose rates were calculated from our data using the values

of mean energy emitted per time integral of activity ( $\Delta$ ) tabulated by NCRP [25]. Since the range of beta particles was small relative to the dimensions of *C. glareolus*, the beta energy was assumed to be entirely absorbed. The internal gamma dose rates were determined using corresponding  $\Delta$ -values [25] with absorbed fractions for photon sources determined for a mass of about 20 g [26]. For internal dose rate calculations, uniformly distributed radiocesium was assumed. The results, listed in Table 3, were in good agreement with those obtained by others for rodents [27, 28].

If, for simplicity, the daily dose was assumed to be sustained for one year (which can be considered the expected mean age of this species), the total absorbed doses ranged between about 1.5 and 14.4 mGy. These values were significantly correlated with the average frequency of MPCEs/1000 PCEs of each population (Pearson,  $r=0.98$ ,  $P=0.021$ ). Fig. 2 shows the linear regression of this correlation.

### CONCLUSIONS

The good agreement between body burdens and ground deposition, frequency of micronucleated cells and estimated doses, provides reasonable evidence for a biological effect of the Chernobyl fallout, even at very low doses. Such an indication was obtained in a previously published study [29] on the induction of genetical damages in wild *Mus musculus domesticus* as a consequence of the Chernobyl fallout in Italy. In this study, the content of  $^{137}\text{Cs}$  and the frequency of chromosomal aberrations exhibited a statistically significant increase during and after the Chernobyl fallout, Fig. 3.

Both these results reveal a measurable cytogenetic effect at very low exposure levels, undoubtedly beyond expectation in *C. glareolus*, a species considered as the most radio-resistant among wild rodents [30, 31]. We are aware that the doses absorbed by the investigated animals appear too low (if compared with extrapolated results of laboratory experiments) to explain the measured biological effects. However, both in the aftermath of Chernobyl [32, 33, 34] and in studies on the environmental exposure near nuclear installations [35], there have been reports of several observations of low-dose effects not compatible with values extrapolated from high-dose measurements.

An increased frequency of genetic damages might occur already at minimal radioactive gradients and the genetic effects reported in this investigation, more than a conclusion, should constitute a basis for further discussions and investigations in other areas in Europe.

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TABLES

Table 1. Concentration of  $^{137}\text{Cs}$  in the animals collected in the four trapping sites. Number of animals (N), mean ( $\bar{x}$ ), minimum (Min), maximum (Max), standard deviation (SD) and standard error (SE).

Site	$^{137}\text{Cs}$ dep. $\text{Bq m}^{-2}$	Species	N	$^{137}\text{Cs}$ , $\text{Bq kg}^{-1}$				
				$\bar{x}$	Min	Max	SD	SE
1	1800	<i>C. glareolus</i>	22	39	2	254	54	11
		<i>S. araneus</i>	5	48	2	133	50	22
2	22000	<i>C. glareolus</i>	22	1031	38	2744	636	136
		<i>S. araneus</i>	25	751	2	2092	582	116
3	90000	<i>C. glareolus</i>	22	5119	800	11100	3315	707
		<i>S. araneus</i>	25	3233	217	7015	1726	345
4	145000	<i>C. glareolus</i>	38	7784	631	32330	8169	1325
		<i>S. araneus</i>	9	6289	1397	12520	4471	1490

Table 2. Frequency of micronucleated polychromatic erythrocytes (MPCEs/1000 PCEs) in the bone marrow of *C. glareolus*.

Site	N	MPCEs/1000 PCEs		
		$\bar{x}$	SD	SE
1	22	1.3	1.4	0.3
2	22	1.5	1.2	0.3
3	22	1.9	1.7	0.4
4	38	2.6	1.4	0.2

Table 3. Estimated total whole-body dose rates from external ( $\gamma_E$ ) and internal ( $\gamma_I$ ) gamma, internal ( $\beta_I$ ) beta, total (Tot) irradiation from  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  and natural background (Bg).

Site	Dose rates ( $\mu\text{Gy/d}$ )									
	$^{134}\text{Cs}$				$^{137}\text{Cs}$				Bg	TOT
	$\gamma_E$	$\gamma_I$	$\beta_I$	Tot	$\gamma_E$	$\gamma_I$	$\beta_I$	Tot	Tot	
1	0.04	0.01	0.02	0.07	0.05	0.01	0.10	0.16	4.0	4.23
2	0.49	0.17	0.61	1.27	0.65	0.25	2.66	3.56	4.0	8.83
3	1.88	0.82	2.96	5.66	2.65	1.27	13.21	17.13	4.0	26.79
4	3.06	1.31	4.70	9.07	4.28	1.92	20.09	26.29	4.0	39.36

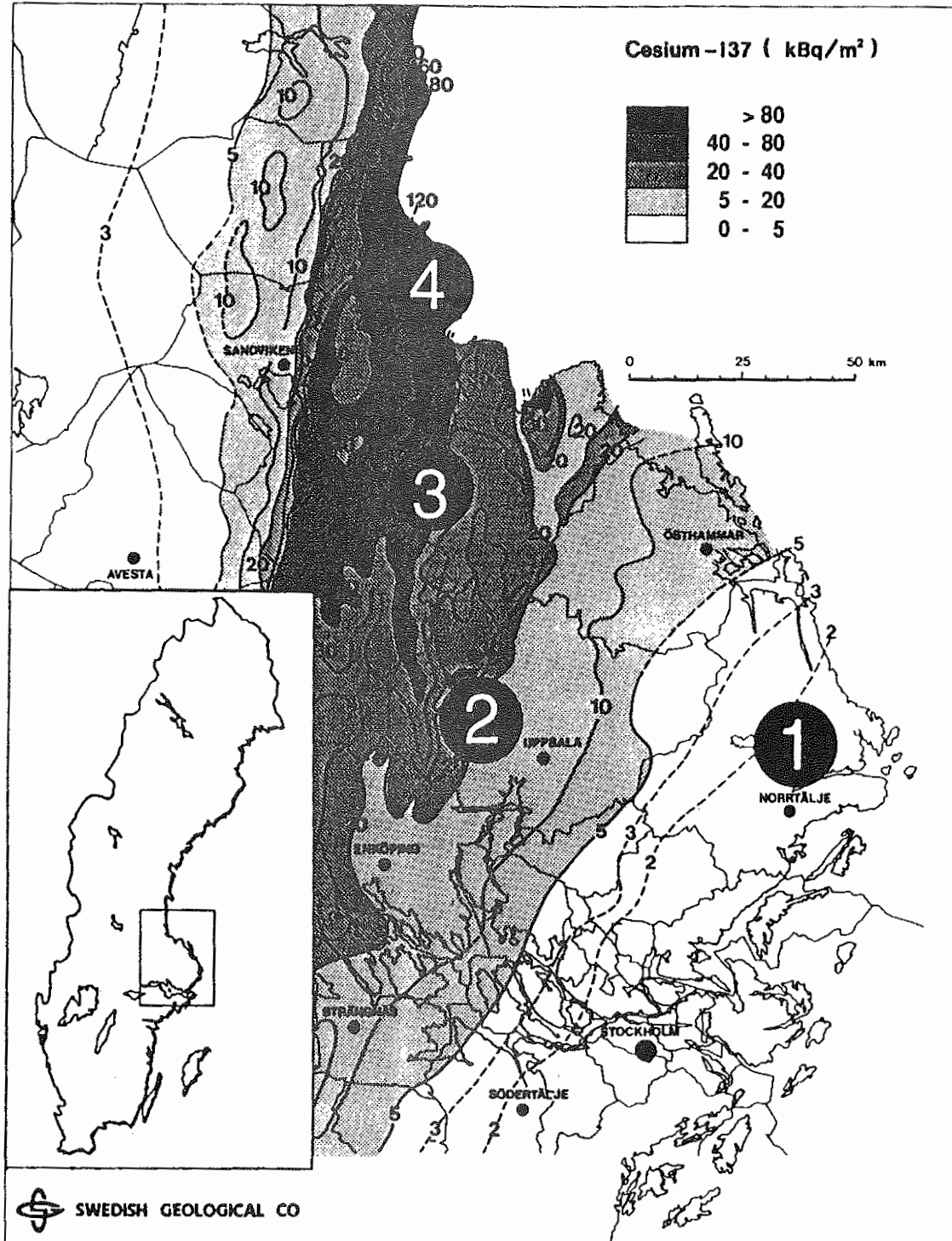


Fig. 1. Aerial survey of <sup>137</sup>Cs ground deposition and trapping sites.



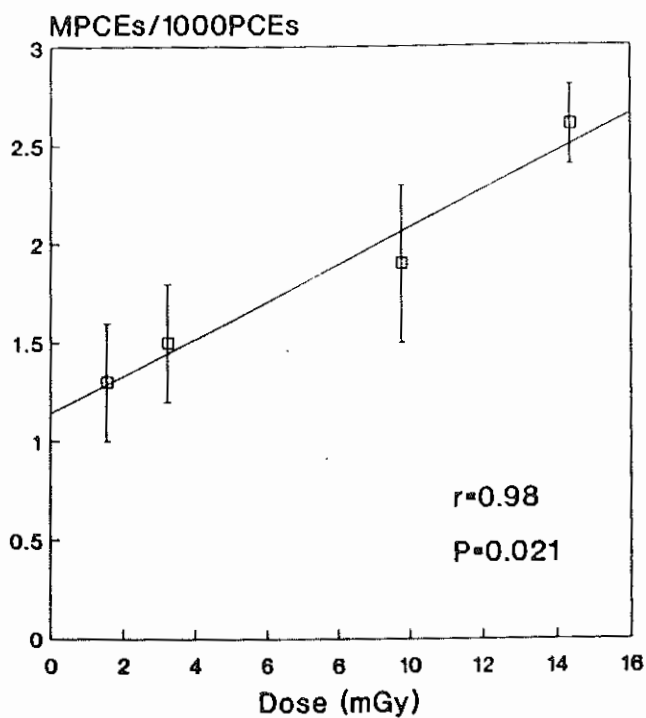


Fig. 2. Linear regression of the frequency of MPCEs/1000 PCEs (with standard error bars) on total estimated doses received by the investigated individuals of *C. glareolus*.

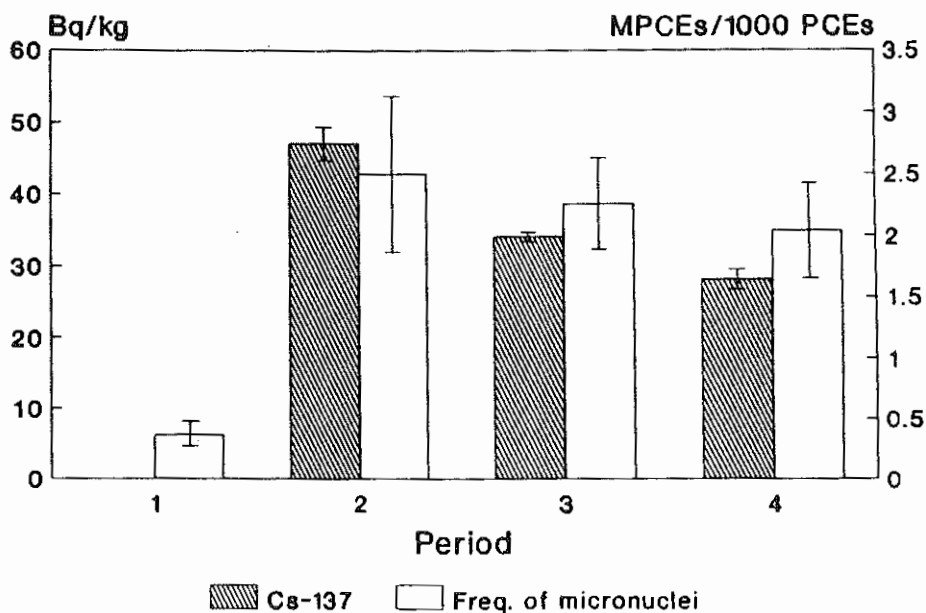


Fig. 3. Content of  $^{137}\text{Cs}$  (with counting error bars) and frequency of MPCEs/1000 PCEs (with standard error bars) in *Mus m. domesticus* as observed in the four trapping periods: 1) before the Chernobyl accident, Oct 1981; 2) during the fallout, May 1986; 3) 6 months later, Oct 1986; 4) 1 year later, May 1987 [29].

**Chernobyl Fallout in England and  
Wales: Countermeasures  
Research and Possible Remedial  
Measures by MAFF**

**L. McDONOUGH, M.G. SEGAL**

Ministry of Agriculture, Fisheries and Food

Abstract

The paper summarises the countermeasures taken by the Ministry of Agriculture Fisheries and Food after the Chernobyl accident to prevent radioactivity reaching man through the food chain. The effectiveness of these countermeasures is discussed, as are some aspects of the Research and Development work carried out within the past four years. A summary of research carried out on possible remedial measures to reduce the impact of the Chernobyl accident, and any future major nuclear accident, on agriculture and the food chain is also given.

A duplicate diet study was carried out in June 1986 in order to gain a clearer indication of actual radiation doses received in different parts of the country from the consumption of contaminated foodstuffs. The maximum dose received was calculated to be 0.1 mSv. The study showed that traditional methods of dose assessment resulted in a significant over-estimate of actual doses received in high deposition areas.

The "Mark and Release Scheme", an intervention system for sheep developed by the Ministry to prevent sheepmeat above the action level from entering the foodchain whilst minimising the impact on the agricultural community, is described. A summary of results of live-monitoring over the past 4 years before movement out of restricted areas is given. Results from a 2-year study at a farm in the restricted area are presented, which show that the levels of contamination present in sheep show very similar trends on a yearly basis, and overall are only dropping slowly.

Research has shown that the contamination of land by radiocaesium in certain areas of England and Wales may continue to be a problem for some time. A main priority of our research is therefore to find ways and means of releasing the currently restricted areas from control. Land management techniques and chemical treatment of both soil and sheep are currently being investigated: preliminary results of some aspects of this work are presented.

### Introduction

The Ministry of Agriculture Fisheries and Food (MAFF) has a statutory duty to ensure the safety of the foodchain and protect the public from the consumption of contaminated foodstuffs. After the Chernobyl accident in 1986 certain measures had to be taken to make sure no foodstuff contaminated to an unacceptable level reached the public and thus to ensure that no member of the public received an unacceptable radiation dose through the ingestion pathway.

This paper describes the countermeasures undertaken by MAFF to discharge our statutory responsibilities and then goes on to look at some aspects of the research work which has been undertaken in the past 4 years.

### Initial Response

Immediately after the accident, MAFF initiated an extensive monitoring programme for fresh foodstuffs of all types across the whole UK. Within a few days, the results of this programme showed that levels of radioactivity in the foodstuffs were below acceptable limits and that no restrictions were necessary; in the case of sheep in certain upland areas of the UK where soils lacked the mineral content to bind radiocaesium and prevent its uptake by sheep, data from our monitoring programme showed that restrictions were required.

A programme of re-assurance monitoring for items such as milk, grass and a wide range of foodstuffs has nevertheless continued ever since with the emphasis being on those areas that received the greatest amounts of contamination. The results of this programme of monitoring have been published and are freely available (1, 2).

### Duplicate Diet study

In addition to this routine monitoring of foodstuffs, MAFF decided to carry out a duplicate diet study in order to determine, in actual

diets consumed by members of the population, the radionuclide contamination resulting from the accident. This allowed a clearer indication of actual radiation doses received by the population rather than relying on predicted doses calculated using monitoring data and consumption rates. (3)

The study was carried out in three different parts of England:

1. A rural area of West Cumbria which represented an area of high deposition. 18 diets were collected.
2. A rural area of South West England which represented an area of low deposition. 16 diets were collected.
3. An urban area in West London representing an area with access to food from a wide range of diverse sources. 16 diets were collected.

Participants in the studies were selected on the basis of their consumption of locally-produced food, and so their diets reflected the extent of contamination of foodstuffs in each area, particularly in 1 and 2. 50% of participants were adults, the remainder being children between the ages of 3 and 5.

The mean Cs-137/134 ratio of 1.84 for all diets indicated that the majority of the activity was Chernobyl derived although slightly higher values were observed in Cumbria (1.91). The committed effective dose equivalents to age 70 from intake of radiocaesium during the study week were calculated. The average of all data showed a dose of  $< 0.34$  uSv, the range being  $< 0.03$  to  $1.9$  uSv.

The intake for a full year following the accident is approximately 50 times that for the study week, leading to a maximum dose of  $0.1$  mSv with the average dose to the population being  $0.02$  mSv.

A comparison of theoretical values, calculated from monitoring data, with those from the diet study is shown in Table 1. This comparison highlights the large overestimate of dose obtained in the high-deposition area using the indirect method of dose assessment, and confirms the need to obtain direct measurements of radionuclide concentrations in foodstuffs when accuracy is required.

The maximum dose of 0.1 mSv resulting in consumption of foodstuffs in an area of high deposition is only a small fraction of the annual limit and well below the average dose of 0.3 mSv calculated to be received from natural radioactivity in food and drink.

#### Restrictions on the movement and slaughter of sheep

As noted earlier, a programme of sheep monitoring showed that, in certain upland areas of the country, caesium levels remained unacceptable, and as a result of this, the movement and slaughter of sheep within certain well-defined areas of Wales, Cumbria and Scotland was restricted towards the end of June 1986 with some 4.2 million sheep out of the national flock of 24.6 million being affected.

Despite most holdings being released from controls within 3 months, certain areas still remained under restriction, and in August 1986, a "Mark and Release" scheme was introduced by MAFF. This scheme allows sheep to be live-monitored within the restricted areas, and has two main advantages in that all animals can be monitored to ensure that they are below the action level before going to slaughter and that those over the limit can be identified, moved out of the restricted area on to clean grazing, causing activity levels to fall rapidly, and re-monitored before being cleared for slaughter.

The use of colour marking with indelible paint to indicate the status of the sheep that have failed still allows the sheep to be moved for breeding or fattening. Marked animals that pass when re-monitored are eartagged and released from slaughter controls.

Restrictions are still in force, and Table 2 gives a summary of results up to the end of April 1990 for the Cumbrian restricted area. The figures for failures are those for the initial monitoring of the animals moving out of the area, and do not take into account the passes on subsequent re-monitoring.

Controls under the Mark and Release scheme ensure that contaminated meat is prevented from entering the market, and give confidence that the foodchain is being fully protected. Further details have been published elsewhere (4).

#### Research Programme

In addition to this continuing monitoring, MAFF has an extensive research and development programme related to the study of the movement of radiocaesium in the environment with particular emphasis on the soil and plant types found in the problem areas, and the subsequent plant-to-animal transfer of the radioactivity. The problem is greatest in upland areas where the soil has a low mineral content and the caesium is fixed in the soil sufficiently well that it remains close to the surface. The caesium is taken up by grass and other plants and is therefore made available to grazing animals.

Research has shown that there is considerable seasonal variation in the levels of contamination. Overall, levels are falling steadily at present, however this trend will fall off as the physical decay of radiocaesium becomes the main factor in the reduction of activity concentrations. At some locations within the restricted area, levels are falling at a much slower rate. This can be illustrated by the results of an experiment carried out on a Cumbrian farm within the restricted area over the period from July 1988 to June 1990.

50 ewes from the farm were eartagged and live monitored every 2 weeks. As many of the tagged animals as possible were gathered and monitored on every visit. Information on any supplementary feeding throughout the winter, and on the movement of the sheep within the farm was noted. Samples of supplementary feed were collected and sent for analysis.

Figure 1 highlights the variation in activity levels throughout the year, and indicates the location of the sheep and when supplementary feeding took place. Both the maximum and mean activity levels are shown. Figure 2 presents the mean activity levels found in the sheep, and superimposes the results for each year showing that the pattern is very similar. The increasing activity in the summer months coincides with the time the ewes spend grazing on the fell. Superimposed on this summer increase are two sharp decreases when the ewes are briefly taken off the fell for shearing then sorting and dipping. The activity starts to decline in the autumn with decreased caesium levels in the vegetation. This trend continues when the ewes are taken off the fell in December reaching the lowest point in May. After lambing, the ewes and lambs are returned to the fell and the sequence is repeated. Earlier data on monitoring of radiocaesium levels in sheep has been published elsewhere (4,5,6).

This information has shown how levels of contamination in animals grazing in the restricted area change throughout the year and has been used to identify the periods during which activity in sheep is at the maximum and minimum. This in turn has led to more efficient use of monitoring resources and advice to farmers concerning sales of sheep. This work has also indicated that the problem is long term and the areas may remain under restriction for some time to come unless some means of reducing caesium activity in herbage is found.

Other research work (7) has confirmed that the problem is long term. Initially it was concluded that a significant difference existed in the behaviour of Caesium deposited from Chernobyl compared with that already present prior to the accident, with Chernobyl-derived caesium exhibiting a higher degree of transfer (8,9). However, more recent work (7) has shown that the availability of aged and Chernobyl radiocaesium from the top 4 cm soil layer is now similar. Table 3 shows the transfer ratios calculated at two sites within the restricted area of West Cumbria.



These findings agree with results of other areas of work and show that in the absence of remedial measures, the physical decay of radiocaesium is likely to be the main factor in the reduction of activity concentrations in uncultivated areas.

Although the Ministry is confident that the public is being protected by the controls in existence, the fact that nearly 600 farms in England and Wales are still under restriction orders continues to be a problem. Therefore, a major priority in our research programme is to identify possible ways to reduce the radioactivity levels by means of intervention; by treatment of either the sheep or the soil in order to bring about the derestriction of these areas more quickly.

The effects of land management on caesium recycling and retention in cultivated upland ecosystems have been investigated (10) with a view to using this information as a basis for understanding the agricultural implications of radiocaesium deposition post-Chernobyl and following any future accidental events.

5 techniques were examined: liming, application of compound fertiliser and manure, drainage and reseedling, at three sites with known histories of land management before and/or after the Chernobyl accident. This work showed that land management practices do affect radiocaesium transfer. It was concluded that practices such as compound fertiliser application and drainage reduce transfer, whilst reseedling significantly increased transfer of both aged and Chernobyl caesium.

Work is also being undertaken to look into chemical methods of reducing soil to plant transfer of radiocaesium. Two approaches have shown considerable promise: addition of agents to bind the caesium thus making it unavailable to plants, and additions of potassium fertiliser, which affects the readiness of plants to absorb caesium. Large scale field tests are currently underway to test the adequacy

of this method under realistic conditions and to determine the impact any treatment may have on the environment (11). Prussian Blue, bentonite, clinoptilolite and potassium salts will be used on small plots of land within the restricted area of West Cumbria. This work will establish whether any of the likely treatments has the desired affect of reducing radiocaesium uptake by plants, and hence sheep. If the work is successful, it may finally lead to a possible means whereby the restrictions can be lifted.

A further approach is to investigate reduction of the transfer of radionuclides to sheep. The addition of bentonite to feed has been found to reduce the caesium activity in the meat significantly (5), but the use of Prussian Blue, administered in the form of boli, seems the most effective treatment. The results of preliminary work with Prussian Blue boli shown in Table 4, look promising and consequently this work is currently being followed by larger-scale field trials in Northern Ireland (12).

#### Conclusions

After the Chernobyl accident, MAFF carried out an extensive monitoring programme and a duplicate diet study, and imposed restrictions on the movement and slaughter of sheep where necessary to ensure the safety of consumers.

Despite the success of the controls imposed, research has shown that the problem is complex and long term. Therefore, one of the main priorities of our research programme is to identify ways and means of releasing the currently restricted areas from control.

Much has been learnt from the research carried out, and the information gained is valuable in giving a greater understanding of the behaviour of radionuclides in the environment, the effects they have on the foodchain, and what we could do to reduce the impact in the event of a future nuclear accident. Although no method of treatment has yet been identified, which would ensure that all

sheepmeat from the affected areas was below the limit of 1000 Bq/kg, and enable the restrictions to be lifted, promising lines of research are continuing with this aim.

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Table 1

Comparison of theoretical dose estimates with those made from the diet study for an area of low and high deposition

Location	Dose Estimate $\mu\text{Sv}$ (1)	
	Theoretical	Diet Study (2)
High Deposition	5.60	0.77
Low Deposition	< 0.40	< 0.12

(1) Both theoretical and actual values are CEDE to age 70 from intake over study week

(2) Mean values

Table 2

Summary of live-monitoring results for sheep moving out of the Cumbrian restricted area under the "Mark and Release" Scheme

	1986 (1)	1987	1988	1989	1990 (2)	Total
No. of animals	28490	73156	92878	104861	18235	317620
No. of failures	3526	2372	739	326	3	6966
% Failure	12	3.2	0.8	0.3	0.01	2.2
Highest Reading Bq/kg	3089	3379	2594	1624	932 (3)	

- (1) Sept - Dec 1986, form the start of the scheme
- (2) Up to the end of April 1990
- (3) Animals beneath 1000 Bq/kg may occasionally fail because of safety margins built into the monitoring technique

Transfer ratios for aged and Chernobyl radiocaesium  
(Cs-134/137 ratio = 0.53)

Area	No. of samples	Transfer Ratio	
		Chernobyl	Aged
1	12	0.41 +/- 0.09	0.42 +/- 0.15
2	30	0.38 +/- 0.07	0.37 +/- 0.07

\* Values taken from ITE project report Radiocaesium deposition and uptake by vegetation in West Cumbria, May 1990

Table 4

% reductions in tissue levels of Cs-137 in sheep from the use of AFCF Boli under experimental and field conditions

No of boli	% reduction of Cs-137 levels	
	Experimental	Field Trial
1	39	23
2	65	37
3	79	-



Fig. 1 LIVE MONITORING AT A FARM IN THE CUMBRIAN RESTRICTED AREA (15th JULY 1988 - 1st JUNE 1990)

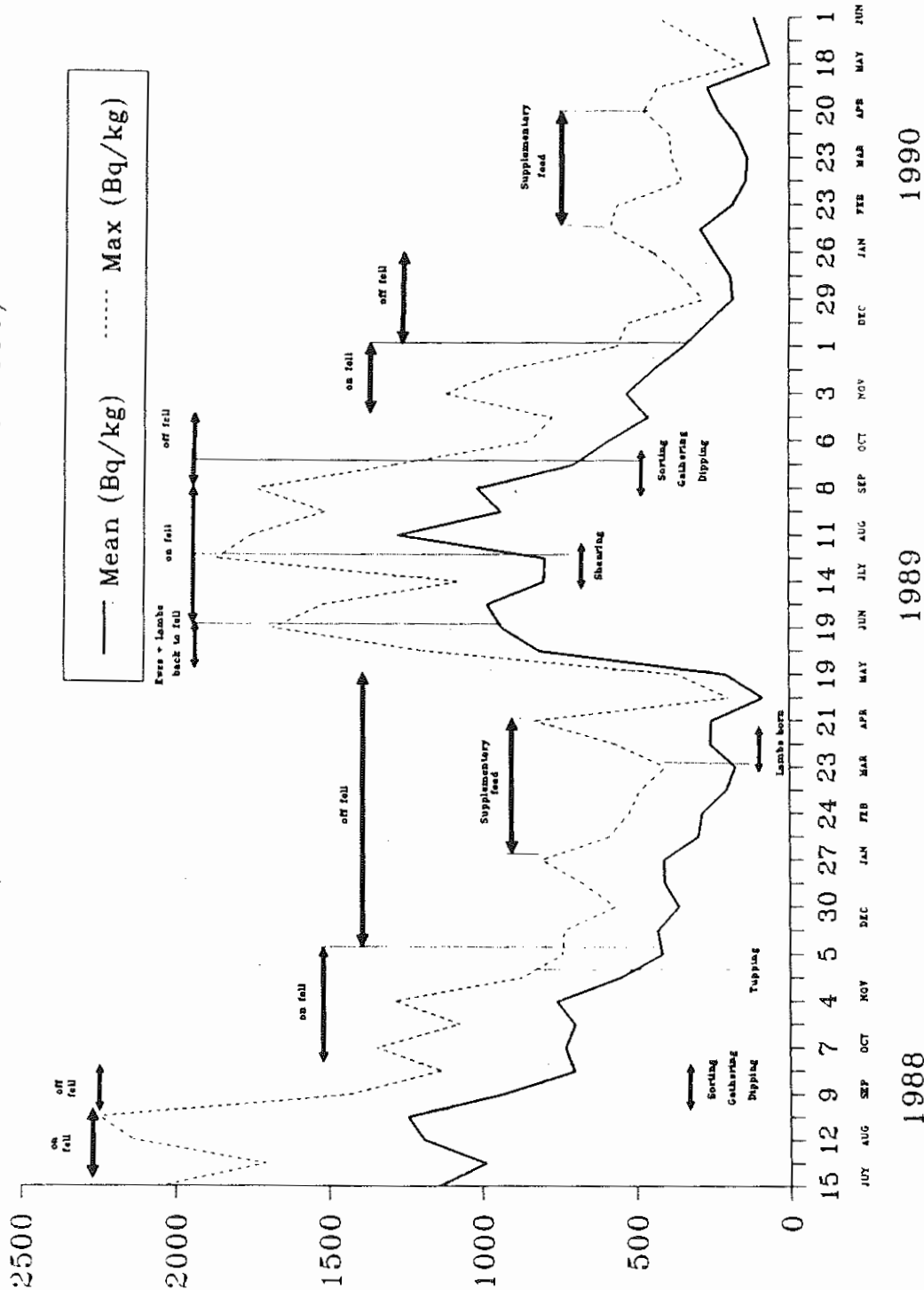
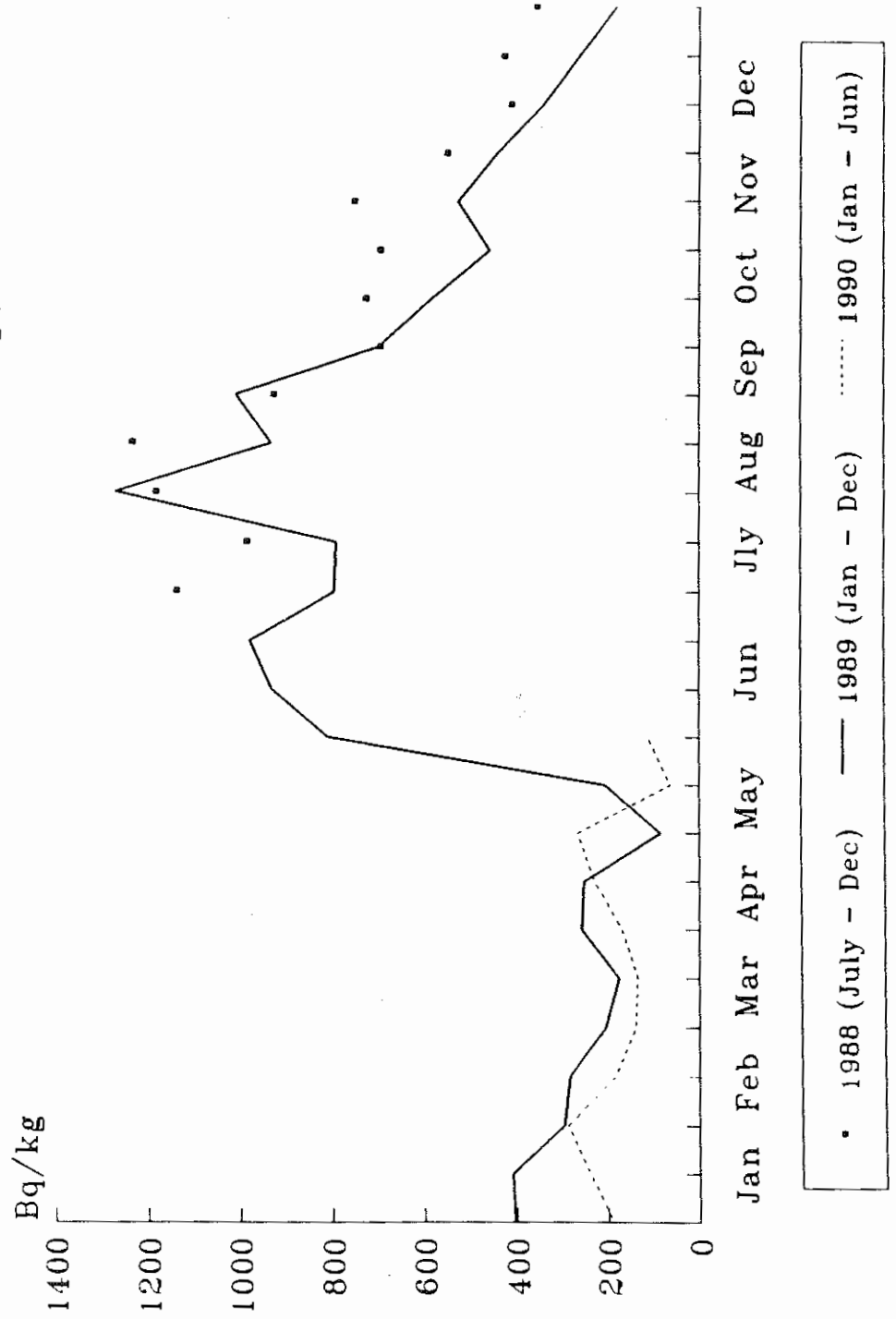


Fig. 2 LIVE MONITORING AT A FARM IN THE CUMBRIAN RESTRICTED AREA (Mean activity of 50 sheep)





# **Analysis of Post-Chernobyl Dose Estimate Based on Modeling and on Body Burden Measurement in CSFR**

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ABSTRACT

Comparison of the early estimates of effective dose equivalents to the Czechoslovak population due to the Chernobyl accident presented in the report to the UNSCEAR has shown that the model calculations based on environmental and food activity measurements give considerably higher estimates than the more direct ones based on whole body activity measurements.

Later analysis was aimed at assessing the validity of both approaches and the reasons of differences in the resulting estimates.

The validity of model calculations was studied on the data provided by the monitoring network of Czech and Slovak Federal Republic. The model calculations for cesium started from the activity deposition on the earth surface data supported by the activity concentration in the air data as well as from the foodstuff activity data. The results of calculation were compared with the data on activity content in the human body.

The validity of the whole body measurement in the reference group of persons used in the comparison as representative for the CSFR was studied using also results of urine measurement in samples of Czechoslovak population.

The discrepancies can be explained among other by generally occurring of not spending the foodstuff in full, by substantial delay in foodstuff consumption, by spontaneous discrimination of some food by inhabitants after the accident as well as by lower absorption of cesium in the intestine than considered in models.

## INTRODUCTION

Contamination of the environment due to the Chernobyl accident has been extensively monitored over the whole territory of CSFR. In later periods after the accident only the cesium radioisotopes have been of interest. More than ten thousand measurements of different components of the environment as well as the whole body measurements have been made in the laboratories of the Czechoslovak Monitoring Network since the accident. (1, 2) These data offered the possibility to compare, validate and improve different approaches used for assessment of the committed effective dose equivalents to the Czechoslovak population in the years 1986-1989. In principle three basic approaches are used:

1. whole body counting in a reference group of inhabitants
2. model calculations starting from data on contamination of food-stuff (Model 1)
3. transport model starting from data on cumulative fallout on the territory of the country (Model 2).

The early estimates of committed effective dose equivalents based on model calculations, as presented in the report to the UNSCEAR (1), gave considerably higher estimates than the more direct ones based on whole body measurements. This was the reason for the study aimed at obtaining more accurate values of input parameters used in models and at evaluation of parameters not taken into account in previous calculations.

## WHOLE BODY MEASUREMENTS

Systematic study of internal contamination of people began immediately after the first passage of contaminated air masses from Chernobyl over the Czechoslovak territory. On the whole body counter (WBC) of the Centre of Radiation Hygiene of the Institute of Hygiene and Epidemiology in Prague equipped by a semiconductor detector a reference group of 40 volunteers (21 women aged from 24 to 65 and 19 men aged from 31 to 63) living in Prague and its vicinity has been measured repeatedly from May, 1986 to the present. The monitoring interval was one month up to September, 1986 then it was extended to two months (3). Since the second half of June 1986, only Cs 134 and Cs 137 have been measurable.

The representativeness of results gained in the reference group

for the whole Czechoslovak populations has been tested by comparisons with persons living in various parts of the country excluding Prague. Persons not living in Prague were measured already since May 1986 parallel to the reference group and the results were compared repeatedly for selective time periods. Significant differences in internal contamination between Prague and the rest of Czechoslovakia were not found. In order to get information on various groups of population including people from remote places and people with different nutrition habits, four nation-wide surveys of internal contamination by Cs 137 based on measurements of its activity in 24-hours urine (4) were carried out in years 1987-1990. Number of samples in individual surveys varied from 85 to 293. The time course of Cs 137 retention in the reference group is shown in Fig. 1, where also the results of surveys based on urine measurements are shown for comparison.

#### FOOD CONSUMPTION MODEL (MODEL 1)

This model (5) starts from the specific and volume activity in the agricultural products which were measured at many units of the Czechoslovak Monitoring Network, namely in laboratories of hygienic stations and some research institutes. Results were collected and evaluated in the Centre of Radiation Hygiene of the Institute of Hygiene and Epidemiology in Prague, being charged with the function of Centre of Czechoslovak Monitoring Network.

The number of analyzed samples was adjusted to the expected contamination level and production in individual regions of CSFR.

#### FOODCHAIN MODEL (MODEL 2)

This model describing the transport of radionuclides in foodchain - which will be described elsewhere (6) - starts from experimental data on surface contamination gained soon after the accident. These data are representative since about 1300 samples of bare surface soil layer were taken in CSFR between 16 and 18 June 1986 and measured with mean value of Cs 137 surface activity  $4,2 \text{ kBq/m}^2$ . The distribution of fallout over the CSFR territory is demonstrated on Fig. 2 (1, 2). The fallout occurred in the beginning of the growing season of plants important both for direct consumption by man (cereals) and for feeding of farm animals (fodder crops).

Direct transport of radionuclides from the air into cereals and fodder crops is simulated using two procedures:

- model describing the dependence of the fraction of cesium transported from the fallout to the grain on the time interval between the fallout and the harvest (7),

- application of interception factor according to Chamberlain (8).

Transport via root uptake was not taken into account in the first vegetation period after the accident, but only in the next periods. Because the territory of CSFR is from the pedologic point of view a mixture of various soil species and types, the mean value of transport factors according to Ng et al. (9) is used for calculation.

The level contamination of milk and meat was estimated using the concentration factor method assuming steady-state conditions resulting in proportionality of the intake by contaminated feed to the specific activity of respective product. In the case of pigs where the long lasting steady-state can generally not be supposed the dynamic model according (10) was used.

The data on production quantities, feeding quantities and distribution of cesium into wheat products and milk products used in model calculations are summarized in Tab. 1, 2, 3 and 4 respectively.

It is presumed that the time between production and consumption of meat is about 25 to 30 days, of milk about 4 days.

#### CONSUMPTION OF FOODSTUFF

The food consumption rates used in both models and summarized in Tab. 5 originate from trade balance data (11). At least two factors may lead to their overestimation:

- estimate of consumption of home produced food which is based on questionnaire surveys of households;

- assumption of full consumption of food bought which is in fact partly not used and thrown off or used for feeding domestic animals.

Consumption of flour and flour products starts in Czechoslovakia normally in November of the year of harvest. For fruits and vegetables the continuous consumption during the year after the harvest is supposed.

#### RESULTS

The intermediate results of calculations by model 2 are presented and



The second main reason for overestimating the real retention by models seems to be the overestimated value of cesium fraction absorbed in human gastrointestinal tract, i.e.  $f_1 = 1$  (12). To show the influence of using a more realistic lower value the retention estimated with  $f_1 = 0,7$  is also shown in Fig. 3. This is in agreement with our earlier speculations on absorption of cesium biologically incorporated in food (3) as well as with data of Moiseev (13) and Piechowski (14) who is reporting the range from 0,5 to 0,9 for  $f_1$ . Due to the similarity in physiology and metabolism of pig and man our choice can be supported by the analysis of Cs 137 transport in pig by Kliment (10) which resulted in estimate  $f_1 = 0,72$  for pig. In the first year, however, even lower absorption is to be considered because of surface contamination of some foodstuff by aerosol particles with hardly leachable cesium (3).

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Tab.1

Production of cereals and fodder			
Species	Vegetation period [d]	Crop yields* [kg/m <sup>2</sup> ]	
Wheat	100	0.44	
Barley	100	0.48	
Corn for silage	95	4.0	
Fodder arable x a:			
Clover 1. reaping	50-55	9.5	85%
2. reaping	35-40		45%
Alfalfa 1. reaping	55-60		45%
2. reaping	40-45	9.1	20%
3. reaping	35-40		20%
4. reaping (↗)	45-50		15%
Pasture grasses			
1. reaping	55-60	4.6	60%
2. reaping	50-55		40%
Fodder arable 1 a	70-100	3.2	

Note. \* in harvested weight  
 ↗ in climatic favourable years

Tab.2

Survey of feeding quantities of farm animals

Cattle

Feed [kg,L/d]	Dairy cattle		Beef cattle	
	Summer	Winter	Summer	Winter
Green fodder	48	-	18	-
Silage	6	26	7	15
Ensilaged hay	2	8	2	3
Hay	-	4	-	2
Straw	3	3	3	3
Cereals	4	4	2.5	2.5

Note. Cereals are mostly a mixture of feed wheat and barley, ratio 1:1; the feeding of cereals started approximately in November of the year of harvest.  
 Green fodder obtained by about June 30 is from 1st reaping, by July 31 from 2nd reaping, and by October 30 from 3rd and further reapings.  
 2/3 of ensilaged hay and hay is obtained in 1st reaping cycle and 1/3 in 2nd cycle.  
 Silage is usually a mixture of green corn and beet (leaves and cutting).  
 Winter period continues from November till April.

Pigs

Month of feeding: Feed [kg,L/d]	1	2	3	4	5	6
Wheat	0.4	0.4	1.1	1.1	1.3	1.3
Barley	0.3	0.3	0.75	0.8	1.3	1.3
Dried milk	till 0.1	0.08	-	-	-	-
Whey	-	2.8	2.5	2.5	2.8	2.8

Note. Specific activity of dried milk is a rule 8.5 times the activity of pasteurized milk

Poultry

Laying hens 110 g/d, broilers 115 g/d cereals and groats

compared in Tab. 6 with the data gained by foodstuff monitoring used in model 1. The final results of both models in terms of time course of intake supposing full consumption (gross intake) are in Fig. 3.

The annual time intergrals of Cs 137 content in humans in consecutive years after the accident are presented in Tab. 7 and compared with the respective whole body counting results.

The time course of cesium retention in adult person estimated by model 2 is demonstrated in Fig. 4 supposing full absorption in GIT ( $f_1 = 1$ ) and incomplete absorption ( $f_1 = 0,7$ ) as well; for comparison the whole body measurement results are also included.

#### DISCUSSION

The comparison of the time courses of gross intakes estimated by food consumption model 1 and foodchain model 2 in Fig. 2 gives evidence of good validity of the model 2 and parameters used in it underestimating the gross intake by approx. 30% only. It has to be reminded that in both models full consumption of foodstuff as shown in Tab. 6 is supposed, however.

The comparison on Fig. 3 of retention estimated by model 2 with the retention determined by whole body counting of the reference group - validity of which was verified by nation-wide surveys - is less satisfactory.

One of the main reasons of overestimating the real retention is the difference between the gross and real intake which we are not able to quantify yet. The foodstuff and cesium in it is not ingested entirely due to losses at food at-home-storage like premature molding and rotting and losses at food preparation like scraping, leaching, etc., as well as losses due to not consuming the prepared food in full and buying some foodstuff for domestic animals feeding. In addition to it the reason for real retention being lower namely in the first time period after the accident could be some self-limitation in consumption of fresh food being supposed by inhabitants to be contaminated.

The overestimation by foodchain model in the first and the second year followed by underestimation in the third year suggests the possible influence of rather long delay in food consumption after production which is, however, not supported by our information on this delay in market network.

Tab.3

<i>Distribution of cesium in the by-products of wheat milling</i>			
<i>(f<sub>1</sub> is the ratio of specific activity in the milling product i and in wheat before milling)</i>			
Product	Ash (mm)	Weight contribution [%]	f <sub>1</sub>
semolina	0.45-0.48	2	0.40
wholemeal flour	0.40-0.45	17	0.43
medium flour	0.50-0.55	27	0.49
fine-ground flour	0.70-0.75	4	0.49
bread flour	0.90-1.70	19	0.67
edible fraction	0.40-1.70	69	0.52
feeding fraction	1.6 - 3.3	31	2.06

Tab.4

<i>Distribution of cesium in the milk by-products</i>			
<i>(f<sub>1</sub> is the ratio of specific or volume activity in the product i and in the pasteurized milk)</i>			
Product	Annual consumption [kg, L] converted to		f <sub>1</sub>
	milk	product	
pasteurized milk	111.1	111.1	1.00
cream	4.5	4.5	0.53
curd	26.0	3.7	1.06
cheeses	52.5	6.3	1.06
frozen products	1.5	2.8	0.53
milk powder	30.5	3.2	9.50
evaporated milk	4.5	1.7	2.56
other	17.8	17.8	1.00

Tab.5

<i>Annual consumption of chief kinds of foodstuff in adults in Czechoslovakia</i>			
Foodstuff	Consumption (kg.L)	Foodstuff	Consumption (kg.L)
Milk (5)	178.8	Fruit (2)	46.0
Beef (1)	21.8	Potatoe	80.0
Pork (1)	39.8	Vegetable (3)	76.0
Poultry	12.0	Egg (4)	17.3
Other meat (1)	3.7	Fat	26.0
Wheat (5)	46.3	Sugar	37.5
Rye (5)	14.1	Other foodstuffs	7.1

Note. 1 - in net weight including viscera  
 2 - without fruits of tropical and subtropical zones  
 3 - comprise 24% leafy, 34% root and 42% other vegetables  
 4 - in net weight  
 5 - effective consumption rate (given by the product of physical consumption and distribution factor of caesium between dairy products and milk and between milling products and cereals before milling)

Tab.6

<i>Summary of selected model and monitored values of specific activity in agricultural products [Bq/kg] and of time integrals of specific activity in continuously produced foodstuff [kBqy/kg.L] in three real years after the accident</i>			
Product	Specific activity [Model 2 / Monitor]		
	1.year	2.year	3.year
Wheat	10.4/16.0	0.3/0.8	0.2/0.1
Barley	8.8/ 7.2	0.3/0.7	0.3/0.4
Vegetable	3.1/ 6.3	2.2/1.1	1.3/0.4
Potatoe	2.8/ 3.0	0.5/0.3	0.4/0.1
Fruit	25.2/23.5	2.7/3.5	1.4/1.6
Foodstuff	Time Integral [Model 2 / Monitor]		
	1.year	2.year	3.year
Milk	4.86/3.98	0.36/0.75	0.03/0.15
Beef	13.00/8.84	2.75/3.31	0.16/0.38
Pork	6.98/7.46	3.52/3.62	0.17/0.38
Number of measured samples	4734	2412	1544

Note. Fruit and vegetable values are weighted means by respective consumption rates

**Tab.7**

<i>Annual time integral of Cs 137 content in humans in three years after the accident</i>			
Period	Annual time integral [kBq]		
	Model 1	Model 2	Whole-body measurements
1. year	12.89	12.84	7.18
M/WBC	1.80	1.79	-
2. year	11.98	11.31	7.93
M/WBC	1.81	1.43	-
3. year	2.91	2.84	3.48
M/WBC	0.84	0.73	-

Note. Parameter M/WBC denotes the ratio of model value to WBC one





Fig.3 Intake of Cs 137 by humans

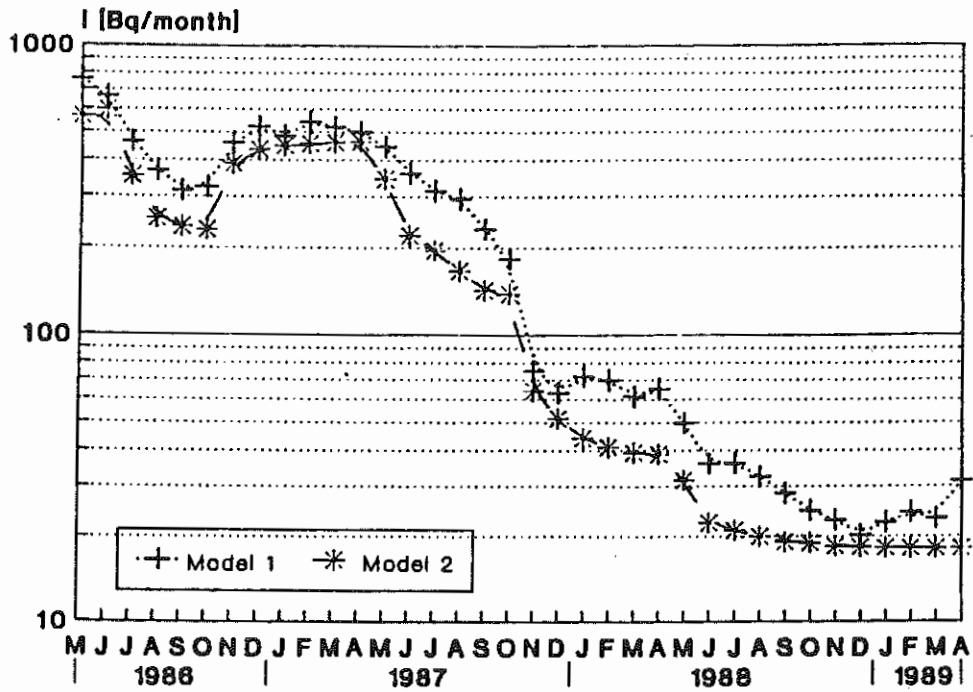
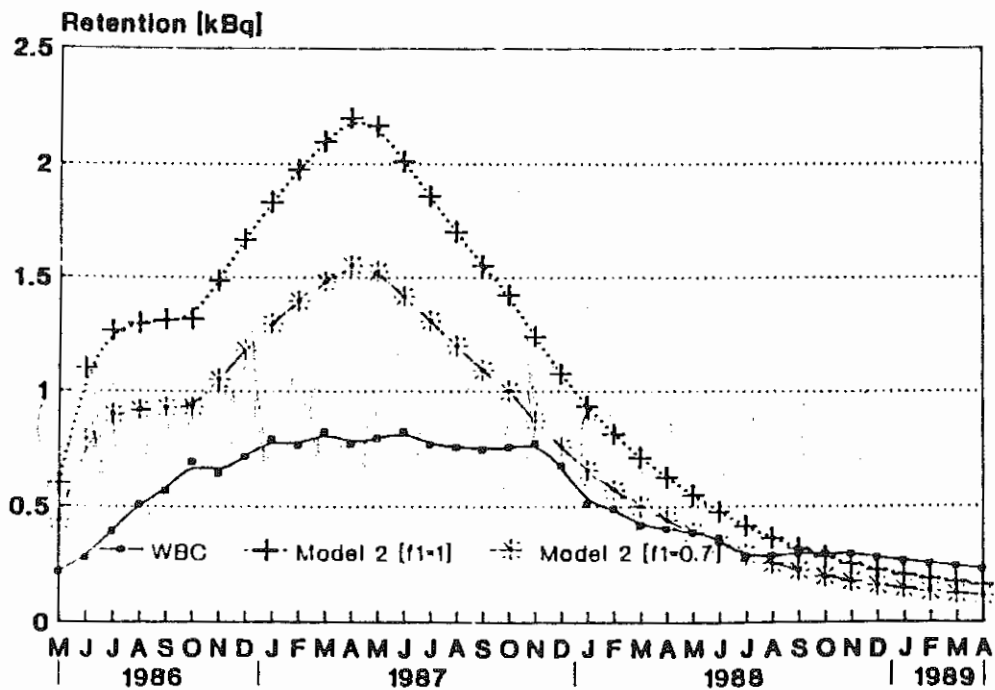


Fig.4 Retention of Cs 137 in humans





Rapporteur Summary of Poster Sessions  
by W.R. Schell, C. Vandecasteele and V.F. Demin

Overview

The poster session of 12 posters provided an opportunity for investigators and delegates to discuss in detail, at a more leisurely pace, the new and significant findings and plans which are developing in their fields. This poster session certainly fulfilled the role of interchange of information between interested participants. Dr. Vandecasteele and Dr. Demin supplied excellent summaries which are included here. As usual, the diversity of the posters captured the essence of the recent work being done in radioecology on the effects of the accidents. Our Soviet colleagues should be encouraged, in future meetings, to present additional posters which can be discussed.

The posters and the papers, which will be included in the proceedings, presented new information on forest ecosystems, both new data on throughfall, chemical speciation and organic composition, and computational models attempting to organize such data into a prediction format. Other posters included important information on soil to plant studies, studies on winter wheat contaminated before harvesting and placed into the pathway: cereal  $\Rightarrow$  bread  $\Rightarrow$  man, studies on the uptake by sheep grazing on contaminated upland pastures, uptake of Chernobyl radionuclides by small mammals and rodents in contaminated zones, transfer of  $^{134,137}\text{Cs}$  in aquatic ecosystems, remediation studies on ways to reduce animal-man uptake through feeding by time and by chemical additions to the diet which remove Cs, studies on the evaluations of some reasons why radioprotection models overestimate the uptake to man during the first two years and underestimate the dose the third year after Chernobyl, discussions of the counter measures being introduced, and finally sewage treatment plants as the repository of man's waste material in two contaminated cities where information on the dose to man can be evaluated.

An overview of the symposium, as encapsulated in the posters presented, might be interpreted in the context that we are living in a global laboratory of radionuclide contaminants and that man's activities, including the nuclear accidents, have increased the natural levels of radioactivity that can effect the total biota. It is important to realize that man is simply one omnivore which must survive under unusual conditions. Unlike the other herbivorous and carnivorous animals, man has the ability to understand and then to take action to control our environment. We must learn as much as possible from accidents

such as these discussed at this symposium. We must increase the studies on nuclear safety and we must prepare prediction models on radionuclides in the environment which can be used to estimate radiation dose in our efforts to protect all life forms. In addition to our goals of predicting and protecting all life forms, we must consider the cost-benefit of remedial action by decontaminating and by counter measures which may be necessary in large zones and/or by isolating certain regions and permitting natural processes of radioactive decay over time to reduce levels to acceptable "living" level standards.

POSTER REVIEW:

Radiological Studies and Modeling. Part 1. Forest Ecosystems

Our understanding of the interception and cycling of radionuclides in the forest has been limited, prior to this week's Seminar where a wealth of new information was presented by our Soviet colleagues. However, investigators in other countries also have made certain advances which treat specific problems and ecosystem mechanisms which now can trace mineral nutrients using the radioactive tracers available from the Chernobyl accident.

We have expected and have tested that the  $^{137}\text{Cs}$  would follow stable potassium in the cycling through the forest, since they are chemically similar elements. However, the  $^{137}\text{Cs}$  tracer has been used by the Belgian scientists Sombre and colleagues to evaluate the seasonal cycle of nutrient transfer in spruce and oak forest ecosystems. This group collected forest vegetation and throughfall waters in the forest, and rainfall in cleared land located nearby. The radioactivity and stable element concentrations were determined in the particulate and soluble fractions. After the Chernobyl deposition in 1986, the radiocesium has been more strongly retained by coniferous forest than by deciduous forest and has, after four years, penetrated into the several tree compartment reservoirs. Leaching of the vegetation by the throughfall waters collected under the trees confirmed the differences between conifer and deciduous trees. On the other hand, radiocesium contamination measured in the different tree components has not yet demonstrated any significant overall decrease in the total radioactivity. In spring, new shoots are contaminated with radiocesium and at the same time, radiocesium levels of the old needles have decreased due to translocation phenomena. In autumn, the radiocesium levels return to the levels measured in the previous year. Some correlations have been tested in order to understand why the leaching of radiocesium is variable with season. No significant correlations were found between  $^{137}\text{Cs}$  and rainfall amounts. However, highly significant correlations were found under the spruce stand between  $^{137}\text{Cs}$  and stable K in the soluble fraction of throughfall waters, particularly during autumn and winter. An explanation of these findings is proposed that during the dormancy season, K and Cs are translocated to o

leaves, trunk and branches. As a consequence, K and Cs are more available to be leached from the vegetation by precipitation. During the growth season, K and Cs are translocated to meristematic tissues where they become less available to be leached. The radiocesium that is leached from the free foliage is deposited on the upper layer of soil for storage.

The temporary reservoir is the contaminated tree tissues which leach slowly by precipitation and transfer  $^{137}\text{Cs}$  to the upper soil layers for storage. These radionuclides will be available, after several years delay, for uptake by the roots. The forest soil ultimately is the sink for the once deposited radionuclides. The question asked by Thiry and Myttenaere is "how do you express the availability of radionuclides for uptake by the root system of forests?" This term is essential in the modeling of forest ecosystems. A common unit used is the distribution coefficient,  $K_d$ , which is defined by the ratio of soluble concentration/particulate concentration. In the forest soil the values can differ greatly with depth because the organic content changes gradually to inorganic soil. In addition, an equilibrium exists between the pore water and these soil fractions.  $K_d$  ratios were calculated using activity of the solid phase expressed as both mass and volume and the activity in the soil solution. The expression of  $K_d$  values in the soil horizons show wide variations when expressed in the normal way of grams sediment. The top organic humus layers expressed in mass activity are considerably higher, 2-10 times, than that expressed as volume activity; in the transition layer with increasing mineral content the difference is smaller, 1.5 times; in the mineral layer the same values are obtained. Extraction using  $\text{NH}_4\text{Ac}$  and  $\text{CaCl}_2$  gave the biologically availability amount and the surface lattice exchangeable amount of Cs by ion exchange. The clay lattice of illite has ideal dimensions for Cs adsorption and thus the bonding in such clays is very strong. The difference between results of Cs extraction with  $\text{NH}_4\text{Ac}$  and with  $\text{CaCl}_2$  suggests that the mineral components present in the upper organic layers play a role in the high Cs retention of the surface soil layers.

The volume rather than mass expression of data provides a better indication of bioavailability. The combination of Cs adsorption on different matrix material provides information on the availability of Cs to tree roots in the forest. Mobilization of Cs may be mitigated by microbial activity in the organic rich litter and humus layers.

#### Radiological Studies and Modeling. Part 2. Aquatic and Terrestrial Ecosystems

The information presented by Gil Corisco and Vaz Carreiro deals with freshwater ecosystems where the results of a laboratory study on the contamination by radiocesium of a bottom feeder fish, the tench or *Tinca tinca*, were obtained. Two contamination pathways have been investigated and compared. When the source of contamination was water the concentration ratio, expressed on a fresh weight basis between fish and water, reaches a

value of about 10 after one month. When radiocesium was provided to the fish as labeled food, *Daphnia magna*, the concentration ratio between fish and prey, on a fresh weight basis, reaches a value of about 0.2 after one month; the authors conclude that the direct uptake of Cs from water is more important than the food pathway regarding the contamination of fish in the short term. The kinetics of the retention of accumulated radiocesium has also been investigated; when contamination occurred through the water, one component of about 7 days and a second component of 87 days half-time were measured. After ingestion of cesium through contaminated food, only one component could be discerned, characterized by a half-time of 61 days; this was similar to the 87 day half-time estimated for the long term retention of contamination from water. All the data reported in this poster are in good agreement with the previously published data related to the metabolism of Cs in fish.

Cesium contamination in Hungarian cereals after the Chernobyl accident has been investigated by Kanyar, et al. The radiocesium contamination of winter wheat which was standing during the accident was found to be proportional to the total deposit; about 1% of the deposited activity was transferred into the grain. The distribution of radiocesium within the grain was also investigated and the results show that most of the activity, approximately 60%, is associated with the bran which represents 20% of the mass of whole grain. This distribution is comparable to the distribution of potassium-40, which is not surprising since Cs and K are known to behave similarly and since the contamination of grain is not the result of direct deposition but mainly arises from translocation of activity deposited on the leaves which were present at the time of the accident. Other cereals were also considered: rye that was, at the time of the accident, in a stage of development comparable to that of winter wheat gives similar results while oat and maize which had been sown shortly before the accident showed lower contamination levels. The summer wheat is much lower in  $^{137}\text{Cs}$  concentrations in Hungary. The committed effective dose equivalent for the population has been estimated for different age classes. The cereals, vegetables and milk are the most important contributors to the dose.

Sandalls raises questions about the behavior of the Chernobyl radiocesium in upland pastures and also may provide some of the answers. The results show that the migration of radiocesium in soil is limited: three years after the accident, the peak of activity is found in the 2 to 4 cm layer and all the Chernobyl radiocesium is still located in the top 9 cm. The concentration in herbage was shown to decrease significantly between 1987 and 1988 for soils characterized by an organic matter content lower than 40% but no statistically significant difference could be observed for soil with the higher organic matter content. He also reports a cyclic variation of radiocesium in the vegetation that

parallels the cycle of potassium, with an increase in spring and a decrease during winter due to leaching from senescent vegetation. A direct proportionality is also observed between radiocesium transfer factors and the soil organic content on one hand and between transfer factors and exchangeable potassium on the other hand as could be expected.

#### Impact Assessment and Remedial Action

Information presented by Crow and co-workers gives preliminary predictions of the radiocesium contamination of upland lambs resulting from the Windscale and Chernobyl accidents. These predictions were obtained using an improved version of the RUINS Model. Several scenarios were considered, namely regarding the grazing density, the time of slaughter and the fact that lambs could be fattened on upland pastures, which is currently unlikely but has been the case for the Windscale accident. The delay between the accident and the time of slaughter in the first year is certainly a key factor in explaining the higher activities which were predicted in lamb meat after Windscale than after Chernobyl. This conclusion is valid for lambs fattened on lowland pastures but also for those that could have been fattened on upland pastures. However, radiocesium levels remain higher in lambs fattened on upland rather than lowland pastures. A finding of note is the concentration levels expected in lambs fattened on upland pastures after the Windscale accident would have required consumption restrictions, if the same Chernobyl action levels would have been used. In 1957, however, sheep were not considered as a critical pathway for dose to man and no measurements of these products were made. The model shows that there is no significant decrease in the radiocesium contamination level in lamb meat four years after Windscale and after Chernobyl.

McDonough et al. has suggested remedial measures to be taken to minimize the contamination from contaminated food stuffs. For sheep, a mark and release scheme is proposed using live monitoring methods for the radiocesium. The results show that there is little reduction in activity over the past four years.

Bucina and colleagues found that the food chain model overestimates the input of dose the first and second year and underestimates it the third year. Their data in Czechoslovakia included repeated whole body measurements of a given population group. They suggest two reasons for the observations. There may be a storage time for important food stuffs causing a delay in consumption, and there is a possibility of overestimation of the radiocesium fraction adsorbed in the GI tract. In the USSR, overestimation of radiation doses by two times could result in additional relocation of thousands of people.

Erlandsson presented a fascinating data set on how radionuclides transfer through human ecosystems of major cities. Lund and Gotheburg were chosen, where area contamination was about 2 kBq/m<sup>2</sup> and the sewage treatment sludge was measured. The

total yearly activity has decreased exponentially from 900 MBq to 100 MBq from 1986-1989. The urban effective half-time for decay of the radiocesium was found to be about 0.9 years.



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