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 I

Report

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Participants' copy

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Volume I

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LEGAL NOTICE

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We are happy to acknowledge this assistance and to record our appreciation of the help provided.

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SUMMARY

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This summary is based on the reports presented for the panel discussion chaired by G. Polikarpov. The chairmen of the previous sessions were asked to summarize the information presented and conclusions of their own sessions.

NOSSACH/GUDIKSEN (Session 2):

The environmental source terms for the Kyshtym (Romanov, Trabalka), Windscale (Eggleton), Chernobyl (Borovoi, Gudiksen) and Three Mile Island (Gudiksen) accidents were discussed. The Kyshtym accident took place on 29 September 1957, in a factory which produced plutonium for military purposes. Liquid radioactive material was stored in cooled metal storage tanks surrounded by concrete. A tank overheated, and there was a chemical explosion due to acetates and nitrates, of strength about 70-100 T of TNT equivalent. The downwind environment was heavily contaminated with about 2 MCi of radionuclides (10% of the contents), principally Sr-90. The interpretation of the Kyshtym accident by western scientists was described (Trabalka). It was suggested that the environmental contamination is due to several accidents which may have occurred over a considerable period of time.

Concerning the Chernobyl accident source term, much more information has now become available since the original reports in 1986. In addition to the dispersion of volatile material (such as caesium), particles of fuel containing transuranic elements were also emitted, which have deposited mainly within a 30 km radius. These are known as "hot" particles. They are considered to present a larger hazard than was originally estimated. The collaboration with scientists from other countries on the source term analysis was welcomed, and should be extended. The estimates of the Chernobyl source term from global environmental measurements was described.

For the Windscale accident, the fire started during a semi-routine operation to release stored (Wigner) energy in the graphite moderator. It was finally quenched by pumping water into the reactor. Some fuel element cans were ruptured and radioactivity was emitted from the cooling air chimney stack. From subsequent environmental measurements it was estimated that this included 27 kCi of 1311, 1 kCi Cs-137 and Po-210. The Three Mile Island accident was discussed; the environmental impact was negligible.

LINSLEY/APSIMON/VICTOROVA (Session 3) :
(This report was presented by G. Linsley, Mrs. Apsimon being unable to attend the panel session)

The atmospheric dispersion and deposition of material released from Chernobyl was described (Borzilov, Rumiantsev, Petryaev, Kerekes, Victorova, Eggleton (for Garland, Stukin), together with a model comparison of the Windscale and Chernobyl dispersions (Apsimon). The Windscale dispersion pattern has been re-analysed (Apsimon) with present-day atmospheric dispersion models, which estimate a release of 30kCi of I-131 with a dry deposition velocity of 0.3 cm/s. Wet deposition was important in both accidents, but it is more difficult to model. Hot particles from Chernobyl were observed in deposit in Byelorussia (Victorova) and in Sweden (Kerekes). They were observed on leaf surfaces by autoradiographic methods, which showed that some material could translocate into the leaf. Some particles appeared to be from fuel elements containing fission products and alpha emitters,

whereas others contained no alpha emitters but mainly Ru-103 and Ru- 106. Little assessment of the risks from hot particles has been made so far.

The resuspension of Chernobyl radiocaesium deposited over Europe was discussed (Garland, delivered by Eggleton). The concentration of resuspended material was related to the local deposition, which varied according to the local rain scavenging of the contaminated cloud. However, the calculated resuspension factor was lower at locations receiving higher deposits, suggesting that other effects were also involved, such as contributions from resuspension from distant high deposits, or perhaps from a stratospheric reservoir.

The resuspension around Chernobyl was also discussed by Stukin. The fractions of radiocaesium released in the accident which was initially deposited in different regions was estimated. Between 1987 and 1989, further measurements were made to assess the possible redistribution of the deposited material. In this way, estimates of the resuspension were made. It was also noted that ploughing the soil and the occurrence of forest fires affected the amounts resuspended.

COUGHTREY/SENIN (Session 4) :

The accumulated deposit of radionuclides in soils and their uptake by plants were discussed, relating to Windscale (Chamberlain), Chernobyl (Korobova, Davydchuk, Kulikov, Yushkov, Davidov, Novikova, Kulakov, Grebenshchikova), Kyshtym (Konoplyov, Pavlotskaya, Romanov, Prister), the comparative effects of Windscale and Chernobyl in Cumbria, UK (Coughtrey), and the global radiation dose from Chernobyl (Bennett). The soil contamination from 137Cs around Windscale was discussed (Chamberlain), although at the time of the accident the main concern was with 131I, and the effects of the Windscale and Chernobyl accidents in areas around Windscale were compared (Coughtrey); by chance, both accidents gave comparable radiocaesium deposits in some areas. The deposition of radiocaesium, Sr-90 and Ce-144, and Ru-106 on to the soil and vegetation around Chernobyl, and its subsequent migration was discussed by several authors (Korobova, Davydchuk, Kulikov, Yushkov, Davydov and Novikova).

These showed that much new information on migration through soils is now available. In some cases, penetration to more than 10 cm depth in months was observed. Uncertainties are still concerned with the role of organic matter, the movement of discrete particles and ground water leaching. However, the distribution pattern around Chernobyl has not changed much since the accident, indicating that the mobility is not large. The cycling of radiocaesium and Sr-90 by trees tends to maintain the soil surface retention in woodlands. The uptake into crops was also described (Grebenshchikova). Plutonium deposition around Chernobyl was described (Kulakov), by measurement of Ce-144 in hot particles and the use of a conversion factor. Of the 600 kg of Pu in the reactor, it is estimated that about 20 kg were released.

Comparative studies of the deposition and migration of radionuclides at Kyshtym and Chernobyl were described (Konoplev, Pavlotskaya and Prister), which indicated that the plant uptake factors were fairly similar. For Pu, the migration through soil depended on its form and on the soil type.

The assessment of the world-wide radiation doses due to Chernobyl, made by UNSCEAR (1988), was described (Bennett).

It was clear that although much new data with application to bio-geochemical theories had become available, some questions remained to be answered.

FOULQUIER/SOBOTOVITCH (Session 5) :

In this session papers were presented on aquatic systems. New data on hydrographic networks including uptake by fish and other organisms around Chernobyl were described (Voitcekhovitch, Rjabov, Senin, Khitrov, Pugachevskiy and Kuzmenko), together with a model description of the Pripyat and Dnieper rivers system (Zheleznyak). A review of the impact of Chernobyl deposit on European fresh water environments (Scandinavia, Germany, Austria, northern Italy, northwest United Kingdom) was given (Foulquier). The vector mechanism was wet deposition, with great variability in distribution. Cs-137 became the most significant long-term contaminant. The transfer through different trophic chains showed different rates, and Cs-137 biological residence times in fish were 200-500 days. Cs-137 depositions from Windscale and Chernobyl were compared in their effects on sediments of a lake near Windscale (Bennett) showing that the accumulation mechanisms were complex.

Marine environmental studies (Baltic and Black Seas) were also described (Kuznetsov, Kulebakina), together with a model of 137Cs in the Black Sea (Egorov). Because of recycling through the food chain, some Cs-137 concentrations in fish were higher in 1988 than in 1987.

The amount of aquatic data is now very large. The main requirements for the future are to assimilate the data, to bring them together in some cohesive way, and to produce interpretations and models, which can be validated.

The interactions of hot particles from Chernobyl with the aquatic environment was discussed (Voitcekhovitch, Sobotovich). Hot particle leaching in different chemical systems and the transfer of 90Sr from hot particles were described. Radionuclides are initially bound in hot particles with relatively insoluble material (eg. UO_2). In podsol and peaty soils this material will be dissolved and radionuclide transfer to the water phase can occur. It is expected that this will peak between 1991 and 1995, providing a new hazard from Sr-90 and Pu. Such pollution will be very long-lived.

KONOPLYA/PARETZKE (Session 6) :

The medical aspects of the three accidents were discussed and compared (Balonov, Buldakov, Dushutin, Konoplya). Many of the conclusions from Chernobyl are still preliminary, since deleterious effects to health can occur many years after the release. The general statement that if humans are protected then the environment is also protected must be regarded with caution. The relative scale of the collective doses from the Windscale, Kyshtym and Chernobyl accidents was estimated to be 1:5:600 respectively. At Chernobyl, increases in the incidence of diseases of the alimentary canal and the osteo-muscular system were noted among those who assisted in clean-up operations.

The effects of the accidents on the terrestrial environment were discussed (Romanov, Rjabtsev, Shevchenko, Yushkov, Tihomirov, Kutlahmedov), and on aquatic organisms in rivers and seas (Tsytsugina).

In general, the session demonstrated that many new data had been obtained, but these must be made more cohesive and linked with models for better evaluation. In studying the effects on human health, it is difficult to discriminate between the effects of the accidents and the baselines, which can be further confused by the effects of chemical pollutants. There is a lack of epidemiological data. More attention should be also paid to the estimates of risks.

TIKHOMIROV/AARKROG (Session 7) :

The effectiveness of countermeasures used in the Kyshtym and Chernobyl accidents was discussed (Rumiantsev, Romanov, Tikhomirov, Arkhipov, Ilyazov), and in the Windscale accident (Baverstock, Jackson). Countermeasures taken in the UK were discussed (Robinson), and a comparison of countermeasures for rural areas (Millan).

Countermeasures can be divided between short-term and long-term. Under short-term comes evacuation of the population, which, in the case of Chernobyl, reduced the doses received by a factor of 10. It also includes decontamination of skin and fur, control of foodstuffs and efforts to reduce environmental migration. Long-term countermeasures include removal of top soil, ploughing, erection of barriers to prevent migration to rivers, stopping agricultural production in contaminated areas. Many methods developed at Kyshtym were subsequently used at Chernobyl, including deep ploughing, using chemical additives to soil including P and K, replacement of one crop by another. The agro-technological methods used at Kyshtym were estimated to have reduced the overall dose by a factor of 100.

SCHELL (Poster sessions) :

Three poster sessions were arranged, including forest ecosystems (presentations by Berg, Sombre, Thiry, Schell), other ecosystems food transfer (presentations by Crout, Kanyar, Gil Corisco, Sandalls), and impact assessments, remedial action (presentations by Mascanzoni, Baeza, McDonough, Erlandsson, Pearce, Bucina)..

Rapporteurs of the poster sessions were respectively W.R. Schell, C. Van De Casteele and V.F. Demin.

The field covered was very diverse, and thus not easy to summarize. The effects on forest ecosystems were considered by models. Soil-to-plant transfer was discussed for winter wheat, and for pastures, followed by uptake in small mammals. In aquatic systems, the Cs-137 transfer to organisms by water was shown to be more important than the transfer via the food chain. Counter-measures taken in contaminated zones were described. Man should be considered as part of the environment, but with the ability to control and modify the environment. The need for more and better predictive models, which also include cost and benefit components, was stressed.

A more extended rapporteur summary is given at the end of volume II.

GENERAL DISCUSSION.

Many speakers made points in the ensuing general discussion, some of which are mentioned below:

Khitrov: Soviet estimates of the Chernobyl Cs-137 source term now agree with those of Livermore. More comparisons between Chernobyl and nuclear bomb measurements should be made. The Chernobyl explosive power is estimated as 10 MT of TNT equivalent, ie. 500 Hiroshimas. Very little migration of the deposited Cs-137 has been observed in the USSR. Most of the activity is with the hot particles; these could be the subject of another conference. The political aspects of the Chernobyl accident have a strong influence on practical scientific solutions. The USSR presentations at the meeting were the work of individuals, and did not represent the views of a single delegation.

Foulquier: It is important not to base radioecological conclusions solely on field observations. Laboratory research must also be done and included in interpretation if fundamental mechanism are to be understood.

Sandalls: A great deal of radioecological information on Cs-137 can be obtained from the study of the disposal of radioactive waste. This should be included in assessment in addition to the effects of accidents.

Eggleton: This meeting has been unique in that western scientists have been able to have free discussions with their Soviet colleagues, who have attended in large numbers and have given their results and opinions without having to follow any imposed rules. Perhaps the Chernobyl accident must be given a little credit for this situation. We leave the meeting with a desire to learn more of the Russion language.

Sinnaeve: There should be future meetings to consolidate and follow up the information presented at this one. We understand that international research centres at Chernobyl and Obninsk are proposed, where scientists from many countries can collaborate in their work on radioecological problems.

Session I

OPENING SESSION

Opening Address

G.G. POLIKARPOV

Institute of Biology of South Seas, Department of Radiation
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Mr Chairman, colleagues of the International Union of Radioecologists, officials of the Commission of the European Communities, our Luxembourg hosts,

On behalf of the Soviet branch of the International Union of Radioecologists, I warmly welcome all participants in the Seminar, the first of its kind to deal with comparative radioecology of the most serious and yet dissimilar nuclear accidents (Kyshtym, Windscale and Chernobyl), and I would like to congratulate the organizers of the seminar for the considerable pains they have taken in preparing and organizing this meeting, which brings together the world's most eminent scientists in this field.

It would have been difficult to find a better and more significant place for the Seminar than Luxembourg. Luxembourg is situated between the first and second major nuclear accidents (Kyshtym, Windscale) and between the second and third such accidents (Windscale, Chernobyl). Luxembourg is also a seat of the Commission of the European Communities. The Grand Duchy of Luxembourg has one of the highest standards of living in the world, and has long been active in international cooperation and the solving of complicated world problems. Luxembourg is the first country to host such a large group of Soviet scientists from three sovereign Republics: Russia, which suffered from the Kyshtym accident in the Urals in September 1957 and whose western regions were hit by the after-effects of the Chernobyl accident in April and May 1986; and the Ukraine and Byelorussia, which were ecologically contaminated more or less in their entirety following the Chernobyl accident.

Irrespective of radioecologists themselves, two approaches to information on nuclear accidents have been used in the past: firstly, complete openness about the radioecological situation, together with full compensation for the population which has been, or may be, affected by the nuclear accident in question and, secondly, complete secrecy with only partial compensation.

It is now clear to everyone, not only to radioecologists but also to politicians, that the main enemy is secrecy, lack of glasnost and bureaucratic attempts to shroud in secrecy events and their consequences which by their very nature cannot be kept secret. There is a Russian proverb to the effect that there are some problems you cannot sweep under the carpet, particularly if the problem is a major nuclear one affecting places up to 100 km away (in the case of Kyshtym and Windscale) or on a global scale (in the case of Chernobyl). In the case of the Windscale accident, however, soon - although not immediately - afterwards the population was provided with full information, costly measures were taken and the permissible radiation exposure level for the population was considerably reduced (0.07 Sv over 70 years instead of 0.35 Sv). This healed the relationship between the population and the nuclear energy industry in the United Kingdom. The experience following the Kyshtym accident was precisely the reverse, because such matters were treated unnecessarily and spuriously as 'top secret' in the Soviet Union until 1989, including after the Chernobyl accident to a considerable extent. The reaction of the public was not long in coming: operating nuclear power plants are being closed down, and building work on a number of new nuclear power plants in the Soviet Union has been stopped. Glasnost is developing and it is to be hoped that it will eventually become a part of everyday life. This is important so that no attempts are made to sweep even just one nuclear accident under the national "carpet". In this way it will be possible to establish a healthy relationship between the general public and the nuclear energy industry.

Although various specialized Ministries exist, there is no reason why individuals should not become members of national and international non-governmental organizations, in which all the specialists are equal and the only thing that matters is their competence in their field of radioecology.

For a long time I suffered from "isolation" in that up to 1989 I was the only Soviet member of the International Union of Radioecologists. On 8 November 1989, an initial orientation meeting of the International Union of Radioecologists (IUR) was held in Sevastopol with the participation of Prof. René Kirchmann (its Secretary), Felix Luykx (a member of its Board of Council), and radioecologists from the Urals

(Sverdlovsk), Kiev (Institute of Cytology and Genetic Engineering) and Sevastopol (Institute of Biology of the Southern Seas).
The meeting decided to set up a Soviet Branch of the IUR with its headquarters in Sevastopol on the shore of the international Black Sea, at the seat of the "A. O. Kovalevsky" Institute of Biology of the Southern Seas under the Academy of Sciences of the Ukraine. This Institute has been conducting broad international research on the World Ocean since late last century (1871). The formation of the Soviet Branch of the IUR was supported by the Scientific Committee of the Academy of Sciences of the Soviet Union with responsibility for Radiobiology, the Radiobiological Society of the USSR, the Academy of Sciences of the USSR, the Academy of Sciences of the Ukraine and other organizations. Soon afterwards people began applying to be active members of the Soviet Branch of the IUR, and so far 73 people have been accepted as active members; we congratulate them on this.

It is clear that the very size of the Soviet Union lends itself to the creation of regional sub-branches of the Soviet Branch of the IUR. The following have been set up so far: one for the European part of the Russian Federation (Chairman: Prof. F. A. Tikhomirov, Moscow), one for the Asian part of the USSR (Chairman: Prof. N. V. Kulikov, Sverdlovsk) and one for the European Republics of the USSR other than Russia (Chairman: Prof. Yu. A. Kutlakhmedov, Kiev). Naturally, it will be important to subdivide further by Republics to create branches for Byelorussia (a proposal to this effect is expected from E. F. Konoplya of the Byelorussian Academy of Sciences, Minsk), the Ukraine and possibly other sovereign Republics of the USSR.

The Soviet Branch of the IUR has undertaken a number of initiatives, the most important being its participation in organizing this Seminar. A General Assembly of the Soviet Branch of the IUR is planned for April 1991 to deal with routine matters of organization and to discuss scientific problems (it may be held in Kiev and possibly partly in Sevastopol - this is a matter which will need to be discussed). The IUR leadership will participate in this General Assembly, which will provide a good opportunity to discuss the results of joint projects with scientists from various countries, as well as ways of training young researchers in the field of radioecology.

I would like to express my admiration for the results of the work carried out over the past four years by the International Union of Radioecologists, and to thank the entire IUR leadership team for their splendid contribution - especially the President (Dr Asker Aarkrog), the Secretary (Prof. René Kirchmann), the Vice-Presidents and the members of the Board of Council. If it had not been for their helpfulness and concern for mutually beneficial cooperation to the common good in the field of radioecology, our achievements would not have been as positive and substantial.

I wish the participants in the Seminar success in their work. I also wish the newly elected President of the IUR, Prof. C. Myttenaere, every success in his work over the next four years.

Thank you.

1 October 1990

G. G. Polikarpov
Welcoming address at the Seminar
on behalf of the Soviet Branch of the IUR,
Luxembourg.

Opening Address

F. LUYKX

Commission of the European Communities
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When on the 2nd December 1942 at 3.48 p.m., on a squash court of the University of Chicago Enrico Fermi announced to his team that "The pile had gone critical", it was the first time in human history that man had controlled the release of energy from the atomic nucleus.

Now, 48 years later controlled release of nuclear energy has become part of our daily life, since over the world more than 400 nuclear power plants are operating and in the European Community 36% of the electricity is of nuclear origin.

But, as for all human activities, also nuclear energy production is linked with risks. Since the beginning of the nuclear age, several accidents have occurred in nuclear installations. Most of these were limited to in-plant consequences and had no impact on the external world.

However three accidents have occurred which had severe consequences for the environment.

The first of these happened on 29 September 1957 in Kyshtym, in the Southern Urals in a nuclear fuel reprocessing plant, where a tank containing highly radioactive waste exploded. In the Western world this accident was unknown until 1976 when Dr. Zhores Medvedev, who is present at this meeting, published his first article on this accident in "The New Scientist".

The second accident occurred about two weeks later on the 11th October 1957 at Windscale, now called Sellafield, in a plutonium producing air-cooled graphite reactor. During a Wigner release of the graphite there was an uncontrolled temperature increase to such a level that the graphite caught fire.

The third accident, known to all of you, happened on the 26th April 1986 in a nuclear power plant at Chernobyl, where as a result of a nuclear excursion reactor-unit no. 4 exploded and the graphite caught fire.

These three accidents have one fact in common: they all resulted in the release of large quantities of radioactive substances into the environment causing contamination of large areas in the Northern hemisphere.

Many studies, especially over the last years, have investigated the nature and the consequences of these accidents.

The Seminar of this week will provide an opportunity to present and to compare the nature of these accidental releases, their atmospheric dispersion and deposition and especially the subsequent transfer of contamination through terrestrial and aquatic ecosystems and the resulting implications for man and his environment.

The specific conditions of each accident being quite different, the seminar will give us the opportunity to put the enormous amount of radioecological data gathered after the Chernobyl accident in perspective with the results obtained after the earlier accidents.

This Seminar, which is organized by the Commission of the European Communities, Directorate-General XI and XII, together with the International Union of Radioecologists and with the cooperation of SCOPE-RADPATH will be of particular interest in that it will provide a considerable amount of information from the USSR, information which previously was either not available or only accessible with great difficulty outside of that country.

About 50 Soviet scientists, coming from the different Republics involved or concerned by the 2 accidents in the USSR, are participating at this meeting to present the latest information available.

On behalf of the Commission of the European Communities and on behalf of the organizers of this seminar it is a honour and a real pleasure for me to welcome our Soviet colleagues here today. I think it is the first time that at a scientific meeting in the E.C. so many Soviet experts are participating. We are convinced that the world-wide exchange of information on the subject, covered at this seminar, will contribute considerably to a better knowledge and understanding of the impact on men of nuclear accidents and, therefore, to a safer and healthier environment.

The importance of the Seminar is reflected by the fact that scientists from over 20 countries are present here today. On behalf of the organizers I wish you all hearty welcome to this meeting. I am convinced that we will have a fruitful Seminar.

Opening Address

A. AARKROG

International Union of Radioecologists
Risø National Laboratory, Roskilde4000, Denmark

M. A. AARKROG, President of the IUR

Ladies and Gentlemen, on behalf of the International Union of Radioecologists, I wish you all a hearty welcome to this CEC-IUR seminar on major nuclear accidents. Nobody likes accidents to happen, those human and economic costs can be very serious indeed. But when nuclear accidents happen, it is the obligation of radioecologists to extract as much information as possible from such an event. This seminar should be seen in this context. If we look at the three accidents mentioned to us by F. LUYKX, we will notice that from a pedagogical point of view they are very useful. The first accident will learn us a lot about the behaviour of Strontium-90 in the environment, that was the Kyshtym accident in the Urals. The Windscale accident told us about iodine-131. The behaviour of this radionuclide in the environment and the Chernobyl accident has first of all learned us about the behaviour of caesium-137 in the environment. IUR is in particular happy to see the 46 soviet scientists attending this meeting because international cooperation within radioecology is one of our major aims. I hope that we shall spent 5 fruitful days together here in Luxembourg.

Session II

ACCIDENT SOURCE TERMS

The Kyshtym Accident: Causes, Scale and Radiation Characteristics

G.N. ROMANOV, B.V. NIKIPELOV, E.G. DROZHKO

ABSTRACT

The Kyshtym accident took place on 29 September 1957 at a plutonium separation plant. The accident was caused by the explosion of dry nitrate and acetate salts in a tank containing highly radioactive wastes as a result of a failure in the cooling system and the consequent self-heating of the wastes. The explosion dispersed approximately 2 million curies of nuclear fission products, of which ^{144}Ce and ^{95}Zr accounted for 91%. Long-lived ^{90}Sr accounted for only 2.7% of the dispersed mixture but was responsible for the long-term radiological hazard within what became known as the Eastern Urals radioactive trail. An area of 300 x 50 km received a minimum contamination level of 0.1 Ci of $^{90}Sr/km_2$, and an area 105 x 9 km a minimum level of 2 Ci $^{90}Sr/km_2$. The spatial distribution of the contamination was fairly typical of models of single-point discharge and dry atmospheric deposition of contaminants; the result was a sharply defined trail axis and a steady falling-off of contamination level both along and across the axis. The maximum contamination was 4 000 Ci of $^{90}Sr/km_2$. The initial exposure dose rate reached 150 μR/h per 1 Ci of $^{90}Sr/km_2$ and was mainly due to ^{95}Zr and ^{95}Nb. The exposure dose over 30 years was 0.5 R/(Ci $^{90}Sr/km_2$), of which 0.42 R/(Ci $^{90}Sr/km_2$) was formed during the first year. As a result of radioactive decay, contamination by all radionuclides decreased over 30 years by more than 30 times, and fell by half in the case of ^{90}Sr, while the exposure dose rate decreased by 2 800 times and radionuclide concentration in the various parts of the environment by 10^3–10^4 times.

All the short-lived radionuclides decayed within the first five years, after which time ^{90}Sr was practically the only factor determining the radiation and radiological characteristics of the Eastern Urals radioactive trail. The processes governing ^{90}Sr migration in the environment and in human food chains determine the radiological consequences of the accident for human beings.

1 Causes and scale of the accident

In 1957, almost at the same time as the Windscale accident in Great Britain, there was a major radiation accident in the Southern Urals resulting in the radioactive contamination of a vast area and leading to the implementation of a series of urgent and long-term measures for the radiological protection of the population. The accident occurred at the first Soviet nuclear installation, located near the town of Kyshtym in Chelyabinsk oblast (region), dedicated to the production of plutonium for military purposes; the installation contained a radiochemical plant for separating the plutonium.

As always, the new technology required the solution of a series of difficult problems. Even today, the processing and storage ofradioactive waste has not been satisfactorily resolved, but in the early history of plutonium production one practical and acceptable method of dealing with radioactive waste was to store it on a long-term basis in water-cooled metal tanks encased in concrete. The heat generated by the decay of radionuclides in the waste was dissipated by a water cooling system.

Corrosion and the failure of monitoring equipment led to a breakdown in the cooling system of a 300 m^3 tank; insufficient monitoring allowed the 70-80 tonnes of highly radioactive wastes stored there, mainly in the form of nitrate and acetate compounds, to heat up. The water evaporated, the sediments dried out and heated up to a temperature of 330 - 350°C, leading on 29 September 1957 at 16.20 local time to the contents of the tank exploding with a force estimated at between 70 and 100 tonnes of TNT.

Of the 20 MCi of radioactive material contained in the tank, approximately 2 MCi was ejected into the air to a height of approximately 1 000 m, forming a radioactive cloud. Fallout from this cloud, blown in a north-easterly direction from the plant by the wind, caused radioactive contamination of areas along the path of the cloud in the Chelyabinsk, Sverdlovsk and Tyumen oblasts. This area was later referred to as the Eastern Urals radioactive trail.

Prior to 1957 there had been no similar instances of radioactive contamination of large areas of the Soviet Union; and this dramatic situation, which demanded rapid action to deal with the consequences of the accident and protect the population, was aggravated not only by the lack of practical skills for coping with accidents of this type, but also a lack of understanding by scientists of the behaviour of radioactive nuclides in the environment, the methods and conditions governing the irradation of people, flora and fauna, and the degree of radiation hazard. Scientific knowledge of environmental radioactive contamination and of irradiation pathways and levels among the population was still basic, and the isolated initial results obtained by Soviet and foreign researchers were classified and unavailable for practical use.

Nevertheless, under these difficult circumstances the first real steps were taken towards evaluating radiation hazards and methods for protecting the population, and these soon became closely linked with measures to restore normal productive and everyday activities in a significant proportion of the contaminated area. In addition, as fate would have it, Soviet researchers were presented with a unique opportunity for field experiments; it was this that brought about the development of radioecology in the Soviet Union.

2. Radiation Characteristics

The radioactive material dispersed by the explosion primarily comprised short-lived radionuclides (Table 1). For a long time following the accident, however, the main radiation hazard was the presence in the mixture of long-lived ^{90}Sr (2.7% of total activity) together with its daughter product ^{90}Y. The composition of the radionuclide mixture was similar to that of the fission products formed in a nuclear reactorafter approximately one year, when all the shortest-lived nuclides have decayed, but with one difference. The method used in the waste reprocessing plant involved concentrating these wastes by means of precipitation with NaOH. With this method, the sediment put into storage after dissolution contained practically all the radionuclides with the exception of caesium which, as a soluble Group 1 element, remained in the alkaline solution and was later concentrated separately.

There was therefore almost no caesium in the radionuclide mixture. This was not taken into account, however, by foreign researchers and subsequently led to incorrect conclusions both in the analysis and, above all, in the assessment of the extent of the consequences.

Table 1

Radionuclide content of released material

Radionuclide	Half-life	Type of radiation	Contribution to activity of mixture
^{89}Sr	51 days	beta, gamma	traces
^{90}Sr + ^{90}Y	28.6 years	beta	5.4 (2.7x2)
^{95}Zr + ^{95}Nb	65 days	beta, gamma	24.9
^{106}Ru + ^{106}Rh	1 year	beta, gamma	3.7
^{137}Cs	30 years	beta, gamma	0.036
^{144}Ce + ^{144}Pr	284 days	beta, gamma	66
^{147}Pm	2.6 years	beta, gamma	traces
^{155}Eu	5 years	alpha, beta	traces
Pu		gamma	traces

At the moment the trail was formed the fallout mixture emitted gamma radiation with a total energy of 7.63 MeV per ^{90}Sr disintegration (adopted as the "benchmark" radionuclide on account of its significant half-life), and beta radiation with an initial energy total approximately three times greater. The gamma radiation of the mixture decreased markedly as a result of the subsequent radioactive decay of short-lived gamma-emitting nuclides (Fig. 1) and now beta radiation alone, almost exclusively from ^{90}Sr and ^{90}Y, is the significant factor in the contamination.

The Eastern Urals radioactive trail was basically formed by the fallout of radioactive material from the passing cloud.

The time at which radioactive substances began to settle on any given point depended on distance from the source and the average speed of the cloud.

The duration of fallout ranged from several minutes at the beginning of the trail to 30-60 minutes at its furthest extent.

Due to the lack of atmospheric precipitation during the formation of the trail, and also the occurrence of periods of dry weather and strong winds until the constant autumn rains began and settled snow cover was established, some redistribution of radioactive material by wind was observed in places during the first four to six weeks; this led to changes in the radioactive contamination in those parts of the trail close to the accident site, where levels of contamination were highest. Thus the trail is wider at the beginning than towards the end, where it "peters out" in an easterly direction.

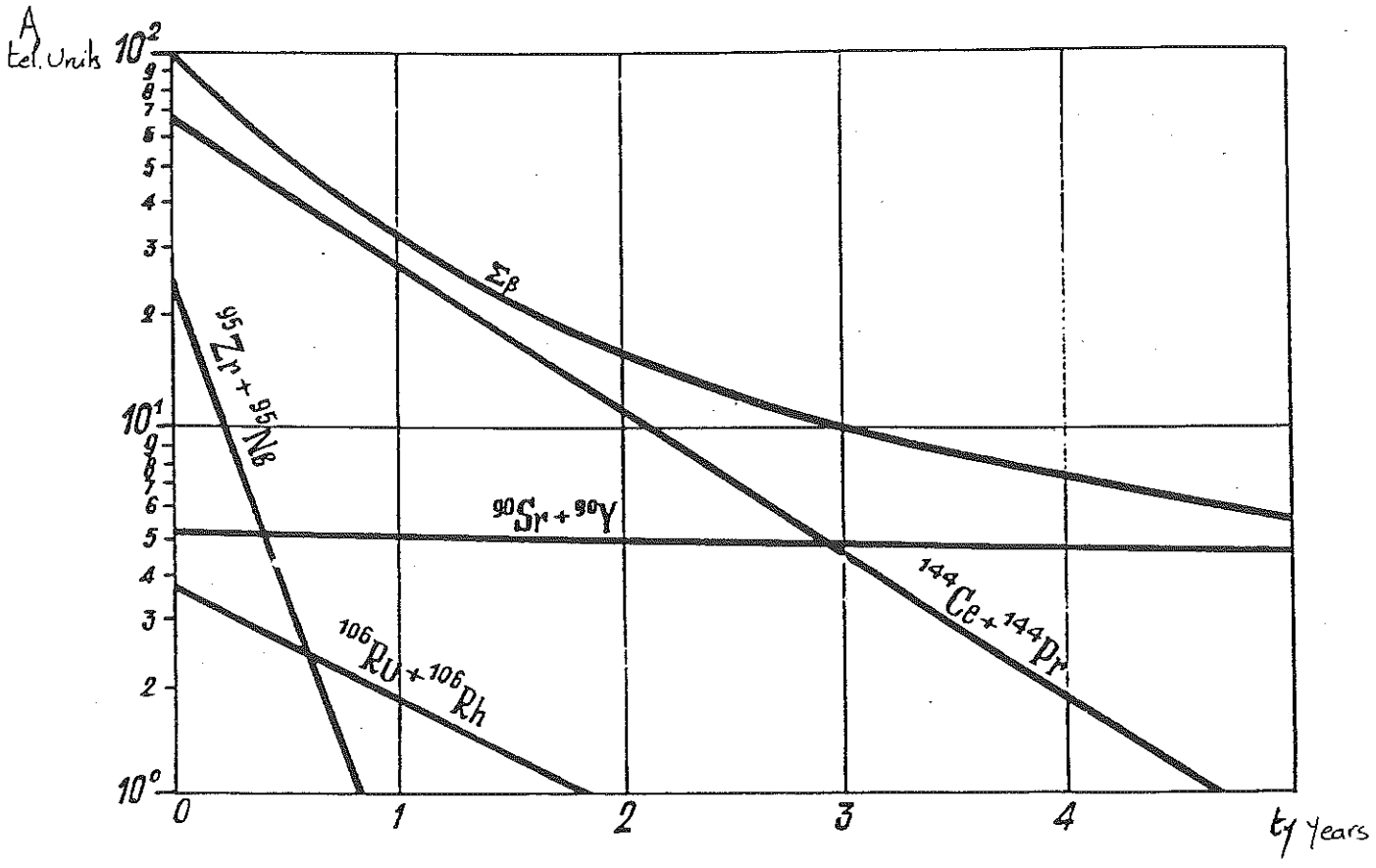

Fig. 1. Radioactive decay of the nuclide mixture in the contaminated area during the first five years

The area within the contamination contour of 0.1 Ci/km_2 of strontium-90 (the minimum level detected, equal to twice the level of overall radioactive contamination by strontium-90 for the given region in 1957) had a maximum length of 300 km, extending close to the town of Tyumen, and was 30-50 km wide (Fig. 2); the 2 Ci/km_2 contour for strontium-90 was 105 km long and 8-9 km wide. A strontium-90 contamination level of 2 Ci/km_2 was considered the maximum safe limit for habitation and was adopted as the official boundary of the Eastern Urals radioactive trail. The total area exposed to that level of radioactive contamination covered approximately 1 000 km_2, while the area within the 0.1 Ci/km_2 contour was some 20 000 km_2.

The Eastern Urals radioactive trail displays quite natural territorial distribution characteristics, namely a pronounced axis along which the contamination level steadily diminishes (from 4 000 Ci/km_2 of strontium-90 at the start to 0.1 Ci/km_2 at its furthest extent). Transverse distribution of contamination is characterized by sharply pronounced maxima along the axis of the trail, exceeding the peripheral density values by 1-4 orders of magnitude. Table 2 shows the territorial distribution of strontium-90 contamination by level.

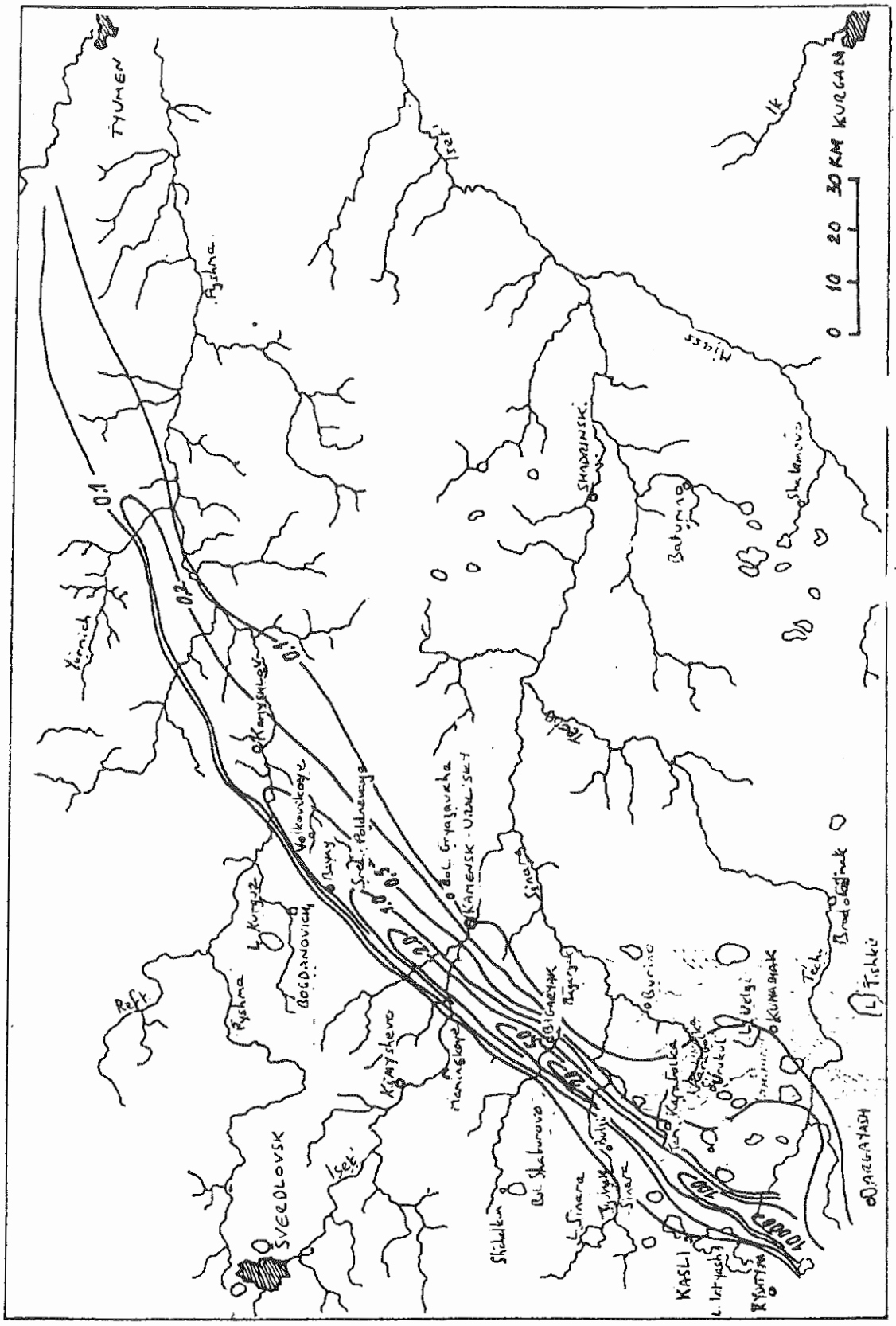

Fig. 2. The Eastern Urals radioactive trail.

Table 2

Contamination levels and areas affected

Strontium-90 contamination, Ci/km_2	Area affected in km_2
0.1–2	15 x 10_3
2–20	600
20–100	280
100–1 000	100
1 000–4 000	17

Note: Boundaries of area contaminated with 0.1 Ci/km_2 of strontium-90 are not completely certain.

The radioactive fallout was not initially immobilized in the environment, and its presence was noted in absolutely everything, including living organisms and foodstuffs. Depending on its location relative to the source, initial levels of radioactive contamination (based on total beta activity) compared with the period prior to the accident increased by 10_2 – 2 x 10^5 times in natural grassland, 1.5 – 3 x 10^4 times in open bodies of water, 25 – 1 000 times in wheat grain and 10 – 2 000 times in cow's milk. The main pathway of radioactivity uptake into crop produce was direct surface contamination.

In the initial stages, the exposure dose rate for gamma radiation in the open at a height of 1 metre was 150 μR/h calculated on the basis of 1 Ci/km_2 of strontium-90; of this amount approximately 90% came from ^{95}Zr and ^{95}Nb. At maximum contamination of approximately 4 000 Ci/km_2 of strontium-90, the initial gamma exposure dose rate was 0.6 R/h (Table 3).

Subsequently, the situation in the contaminated area underwent considerable changes. The basic factors which influenced and continue to influence the situation are as follows:

- the radioactive decay of gamma-emitting nuclides;
- the redistribution of radioactive substances in natural systems, including working down into soil and bed sediments;
- biogeochemical migration of radionuclides;
- human economic activity, including measures for the radiological protection of the population.

The general dynamics of the radiological situation in the area of the Eastern Urals radioactive trail are illustrated in Table 3.

Table 3

Dynamics of radiological conditions within the Eastern Urals radioactive trail

Radiological situation indicator	Number of years following accident					
	0	1	5	10	25	75 (forecast)
Contamination level (relative units)						
As regards total beta activity	1	0.34	0.057	0.043	0.030	0.0088
As regards ^{90}Sr	1	0.96	0.89	0.78	0.52	0.16
Gamma radiation exposure dose rate at a height of 1 m, $\frac{\mu R/h}{Ci\ ^{90}Sr/km_2}$	150	8.7	0.33	0.15	0.053	0.017
Gamma radiation exposure dose at a height of 1 m, $\frac{R}{Ci\ ^{90}Sr/km_2}$	0	0.42	0.49	0.49	0.50	0.50
Total concentration of radionuclides (relative units)						
Grass	1	0.1	0.01	0.004	0.001	0.0001
Grain	1	0.2	0.01	0.008	0.002	0.001
Milk	1	0.1	0.01	0.008	0.005	0.001
Water (in lakes)	1	0.03	0.01	0.0075	0.003	0.0005

Due to radioactive decay, the contamination level of the radionuclide mixture was over 30 times less after 30 years; levels of strontium-90 had reduced by half. For that reason, gamma-radiation energy fell from 7.6 to 0.004 meV per ^{90}Sr disintegration; this led to a 2 800-fold reduction in the exposure dose rate for gamma radiation at a height of 1 m (allowing for migration down into the soil). The gamma radiation exposure dose, amounting over 30 years to 0.5 R calculated on the basis of 1 Ci/km_2 of strontium-90 and almost all formed within the first year following the accident, has up until now increased by only 16%. This means that gamma irradiation of people and flora and fauna in the contaminated area was prevalent throughout the first 12-18 months. The concentration of all radionuclides in various parts of the environment, including agricultural produce, declined during this period by hundreds or thousands of times, with the maximum decrease taking place during the first 5 years. Since then, ^{90}Sr has been the only factor governing the radioactive contamination of all flora and fauna and inanimate organic matter, and the subsequent decline in levels of radioactive contamination was conditional on the underlying mechanisms governing the behaviour of ^{90}Sr in the environment.

3. Population irradiation doses

There were several pathways involved in irradiation of the population in the contaminated area (totalling some 270 000 people).

External irradiation of the whole body and of internal organs was governed by:

1) gamma irradiation from the passing cloud of released material;

2) gamma irradiation from contaminated soil and dwellings;

3) beta and gamma irradiation from contaminated skin and clothing.

Internal irradiation stemmed from intake of radioactive substances into the organism with inhaled air or in food and water, and the resulting short- or long-term (in the case of ^{90}Sr) presence of radionuclides in human tissue and organs.

In the initial stages, external irradiation was the predominant factor in the contaminated area; later, internal irradiation came to dominate, due to ^{90}Sr intake via food and its deposition in human bones. The long-term (over 30 years) formation of radiation doses may be divided into two periods: the "acute" or initial 12-18 month period of mainly external irradiation and the later one, with predominantly internal irradiation. At the time the cloud passed, external irradiation was estimated at 0.13 mrem/(Ci $^{90}Sr/km_2$). Internal irradiation of the lungs, caused by the inhalation of radioactive substances, was estimated at 5-300 mrem/(Ci $^{90}Sr/km_2$) throughout the subsequent period of activity in the lungs, depending on the degree of solubility of the radioactive substance in pulmonary fluid.

Most of the external irradiation dose to the public was received in the "acute" period (Table 4). Of the overall dose over 30 years - 260 mrem/(Ci $^{90}Sr/km_2$) - more than half (160 mrem) was absorbed during the first 120 days, and approximately 90% during the first two years. The most critical organs were in the gastro-intestinal tract; these received the largest doses of internal irradiation during the "acute" period. Of the 30-year irradiation dose for the gastro-intestinal tract - 2 rem/(Ci $^{90}Sr/km_2$) - 12% was absorbed during the first ten days, and 80% during the first year. During the "acute" period radioactivity also accumulated in the bone tissue and red marrow due to deposition of ^{90}Sr in the bones; the dose absorbed in these tissues increased from 9 and 3 mrem respectively after the first month to 720 and 220 mrem/(Ci $^{90}Sr/km_2$) at the end of the first year.

Table 4

Mean population irradiation doses since the formation of the Eastern Urals radioactive trail, in mrem/(Ci $^{90}Sr/km^2$)

Irradiation pathway Irradiated organs and tissues	10 days	30 days	120 days	1 year	2 years	5 years	10 years	15 years	20 years	30 years
Dose equivalent										
External irradiation	33	67	160	220	230	250	260	260	260	260
Internal irradiation:										
Gastro-intestinal tract	250	480	1 200	1 600	1 800	1 900	2 000	2 000	2 000	2 000
Bone	3.1	9.4	46	720	1 500	3 300	5 400	6 600	7 300	8 000
Red marrow	0.97	2.9	14	220	490	1 000	1 700	2 000	2 300	2 500
Effective dose equivalent	93	180	450	700	780	920	1 100	1 100	1 200	1 200

The leading role of ^{90}Sr in the internal irradiation dose among the population and, to a significant extent, in the effective dose equivalent stemming from this irradiation, began to become apparent in the second year after the accident. Most of the foodstuffs consumed by the rural population were produced locally, chiefly on private plots, and ^{90}Sr intake derived mainly from consumption of locally produced milk, meat, potatoes and vegetables. Some 50-70% of ^{90}Sr intake came from milk, 5-25% from meat and 15-45% from potatoes and vegetables. There were no great changes in this intake ratio during the later period, although the radiological measures taken to protect the population, together with natural processes influencing ^{90}Sr availability for plants, led to a systematic fall in produce contamination levels and, finally, to a constant decline in the annual intake of ^{90}Sr in human beings via foodstuffs (Fig. 3). The ^{90}Sr content in foodstuffs halves every 5.5 years. For these reasons, the intensity of ^{90}Sr intake into the organism and its deposition in the bones have declined over the course of time, leading to lower increases in the dose rates for bones and red marrow over the last 15-20 years. Over 30 years the dose to bone tissue, within the 1 Ci/km$_2$ (^{90}Sr) contour, was 8 rem, and to red marrow 2.5 rem; in each case, half of the dose accumulated during the first 6-7 years. Over the course of 30 years the effective dose equivalent amounted to 1.2 rem/(Ci ^{90}Sr/km$_2$); external irradiation accounts for 22% of this dose, internal irradiation of bone tissue for 21% and internal irradiation of red marrow 28%.

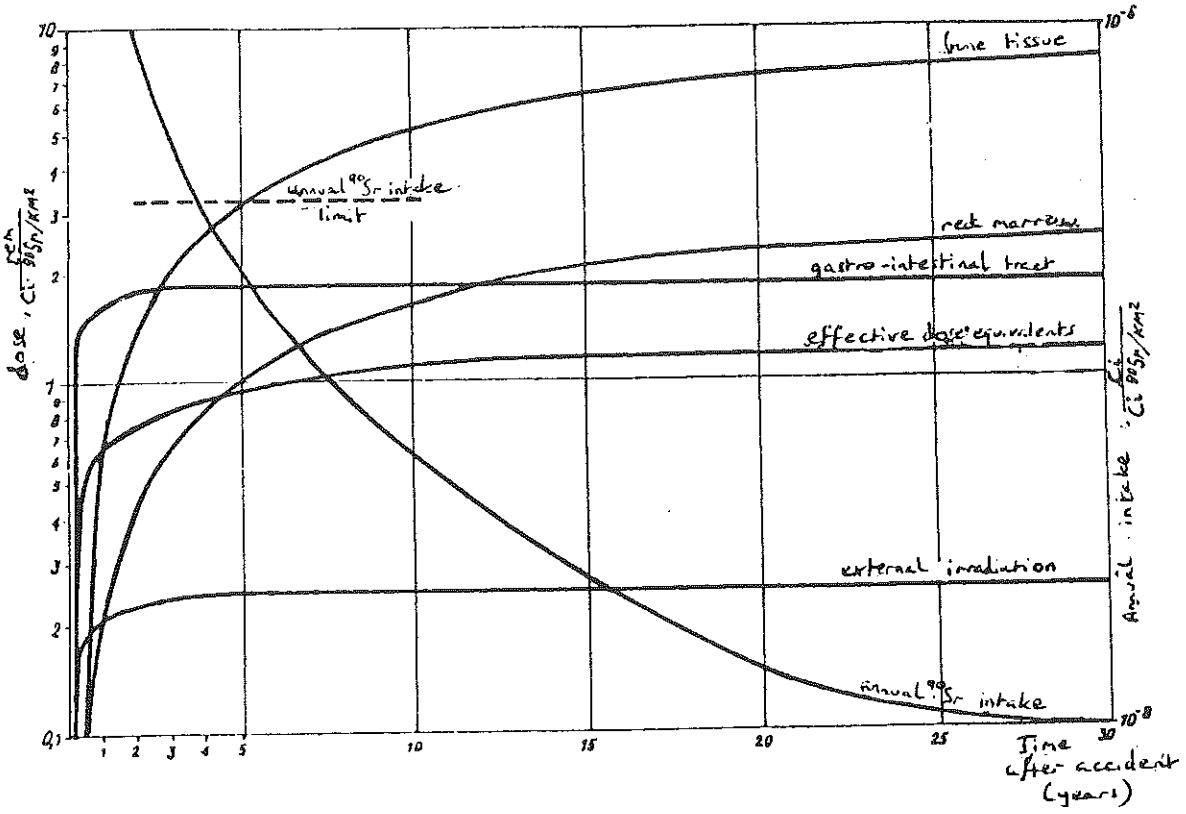

Fig. 3: Dose burdens in the non-evacuated population.

Despite the lack of experience with radiation accidents, especially those involving intense radioactive contamination of large areas, the strategy and tactics adopted for the urgent (and then the systematic) measures taken for the radiological protection of the population within the Eastern Urals radioactive trail appeared to be correct and even, from a present-day point of view, unnecessarily very much on the safe side.

The main urgent measures taken immediately after contamination of the area included the following:

1. Evacuation of nearby population centres where the potential external irradiation dose might have exceeded 100 rem during the first month.

2. Medical treatment for evacuees, replacement of clothing, introduction of a ban on the removal of personal effects and stocks of food by people in this category.

3. Introduction of radiation and dosimetric checks in the most contaminated area, accompanied by restrictions on access to that area.

Urgent evacuation (which in effect meant resettlement) was carried out within the first ten days from the four villages closest to the plant and housing some 1 100 people. The evacuees were accommodated in uncontaminated towns and villages and given housing and work. Irradiation doses for evacuees are given in Table 5.

The subsequent systematic measures to reduce population irradiation levels during the "acute" period included the following:

1. Monitoring radioactive contamination levels in foodstuffs and agricultural produce, destruction of produce containing levels higher than permitted and a guaranteed supply of uncontaminated foodstuffs to replace them.

2. Further evacuation of the population.

3. Introduction of restrictions on public access to - and economic activity in - part of the contaminated area.

4. Decontamination of populated areas and agricultural land.

The monitoring of contamination levels in produce, and the destruction of that produce, were dictated by the need to ensure an immediate reduction in human intake of radioactivity via food. The alternative solution to this problem - the systematic and long-term guaranteed supply of uncontaminated foodstuffs from other regions - was unrealistic: if people were forbidden to grow and consume agricultural produce there would be no point in their staying on in the countryside. On the basis of the provisional standard worked out - a permitted annual ^{90}Sr intake of 1.4 μCi/year - the need for continuous radiation monitoring and, if need be, destruction of produce, was recognized. The monitoring covered territory with a minimum contamination level of 0.5-1 Ci/km_2 of ^{90}Sr, an area of approximately 1 000 km_2 (50 population centres). During the first two years, more than 10 000 tonnes of various types of produce were destroyed.

Given the impossibility of completely replacing contaminated foodstuffs with "clean" foodstuffs in towns and villages where ^{90}Sr intake via food exceeded the provisional permitted annual level, the decision was taken to carry out a further systematic evacuation of the population from areas where contamination exceeded 4 Ci/km$_2$ of ^{90}Sr.

Resettlement priorities were established on the basis of the local contamination level and the degree of economic exploitation of the surrounding land. The resettlement was begun 8 months (and completed 18 months) after the initial contamination. In all, together with the urgent resettlement, more than 10 000 people from 23 rural towns and villages were moved (Table 5).

Table 5

Resettlement measures and their role in reducing irradiation levels in the population

Index	Population groups					
	I	II	III	IV	V	Total
	Urgent resett-lement	Syst	ematic	resetti	ement	
Number of towns and villages	4	1	5	7	6	23
Number of people (x 1 000)	1.1	0.3	2.0	4.2	3.1	10.7
Mean contamination level in Ci $^{90}Sr/km^2$	500	65	18	8.9	3.3	
Duration of post-contamination residence prior to resettlement in days	10	250	250	330	670	
Mean irradiation doses received prior to resettlement, in rem						
Dose equivalent						
External irradiation	17	14	3.9	1.9	0.68	
Internal irradiation						
Gastro-intestinal tract	150	98	27	13	5.4	
Bone	1.6	10	2.8	5.8	4.4	
Mean effective dose equivalent	52	14	12	5.6	2.3	

Table 5 shows that the maximum mean dose received by evacuees over a period of 30 years amounted to an effective dose equivalent of 52 rem, and 150 rem to the gastro-intestinal tract. The corresponding minimum mean doses were 2.3 and 4.4 rem, which is close to the level of irradiation doses in the non-evacuated population.

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One Western Perspective of the 1957 Soviet Nuclear Accident

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ABSTRACT

Much information about a 1957 chemical explosion of high-level radioactive waste at the Chelyabinsk-40 (Ch-40) plutonium-production center in the Urals is available, but seeming inconsistencies and the complex history of Ch-40 limit its interpretation.

The total radioactivity released was 20 MCi and the combined activity of ^{90}Sr and ^{137}Cs released was 1 MCi, comparable to releases from the 1986 Chernobyl reactor accident. In contrast to the latter, however, only about 10% of the 1957 release was more widely dispersed and deposited over an area of about 20,000 km^2 along a $\geq$300-km-long track. Further, ^{137}Cs comprised about 80% of the combined activity of ^{90}Sr and ^{137}Cs from Chernobyl, but only a small fraction of that from the 1957 accident.

Nearly 11,000 persons were relocated from 23 populated places in a 90-km-long (700-km^2) area containing $\geq$2-4 Ci/km^2 ^{90}Sr; 24% of this area is still uninhabitable--now dedicated for radioecological research. Over 1100 people were evacuated within 7-10 d from an "extreme evacuation zone", and the rest were relocated in stages over 250-670 d. Acute radiation effects were observed in farm animals and natural ecosystems in and near the "extreme evacuation zone" but not, reportedly, in humans. Temporary agricultural restrictions were also applied to the 700-km^2 area ($\geq$2-4 Ci/km^2 ^{90}Sr), but, in the remainder (<2 Ci/km^2 ^{90}Sr), restrictions were not applied and impacts appear to have been slight.

Releases from Ch-40 contaminated the Techa River drainage and required evacuation of $\geq$7500 persons prior to 1957. A small reservoir (Lake Karachay), containing 120 MCi of radioactivity from early intermediate-level waste disposal practices, was the source of additional releases. There are also a number of unconfirmed reports of reactor accidents at Ch-40. The extent to which other releases contributed to reports of casualties attributed to the 1957 accident and/or to the need for a massive hydrologic isolation system at Ch-40 is not yet clear.

INTRODUCTION

Much information about a 1957 explosion of high-level radioactive wastes (HLW) and other happenings at the Chelyabinsk-40 plutonium-production complex (Ch-40; also called the Kyshtym nuclear complex) in the eastern Urals has been released by the Soviet Union prior to the seminar in Luxembourg [1-18]. However, this information has not been reported in a clear and consistent manner. Among other things, this may indicate that the enumeration of events, including accidents, is not yet complete. A 1974 Soviet report, which was given a wide distribution in 1990 [7], provides information not currently available elsewhere, but also contains serious errors and inconsistencies that limit its usefulness.

BACKGROUND

In our analysis of the 1957 accident [19], we identified an area near Ch-40 in which over 30 communities had disappeared (Fig. 1) and identified a major hydrologic isolation system, consisting of several cascaded reservoirs and canals (Fig. 2), apparently designed to limit the spread of radioactive contamination down the Techa River. We hypothesized that the narrow, southwest-northeast arm of this area had been contaminated by an aerosol resulting from the accident, but that the southwest-southeast arm containing the hydrologic isolation system had been contaminated primarily by a liquid release(s), possibly including earlier chronic releases. The situation is even more complex than we imagined in 1979.

Most of the communities from which the human population was relocated were very small (villages and collectives <2000 persons), but five had populations ≥2000. Locations of the larger communities are indicated by symbols within the fully enclosed dashed area in Fig. 1. Proceeding from the northeast to the southeast end of the dashed area, these were: Boyevka, Yugo-Konevo, Russkaya Karabolka, Metlino, and Asanovo. The villages of Metlino and Asanovo each were located near dams of reservoirs in the hydrologic isolation system downstream from Lake Kyzyltash, a large natural water body in the Techa River system north of Ch-40 (Fig. 2). Lake Kyzyltash was itself contaminated through its use as a cooling water source for the reactors at Ch-40 [18, 19, 21].

Releases originating in 1948 from Ch-40 resulted in contamination of the Techa River that required relocation of ≥7,500 persons, including the inhabitants of Metlino and Asanovo, prior to 1957, and initiation of the hydrologic isolation system [4, 17, 18, 20]. All persons in communities

from Metlino downstream to Muslyumovo (see Fig. 3) were relocated. These releases, amounting to 3 MCi (110 PBq) in 1949-1952 (25% ^{90}Sr + ^{137}Cs), appear to have resulted in large part from a lack of early waste treatment capability and the storage of radioactive wastes in open, unlined earthen reservoirs [17-20; G. N. Romanov, pers. comm., October 19, 1990].

The villages of Russkaya Karabolka, Yugo-Konevo, and Boyevka were part of a group of 23 populated points (10,730 inhabitants [6, 8, 14, 18]) that were evacuated in 1957-1959, following the HLW explosion that took place on September 29, 1957. These villages were located within a 700-km^2 "sanitary-protective zone" (Fig. 1) in which the ^{90}Sr aerosol deposition was $\geq$4 Ci/km^2 (0.15 TBq/km^2) [7, 14]. The most heavily contaminated part of this zone was designated as the "extreme evacuation" zone (Fig. 2), from which the residents were removed within 7-10 d [7]. A much larger area (est. 15,000-23,000 km^2) was contaminated at levels of 0.1-2 Ci/km^2 ^{90}Sr (Fig. 3). Its boundary was the level of ^{90}Sr deposition from the 1957 accident equal to the background level (about 0.1 Ci/km^2) produced by global fallout from nuclear weapons testing [7, 11].

A small reservoir at Ch-40 (L. Karachay; note the small unnamed water body south of L. Kyzyltash in Fig. 2), containing 120 MCi (4400 PBq) of radioactivity from early intermediate-level radioactive waste disposal practices (10, 21), was the source of additional releases--both liquid and aerosol--and is the focus of intensive remedial actions [17]. In 1967, about 600 Ci (22 TBq) of radioactivity (primarily ^{137}Cs and ^{90}Sr; ^{137}Cs:^{90}Sr ratio 3:1) in dry contaminated soils or sediments from its shoreline were reportedly dispersed by the wind (during a tornado?) over an area of 1,800 km^2 [17]. The resulting contamination (maximum ^{90}Sr level 10 Ci/km^2) was reportedly superimposed on the area contaminated by the 1957 accident. We think that this may have expanded the overall area of contamination, producing the bulge at its southeastern extremity (see Fig. 3), because this feature is not shown on maps in [7].

This area of the Urals reportedly received elevated nuclear weapons fallout in May 1958 from an underground test on Novaya Zemlya [3]. The resulting contamination was apparently insignificant compared to the levels produced by the 1957 accident and other releases from Ch-40.

Contrary to information available previously in the West (22-25), early Soviet reports indicated that none of the releases, including the 1957 HLW explosion, had produced casualties or had involved reactor accidents. However, there were deleterious health effects among those people exposed

to contamination of the Techa river before 1957. A population ≥12,800 persons living along a 230-km stretch of the Techa River received significant radiation doses: Mean effective radiation dose equivalents in exposed communities 3.6-140 rem (0.036-1.4 Sv). An excess incidence of leukemia amounting to 14-23 cases was reported [18].

Earlier indications that Ch-40 was operated "under extremely difficult conditions". . . which had a deleterious effect on the health of the staff" [1] have now been confirmed. A significant fraction of the work force received annual radiation doses of 100 to >400 rem (1 to >4 Sv) during its early history (1949-1951), and the consequences have been described [9].

There are a number of reports--all unconfirmed and strongly denied by our Soviet colleagues--of accidents associated with reactor operations at Ch-40 (e.g., 22-25), some from seemingly authoritative sources such as Igor Kurchatov (R. Wilson, Harvard Univ., Cambridge, Mass., pers. comm., June 8, 1981) and Andre Sakharov (24). These reports include two contemporary newspaper articles (25) which detailed evacuations and casualties resulting from a February 1958 release produced by the breakdown of a filter system.

There are too many reports of casualties (22-25) and/or other accidents associated with Ch-40 to dismiss these entirely on the basis of current information. The question of whether casualties resulted from the 1957 HLW explosion or its aftermath may also be open.

CHARACTERISTICS OF THE AFFECTED REGION

The total area contaminated by the 1957 HLW explosion covered 15,000-23,000 km^2 (parts of the Chelyabinsk, Sverdlovsk, and Tyumen' provinces) (Fig. 3), contained 217 communities, and included a 1957 population of about 270,000 persons [7, 8, 11]. The nationality of the inhabitants was primarily Russian (about 75%); most of the remainder had Tartar or Bashkir roots [7]. Only one major city (Kamensk-Ural'skiy; 1959 population 141,000) was inside the 1957 deposition zone (Fig. 3). Fig. 1 in [7] places part of Kamensk-Ural'skiy in the "sanitary-protective zone," but appears to be in error (see Fig. 23 in [7] and Fig. 1 in [11]).

The contaminated areas identified in Figs. 1-3 were largely rural in character: Cropland, pastures, and hay fields reportedly covered 60% of the area [7], but also see [12]. The remaining area was approximately divided between forests and natural aquatic ecosystems (lakes, bogs, and rivers). The primary natural terrestrial ecosystems in the deposition zone are forest-steppe over the initial 100 km (i.e, nearest to the accident site)

and southern taiga in the remainder [26]. Principal canopy species in the forest-steppe were birch (B. verrucosa), pine (P. silvestris), or admixtures. There were significant stands of pine over the first 30 km of the deposition zone resulting from the 1957 HLW explosion [7]. See, e.g., [7, 12, 14, 26, 27] for more on the environmental setting (climate, soils, characteristics of aquatic ecosystems, etc.) and socioeconomic factors.

THE CAUSE OF THE 1957 ACCIDENT

The Soviet commission which investigated the 1957 accident concluded that the most likely cause was a chemical explosion of stored HLW. The measuring and control system for the tanks failed, its design and high radiation fields prevented repair, and the stored HLW wastes overheated and began to evaporate. Since the tanks were cooled externally and were entirely immersed in water, they gradually floated as liquids evaporated. Resulting leaks in several tanks led to contamination of the cooling water, forcing adoption of an ineffective regime of periodic cooling [10]. Conditions deteriorated, leading to the explosion, which demolished the tank, blew off a 2.5-m-thick concrete plate ([29]; said to be 25-m-thick in [10]) covering the cell [10], and left a "crater" [17]. The energy for the explosion and resulting dispersal of radioactive materials was thought to have been about 75 t of trinitrotoluene (TNT)-equivalent, provided by an acetate-nitrate reaction in the drying, concentrated wastes (Table 1).

The tank in which the explosion took place was one of 20 stainless-steel tanks, each isolated in a concrete-walled compartment ("canyon-cell"), located in a large, rectangular, reinforced-concrete structure. (Other information indicates that the explosion took place in a 300-m^3 concrete tank [2, 3, 8] located in a 16-tank HLW storage complex [28].) Discharges of HLW to this storage complex and of intermediate-level radioactive wastes to L. Karachay both began in 1953 [10].

The accident scenario was also described by Y. I. Mikerin, manager of the Ch-40 reprocessing plant. He said that the cooling system, reportedly internal, in one of the tanks began to leak and was shut off in 1956. More than a year lapsed before a spark from a control device detonated the salts and "obliterated the tank" [30]. Some of the details provided by Mikerin are inconsistent with [10], but it is his timing of events leading to the explosion that provokes the most serious questions (discussed below).

THE RADIOACTIVITY RELEASED

The total radioactivity released by the 1957 HLW explosion was about 20 MCi (740 PBq) (Table 1), and the combined activity of ^{90}Sr-^{90}Y plus ^{137}Cs released was 1 MCi (37 PBq) [2, 3, 8, 10]. These amounts are comparable to the releases from the Chernobyl reactor accident, 50 and 2 MCi (1850 and 74 PBq), respectively [32]. Whereas ^{137}Cs represented about 80% of the activity of long-lived materials from Chernobyl, however, it reportedly comprised a small fraction of the 1957 release.

The reported composition of the radionuclide mixture is also shown in Table 1. The major constituents were ^{144}Ce-^{144}Pr (half-life 284 d), which was the principal source of the radiation dose to biological surfaces (i.e, from beta particles) during the first year after the accident ("acute" phase of irradiation). Next in importance were ^{95}Zr-^{95}Nb (half-lives 65 and 35 d, respectively), which delivered most of the external gamma-radiation dose during this same period. The principal source of long-term exposures to humans as well as ecosystems was ^{90}Sr (half-life 28.6 y). Consequently, the areal extent of contamination is referenced to this radionuclide [1, 5-8, 11-18]. Notably reduced in its contribution relative to typical HLW is ^{137}Cs (Table 1). However, there are also large differences in the ^{90}Sr-^{90}Y:^{137}Cs activity ratios given in Soviet reports, i.e., values of 150:1 or 7:1 (Table 1)--as opposed to a ratio of about 1:1 in unseparated HLW [19].

Information provided in earlier Soviet radioecology publications [19] is more consistent with the higher ratio of ^{90}Sr-^{90}Y:^{137}Cs (150:1) given in Table 1. Although we have been assured that the lower value of 7:1, obtained from [7], is in error (G. N. Romanov, pers. comm., October 19, 1990), we do not yet have an explanation for the discrepancy. It is clearly the ^{137}Cs data that are the source of the error because the differences in reported percentages of ^{90}Sr-^{90}Y are slight (Table 1).

We cannot attribute the discrepancies between values for ^{137}Cs in Table 1 to differences in analytical methods because gamma spectrometry was reportedly utilized for ^{144}Ce-^{144}Pr, ^{137}Cs, ^{106}Ru, and ^{125}Sb (?) [33]. The discrepancies between [7] and other sources need explanation.

The ^{95}Zr-^{95}Nb content shown in Table 1 (20-25%) is reasonably consistent among references. However, the ^{95}Zr-^{95}Nb content is highly atypical of materials that had been cooled ($\geq$100 d), reprocessed, and then stored for >1 y--as suggested by Mikerin [30]--(see, e.g., Table 1 in [19]). Because of very short half-lives, these two isotopes do not usually

exceed 20% of the radioactivity in plutonium-production wastes at ≥1 y.

With the exception of a slightly lower ^{106}Ru-^{106}Rh content than expected, the reported composition is reasonably close to that of 1-y-old reprocessed wastes from which ^{137}Cs had been chemically separated [19]. The latter appears to have occurred [3; also G. N. Romanov, pers. comm., October 19, 1990], but details of the separation process and of the chemical composition of the resulting HLW have not been reported. The radiochemical composition also agrees more closely with that in an earlier Soviet study designed to determine criteria for evacuation of areas accidentally contaminated by 200-350-d-old fission products released from radiochemical separations plants [31]. This study was rather obviously based on operational experience following the 1957 accident and was cited by Soviet radioecologists [19]. The waste characteristics in [31] are also in agreement with the external dose rates reported in [11], but not with Mikerin's accident scenario. Thus, this discrepancy needs to be resolved.

THE CONSEQUENCES OF THE 1957 HLW EXPLOSION

Effects at Chelyabinsk-40

Most (90%) of the 20 MCi released by the 1957 accident reportedly "fell out" near the explosion site and very high contamination levels resulted. External exposure rates were initially >400, 20, and 3 R/h at distances of 0.1, 1, and 3 km, respectively, from the explosion "crater" [17]. Based on data in [11], these exposure rates could correlate with ^{90}Sr-^{90}Y levels of roughly 40 mCi/m^2 to >5 Ci/m^2--assuming little variation in fallout composition as a function of distance; total concentrations would have been about 20 times higher (Table 1). Yet decontamination and rehabilitation of the waste storage site, as well as the remainder of the Ch-40 site, was accomplished largely during late 1957 and early 1958 [10].

Although the external dose rate fell significantly (est. factor ≥7) during the six months following the explosion because of radioactive decay and environmental migration, radiation levels close to the explosion site would still have been so high that we surmise that the rehabilitation effort was carried out at great individual cost. This effort brings to mind the dedication and heroism of those who controlled the fires at the Chernobyl reactor. Much more information about the remedial actions undertaken under such extreme conditions would be extremely beneficial.

Given the magnitude of the explosion and leaks from several HLW tanks

before the explosion [10], it seems plausible that other tanks contributed to radioactive releases from the site [e.g., [28]), if only in liquid form. The location of the explosion site and its current condition, e.g., an accounting for the 0.9 MCi of ^{90}Sr-^{90}Y plus ^{137}Cs that "fell out" nearby, have not been reported. Perhaps the greatest concern, however, is whether casualties occurred among those who rehabilitated the HLW storage complex after the accident. This is at least one plausible explanation for the many unconfirmed reports of casualties associated with Ch-40 (22-25).

Environmental Impacts Away from Chelyabinsk-40

A plume of finer particulates containing about 2 MCi (74 PBq) was initially estimated to have been lofted about 1-2 km [7]; reconstructions now indicate a maximum height of 1 km [2, 3, 10]. Virtually all of this was deposited within 11 h along a ≥300-km-long track to the north-northeast and over an area of 15,000-23,000 km^2 [1, 2, 5-8, 10]. Details of the aerosol dispersal characteristics and approximate ground deposition patterns are presented in Table 2. These data include some expansion of the original depositional areas by wind dispersal, principally during 1957 and 1958. Tabulations of gamma-radiation exposure rates as a function of time and surface contamination levels are also provided in [7] but appear to be erroneous (see later discussion).

There are significant inconsistencies in Soviet data for (1) the areal extent of contamination, (2) habitation of contaminated areas, (3) the dynamics of evacuations (Table 3). The data in Table 3 are given as a function of ^{90}Sr concentration (reference), but it should be emphasized that the total initial activity deposited was 37 times greater (Table 1).

Maximum ^{90}Sr concentrations in the far-field deposition zone (i.e, ≥5 km from the explosion site) reportedly were either 4000 or 10,000 Ci/km^2; the latter concentration probably includes the ^{90}Y daughter activity. Inclusion of ^{90}Y daughter activity by a number of authors (e.g., [7, 17]) appears to explain some, but not all, of the inconsistencies in Table 3. Estimates of the total area contaminated range from 15,000 to 23,000 km^2, in part because of uncertainties in the 0.1 Ci/km^2 ^{90}Sr isopleth [11].

Isopleths of ^{90}Sr concentration (^{90}Y daughter activity not included) in [11] appear to be the most comprehensive available, although the level of resolution is still very coarse and ^{90}Sr concentrations are not matched with human populations (Table 3).

Despite the uncertainties implied in Table 3, the population was

apparently evacuated from 23 villages contaminated with ≥2-4 Ci/km^2 ^{90}Sr (≥2 Ci/km^2 near the "extreme evacuation zone" and in the intermediate part of the "sanitary-protective zone", and ≥4 Ci/km^2 ^{90}Sr at the far end of the "sanitary-protective zone"; G. N. Romanov, pers. comm., October 19, 1990). Four villages were mistakenly evacuated from areas with contamination levels 2-4 Ci/km^2 ^{90}Sr (G. N. Romanov, pers. comm., October 6, 1990). This explains in part why [7] indicates that only 19 villages were evacuated and why differences in the total evacuated (e.g., 10,180 vs 10,730) exist. One of these villages, inhabited by 554 persons, was evacuated at 7-10 d after the 1957 explosion: The apparent source of the difference between the reported figures of 1154 and 600.

Estimates of the number of people evacuated in the second wave at 250 d (≥100 Ci/km^2 along main axis; mean ^{90}Sr level reportedly 65 Ci/km^2) appear to range from 280 to 1500, based on information in [1, 5-8, 14], but the correct number appears to be 280. Other groups, inhabiting areas with successively lower levels of ^{90}Sr contamination (≥18 Ci/km^2), were relocated from 250-670 d following the accident. A third group of 2000 persons was relocated at 250 d; a fourth group, numbering 4200, at 330 d; and, finally, a fifth group of 3100 at 670 d, according to [1, 6, 8, 14].

Data in [18] indicate that an additional 220 persons were relocated between 250-330 d (6700 vs 6480), resulting in a total evacuation of 10,854. However, we have been assured that these figures are in error (G. N. Romanov, pers. comm., October 19, 1990).

Such inconsistencies are puzzling and raise needless concerns about the quality of the information. Based on current information, it appears that a total of 10,730 persons were relocated from 23 populated places, 19 of which were inside the ≥4 Ci/km^2 ^{90}Sr concentration isopleth of the 700-km^2 "sanitary-protective zone." Of this total, 1154 people were evacuated within 7-10 d from four villages, three of which were inside the "extreme evacuation zone" shown in Fig. 2. The remaining 9580 persons, inhabiting areas with progressively lower levels of radioactivity, were moved out in stages over 250-670 d [1, 6, 8, 14]. For comparison, ≥115,000 persons were rapidly evacuated from a 3600-km^2 area (and from other areas of isolated heavy contamination) after the 1986 Chernobyl accident [1, 32].

Conditions in the Extreme Evacuation Zone

The maximum areal ^{90}Sr concentrations in four villages (50-80 homesteads each [7]), from which 1154 inhabitants were evacuated in 7-10 d,

were reportedly 1000 Ci/km^2 (total activity about 40,000 Ci/km^2) (G. N. Romanov, pers. comm., October 19, 1990). References [1, 6, 14] place these four communities within a 20-km^2 area in which the ^{90}Sr concentrations along the main axis of the deposition zone were $\geq$1000 Ci/km^2 (up to 4000 Ci/km^2 according to [11]) and the mean level was 500 Ci/km^2. Because only three villages were actually inside the "extreme evacuation zone" (Fig. 2), the mean concentration of ^{90}Sr for these three sites was significantly >500 Ci/km^2, probably close to 1000 Ci/km^2--consistent with information from [7] and from radioecology studies [7, 35].

The reconstructed external exposure rate, 0.15 R/h at 1000 Ci/km^2 ^{90}Sr [11]--significantly lower than the values measured at the three villages the "extreme evacuation zone" in 1957, 0.61-1.44 R/h [7]--agrees with our independent calculations. If the instrumentation used to measure external exposure rates in 1957 was not shielded to exclude beta radiation, this could have resulted in significant errors, perhaps providing the explanation for differences between measured and calculated values.

In Berdenish, the community closest to the accident site, cattle reportedly accumulated body burdens on the order of 1-5 Ci (37-190 GBq) on the first day. By day 11, cattle and sheep had concentrations of 1-4 mCi/kg (0.037-0.15 GBq/kg) in hides or wool. Concentrations in forage grasses from all three villages averaged about 9-10 mCi/kg at 9-12 d after the accident. The cumulative doses through day 12 from gamma radiation were 135-290 rad (1.35-2.9 Gy), but these values appear to be too high by factors of 4-10. The maximum estimated doses from beta radiation to the small intestine were 150-760 rad (1.5-7.6 Gy) by the 12th day, but are also in question (G. N. Romanov, pers. comm., October 19, 1990). Data for other livestock (goats, poultry), also provided in [7], are similarly questionable. Loss of farm animals, exhibiting symptoms of acute radiation sickness, reportedly began within 9-12 d at all three sites [7].

In a community located farther from the accident site (Russkaya Karabolka; populated point No. 4) and contaminated at a level of 4500 Ci/km^2 (total activity; 0.76 mCi/kg in forage grasses), no mortality was experienced in farm animals over a period of 6 mo. After removal from the contaminated area, no differences from control animals were observed [7].

Despite concerns about the dosimetry and radionuclide concentration measurements in [7], one point is clear: Some farm animals within the "extreme evacuation zone" received effective whole-body or intestinal radiation doses on the order of 1000 rad (10 Gy) or more in 9-12 d after

the 1957 accident, as demonstrated by the the onset of the acute gastrointestinal syndrome [32]. Thus, doses from beta radiation to intestines or other organs or tissues reported in [7] are significant underestimates. Unfortunately, we currently have no information on Soviet dosimetric methodology in any of the reports released thus far.

Radiation Effects on the Human Population

The 1154 persons evacuated after the 1957 accident after 7-10 d reportedly received average effective-radiation-dose equivalents (ERDEs) of 52 rem (0.52 Sv; 150 rem (1.5 Sv) to the digestive tract) by the time they left the area. About one-third of this total, 17 rem (0.17 Sv), was derived from external exposure. It should be clearly recognized that these figures contain significant uncertainties and in fact are based on dose reconstructions, not on original measurements--for obvious reasons.

Further, 554 of the 1154 individuals inhabited a village contaminated to a level 2-3 orders of magnitude lower and should have received relatively low doses (est. <1 rem). The reported average values of 52 and 17 rem, respectively, thus do not necessarily provide meaningful estimates of doses for the inhabitants of the three remaining villages contaminated at about 1000 Ci/km^2 ^{90}Sr (40,000 Ci/km^2 total activity). Recall that some farm animals in these three villages had begun to die from acute radiation sickness at about the time the human population was evacuated.

Two hundred eighty of the 2280 (or possibly 2500 [18]) individuals evacuated in the second wave (around 250 d) from the area north of Lake Uruskul' had received average ERDEs of 44 rem (14 rem from external exposure) by the time they were moved, while the remainder received ERDEs of 12 rem (3.9 rem from external exposure). These dose estimates also may not be representative for the reasons given above; doses to the inhabitants of villages evacuated from areas contaminated at <4 Ci/km^2 ^{90}Sr have also been included in these averages. The remaining population was relocated at intervals of 330 d (4,200 persons, but see [18]) and 670 d (3,100 persons), respectively. Individuals from these latter groups had accumulated ERDEs $\leq$5.6 rem by the time they left [1, 6, 8, 14].

Three reports stated that all of these dose estimates should be doubled because of uncertainties created by non-uniform ground deposition (see [7]) and/or exposure conditions [5, 6, 8]. What were the maximum and mean doses to the 600 persons who lived in the "extreme evacuation zone" if the average ERDEs for 1154 persons evacuated at 7-10 d were actually 100 rem (1

Sv) based on doses ≤1 rem to 554 persons? Could some doses to individuals have exceeded 400 rem (4 Sv)?

The highest doses (about 100 rem or 1 Sv [7]) were supposedly delivered to soldiers who were on guard in the area after the accident, but there are significant uncertainties in these dose estimates. Medical examinations of 153 soldiers in the first month after the accident reportedly revealed no signs of acute radiation sickness; details are provided in [7].

Studies of both evacuated and resident exposed populations conducted over the past 30 years have revealed no significant radiation effects (1, 6-8, 14). However, these results should perhaps be regarded as tentative, subject to further review, because major questions exist about dosimetry (see above), methodology, and--in some cases--the suitability of control populations (see, e.g., [6, 8]).

For example, a relatively small fraction of the population evacuated during the first year was subjected to medical examinations (none before 9 months) and only qualitative conclusions about negative findings related to radiation sickness are available [7]. Reportedly, there were no skin burns [1, 5-8, 14], contrary to other reports [22, 25]. However, certain acute radiation effects would not necessarily have been evident at the time of evacuation (latent period for radiation burns) or after 9 months (healing, recovery of blood parameters; see e.g. [36]). Lymphopenia might have persisted, but this index was not measured. Although there are reasons why acute effects might not have occurred (e.g., shelter and confinement, restricted contact with contaminated surfaces, hygienic measures), we do not have enough information at this point to make such a determination.

Long-term follow-up studies likewise appear to have included only 10-25% of the 10,730 persons evacuated following the accident [1, 6-8, 14], although 1054 persons from the group evacuated at 7-10 d were included in a 25-y leukemia study [18]. Although the leukemia incidence was elevated relative to controls--in fact comparable to that in the population most highly exposed to releases via the Techa River--the result was not statistically significant owing to the small population size. A large fraction of the sampled population received very low doses (est. <1 rem (<0.01 Sv); see earlier discussion), possibly compromising the outcome.

Beyond the perimeter of the area that was evacuated (700 km^2; ≥2-4 Ci/km^2), effects appear to have been relatively slight. For example, there was a 4-y restriction on agriculture over a 300-km^2 area (contaminated at 2-4 Ci/km^2), primarily in Sverdlovsk Province [1, 5].

Persons living in the area with measurable contamination, but outside the most highly contaminated zone (105-km long, 1000-km^2 area, $\geq$2 Ci/km^2 ^{90}Sr; Fig. 3), for 30 y following the accident reportedly received ERDEs on the order of 1-10% above the dose from natural background radiation (1, 6, 37), well within the variability of the background dose (37).

Ecological Effects

The radioactive release from the 1957 accident was the first from a nuclear installation to produce major changes in the exposed ecosystem. Damage to pine trees was observed over the initial 30 km [7] of the deposition zone at ^{90}Sr concentrations >40 Ci/km^2 [13]. In places where ^{90}Sr levels were >40 Ci/km^2 and doses to tree crowns were >0.5 krad (5 Gy) during the first year of exposure, radiation damage to pines was expressed to varying degrees over the two-year period following the explosion. These effects included incomplete loss of needles, developmental defects, lowered growth, a variety of morphological and physiological aberrations, decreased viability of pollen and seeds, and phenological shifts (delays in opening of buds and flowers, etc.) [7, 13].

According to [13], all pines perished by autumn 1959 in areas with a level of contamination $\geq$180 Ci/km^2 ^{90}Sr, in which needles received doses of $\geq$3-4 krad ($\geq$30-40 Gy; affected area about 20 km^2). However, according to [7], pines were killed in areas contaminated with $\geq$1750 Ci/km^2 ^{90}Sr (3500-4000 Ci/km^2 ^{90}Sr-^{90}Y) in which they received doses $\geq$5 krad ($\geq$50 Gy) during fall-winter 1957-1958. This appears to be yet another example of the errors in [7] (G. N. Romanov, pers. comm., October 19, 1990). Thus, the data in [13] appear to be the best summary of ecological effects published prior to the Luxembourg seminar.

Birches were killed completely on plots with a contamination level of about 4000 Ci/km^2 ^{90}Sr (covering about 5 km^2), where meristem buds received doses above 20 krad (200 Gy) after the accident. At lower doses, parts of the crowns were withered, underdeveloped leaves appeared, and phenological shifts were observed over a period of 4 years. Radiation damage was observed to birches over an area of 17 km^2 [13].

The herbaceous vegetation was killed in its entirety at concentrations comparable to those that were required to kill birches, but the lethal doses accumulated at or near the soil surface at such concentrations were much greater (150-200 krad (1500-2000 Gy); area about 5 km^2). Partial destruction of the herbaceous cover occurred at ^{90}Sr concentrations $\geq$1500

Ci/km^2 (area about 15 km^2; absorbed doses to renewal buds of perennials located near the soil surface ≥20 krad (≥200 Gy)). At ^{90}Sr levels below 1000 Ci/km^2, mature plants did not die, but seeds experienced losses in germination and morphological changes were observed over a period of 2-3 years. Secondary ecological effects also occurred [13].

Mixed pine-birch stands in which pines received absorbed doses to the crowns in excess of 4 krad (40 Gy) were transformed to pure birch stands [7, 13]. In these altered stands, the herbaceous cover expanded greatly and "Il'enko [39] established that deposition of ^{90}Sr under the forest canopy in concentrations of [600 to 3400 Ci/km^2 ^{90}Sr-^{90}Y] did not produce noticeable effects on the life spans of small rodents" [38].

These effects on life spans of small rodents clearly do not apply to the fall-winter period following the accident, during which all birds and mammals could have received lethal doses from continuous residence in areas with ^{90}Sr levels ≥1000 Ci/km^2. Significant early effects on populations of certain invertebrates inhabiting the upper soil or forest litter layers (e.g., earthworms, myriapods, and mites) were observed in areas contaminated above 100 Ci/km^2. These effects persisted for many years in areas with concentrations ≥1000 Ci/km^2 where areas were large enough (several km^2 or more) that recolonization from less-contaminated areas could not overcome the effects of radiation exposure [13].

The results reported by Il'enko illustrate that most effects were temporary because of the potential for migration of animals from areas of lower or no contamination to replenish those lost during the "acute" exposure phase following the accident. This was especially true for larger mammals (roe deer, elk, wolves, lynx, hares). In the case of the latter, however, other factors included the elimination of hunting in a part of the contaminated area, but, more importantly, the increase in available habitat associated with removal of land from agriculture and other disturbances (habitation, fishing, tree cutting, foraging for wild foods) [7, 13].

Some sublethal effects were noted (e.g., increased radioresistance in small rodents from areas of ^{90}Sr concentrations ≥100 Ci/km^2 [40], increased frequency of mutations in plants living in areas with ^{90}Sr levels 1-10 Ci/km^2 and in field mice from areas with ^{90}Sr levels 10-100 Ci/km^2 [13]), but these effects would not have been expected to play a major role in determining the survival and fitness of exposed populations.

At least 13 of the 30 lakes within the deposition zone from the 1957 accident were studied, although one (L. Uruskul': Lake B in [7]; Lake

Number 2 in [34]) seems to have been a focal point [19]. Three of four communities (points 1, 2, and 3 in [7]) evacuated within 7-10 d after the 1957 accident were located on the shores of the two lakes (Berdenish and Uruskul'; Fig. 2) that received the highest levels of aerosol deposition (about 800 Ci/km^2 ^{90}Sr) [7, 35]. The remaining lakes within the deposition zone were contaminated at levels ≥100 times lower [7]. The principal focus of these radioecological studies was environmental transport, partitioning, and food-chain behavior of radionuclides (particularly ^{90}Sr; see references in [19]). Major findings on the environmental behavior of radionuclides in aquatic and terrestrial systems contaminated by the 1957 accident are summarized in [2, 7, 12].

Fish in the most heavily contaminated lakes received doses of 4 krad (40 Gy) from exposure to radioactivity in the water column during the fall-winter period following the accident. However, peak radionuclide concentrations (and dose rates) were rapidly reduced by incorporation of radioactive materials into the bottom sediments. The most vulnerable species were herbivorous fishes such as carp and goldfish, which spend much of their existence near the sediment-water interface. Doses to eggs of these species exceeded lethal levels (≥1 krad (≥10 Gy)) during the winter period of 1957-58 and within 2-3 y resulted in diminished reproduction. However, by 1960, no ecological effects on carp or goldfish were observable. One study conducted about 15 y after the accident detected no effects on growth rate of pike (Esox lucius) in one of the lakes at a dose rate of 0.45 rad/d (4.5 mGy/d) [41]. Effects on plankton, invertebrates, or aquatic plants were reportedly not detected [13].

Remedial Measures

Protective measures implemented on the contaminated area may be divided into three basic groups:

(1) extreme protective measures (evacuation of the population from the most highly contaminated part of a "sanitary-protective zone");

(2) measures to ensure the long-term safety of the resident population of the contaminated area (evacuations from the remainder of and restrictions on access to the sanitary-protective zone, extensive monitoring system for food products, controls on consumption of foods produced in areas of contamination); and

(3) measures to ensure the safety of agricultural products from the contaminated area (controlled agricultural production through special

state-run farm system; limits on production of foods and forest products as a function of type, areal contamination level, and time; controls on exports to other areas) [7, 15, 16].

Initially, restrictions on use of contaminated water bodies (harvesting fish, use of aquatic plants as feed for domestic livestock) were imposed in areas with ^{90}Sr levels ≥ 2 Ci/km^2, but, by 1970, the incorporation of ^{90}Sr by bottom sediments was sufficient to permit normal fish-culture practices in lakes on the periphery of this area [15].

Decontamination of agricultural and industrial areas, as well as populated points, was largely accomplished between 1-1.5 y after the accident by a mechanized detachment created for this purpose [14, 16]. Plowing, using special soil-working equipment and plows, was practiced in agricultural areas (20,000 ha in 1958-1959; deep plowing to >50 cm over 6200 ha in 1960-1961 [1, 8]) to reduce the level of contamination in the surface layer (root zone). Removal and disposal of the contaminated surface layer was also carried out, principally in industrial areas and populated locations, utilizing special soil removal mechanisms and vehicles (e.g., road-scrapers [42]). Chemical treatments of the soil were explored, in combination with tillage practices, but relatively little was known about their efficacy and usage [42, 43] prior to the Luxembourg seminar. Information on in situ remedial measures, particularly for environmental decontamination or contamination control, is still incomplete.

At the Chelyabinsk-40 complex itself, the principal methods employed in terrestrial areas included: (1) covering contaminated soil with clean soil and (2) removal and burial of the contaminated layer of soil. Altogether 3.5×10^5 m^3 of contaminated soil were removed and 4.0×10^5 m^3 of clean soil used to cover contaminated areas. Decontamination solutions were used for washing asphalt roads and some areas were decontaminated using road-building equipment (e.g., scrapers) with special shielding. However, significant areas of contamination still remain [17].

The most highly contaminated (170-km^2) portion of the 1957 deposition zone is still unsuitable for human habitation, agriculture, or forestry. It is now a radioecological reserve (with accompanying research station), set aside for long-term studies of the cycling and effects of radionuclides (1, 44). A major work entitled "Guide to the Planning and Implementation of Measures Designed to Reduce the Negative Radiological and Radioecological Consequences of Accidents Going Beyond the Design Basis Accident and Involving Releases of Radioactivity to the Environment" is

reportedly nearing completion by the staff of the environmental research station [44; G. N. Romanov, pers. comm., October 19, 1990].

The most massive remedial action effort in evidence at the Ch-40 site is a major hydrologic diversion system consisting of bypass canals and a cascade of 4 reservoirs (total surface area 65 km^2 [17]) (Fig. 2). The first reservoir was completed in 1951 and the fourth in 1964, well after the 1957 accident and the initiation of waste disposal into L. Karachay.

The bypass canal through L. Berdenish (Fig. 2) does not appear on the Soviet map of the hydrologic isolation complex provided in [17] and the dams on the first two reservoirs downstream from L. Kyzyltash (Fig. 2) are not shown on our 1973 map of this area. This suggests that significant changes were made in the design and operation of this system over time, even though hydrologic isolation of early waste discharges to the Techa River was the principal driving force. Further, there is a hydrologic connection between L. Karachay and the third reservoir in the cascade through another small water body (see [19, 45] for potential background on the latter) (Fig. 2). Given the existence of this hydrologic connection, the potential for additional liquid releases from L. Karachay and/or this smaller water body during 1953-1964 seems plausible on the basis of current information. Thus, it appears that the hydrologic isolation system may have been designed or modified to serve multiple purposes--including remediation of contamination from the 1957 accident. More information on its purposes and efficacy could be very useful.

Observations of artificially elevated calcium concentrations (110-226 mg/L [46, 47]) in two of the reservoirs from radioecology studies [19] raise the question whether these are related to contamination control and/or remediation [43]. In situ decontamination of the isolated systems (i.e., remobilization of sorbed ^{90}Sr by increasing the concentration of its chemical analogue calcium in the water column) is one possibility, seemingly supported by the very low sediment-water concentration ratios for ^{90}Sr in the third and fourth reservoirs in the cascade (17). Further, the reservoir cascade only accounts for about 50% of the ^{90}Sr and ^{137}Cs reportedly intercepted through 1952 (about 0.75 MCi [17, 18]; 38-y decay), and there may have been later additions, as described above. More information on Soviet experience in this area could be extremely valuable.

CONCLUSIONS AND RECOMMENDATIONS

A complete assessment of the environmental impact of the radionuclides

released by the 1957 accident at Ch-40 must await (1) elimination of seeming inconsistencies in information, (2) more detailed information in several key areas, and (3) better understanding of the nature of and interactions among all significant radioactive releases from Ch-40. Thus, we need answers to the following questions:

(1) How and when will we get an authoritative, internally consistent set of complete information on the 1957 accident, sufficient to address the specific concerns and questions raised by our review? Why was material from reference [7] so widely circulated, given its obvious errors and inconsistencies? When will we have a map of the contamination zone showing appropriate details on radioactivity and population centers, including evacuation dates, radioecological, and topographic features?

(2) If there were no casualties among evacuees after the 1957 accident, why are there so many reports of accident-related casualties, particularly burns, associated with Ch-40? Were there serious radiation effects on workers who rehabilitated the HLW explosion site? Were there other causes, including reactor accidents--as yet unreported? Was medical treatment for Ch-40 radiation workers significant enough to have been misinterpreted as the result of an accident(s)?

(3) What was the history of all significant radioactive waste operations and accidents at Ch-40 leading to environmental contamination? To what extent did accidental releases contribute to the need for the massive hydrologic isolation system? Is this system being decontaminated?

(4) What is the status of the comprehensive Soviet report on reducing the consequences of radiation accidents? Will this report also cover releases to the Techa River and from the Karachay reservoir?

We expect that the continuing declassification of Soviet information could in time result in important new revelations about releases of radioactivity from Ch-40 and their consequences. The task at hand is to obtain timely answers to the questions we have posed. We do not think that this task can be accomplished without access to Ch-40 for scientists both knowledgable and free enough to explore its many aspects in order to lay to rest major concerns about the events of 30-odd years ago, as well as their consequences. As scientists involved in evaluating hazards associated with radioactive releases to the biosphere, we continue to urge that all pertinent information on Ch-40 be shared with others concerned with achieving such objectives.

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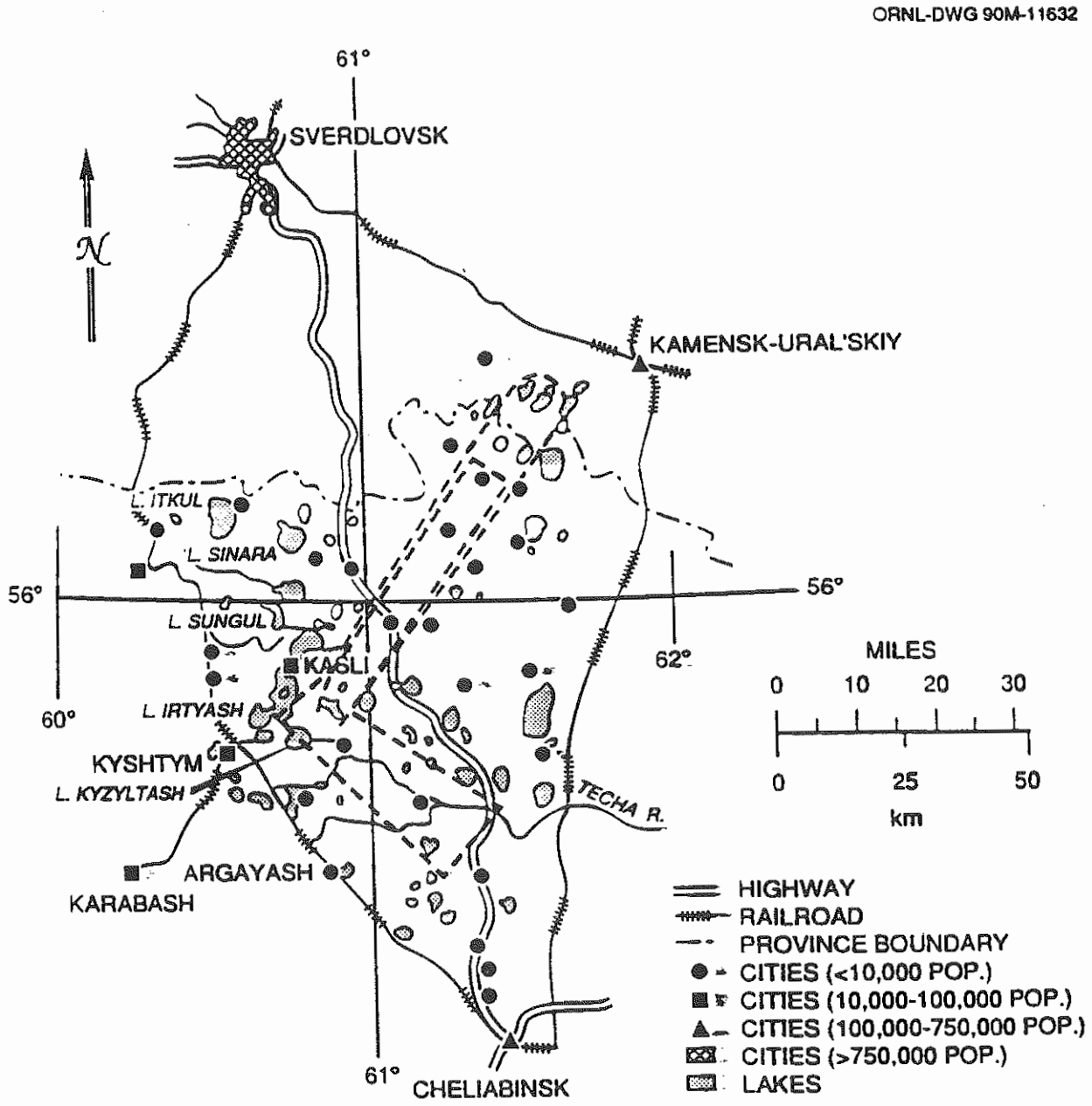

Figure 1. Map based on pre-accident geographic features (redrawn from [18]) of geographic region in which a Soviet catastrophe involving high-level nuclear wastes occurred in 1957. Area fully enclosed by heavy dashed line indicates zones which exhibited extensive changes (in population centers and surface hydrologic features) on later maps. Finer dashed lines represent the reported extent of the "sanitary-protective zone" (700-km^2 area; $\geq$4 Ci/km^2 (0.15 TBq/km^2) ^{90}Sr) which was established by Soviet authorities after this accident and from which the population was relocated (taken from Fig. 23 in [7]).

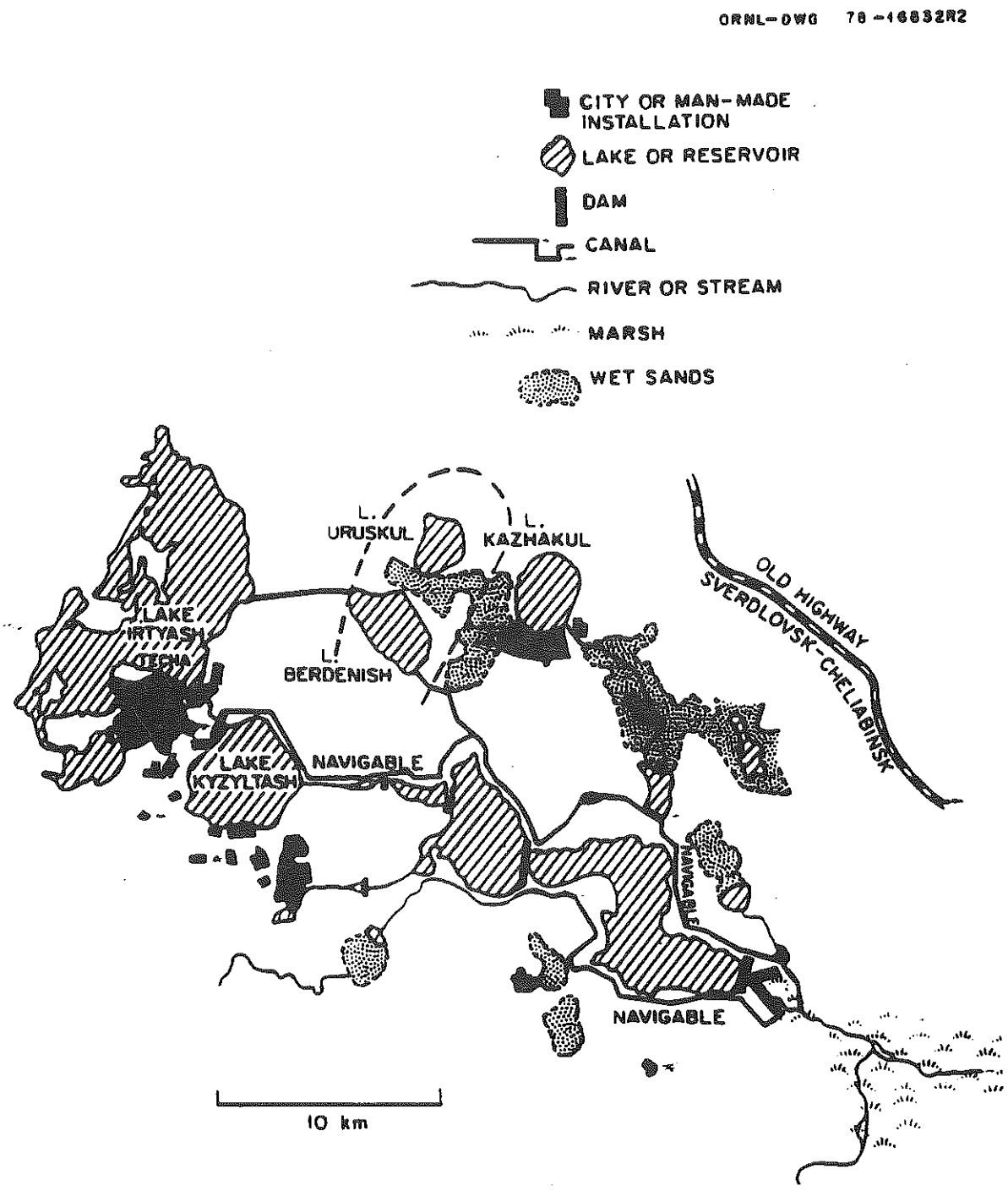

Figure 2. Reservoir-canal system constructed to reduce the hydrologic transport of radioactive materials down the Techa River system. Heavy dashed contour line represents the reported extent of the "extreme evacuation zone" ($\geq$1,000 Ci/km^2 (37 TBq/km^2) ^{90}Sr) within the "sanitary-protective zone" which was established by Soviet authorities after the 1957 nuclear waste explosion, and from which the civilian population was evacuated within 7-10 d (from Fig. 23 in [7]).

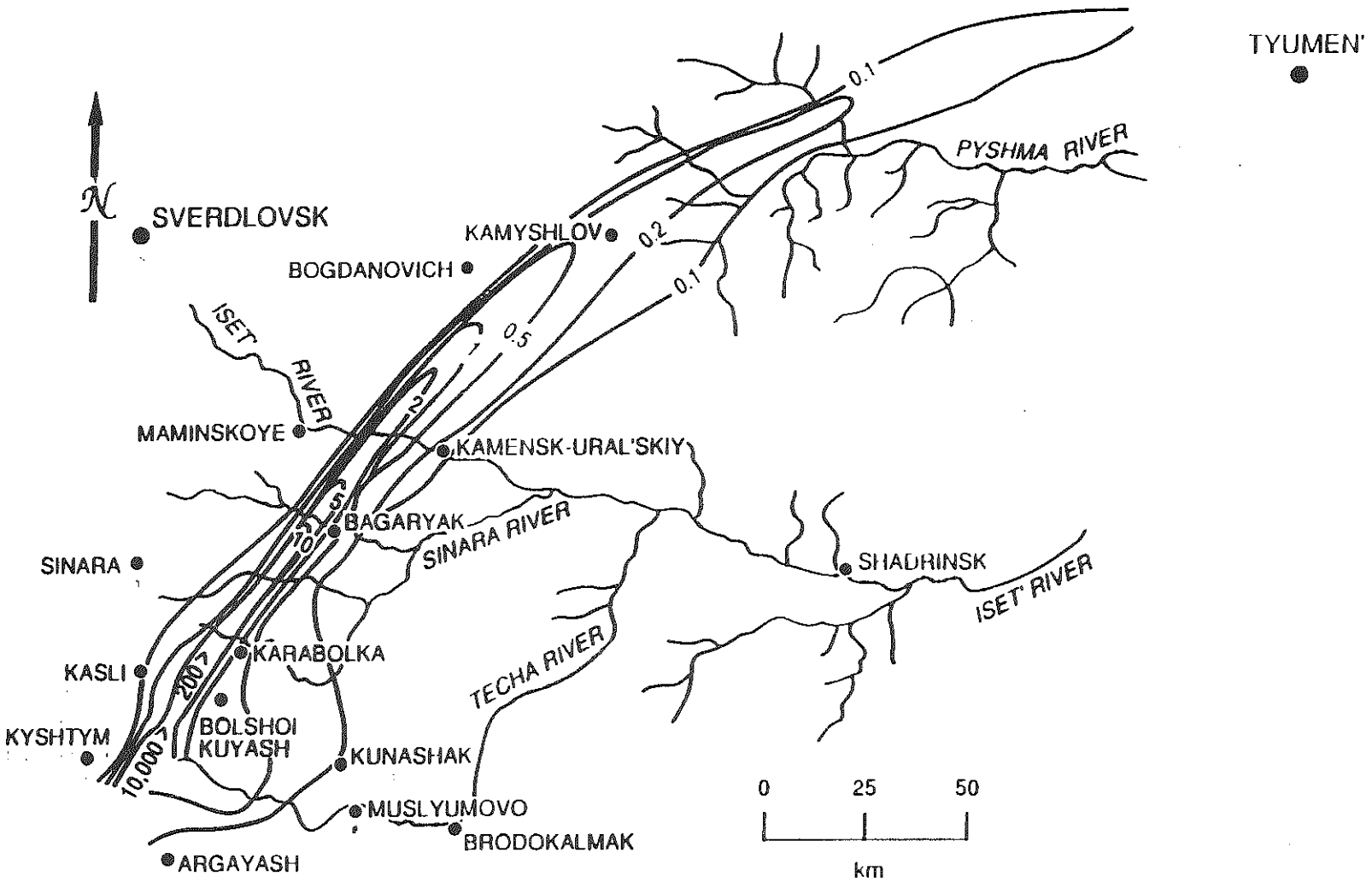

Figure 3. Isopleths of ^{90}Sr concentration {Ci/km^2 (0.037 TBq/km^2)} resulting from high-level nuclear waste explosion in 1957 (15,000-23,000 km^2 at $\geq$0.1 Ci/km^2) and another release in 1967 that contaminated an additional 1800 km^2 (note large bulge at the southern end of the contamination zone).

Table 1. Characteristics of the 1957 high-level waste explosion and the materials released[a]

Source	Release mechanism	Energy of explosion	Time of occurrence
250 m^3 of high-level liquid wastes in 300-m^3 stainless-steel tank[b]	Chemical explosion following failure of tank cooling system: Ignition of acetate-nitrate concentrate	Equal to about 75 t of TNT; soluble nitrates: 70-80 t	16:20 local time, 29 September 1957[b]

Radioactivity released[c]
2×10^7 Ci (740 PBq) of mixed fission products; 10% was more widely dispersed in aerosol form

	Contribution to total activity of the release (%)	
Radionuclide	References [1, 5, 8, 10]	Table 18 in [7]
^{89}Sr	Traces	2.6
$^{90}Sr + ^{90}Y$	5.4	7.0
$^{95}Zr + ^{95}Nb$	24.9	22
$^{106}Ru + ^{106}Rh$	3.7	3.5
^{137}Cs	0.036	1
$^{144}Ce + ^{144}Pr$	66	64
Remainder	Traces	<12
^{90}Sr-^{90}Y:^{137}Cs Activity ratio	150:1	7:1

[a]Source: Reference [7], except where noted.

[b]Source: Reference [10], but also see references [2, 8].

[c]Sources: References [2, 3, 7, 8, 10]).

Table 2. Characteristics of the atmospheric release and its dispersal[a]

Atmospheric conditions	Winds		
	Direction	Altitude (m)	Velocity (m/s)
Relatively stable; no precipitation	Southwesterly, changing to easterly at higher levels	10 to 12	5
		500	10

Plume Dispersal Characteristics	Initial			
	Height	Diameter	Velocity	Direction
Effective conditions	≤1 km[b]	2 km?	35 km/h	North-northeast

	^{90}Sr concentration[c] (Ci/km^2 (0.037 TBq/km^2))				
Ground deposition-population patterns[d]	>1000	>100	10-100	1-10	0.1-1.0
Area (km^2)	20	200	200	1000	21,600
Number of population centers	3	3	10	58	146
1957 population	1100	1500	3500	12,000	253,000

[a]Source: Reference [7], except where noted.

[b]Based on recalculations [2, 3, 8, 9]; given as 1.9 km in [7].

[c]Apparently ^{90}Sr + ^{90}Y.

[d]Maxima along north-northeast axis, but with assymetric distribution perpendicular to axis approximating a skewed Gaussian curve (area of contaminated zone southeast of axis 2 to 3 times that northwest of axis).

Table 3. Reported areas and populations of the contaminated region

^{90}Sr concentration[a] (Ci/km^2 (0.037 TBq/km^2))	Area (km^2)	1957 population	References
International Atomic Energy Agency reports and others			
≥0.1	15,000	270,000	[1, 5, 6, 8]
≥2	1000	10,180-10,854 (Evacuated zone-23 communities)	[1, 5, 6, 8, 18]
≥100	120	280-2100	[1, 5, 6, 8]
≥1000	20	600-1154	[1, 5, 6, 8]
Declassified 1974 Soviet report by Burnazyan et al.			[7]
≥0.1	23,000	270,000	
≥1	1400	17,000	
≥4	700	10,000 (Evacuated zone-19 communities)	
≥10	400	5000	
≥100	200	1500	
≥1000	20	1100	
≥10,000	0.5-2	?	
May 1990 articles in Soviet journal Priroda			
0.1-2	15,000-23,000	---	[11]
≥4	700	10,730 (Evacuated zone-23 communities)	[14]
2-20	600	---	[11]
20-100	280	---	[11]
100-1000	100	---	[11]
1000-4000	17	---	[11]

[a]Concentrations reported in [7] appear to be $^{90}Sr + {}^{90}Y$.

The Chernobyl Source Term

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ABSTRACT

The report contains the results of recent estimates of the quantity and composition of the radionuclides contained in Chernobyl Reactor No 4 prior to the accident. Different approaches to determining the release of radioactivity from the reactor are discussed, with particular reference to the release of fine fuel particles and of caesium-137. Data on the release of radioactivity after completion of the containment structure ("sarcophagus") are included.

The report also examines typical radioactive release effects and discusses the effectiveness of methods for investigating contaminated areas.

1. Data on the amount and composition of the radioactivity released during the Chernobyl accident were summarized for the first time in the report presented to the IAEA post-accident review meeting in August 1986. [1] Discussion and refinement of the data have continued since that time (see [2], [3], [4] etc.).

   This paper also sets out to discuss this question and to make a more precise estimate of the amount of radioactive products released. For this the authors used the following new information:

   - improved calculations of the radionuclide content of the fuel in Reactor No 4 prior to the accident (1989-1990) [5];
   - analyses of soil contamination (1986);
   - data on the condition of the nuclear fuel left in the destroyed reactor (1988-1990) [6], [7];
   - data on the escape of radioactivity from the containment structure ("sarcophagus") around Reactor No 4 (1987-1990) [8].

2. The No 4 reactor at Chernobyl began operating in December 1983 and by 26 April 1986 had operated for 865 calendar days (715 effective days). The fuel - enriched uranium dioxide - was contained in 1 659 fuel elements, giving a total charge of 190.2 tonnes of uranium. Average core burn-up was 11 MWd/kg U.

   Calculation of the radionuclide inventory in the core was done on an element-by-element basis. Firstly the relationship of a specific quantity of particular isotopes to burn-up was calculated for a single fuel element. Then, on the basis of burn-up data, the quantity of isotopes produced in each fuel element was calculated.

   Table 1 contains data on the core's inventory of the most important, long-lived and biologically significant radionuclides.

Table 1

Radionuclide	Half-life	Mass (kg)	Activity	
			Bq	MCi
Strontium-90	29.12 years	4.3 (+1)	2.2 (1)	5.9
Ruthenium-106	368.2 days	6.9	8.6 (17)	2.3 (+1)
Antimony-125	2.77 years	5.1 (-1)	1.9 (16)	5.2 (-1)
Caesium-134	2.07 years	3.2	1.5 (17)	4.1
Caesium-137	30.17 years	8.1 (+1)	2.6 (17)	7.0
Cerium-144	284.3 days	3.3 (+1)	3.9 (18)	1.1 (2)
Plutonium-238	86.4 years	1.5	9.4 (14)	2.5 (-2)
Plutonium-239	24 110 years	4.12 (+2)	9.5 (15)	2.6 (-2)
Plutonium-240	6 553 years	1.76 (+2)	1.5 (15)	4.0 (-2)
Plutonium-241	14.7 years	4.9 (+1)	1.8 (17)	5.0
Plutonium-242	3.76 (5) years	1.4 (+1)	2.1 (12)	5.6 (-5)
Americium-241	433 years	1.1	1.4 (14)	3.7 (-3)
Americium-243	7.38 (3) years	7.3 (-1)	5.4 (12)	1.5 (-4)
Curium-242	162.8 days	2.6 (-1)	3.1 (16)	8.3 (-1)
Curium-244	18.11 years	6.0 (-2)	1.8 (14)	4.8 (-3)

Figures in brackets to be understood as follows: 3.76 (5) = 3.76×10^5 years

The alpha emitters in this table are the isotopes of plutonium (238, 239, 240), americium (241, 243) and curium (242, 244).

Immediately after the accident curium-242 was the determining factor for alpha activity in the nuclear fuel, whereas plutonium isotopes are now predominant. However, as a result of the beta decay of ^{241}Pu (half-life 14.7 years), ^{241}Am (half-life 433 years) is beginning to accumulate, and after ten years its activity will account for approximately 50% of total alpha activity (Fig. 1).

The beta and gamma emitters (apart from ^{241}Pu) were ^{90}Sr (beta-emitter only), ^{134}Cs, ^{137}Cs and ^{144}Ce. Caesium-137 and strontium have a half-life of approximately 30 years and their activity decreases by an order of magnitude every 100 years.

3. Three approaches were used for determining the amount and composition of radioactive releases. The first used data on release dynamics, gathered during the active phase of the accident (26 April - 6 June 1986) and afterwards. It has already been stated several times that, for many objective reasons, using this method to assess releases was very inaccurate. This is clear from the well-known diagram (Fig. 2) presenting the results contained in [1]. Attempts are currently being made to reconstruct and reanalyse the data, but the work is not yet complete. One of the most important conclusions from these measurements is that at all stages of the accident the release of radioactivity was in the form of finely dispersed fuel particles, with the exception of inert gases and a number of highly volatile substances (iodine, caesium, tellurium and ruthenium). Initial results from the analysis of air samples taken around the damaged reactor had already shown this.

4. It was precisely this conclusion which led to relatively rapid success with the second method - assessing releases by investigating soil contamination.

Wide-scale measurements of soil, water and air contamination began during the first days following the accident. It must be pointed out that the task of detecting and quantifying radionuclide contamination over an area of tens of thousands of square kilometres was extremely laborious. For example, quantifying the fallout of plutonium or strontium-90 isotopes was done by taking soil samples and conducting complicated and time-consuming radiochemical analyses.

The stable relationship between the quantity of radionuclides in the fuel released from the No 4 reactor and between alpha and gamma activity made for a simple solution to the problem of quantifying environmental contamination by various biologically significant radionuclides.

For example, plutonium alpha activity was determined through correlation with cerium gamma activity using the formula:

$A(Pu)=K_c A(^{144}Ce)$,

where A(Pu) = total activity of ^{238}Pu, ^{239}Pu and ^{240}Pu; K_c = correlation coefficient, and $A(^{144}Ce)$ = ^{144}Ce activity.

On 26 April 1986 the K_c value was approximately 9×10^{-4}.

Since the K_c value depended to a relatively small extent on the scale of fuel burn-up, and fuel element burn-up differed only slightly from the average, the correlation coefficient for all the reactor fuel may be considered constant (see Fig. 3).

The methods used to assess fuel contamination in the various areas were as follows:

- firstly, measurement of the gamma dose over the areas affected using aerial gamma surveying (a first, rough approximation using the relationship of the dose rate to the amount of fuel);

- secondly, analysis of soil samples using semiconductor gamma spectrometers (more precise measurements using the correlation coefficient for cerium-144);

- finally, painstaking radiochemical analyses (verification of the K_c coefficient for a given area).

The report in [1] gave information on the amount of radioactive substances released from Reactor No 4.

The basic conclusions of the report were as follows:

- almost all radioactive inert gases and a significant amount of iodine were released from the reactor;

- 13 (±7)% of the caesium was released, as was

- 3 (±1.5)% of the fuel containing fission products and transuranic elements.

The I. V. Kurchatov Institute of Atomic Energy compiled the preliminary estimates of fuel distribution given in Fig. 4.

Between August and November 1986 a series of thermal measurements was taken on the surface of (and around) the ruined reactor. Comparisons of the actual amount of heat escaping with estimates led to the conclusion that no less than 90% of the fuel was still inside the containment structure.

The amount of fuel released has been calculated more exactly over the past few years, and the debate on this issue can be considered closed. Results calculated on the basis of a data bank, containing information on tens of thousands of soil samples, now give the amount of fuel released as 3.5 (±0.5)%.

Again it must be stressed that what is being discussed here is not the fuel released from the active zone or the reactor cavity (and which is still located within the No 4 reactor unit), but only that proportion outside the containment structure. Incorrect interpretation of what was meant by "release" has already led on more than one occasion to unfounded rumours and statements.

6. A third method of assessing the releases is to study the amount and composition of the fuel remaining within the containment structure enclosing Reactor No 4. This became possible only after a great deal of work by the Kurchatov Institute's Complex Expedition, which enabled the main fuel masses to be assessed [9]. Most investigative work to date has been done on the fuel-containing masses situated beneath the reactor. Most of this is a kind of lava - consisting of melted and resolidified SiO_2 (60-80%), traces of Mg, Fe and Al, and up to 20% nuclear fuel (Fig. 5). The fuel released into the central reactor hall and buried under the cascade wall of the containment structure has received less attention (Fig. 6).

   As things stand at the moment the proportions of released and remaining fuel can be assessed to within a few percentage points only. It is possible, however, to make some assessments concerning the release of such a biologically significant radionuclide as caesium-137.

7. Estimates of caesium-137 release differ significantly. The report in [1], for example, gives a figure of 13 ±7%.

   The experts concluded [10] that total ^{137}Cs release amounted to 1.9 MCi, i.e. 27% of the total amount.

   An article in "Pravda", based on detailed maps of caesium-137 fallout, gives a figure of 15% [11], while [5], [4] and [12] give figures of between 1.5 and 2 MCi or 21-28%.

   Table 2 shows the caesium content in the fuel-containing masses on the lower levels of Reactor No 4, where some 75% of the fuel is located.

Table 2

Caesium content of fuel-containing masses inside the containment structure (see Fig. 7)

Area	Elevation (metres)	Amount of fuel (U, t)	$^{137}Cs/^{144}Ce$ activity ratio (extrapolated back to 26.04.86)
1	2	3	4
Pressure suppression pool			
1st floor	0	1.5 ±0.5	0.022
2nd floor	3	11 ±4	0.020
Steam distribution lines	6	23 ±7	0.020
Lower room No 305 2	9	77 ±25	0.022
Corridors, rooms, steam release valves	9	22 ±7	0.021
Total amount on lower levels		135 ±30	Weighted average 0.021

The calculated activity ratio of $^{137}Cs/^{144}Ce$ is 0.064, while the weighted average is 0.021, i.e. three times less. Caesium-137 release from the lower levels of the reactor should therefore have amounted to 3.3 ±0.7 MCi or 47 ±10% of the total amount contained in the reactor.

This raises the question of whether this quantity of radioactive caesium was ejected outside the area where the containment structure now stands and beyond the plant site, or whether a proportion of the caesium is still inside and around the building.

Samples taken from the contaminated upper levels of Reactor No 4 are rich in caesium-137. The degree of enrichment varies widely, but averages 1.5 times in relation to cerium-144. This figure is confirmed by measurements taken during helicopter flights over the containment structure using a semiconductor spectrometer.

Assuming that the radioactivity remaining in the upper levels is enriched with 1.5 times the amount of caesium, this means that the amount outside the containment structure is 2.3 ±0.7 MCi, or 33 ±10% of the original amount.

Further work centring on the containment structure will no doubt result in a more precise estimate.

With regard to caesium fallout on and around the plant, the answer lies in measuring soil samples taken at different distances from the reactor (data are for May 1986, as the area around the reactor was later covered with a layer of hardcore and concrete, and the area around the plant was also decontaminated). Samples were taken in three main directions (north, west and south), and the averaged data are given in Table 3.

Table 3

Ratio between caesium-137 and cerium-144 in soil samples (extrapolated to 26 April 1986; calculated ratio is 0.064)

Distance from reactor (km)	Caesium-137/cerium-144 ratio
1	2
0.05	0.068
0.6	0.078
3	0.068
30	0.19
60	7.5

Thus the ratio increases with distance, and at a distance of 3 km it corresponds to the calculated fuel ratio. It follows that there were no significant "caesium hot-spots" near Reactor No 4.

8. One of the main reasons for building the containment structure was to prevent radioactive contamination of the environment by any type of dust fallout.

Four years' experience has shown this approach to be generally successful.

However, the difficult conditions under which the containment structure was built made it impossible to seal the structure completely. In particular, there are many gaps in the upper part of the structure and in its roof. In addition, hatchways are built into the roof so that measuring apparatus can be inserted into the central reactor hall above the debris. Finally, there is natural ventilation through the stack shared by Reactors No 3 and 4.

All the above-mentioned openings are routes through which radioactive dust and gas can escape from the containment structure into the environment.

Such escapes were observed fairly frequently in 1987 and 1988, and were linked to work being carried out on the ruined reactor or to specific meteorological conditions. One typical example is the sharp increases (by hundreds of times) in the ruthenium-106 concentration in air (within the containment structure and on the plant site) recorded several times during the hot summer of 1987, although overall aerosol activity did not exceed the permissible concentration limits for personnel. These increases correlated with heavy showers and high air temperatures. The mechanism responsible for this effect is not known in detail, but the most likely explanation is that ruthenium-106 and 103 (possibly in the form RuO_4) adsorbed into the porous materials of the debris were released into the air through the evaporation of rainwater.

Contamination of the air masses from the containment structure now appears to be the result of a series of processes, as follows:

- dust rising from the surface of the debris in the central reactor hall, the reactor space and other areas within the containment structure;
- dust formed by drilling and construction activities;
- dust formation and release due to the collapse of structural elements within the containment structure.

A series of measures has been developed and partly implemented to reduce dust formation and release: strengthening of structural elements, dust suppression, wet drilling, etc.

One important task is to organise the careful monitoring of radioactive aerosol releases into the environment.

With the experimental data available to date, only estimates of these can be made. Plans are in hand, however, which will lead to a significant increase in accuracy and speed of response when monitoring radioactive aerosol escape.

The following experimental data can be used to assess the total amount of radioactivity released:

- measurements of radioactivity released through the stack shared by Reactors Nos 3 and 4;
- measurements of the concentration and radionuclide composition of aerosols contaminating the air on the site and within the containment structure;
- measurement of the radioactive aerosols escaping through openings in the containment structure.

A brief examination of each method follows.

Airflow through the stack is by natural ventilation, and varies between 10 and 40 000 m_3/h. Aerosol concentration is measured by filtration in a special sampling line.

After a specified exposure time, gamma spectrometry analysis is used to measure aerosol contamination in the filters.

Table 4 gives the results of the total activity measurements for four identified isotopes (^{137}Cs, ^{134}Cs, ^{144}Cs and ^{106}Ru) averaged over a month.

Table 4

Activity (10-3 Ci/day)

Month	1	2	3	4	5	6
Year						
1988	0.38	0.20	0.20	0.22	0.23	0.197
1989	0.26	0.22	0.21	0.14	0.14	0.26
1990	0.05	0.03	0.04	0.03		

Month	7	8	9	10	11	12
Year						
1988	0.22	0.10	0.053	0.93	0.34	0.69
1989	0.14	0.92	0.20	0.054	0.07	0.07

The total activity escaping from the containment structure through the ventilation stack amounted to 0.08 Ci in 1989.

The next source of information is measurement of the concentration and radionuclide composition of aerosols in the air on the site of Reactor No 4.

As an example, average concentrations of radionuclides in the air at the site in the first half of 1990 were as follows:

Ruthenium-106	-	1.4×10^{-16} Ci/l
Cerium-144	-	3.6×10^{-16} Ci/l
Caesium-137	-	3.2×10^{-16} Ci/l
Caesium-134	-	0.7×10^{-16} Ci/l

It is difficult to relate these data to the radioactivity escaping from the containment structure, since the contribution from dust rising from the ground surface has not been determined. It should be pointed out, however, that the amount escaping is not higher than the above and does not represent a danger to human beings near the containment structure.

Direct measurement of radioactive aerosols escaping from the containment structure roof hatches began at the end of 1989.

Two vertical and two horizontal screens were fitted over each of four hatchways in the roof of the containment structure, each measuring 60x60 cm.

The average activity accumulation rate on the horizontal screens (lower side) does not exceed 0.3 Ci/km_2 per day. Assuming that the radioaerosol escape rate from all gaps in the containment structure is the same, and taking into account the fact that the total surface area of all openings is currently estimated at approximately 1 200 m2, this gives a total of not more than 0.2 Ci/year for the release of radioactivity from the containment structure.

Summarizing the data shows that in static conditions the release of radioactivity from the containment structure is negligible, not exceeding 0.3 Ci per year (with plutonium accounting for 0.6-0.8% of total activity).

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CAPTIONS

Fig. 1: Time dependence of alpha-activity of fuel in Reactor No 4 following the accident.

Fig. 2: Release of radioactivity beyond the confines of reactor No 4 during the active phase of the accident (measurement error indicated by shaded area).

Fig. 3: Amounts of fuel rods with different Pu/^{144}Ce activity ratios, showing the stability of the correlation coefficient.

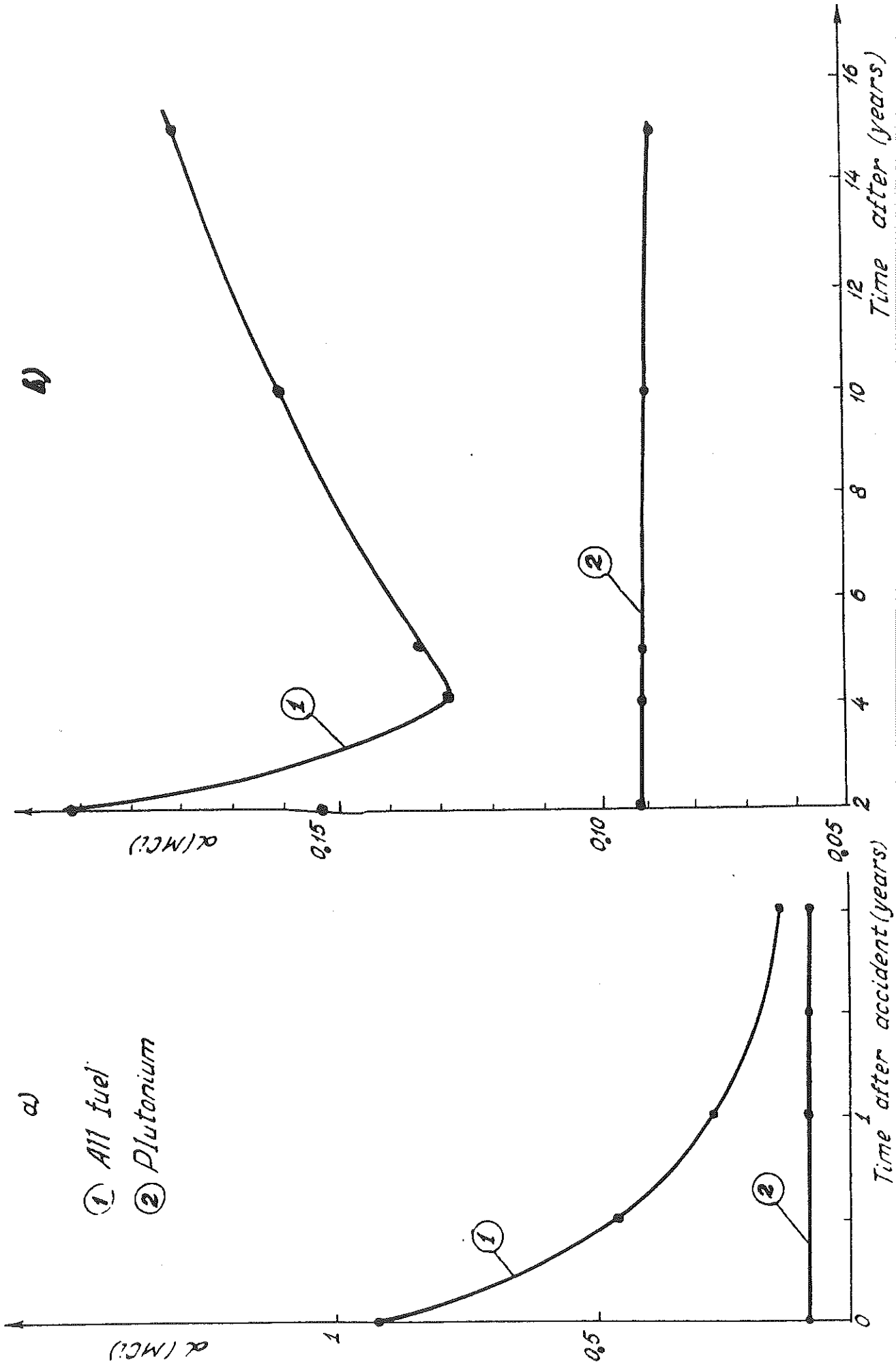
Fig. 4: Amount of fuel released (initial assessments) as % of the total load

> 96% - reactor building;
< 1.5% - adjacent 80 km zone;
< 1.5% - rest of USSR;
< 0.3% - Chernobyl NPP site;
<< 1% - outside USSR.

Fig. 5: Solidified lava flow from steam release valve.

Fig. 6: Central reactor hall.

Fig. 7: Distribution of fuel-containing masses in the lower levels of Reactor No 4.



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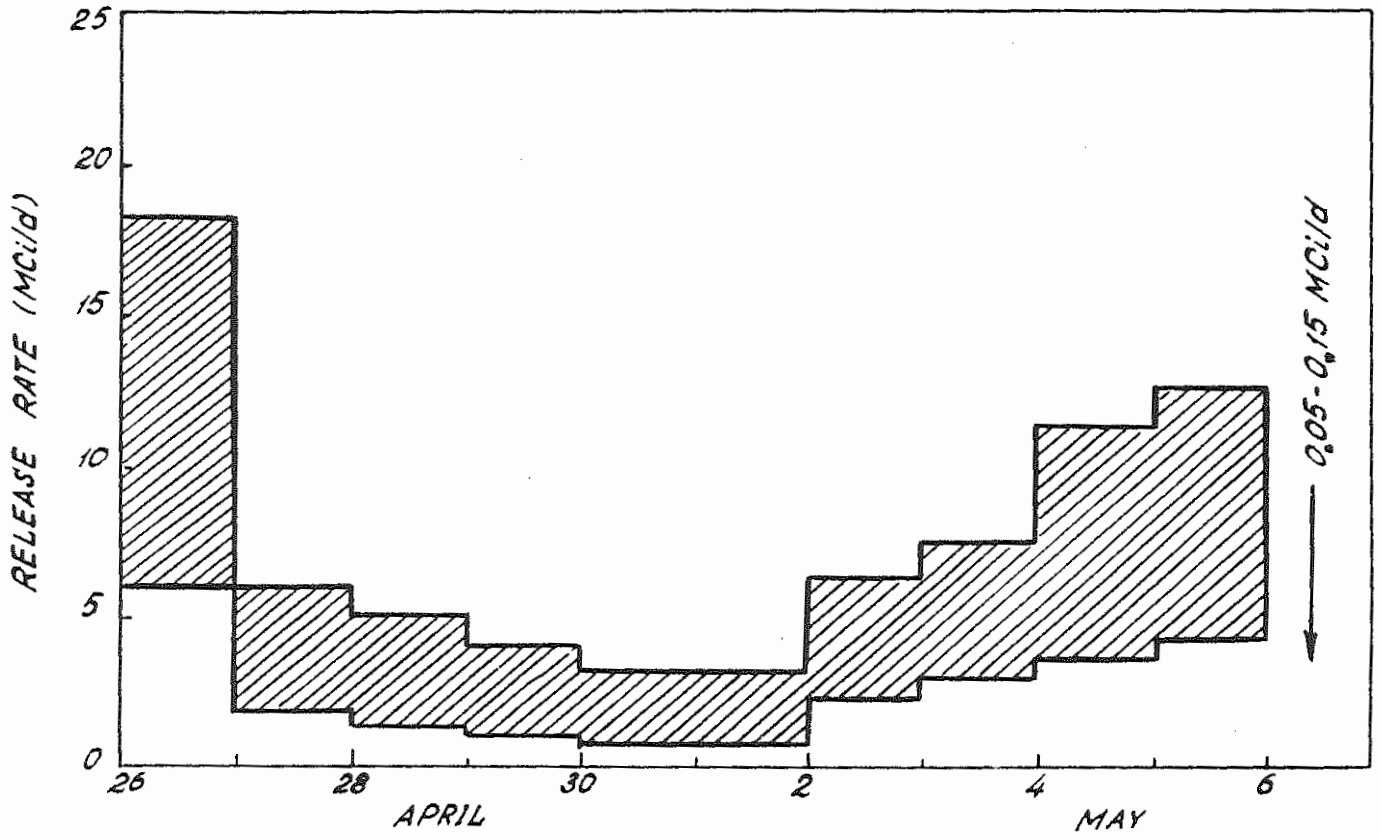

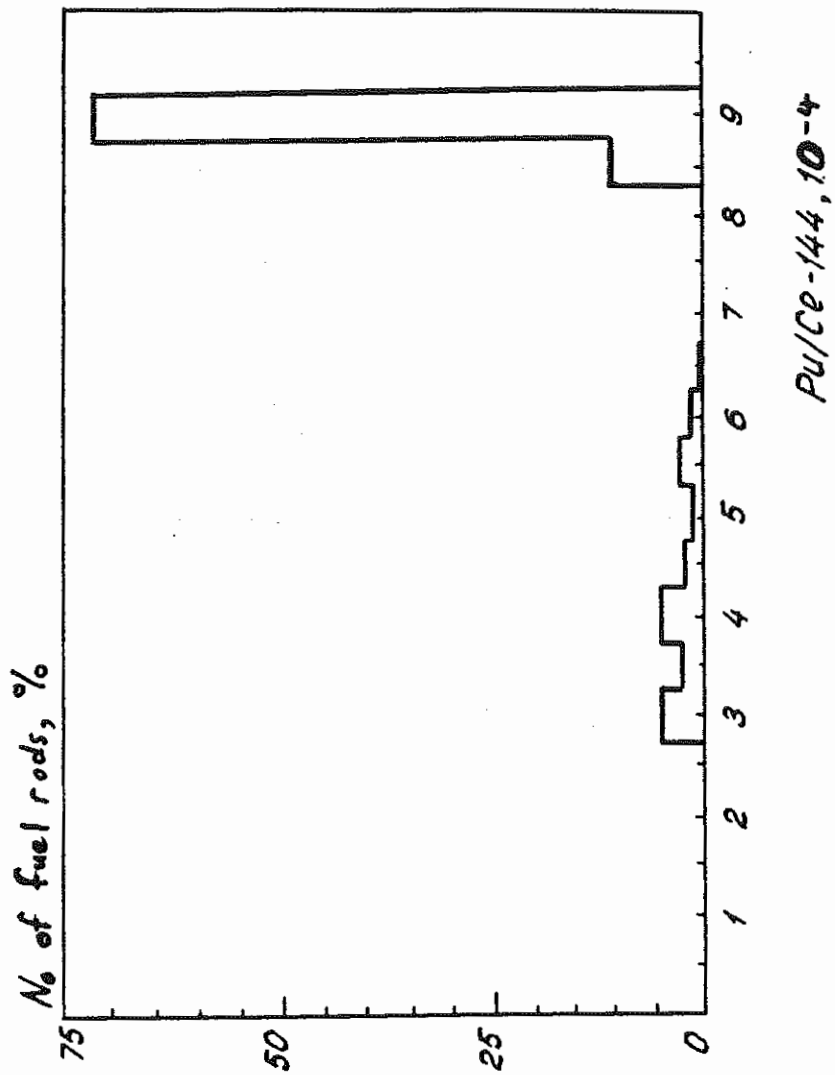

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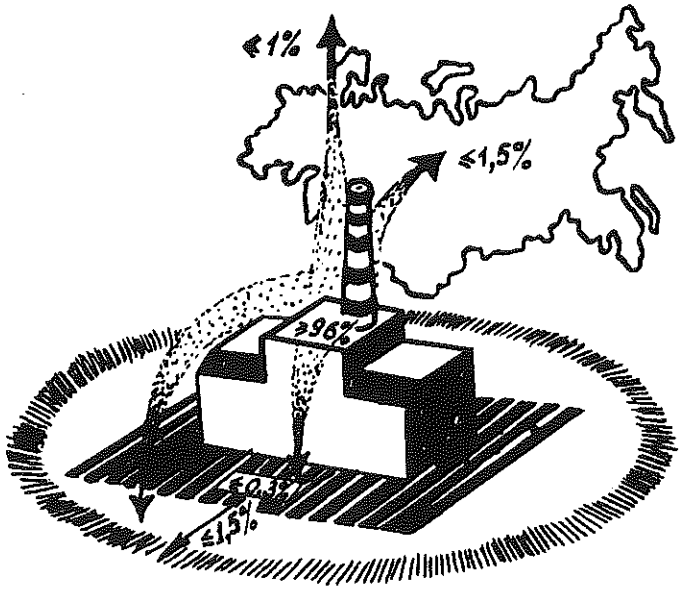

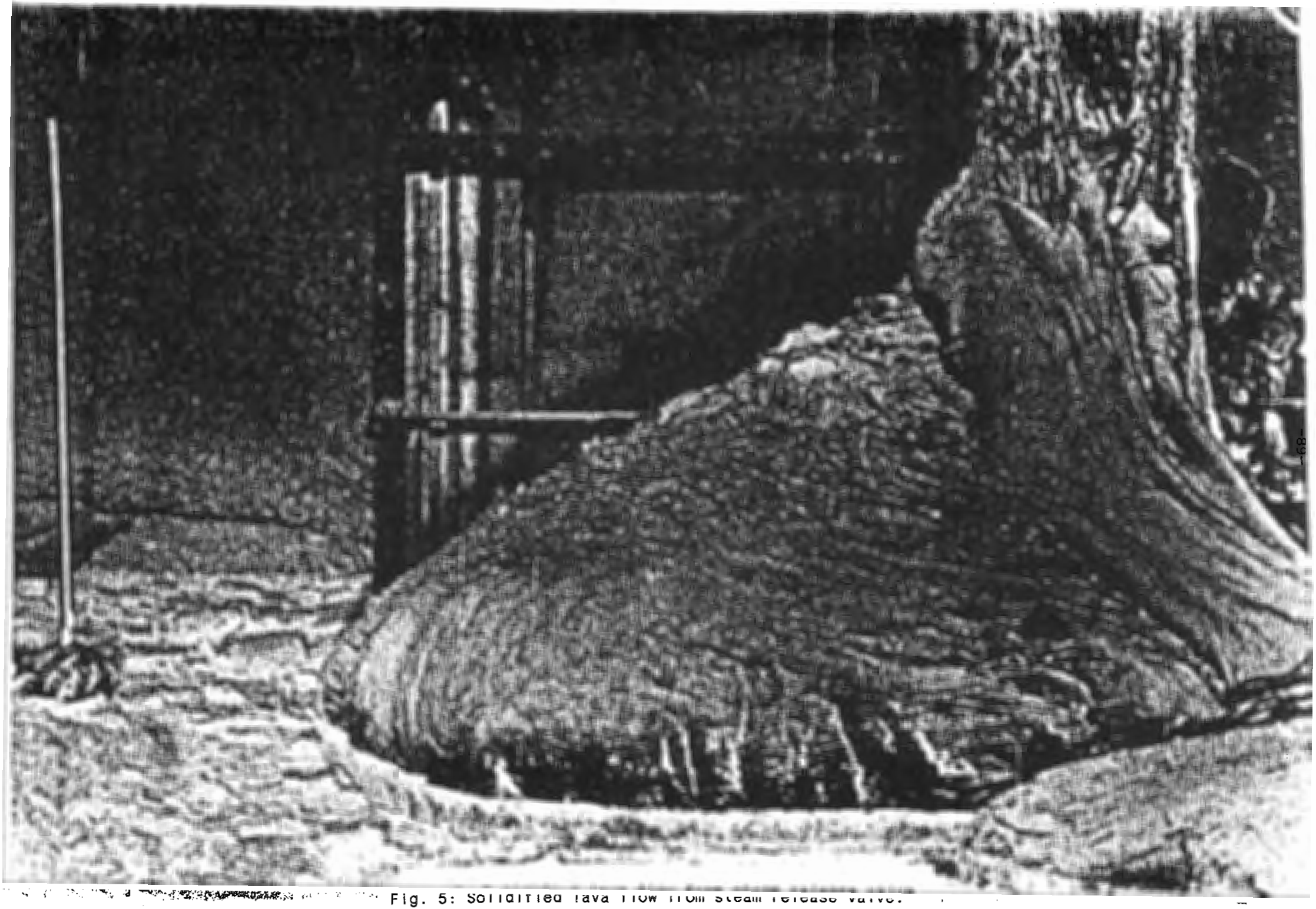

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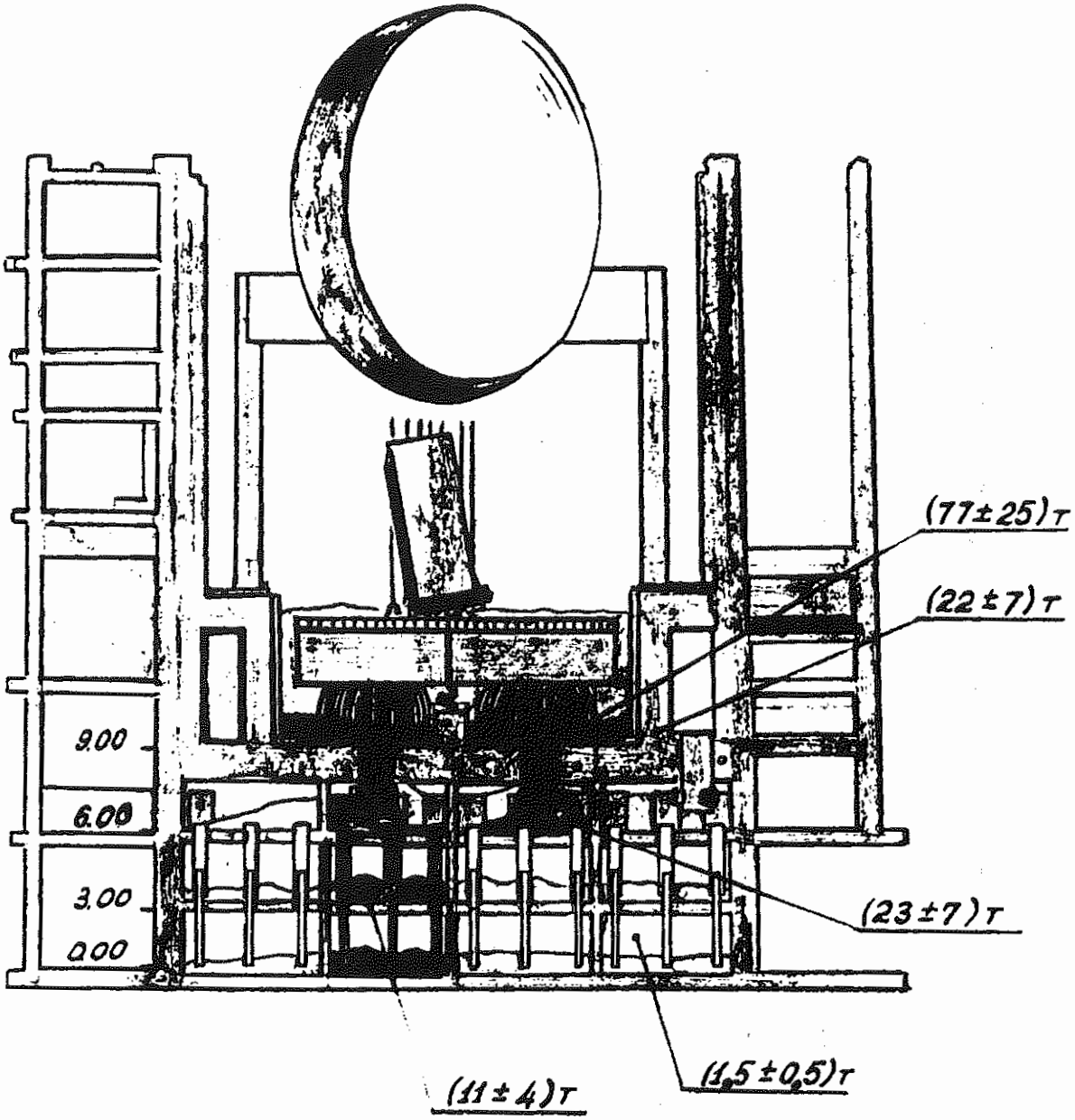

$\Sigma = (135 \pm 30)$ т

0.00; 3.00 – pressure-suppression pool

6.00 – steam distribution lines

9.00 – lower room No 305/2, other rooms

Chernobyl Source Term Estimation

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ABSTRACT

The Chernobyl source term available for long-range transport was estimated by integration of radiological measurements with atmospheric dispersion modeling and by reactor core radionuclide inventory estimation in conjunction with WASH-1400 release fractions associated with specific chemical groups. The model simulations revealed that the radioactive cloud became segmented during the first day, with the lower section heading toward Scandinavia and the upper part heading in a southeasterly direction with subsequent transport across Asia to Japan, the North Pacific, and the west coast of North America. By optimizing the agreement between the observed cloud arrival times and duration of peak concentrations measured over Europe, Japan, Kuwait, and the U.S. with the model predicted concentrations, it was possible to derive source term estimates for those radionuclides measured in airborne radioactivity. This was extended to radionuclides that were largely unmeasured in the environment by performing a reactor core radionuclide inventory analysis to obtain release fractions for the various chemical transport groups.

These analyses indicated that essentially all of the noble gases, 60% of the radioiodines, 40% of the radiocesium, 10% of the tellurium and about 1% or less of the more refractory elements were released. These estimates are in excellent agreement with those obtained on the basis of worldwide deposition measurements.

The Chernobyl source term was several orders of magnitude greater than those associated with the Windscale and TMI reactor accidents. However, the ^{137}Cs from the Chernobyl event is about 6% of that released by the U.S. and U.S.S.R. atmospheric nuclear weapon tests, while the ^{131}I and ^{90}Sr released by the Chernobyl accident was only about 0.1% of that released by the weapon tests.

INTRODUCTION

The radioactivity released into the atmosphere by the Chernobyl-4 nuclear reactor accident on April 26, 1986 was detected at numerous locations throughout the Northern Hemisphere. The release may be thought of as consisting of two components. One, was the activity released during the initial explosion on the first day of the accident, and the second component involving the material released during the subsequent fire which released radioactivity into the atmosphere until May 6, 1986. The radioactive material contained a wide spectrum of fission and activation products. The most important radionuclides of interest were ^{131}I, ^{137}Cs, and ^{134}Cs due to their abundance as well as their radiological and chemical characteristics. Atmospheric dispersion modeling was used to determine the evolution of the spatial distributions of the airborne radioactivity as it was transported over Europe and subsequently over the Northern Hemisphere.[1] Integration of these calculated distributions with the measurements of radioactivity yielded estimation of the source term for those radionuclides that were available for long-range transport and were measured in the airborne debris collected outside the Soviet Union. The release rates for radionuclides that were not generally measured in environmental samples were derived by first estimating the core's radionuclide inventory with a nuclear physics model, and then using the measured radionuclides as tracers of chemical transport groups, groups of radionuclides with similar chemical and physical behavior, to acquire the release fractions.

This paper discusses the hemispheric dispersion of the material, our estimates of the source term available for long-range atmospheric transport, and a comparison of the activity released by the Chernobyl accident with those for previous nuclear events.

SOURCE TERM ESTIMATION FOR ENVIRONMENTALLY MEASURED RADIONUCLIDES

The source term for ^{137}Cs and ^{131}I were acquired by means of optimizing the agreement between the spatial and temporal distributions of the radionuclides predicted by hemispheric-scale atmospheric dispersion modeling with measured air concentrations throughout Europe, Kuwait, Japan, and the U.S. A three-dimensional particle-in-cell

transport and diffusion model, driven by Northern Hemispheric wind fields supplied by the U.S. Air Force Global Weather Central, was used to perform the analysis. The model is based on the concept of generating a large number of marker particles to represent the radioactivity.[2] The particles are injected at the source point and subsequently transported within the three-dimensional Eulerian grid mesh by means of a transport velocity applied to each particle. This transport velocity consists of a wind velocity provided at each grid point and a diffusion velocity. In addition, gravitational settling and dry deposition velocity vectors as well as radioactive decay may be applied to the particles, as appropriate. Wet deposition, however, is not included in the model due to the unavailability of accurate precipitation fields over the Northern Hemisphere.

Air concentration measurements of radioactivity throughout the Northern Hemisphere revealed the presence of fresh fission products up to heights of about 7 km within a few days of the initial explosion. This suggests that some of the radioactivity released by the explosion and the subsequent fire within the reactor core must have been transported to heights well within the middle troposphere. This high altitude presence of the radioactivity may have been due to a variety of factors such as the thermal energy associated with the releases, rapid atmospheric mixing due to the presence of thunderstorms in the vicinity of the Chernobyl area during the first day of the accident, or the possible uplifting of the radioactive debris over a warm front situated between Chernobyl and the Baltic Sea. Consequently, the source term used in the model calculations included an upper-level cloud simulating the initial explosion and a lower-level cloud for the ensuing fire.

By integrating the model calculated cloud arrival times and duration of peak concentrations over Europe, Japan, Kuwait, and the U.S. with the measured air concentrations of specific radionuclides, it was possible by an iterative process to derive the most accurate temporal and spatial evolution of the radioactive cloud over the Northern Hemisphere. This evolution is illustrated in Fig. 1 by the model generated particle distribution patterns depicted over the Northern Hemisphere 2, 4, 6, and 10 days after the initial release. Note that initially the activity near the surface traveled in a northwesterly direction toward Scandinavia, passing over the northeastern corner of Poland

en route. The activity distribution continued its expansion into Scandinavia, while at the same time moving southwesterly through Poland toward central Europe. The upper part of the cloud headed in a southeasterly direction with subsequent transport across Asia to Japan, the North Pacific, and the west coast of North America. This integration of observed air concentrations with those calculated permitted the derivation of the total amounts of ^{137}Cs and ^{131}I released, as shown in Table 1. This approach indicates that 89 PBq of ^{137}Cs and 1300 PBq of ^{131}I (decay-corrected to 29 April 1986) were released. The release rates of other radionuclides that were measured in airborne radioactivity, also given in Table 1, were acquired by calculating their ratios relative to ^{137}Cs from measurements of airborne radioactivity within Scandinavia.

These estimates are consistent with those obtained by other investigators on the basis of worldwide deposition or atmospheric dispersion modeling.[3–9] This is illustrated by utilizing data reported by UNSCEAR.[10] Since some investigators accounted only for the ^{137}Cs reported over Europe or the Soviet Union, it was necessary to normalize their estimates to worldwide deposition. Using the UNSCEAR assumption that 43% of the ^{137}Cs was deposited within the Soviet Union, 38% over Northern Europe, 8% in the oceans, and the remainder within other parts of the Northern Hemisphere, one may translate the various estimates to the total amount of ^{137}Cs released into the atmosphere by the Chernobyl accident. A comparison of these estimates is given in Table 2. Note that most estimates are within about 20% of our estimate of 89 PBq and the USSR estimate of 86 PBq is almost identical.

SOURCE ESTIMATION FOR RADIONUCLIDES NOT GENERALLY MEASURED ENVIRONMENTALLY

The source term estimation process for radionuclides that were largely unmeasured in environmental samples collected outside the Soviet Union involved (1) estimating the core inventories of all radionuclides of interest, (2) using the radionuclides listed in Table 1 as tracers of various chemical transport groups, and (3) calculating the fractional release rate associated with each chemical transport group.

The inventory of specific radionuclides in the RBMK-1000 Soviet reactor core at the time of the accident was estimated by utilizing a modified version of a reactor core

model that solves a large number of rate equations to simulate fission, radioactive decay, nuclear transformations, and neutron reactions in order to calculate build-up and decay of the numerous radionuclides residing in the core.[11] Input to the model included data on the reactor's operating history obtained from the Soviet report presented at the IAEA Experts' Meeting in Vienna.[4]

The Chernobyl-4 reactor is thought to have started operation during December 1983 and operated continuously for about 875 days until 26 April 1986. At the time of the accident, the core-averaged fuel burn-up was 10.3 MWd/kg. The reactor is capable of being refueled during operation and utilizes 2% enriched fuel. To reconstruct its operating history, we assumed (1) an average load factor of 80%, (2) an 8 month start-up period for reaching full power, (3) a continuous refueling schedule of 3% fuel replacement per month commencing in August 1985 to give a core loading at the time of the accident of 75% original fuel, and (4) a core-averaged neutron energy spectrum similar to that in a commercial PWR, but at a lower power density. On the basis of these assumptions, one obtains the fuel burn-up rate given in Fig. 2. To ascertain that these assumptions are reasonable, it is useful to compare the calculated activity ratios of certain radionuclides residing in the core at the time of the accident with those measured in air samples collected in Scandinavia. This comparison is illustrated in Table 3 for isotope ratios of the same chemical elements to minimize atmospheric fractionation effects. The agreement between the measured and calculated values appears to be reasonable in view of the roughly 20% uncertainty associated with the measured ratios. In view of this agreement, it is possible to estimate with reasonable confidence the core's inventory of a large suite of radionuclides at the time of the accident. The inventory of the radionuclides of primary interest from an environmental impact point of view is given in Table 4. A review of the data in the table reveals that we estimate the core to have contained 2300 PBq of ^{131}I, 210 PBq of ^{137}Cs, and 140 PBq of ^{134}Cs. These values have been decay-corrected to 3 days after the accident.

From the releases given in Table 1 and the core inventories listed in Table 4, we obtained the fraction released and available for long-range transport for those radionuclides that were measured in the airborne radioactivity sampled outside the Soviet

Union. These release fractions may be extended to a host of unmeasured radionuclides by using those listed in Table 1 as tracers of their corresponding chemical transport groups as defined by the USAEC reactor safety study.[12] This is illustrated in Table 5 which depicts the Chernobyl release fractions. These reveal that essentially all of the noble gases, 60% of radioiodine, and about 40% of the cesium radionuclides were released. This was followed by the release of about 10% of the tellurium and 1% or less of the more refractory elements. These release fractions agree well with those reported by the Soviet Union when considering their estimates of the reactor core inventory of radionuclides at the time of the accident and the activity that was transported outside the Soviet Union. The release fractions are approximately 70% of the most likely estimates derived from the WASH-1400 study for a major meltdown of a light- water reactor, but are well within the ranges of potential release fractions associated with such a meltdown. A comparison with the release fractions derived for the 1957 Windscale reactor accident in the U.K. reveals that considerably higher fractions of iodine and cesium radionuclides were associated with the Chernobyl accident.[13]

On the basis of the release fractions given in Table 5 and the core inventories listed in Table 4, we obtained estimates of the total activity released for radionuclides of interest that were not generally measured. These are given in Table 6. Most notable is ^{90}Sr whose release rate is estimated from Table 6 to be only about 1% of that for ^{137}Cs due to its low volatility. This seems in reasonable agreement with a few measurements in western Europe.

COMPARISON OF CHERNOBYL WITH OTHER NUCLEAR EVENTS

In order to place the radioactivity released from the Chernobyl reactor in perspective with radioactivity releases from previous nuclear events, it is useful to compare the Chernobyl release with the radioactivity estimated to have been produced by the U.S. and the U.S.S.R. atmospheric nuclear weapons testing programs, as well as with the releases associated with the TMI and the Windscale reactor accidents. However, one should be aware that a complete comparison of the radiological impact of the atmospheric weapon testing programs with that produced by the Chernobyl reactor accident

is very difficult. This is because the weapon tests produce different mixtures of radionuclides, have different injection heights in the atmosphere, and were conducted at isolated sites far from population centers. Nevertheless, it is still useful to compare the data in Table 7, which shows for the most notable radionuclides of interest the amounts released by the weapon tests as well as the TMI and Windscale reactor accidents. A comparison of the data shows that with the exception of ^{134}Cs, the activity released by the Chernobyl accident is minor relative to the weapon test releases, which are based on 225 MT (megatons of TNT equivalent) of fission. The 89 PBq of ^{137}Cs released by the Chernobyl event is only 6% of that produced by the weapon tests, while the remaining radionuclide releases represent less than 1% of the corresponding weapon test releases. However, since ^{134}Cs is is not significantly produced in weapon tests, the Chernobyl contribution is at least 30 times greater than that produced by the weapon testing programs. One may also note that the Chernobyl releases were greater by at least several orders of magnitude than those associated with the Windscale and TMI reactor accidents.

SUMMARY

We utilized Northern Hemispheric scale atmospheric dispersion modeling in conjunction with radiological mesurements to estimate the source term for the radionuclides measured in the airborne radioactivity. By using a nuclear physics model to calculate the expected core radionuclide inventory at the time of the accident and WASH-1400 chemical transport groups, it was possible to estimate the source term available for long-range transport for radionuclides that were largely unmeasured. Thus, we estimated that essentially all of the noble gases, 60% of the radioiodines, 40% of the radiocesium, and 10% of the tellurium, and about 1% or less of the more refractory elements were released.

The source term for the Chernobyl accident was several orders of magnitude greater than those associated with the Windscale and TMI reactor accidents, while our estimates of the ^{137}Cs released by the Chernobyl event is about 6% of that released by the U.S. and U.S.S.R. atmospheric nuclear weapon tests.

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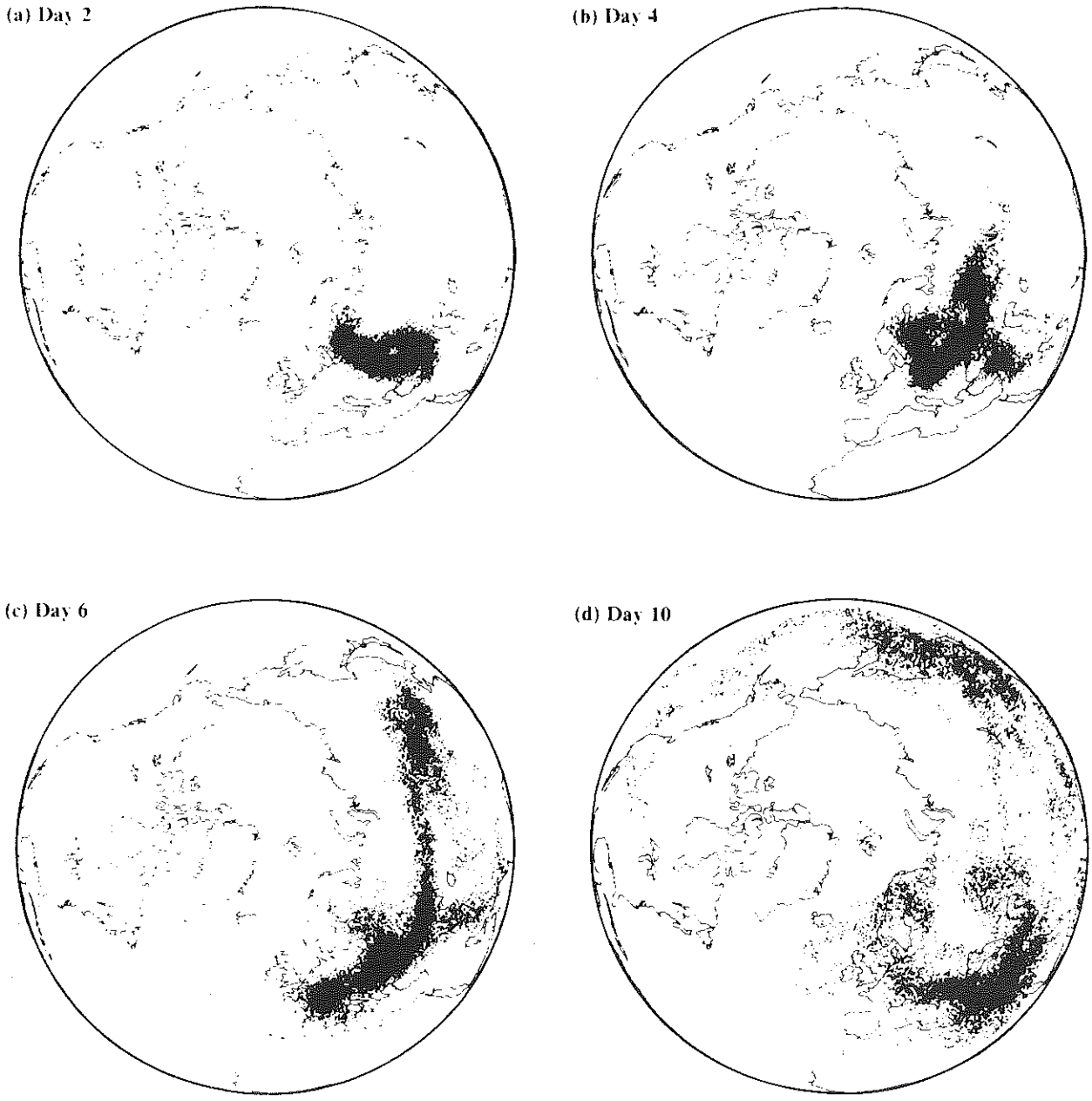
FIGURE CAPTIONS

Fig. 1. The calculated spatial distributions of the radioactive cloud over the Northern Hemisphere on selected days after the explosion on 26 April 1986.

Fig. 2. The estimated fuel burn-up rate of the Chernobyl-4 reactor core.



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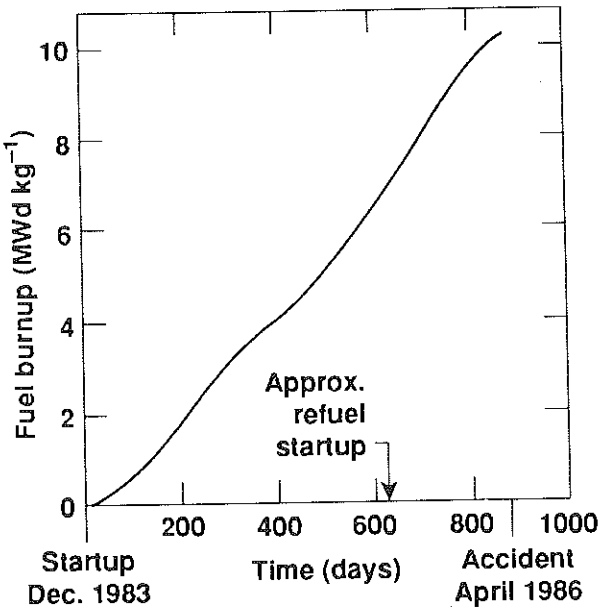

Fig. 2. The estimated fuel burn-up rate of the Chernobyl-4 reactor core.

Table 1. Estimated releases of radionuclides from the Chernobyl event. The activities are decay-corrected to 3 days after the accident; 1 PBq = 10^{15} Bq. The uncertainties associated with these releases are about 30%.

Activity Released	
Radionuclide	PBq
^{95}Zr	8.5
^{103}Ru	27
^{106}Ru	6.3
^{131}I	1300
^{133}I	300
^{132}Te	200
^{134}Cs	48
^{136}Cs	20
^{137}Cs	89
^{140}Ba	37
^{141}Ce	8.5
^{144}Ce	5.2

Table 2. A comparison of estimates provided by various investigators of the total ^{137}Cs activity released by the Chernobyl reactor accident. The normalization factors, obtained from UNSCEAR (1988), account for the worldwide distribution of radioactivity.

Investigators	Geographical Area	Estimated Activity Released (PBq)	Normalization Factor	Total Estimated Released (PBq)
Anspaugh et al.	worldwide	98	1	98
U.S.S.R.	U.S.S.R.	37	1/.43	86
LLNL estimate	worldwide	89	1	89
ApSimon et al.	Europe	39	1/.38	103*
van Egmond & Seuss	Europe	50	1/.38	132*
Cambray et al.	worldwide	70	1	70
Sorensen	worldwide	100	1	100
Aarkrog	worldwide	100	1	100
UNSCEAR 1988	worldwide	70	1	70

* There is some double counting because these estimates account for deposition in the Soviet Union and the normalization factor does not.

Table 3. Fission product activity ratios (decay-corrected to 26 April 1988) measured in environmental samples collected within Scandinavia and predicted by the core inventory model. The latter are core-averaged values.

Activity Ratio	Measured	Model Estimates
$^{103}Ru/^{106}Ru$	4.2	4.0
$^{134}Cs/^{137}Cs$	0.54	0.65
$^{136}Cs/^{137}Cs$	0.27	0.40
$^{134}Cs/^{136}Cs$	2.4	1.7
$^{131}I/^{133}I$	0.56	0.62
$^{141}Ce/^{144}Ce$	1.6	1.6

Table 4. Model-derived estimates of the core inventory of selected radionuclides. The activities are decay-corrected to 3 days after the accident. The uncertainties associated with these inventories are about 30%.

Inventory	
Radionuclide	PBq
^{85}Kr	20
^{89}Sr	2800
^{90}Sr	160
^{91}Y	3600
^{95}Zr	4600
^{99}Mo	2400
^{103}Ru	3900
^{106}Ru	1000
^{127}Sb	180
^{127m}Te	37
^{129m}Te	130
^{131}I	2300
^{131m}Te	67
^{132}Te	2100
^{133}I	440
^{133}Xe	4700
^{133m}Xe	96
^{134}Cs	140
^{136}Cs	70
^{137}Cs	210
^{140}Ba	4500
^{140}La	5000
^{141}Ce	4600
^{144}Ce	3000
^{147}Pm	460

Table 5. Estimated percentage releases of radionuclides available for long-range transport from Chernobyl compared with the USAEC reactor safety study (WASH-1400) and the Windscale reactor accident. The uncertainties of the Chernobyl percentage releases are about 40%.

		Chernobyl	WASH-1400 (%)		Windscale
Chemical Group	Tracers	(%)	Range	Best Estimate	(%)
Noble Gases[a]	^{133}Xe	>90	50–100	90	100
Halogens[b]	^{131}I,^{133}I	60	50–100	90	20
Alkali Metals[c]	^{134}Cs,^{136}Cs,^{137}Cs	40	40–90	80	20
Tellurium Group[d]	^{132}Te	10	5–25	15	20[i]
Alkaline Earths[e]	^{140}Ba	0.8	2–20	10	0.2
Noble Metals[f]	^{103}Ru,^{106}Ru	0.7	1–10	3	2[i]
Rare Earths[g]	^{141}Ce,^{144}Ce	0.2	0.01–1	0.3	0.2
Refractory Oxides[h]	^{95}Zr	0.2	0.01–1	0.3	0.2

[a] Xe, Kr

[b] I, Br

[c] Cs, Rb

[d] Te, Se, Sb

[e] Ba, Sr

[f] Ru, Rh, Pd, Mo, Tc

[g] Y, La, Ce, Pr, Nd, Pm, Sm, Eu, Np, Pu, Cm

[h] Zr, Nb

[i] Sb was estimated at 0.2%[13]

[j] Rh and Pd were estimated at 0.2%[13]

Table 6. Estimated releases of radionuclides that were largely unmeasured in airborne radioactivity collected outside the Soviet Union. The activities are decay-corrected to 3 days after the accident.

Radionuclide	Activity Released (PBq)
^{85}Kr	≥18
^{89}Sr	22
^{90}Sr	1.3
^{91}Y	7.2
^{99}Mo	17
^{127}Sb	18
^{127m}Te	3.7
^{129m}Te	13
^{131m}Te	6.7
^{133}Xe	≥4200
^{133m}Xe	≥86
^{140}La	10
^{147}Pm	0.9

Table 7. Comparison of Chernobyl with other nuclear events. Except for TMI, the activities are decay-corrected to 3 days after the events.

	Radioactivity Released (PBq)			
Nuclide	Chernobyl	Weapon Tests	Windscale	TMI
^{137}Cs	89	1500	0.04	ND+
^{134}Cs	48	<1.5	0.001	ND
^{90}Sr	1.3	1300	2.2×10^{-4}	ND
^{133}Xe	≥4200	2×10^{6}	14	370
^{131}I	1300	7.8×10^{5}	0.6	0.001

+ not detected

A Review of Source Term and Dose Estimation for the TMI-2 Reactor Accident

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ABSTRACT

The TMI-2 nuclear reactor accident, which occurred on March 28, 1979 in Harrisburg, Pennsylvania, produced environmental releases of noble gases and small quantities of radioiodine. The releases occurred over a roughly two week period with almost 90% of the noble gases being released during the first three days after the initiation of the accident. Meteorological conditions during the prolonged release period varied from strong synoptic driven flows that rapidly transported the radioactive gases out of the Harrisburg area to calm situations that allowed the radioactivity to accumulate within the low lying river area and to subsequently slowly disperse within the immediate vicinity of the reactor. Meteorological and radiological data, collected throughout the Harrisburg area by numerous organizations, were used in conjunction with atmospheric dispersion modeling to define the time and spatial evolution of the radioactive plume structure for assessing the environmental impact of the release.

The results reported by various analysts, revealed that approximately 2.4–10 million curies of noble gases (mainly Xe-133), and about 14 curies of I-131 were released. During the first two days, when most of the noble gas release occurred, the plume was transported in a northerly direction causing the most exposed area to lie within a northwesterly to northeasterly direction from TMI. Changing surface winds caused the plume to be subsequently transported in a southerly direction, followed by an easterly direction. Thus, the total dose pattern was governed by the complexities inherent in the temporal evolution of the source term, the changing meteorology and the terrain.

The calculated maximum whole body dose due to plume passage exceeded 100 mrem over an area extending several kilometers north of the plant, although the highest measured dose was 75 mrem. The collective dose equivalent (within a radius of 80 km) due to the noble gas exposure ranged over several orders of magnitude with a central estimate of 3300 person-rem. The small I-131 release produced barely detectable levels of activity in air and milk samples. This may have produced thyroid doses of a few milirem to a small segment of the population.

INTRODUCTION

On March 28, 1979 the Three-Mile-Island Unit 2 nuclear power reactor experienced a severe fuel damage accident that resulted in the release of fission products from the core into the containment atmosphere. Some of the fission products escaped from the primary containment by means of the Makeup and Purification System that transported primary coolant into the auxiliary building. Outgassing of the primary cooling water into the auxiliary building atmosphere permitted volatile fission products to enter the building ventilation system leading to environmental releases of noble gases and small amounts of radioiodine.

The amounts of specific radionuclides released into the atmosphere and the associated environmental consequences were extensively investigated by numerous organizations. The primary studies of interest here were conducted by the plant operator, General Public Utilities (GPU); the Department of Energy (DOE), the AD HOC Interagency Study Group that included participation by the Nuclear Regulatory Commission (NRC), the Department of Health, Education and Welfare (HEW) and the Environmental Protection Agency (EPA); and, the task forces supporting the President's Commission on the Accident at Three Mile Island.

These studies were based on extensive data gathered during the accident and our knowledge of the physical processes governing the behavior of radionuclides in the environment whenever data were lacking. The data were acquired by radiation measurements within the plant, thermoluminescent detectors (TLD) placed in the environs surrounding the facility, environmental radiation measurements made by both surface and airborne detection systems, radionuclide analysis of numerous environmental samples, and meteorological data from the site and the surrounding area. These data were used either independently or in conjunction with atmospheric dispersion modeling to estimate the time evolution of the source term from the auxiliary building vent, and the spatial and temporal evolution of the integrated dose pattern over the Harrisburg region.

This review, which summarizes the major findings of these studies, reveals that the environmental and health impact to the surrounding population was minimal. However,

the details of the source term, the population and individual dose estimates may differ by factors of three or four from one study to another. This paper presents the authors views of the most likely consequences of the accident and a credible range of uncertainty associated with these estimates.

NOBLE GAS SOURCE TERM ESTIMATION

Several independent attempts were made to estimate the magnitude of the noble gas releases from the auxiliary building vent where essentially all of the atmospheric releases occurred. Unfortunately, the normal vent monitor, which was designed to measure routine operational releases, exceeded its saturation limit early in the accident with attendant loss of valuable source term information. Thus, it was necessary to utilize various indirect approaches to estimate the release magnitudes. The GPU derived noble gas source term estimate was based on a combination of numerical modeling techniques and in-plant and environmental radiation measurements.[1] This involved a multistep approach that included (1) reactor core radionuclide inventory modeling and analysis of air samples collected from the building vent system to define the isotopic mix of the noble gases, (2) analysis of temporal variations of the output from area monitors situated within the auxiliary building to indicate the relative release rate as a function of time since the vent monitoring system became saturated during the high release period, (3) atmospheric dispersion modeling based on a relative source rate and on-site meteorological measurements to calculate the gamma dose to ground level receptors, and (4) optimizing the agreement between the calculated dose rates with the environmental TLD measurements by source term scaling. This process produced the estimated total noble gas release rates shown in Fig. 1. Combining these release rates with the time dependent isotopic composition, and integrating with time produced the estimated noble gas radionuclide specific total activity releases shown in Table 1. The results indicate that about 10 million curies of noble gases were released during the period from March 28 through April 30. Approximately 80% of the noble gas activity released was due to Xe-133. A review of the temporal variation of the release revealed that about 66% of the total activity was released during the first day and a half of the accident, while

another 22% of the total activity was released during the following two days; from 1700 on March 29 to 1600 on March 31. Essentially all of the release had occurred by April 6.

A different approach for source term estimation was taken by the Task Group of Health Physics and Dosimetry of the President's Commission on the Three Mile Island Accident.[2] Their approach focused on a careful analysis of the response of a building area radiation monitor, situated near the ventilation ducts that led to the stack vent where the radiation levels were sufficiently low to avoid detector saturation. Intercomparison of this detector's response with that of the vent monitor at various radiation levels below the vent monitor's saturation limit, permitted extrapolation of vent radiation levels above the vent monitor's saturation level. This analysis led to a total noble gas release of 2.4 million curies.

The uncertainty associated with the noble gas release estimates are within a factor of 4. The 10 million curie estimate, which represents about 8–10% of the noble gas core inventory at the time of the accident, seems to be in reasonable agreement with more recent information on the post accident inventory and distribution of radionuclides within the TMI-2 reactor system. This study revealed that 91% of the Kr-85 could be accounted for within the containment atmosphere, the previously melted fuel, and the in-tact fuel-rods; thus, inferring a 9% loss to the atmosphere.[3]

EXTERNAL DOSE ESTIMATION

Several approaches were taken to estimate the radiation dose received by the affected population due to the release of noble gases. This included several independent studies based on atmospheric dispersion modeling in conjunction with the environmental radiation measurements as well as spatial interpolation of the TLD measurements at a limited number of locations surrounding the site. The various dispersion modeling efforts used a variety of models that ranged from Gaussian to complex three-dimensional models.

Meteorological data were available from the on-site tower as well as from several local sources. The meteorological conditions during the first five days of the accident, when the highest release rates occurred, consisted mainly of up and down-river flows. From

the morning on March 28, when the release was initiated, until mid-afternoon on March 29, the winds were primarily from the southeast- southwest direction at approximately 3 m/s. Subsequently the winds rotated to a northeast-northwest direction with an average speed of 1–2 m/s. Calm and highly variable conditions were observed during the night of March 29-30. These calm and variable conditions continued until the evening of March 30 when strong and steady southerly winds of about 3 m/s returned. On April 1 the winds rotated into the westerly to northwesterly directions with speeds generally ranging between 1–3 m/s.

Atmospheric dispersion modeling, based on these meteological conditions, permitted elucidation of the temporal evolution of the time-integrated dose pattern. This is illustrated in Fig. 2 which shows the evolving integrated dose pattern.[4] These results were generated by means of a three-dimensional mass-consistent wind field model coupled with a particle-in-cell transport and diffusion model using a normalized one million curie release that varied in time according to that shown in Fig. 1. The figure shows how the dose pattern was quickly and predominantly established by the generally south-to-north flow during the initial release period. Thereafter, the low levels of release resulted in only relatively minor but discernible changes in the initial dose pattern. Note particularly the southward extension of the pattern from March 29 to March 30, the east and southeast spread from March 30 to March 31, and finally the "diffusion-like" effect of nine days of synoptic and diurnal meteorological variations with a small source term.

Integration of atmospheric dispersion patterns with the environmental radiation measurements permitted the estimation of the total integrated dose to the exposed populations. Using the DOE aerial radiation measurements made within the plume on a regular basis over a two week period, resulted in the DOE integrated dose pattern shown in Fig. 3.[5] Note that the highest doses occurred in the areas immediately north of the plant with secondary nodes extending in the southeast and easterly directions. A similar pattern was generated by GPU after integrating the TLD measurements with their finite plume dispersion modeling.[1] These studies indicate that a total dose of 100 mrem was exceeded over an area extending several kilometers in a northeast to

northwest direction from the plant, although the highest measured off-site dose by the TLD network was 75 mrem.

Integration of the dose pattern with the population distribution throughout the affected region led to an assessment of the collective dose equivalents. The most credible estimates are given in Table 2. The estimates resulting from analysis of the DOE aerial measurements was 2000 person-rem[5] in contrast to 3300-3400 person-rem obtained by the AD HOC Interagency Study Group on the basis of spatial interpolation of the TLD measurements by either atmospheric dispersion modeling or by inverse distance scaling.[6,7] The GPU obtained 3300 person-rem by combining dispersion modeling with the 10 million curie source term and the TLD measurements.[1] The lowest collective dose estimate, 500 person-rem, was derived on the basis of atmospheric dispersion using the 2.4 million curie source term.[2] Thus, the central estimate is about 3300 person-rem with a range of several orders of magnitude.

IODINE RELEASE

The release of small quantities of radioiodine was detected by analysis of air samples collected by filter and charcoal samplers situated in the building ventilation system. These analyses revealed the estimated I-131 release rates from March 28 to April 25, 1979, given in Fig. 4, which led to a total release of 14 curies of I-131. An additional 2.6 curies of I-133 was also released. It is of interest to note that the I-131 release rates do not decrease rapidly like those for the noble gases shown in Fig. 1, but stays reasonably constant throughout the measurement period. This is most likely due the fairly constant evaporation rate of the iodine from the contaminated water that flooded the auxiliary building. The fraction of iodine released is extremely small due to its preferential retention in the water and the subsequent plateout within the building.

On the basis of these release rates and atmospheric dispersion modeling, the highest adult thyroid dose due to inhalation was estimated to be about 7 mrem at a distance of 2400 m from the plant, and the collective thyroid dose for the two million people within an 80 km radius was estimated to be about 180 person-rem. Measurements of iodine air concentrations suggested that these estimates have an uncertainty of about a factor of 4 and are most likely to be higher than indicated by the measurements.

Milk samples were collected at numerous dairies to evaluate the I-131 concentrations. Of approximately 250 samples collected, less than half of the samples showed detectable concentrations. The highest measured concentration in milk was 41 pci/l and the average concentration was less than 20 pCi/l.[5] This potentially could have resulted in a child thyroid dose of about one millirem.

CONCLUSIONS

Extensive environmental monitoring of radioactivity throughout the Harrisburg area during the TMI-2 accident revealed that the environmental and health implications of the noble gas and iodine releases were minimal. Approximately 2.4–10 million curies of noble gases (mainly Xe-133) were released; the higher value being the most likely. The collective dose resulting from the release to the 2 million people living within a radius of 80 km of the plant was about 3300 person-rem. This represents about 1% of the normal annual background radiation dose for that area. The average dose to an individual living within 8 km of the plant was estimated to be about 10% of the annual background dose. The maximum estimated dose received by an off-site individual was about 75 mrem.

The 14 Ci release of I-131 resulted in barely detectable levels of I-131 in air and milk samples collected during the accident. These low levels of activity may have produced thyroid doses of a few millirem to a small segment of the surrounding population. Thus, no detectable health impacts due to radiation exposure were expected to occur as a result of the accident.

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FIGURE CAPTIONS

Fig. 1. Estimated noble gas release rates from the TMI-2 accident.

Fig. 2. Normalized calculated integrated dose patterns in units of millirem due to TMI-2 noble gas release on (a) March 29 (24-h integration), (b) March 30 (48-h integration), (c) March 31 (72-h integration), and (d) April 7, 1979 (240-h integration). The patterns are based on the release of one million curies of Xe-133.

Fig. 3. Estimated dose pattern derived from the DOE aerial measurements from March 28–April 3, 1979. The units are in millirem.

Fig. 4. Estimated I-131 release rates.

TABLE HEADINGS

Table 1. Estimated noble gas releases (megacuries).

Table 2. Estimates of collective dose equivalents due to noble gas releases from TMI-2 accident.

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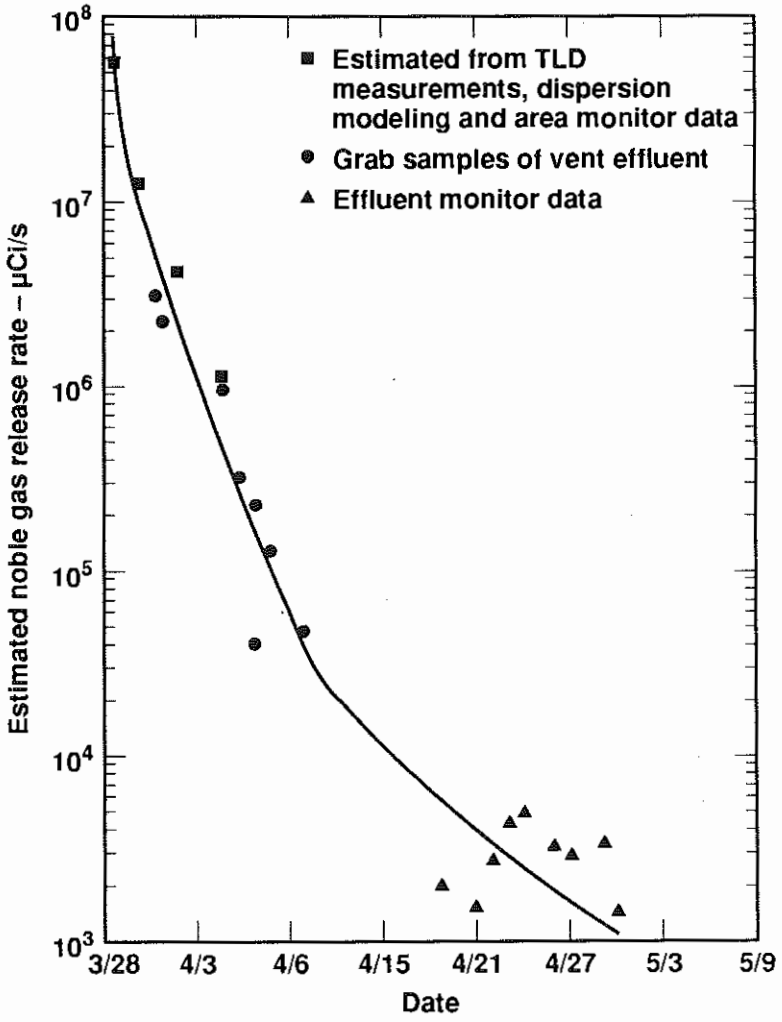
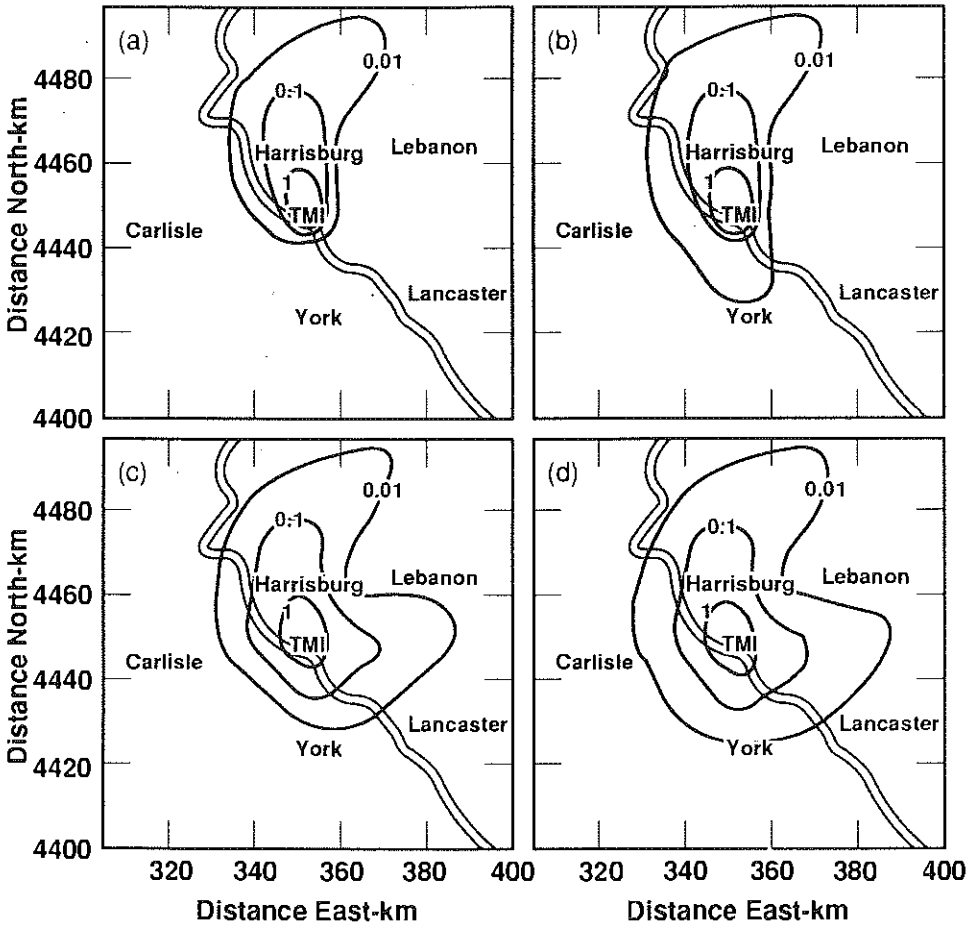


Fig. 1. Estimated noble gas release rates from the TMI-2 accident.



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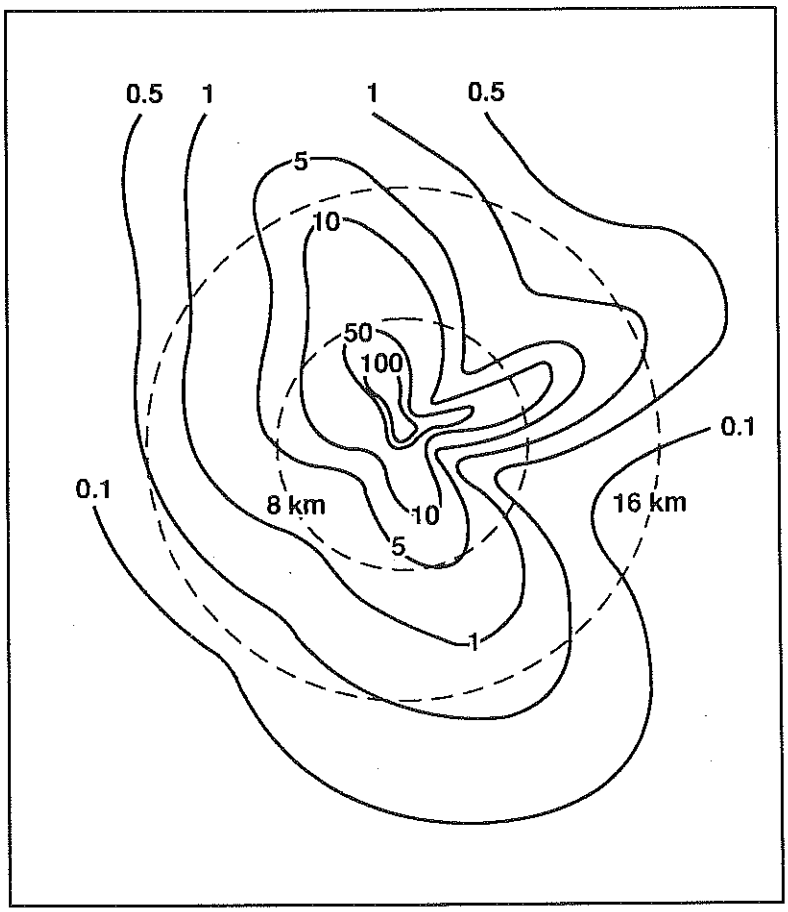

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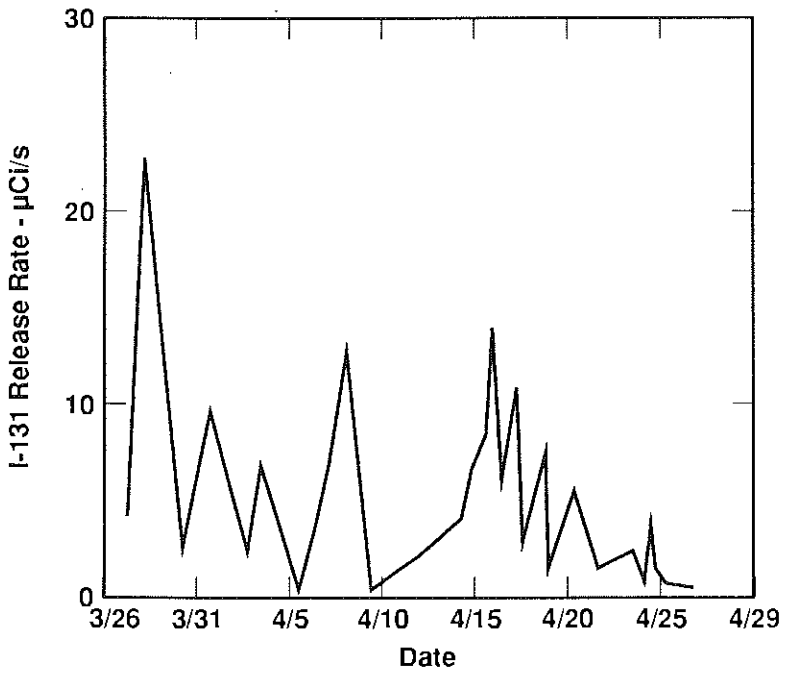

Fig. 4. Estimated I-131 release rates.

Table 1. Estimated Noble Gas Releases* (Megacuries)

	Period					
Isotope	3/28–3/29	3/29–3/31	3/31–4/3	4/3–4/6	4/6–4/30	Total*
Xe-133	4.9	2.1	1.1	0.27	0.015	8.3
Xe-133m	0.12	0.039	0.015	0.0019	—	0.17
Xe-135	1.5	0.077	0.0014	0	0	1.5
Xe-135m	0.14	0.0013	0	0	0	0.14
Kr-88	0.061	0	0	0	0	0.061
Total in Period	6.6	2.2	1.1	0.27	0.015	10.2
% of Total	65	22	10.8	2.6	0.15	100
Cumulative	65	86	97	99.8		

*To 4/30/79

Table 2. Estimates of Collective Dose Equivalent Due To Noble Gas Releases From TMI-2 Accident.

		Person-rem	
Group	Technique	Best Value	Range
DOE	Aerial measurements	2000	
GPU	TLD measurements and dispersion modeling	3300	1650–6600
ADHOC Group	TLD measurements and dispersion modeling	3400	
ADHOC Group	Spatial interpolation of measurements	3300	1600–5300
Pres. Commission	Source term and dispersion modeling	500	50–5000

Twenty-Five Years Before Chernobyl

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Our international Seminar is to deal exclusively with the consequences of three major nuclear accidents: Kyshtym, Windscale and Chernobyl.

For this reason, I had originally not planned to present this paper, which deals with another major incident. However, while attending this meeting of the International Union of Radioecologists, I felt that such a representative group of specialist radioecologists from 20 countries of the world would be interested to hear about the serious accident which took place on a nuclear submarine in 1961, 25 years before Chernobyl. This accident could also have led to a world ecological catastrophe comparable with that of Chernobyl. Fortunately, however, it was possible to prevent this, but at a high price - the loss of several lives. The accident took place in the nuclear reactor of the first Soviet missile-carrying nuclear submarine.

Until July 1990 it was forbidden even to mention this event. However, times change, and international cooperation, including cooperation in the scientific sphere, is being extended and consolidated.

We managed to have two articles published on this subject in the Pravda newspaper under the heading "Top secret". The first article was entitled "25 years before Chernobyl" and the second "The atom did not explode in the sea".

I doubt whether all European and American scientists subscribe to the main newspaper of our country. What is more, the newspaper articles were not long enough to include all the details and, as I witnessed the events of 1961, I would like to give you first-hand information myself.

At 4 a.m. on 4 July 1961 there was a depressurization in the primary coolant circuit in one of the two water-water reactors on the missile-carrying submarine in the northwestern part of the Atlantic Ocean several thousand kilometres from the Soviet Union. This was caused by a crack in the piping of the system controlling the reactor. This piping was connected to the primary coolant circuit. In reactors of this type the water in this circuit is a moderator and coolant at the same time, has a mean temperature of around 300°C and is kept at a pressure of several hundred atmospheres with the aid of a special gas-based high-pressure system.

As a result of the depressurization the pressure in the primary coolant circuit of the stricken reactor fell to approximately atmospheric pressure and the water boiled and evaporated.

This meant that the core had no cooler. The reactor protection system functioned correctly, and the emergency shutdown rods (compensatory lattice) dropped down into the lower part of the core and the reactor became subcritical. In other words, the reactor was shut down automatically, and the operators then carried out all the necessary work on the systems of the damaged reactor. In this way, the possibility of an uncontrolled chain reaction in the core of the stricken reactor was averted at the beginning.

But as they say, "it never rains, but it pours": a fire broke out in the compartment of the stricken reactor but was fortunately extinguished before it spread. The submarine surfaced for the first time after many weeks at sea.

In addition, the loss of pressure in the primary coolant circuit and the fire caused radioactivity to spread through the compartments of the submarine.

I cannot give you detailed data on the gamma, beta and alpha activity at the moment. Suffice it to say that the total contamination level in the submarine's control room reached 50 R/hour and in some parts of the reactor compartment the contamination level was extremely high, but the conditions made it extremely difficult to obtain precise values.

Naturally, the compartments of the submarine were hermetically sealed, but it was not possible to cut off all links between them.

However, the greatest danger was that the temperature of the fuel elements in the shut-down, subcritical reactor, which had been working at 100% capacity for many weeks prior to the accident and whose core had been left without cooling, began to rise as a result of fission and the absence of heat removal.

Normal afterheat removal from the shut-down reactor was not possible, and the spraying of the core was ineffective. The temperature on the surface of the fuel elements rose inexorably to over 800°C. There was a serious danger that the fuel elements would melt as a result of afterheat. Melting of the fuel elements could have made the reactor core unstable. There was the possibility of an uncontrollable reaction. Even if this had not happened, there was a possibility that the core and the reactor vessel would be destroyed, with all the resulting consequences for the crew of the submarine and the environment.

If the stricken reactor had come to rest on the ocean bed, it could not have produced an ecological disaster on the scale of Chernobyl for many objective reasons of which you will be aware, but it would nevertheless have been a catastrophe with global consequences.

In order to prevent this, a temporary emergency system for cooling the core with fresh water, of which the submarine had a reserve, was planned and implemented for a duration of several hours. A number of weapons systems on the submarine had to be dismantled in order to have sufficient piping for this system. It was necessary to carry out welding work in the reactor compartment itself. The operators who carried out this work were first-rate specialists who were fully aware of the risks involved.

The system proved to be effective, as it succeeded in stopping the rise in temperature of the fuel elements and prevented their meltdown and the explosion of the core, but at the loss of several lives.

Seven people died one week later. These were Lieutenant Boris Korchilov and petty officers and seamen Boris Ryzhikov, Yury Ordochkin, Yevgeny Kashenkov, Semen Penkov, Nikolay Savkin and Valery Kharitonov. Three weeks after the accident Lieutenant Commander Yury Povetev died.

This is the information I wanted to give you on this previously unknown major accident which could have caused a major ecological disaster.

I doubt whether all military nuclear installations, in particular submarines, will be taken out of use in the near future. However, although the situation in the world is by no means perfect, things are certainly improving, which gives us grounds to hope that military sources of radioactivity and all potential sources of radioactive contamination of the environment will be limited and will one day disappear altogether.

Session III

ATMOSPHERIC DISPERSION, RESUSPENSION, CHEMICAL AND PHYSICAL FORMS OF CONTAMINATION

Atmospheric Dispersion Models in Assessment of Accidental Releases (illustrated by the Windscale and Chernobyl Accidents)

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Abstract:

Analysis of the pattern of dispersal and deposition of radionuclides, using the MESOS long-range atmospheric transport model, will be described for both the Windscale accident of 1957, and the Chernobyl accident of 1986. Deductions about source terms will be discussed, and limitations of the techniques used. New developments in models for use in any future accident situation will also be reviewed.

Introduction

Under article 37 of the Euratom treaty member states of the European Community are required to consider the transfrontier consequences of a nuclear accident in another country when planning a new nuclear plant. In this connection we developed techniques for assessing such consequences in a probabilistic manner by treating a large number of hypothetical accidents of varying release duration and in different meteorological conditions. This involved construction of a computer model, MESOS, for atmospheric transport and deposition of different nuclides out to long distances. This rather simple model has been applied to study both the Windscale and Chernobyl accidents, and the results will be described in this paper together with some of the difficulties and uncertainties.

The MESOS model, for application to large numbers of situations, was necessarily rather simplified and limited by the data used. Since the Chernobyl accident new models have been developed, suitable for use in any future emergency. These techniques will also be described, and the ongoing work to improve them, especially in critical situations (for example in frontal systems where wet deposition is likely to occur).

The MESOS model

Before embarking on the two accidents a brief description of the MESOS computer model, which we have used as a tool in analysing the pattern of environmental contamination, seems appropriate. MESOS uses meteorological data to simulate dispersal of the release downwind. Thus it follows in detail the trajectories and history of puffs released at 3 hour intervals, assuming that meterial released in between fans out along intermediate pathways. The vertical dilution depends on the evolution of the atmospheric boundary layer, particularly changes in depth of the turbulent mixing layer above the ground Separate dry deposition and wet deposition in precipitation are calculated, as well as time-integrated atmospheric concentrations of each nuclide. The meteorological input comprises standard observations from synoptic weather stations reporting routinely, including the "present weather" code to indicate rainfall and its character and intensity. Thus the model is designed for *a posteriori* investigation, not in forecasting mode during an emergency.

By comparing radiological measurements with model estimates it is possible to work backwards to assess the source terms, as is illustrated in this paper.

THE WINDSCALE RELEASE:-The pattern of release

The release from Windscale took place over two days starting around 8.10am on the

morning of 10 October 1957,when stack monitors first showed an increase of activity. The accident resulted from the uncontrolled release of Wigner energy within the graphite moderator blocks, and by the end of that day 150 channels of fuel were involved in the resulting fire. The decision was taken to douse the pile with water, and accordingly water was switched on at 8.55 am the next day, 11 October.

The nuclide most widely monitored was I-131, and it is this nuclide which our own studies have concentrated on. Although stack monitors were in place the total quantities released are still somewhat uncertain. However it seems probable that most of the radioactivity escaped in two main peaks, the first occurring in the final hours of 10 October when the maximum number of fuel channels was involved; and the second immediately after water was poured on the pile the next day. Over the rest of the accident release rates were variable but much lower. The results presented here are based on the 2 release patterns shown in figure 1, and show the sensitivity to the assumptions made about the pattern of release. The first considers just the peak periods of release; the second a more distributed pattern of release. The consistency of these assumed release patterns with radiological measurements will be discussed below.

The meteorological situation

The meteorological situation has been discussed by Crabtree (1959), and figure 2 shows the synoptic charts reproduced from his paper. At midday on 10 October a cold front lay to the north-west of Windscale, gradually moving south-east and crossing Windscale shortly after midnight. The release can roughly be divided into two parts; that released ahead of the front, and that released following passage of the front. Ahead of the front geostrophic winds were light and south to south-westerly over Windscale. During the afternoon of 10 October the situation was fairly straightforward, but at dusk an inversion formed in the lowest layers of the atmosphere. Winds at the surface differed from those higher up, with local complications such as shallow katabatic winds. However the release took place from the top of a stack 120 metres high, at about the same height as the inversion. It seems likely that most of the activity released at this time was carried away to the north-east just above the inversion. After passage of the front the wind direction veered to the north, giving rise to a plume of radioactivity southwards down the Cumberland coast. Subsequently the front moved on to the south-east across England, gradually decaying and disappearing. After passage of the front the centre of an anticyclone moved slowly across south-east England to the Continent, with moderate westerly winds to the north over the northern half of England and the North Sea.

Scattered light showers were recorded in north-west England during the period from 18.00 hours on 10th to 06.00 hours on 11 October, and from records at Barrow it is known that most of the fall occurred between 17.00 and 19.00 hours on 10 October, before the first peak in the release. Chamberlain ((1959) and Crabtree(1959) note that no rain fell to the East of the Pennines. It was therefore decided that rain was unlikely to be important, and wash-out in precipitation was ignored, although there may have been some hill-fog and slight drizzle giving enhanced deposition over high land to the south of Windscale.

Trajectory analysis

Unfortunately the detailed meteorological data required for the MESOS program was not available for as long ago as 1957. Trajectories were thereore plotted by hand using the hourly presssure charts in the archives department of the UK Meteorological Office. Inevitably these are somewhat subjective, and there are considerable uncertainties in the neighbourhood of the frontal zone, although this was a weak and decaying system.Various sensitivity studies were conducted to test the stability of the trajectories, and comparison of arrival times at various locations was in fair agreement with trajectory predictions (see figure 3).

The handplotted trajectories spanning the period of the accident are shown in figure 4. The three trajectories in figure 4a are for the early part of the release period on the morning of 10 October, and move slowly off to the north-east across England before swinging south towards continental Europe; the earlier the start of the trajectory the further east the trajectory moves. If much radioactivity had been released during this period then, as shown later, it would be expected that fairly high levels of radioactivity would have been detected to the north-east across England, since there was no inversion during the day to inhibit transport down to the ground. Measurements of radioactivity on grass (Chamberlain and Dunster 1958) indicate relatively low levels of contamination in this direction, and therefore it seems that little could have been released until later on 10 October. However small amounts emitted during this period could have been responsible for the relatively early times at which monitoring stations in Europe registered increased levels of radioactivity. The arrival of radioactivity was registered almost simultaneously in London, Mol and Antwerp at about 18.00 hours on Friday 11 October (see figure 3) and is consistent with the plotted trajectories .

Trajectories plotted, starting on the evening of 10 October, are shown in figure 4b. It can be seen that these do not reach Europe but circle round the anticyclone over south east England before leaving the map area to the east early on 14 October. These trajectories reach London at about the time when maximum levels were recorded there at about 02 hours on 12 October, these levels falling by an order of magnitude by mid-day on 12th. The trajectories seem to leave the map rather too early to the east since radioactivity was not detected on filters in Norway until 15 October; but trajectories become increasingly inaccurate with longer travel times, and material could easily have lingered in the anticyclonic system for longer.

Trajectories of puffs released after passage of the front when the wind veered to the north are shown in figure 4c. These move south along the west coast and loop round before penetrating to southern England and turning back over Wales. Relatively low levels were detected in North Wales and it seems probable that these trajectories wuld not have looped back so far north. The topographical features of Wales and the Pennine chain may also have played some role as barriers channelling the flow.

Integrated atmospheric concentrations

Time-integrated atmospheric concentrations were calculated across northern Europe for both emission patterns shown in figure 2, by modelling the dispersion and depletion along the appropriate trajectories, assuming a basic dry deposition velocity of $3x10^{-3}m.s^{-1}$ and a radioactive half-life of 8.1 days for I-131. Contours of I-131 integrated concentrations resulting from the simple two-peaked release, pattern (a), in figure 2, are shown on the small and large scale maps in figures 5 and 6 respectively. The calculated contours within the smaller map area show the dominance of the second peak in the release, resulting in the plume down the Cumberland coast spreading south over Lancashire. Radiaoctivity from the first peak travels above the inversion without reaching ground level until fumigation occurs the following morning over North Yorkshire. This plume is partly visible in the top right hand corner of figure 5 but is clearly seen on the larger scale map in figure 6. Unfortunately no measurements are available over North Yorkshire to confirm this prediction. The contours in the bottom corner of figure 5 show the effects of the second pass of activity over south Lancashire after circulation round the anticyclone. This recirculation is seen on a larger scale in figure 6 with radiocativity eventually drifting over the North Sea. The simple release pattern gives no exposure over the Continent due to the exclusion of emission in the early phase of the accident. There is also a tendency to

underestimate air concentrations across England.

The MESOS calculations using the release pattern (b) are shown in figures 7 and 8. When early releases are included the agreement between calculations and measurements in Europe improves considerably; the agreement across England is also good. This release pattern produces higher concentrations to the north-east of Windscale. This is consistent with measurements of deposition on grass in the locality of Windscale but conflicts with the fact that no activity was measured at Newcastle. Concentrations near the coast of continental Europe are in reasonable agreement with measurements, but are too low inland. Agreement would have been better if the trajectory initiated at 06.00 on 10 October had been followed further; it can be seen from figure 4 that this was only tracked until just after crossing the coast. Subsequent plotting implies that this trajectory became stationary in the anticyclone until about 03.00 on 13 October, and this would have led to higher concentrations.

Deductions

Comparison of calculated and measured time-integrated concentrations of I-131 are consistent with a release of about 30,000 Ci of I-131 (~ 10^{15}Bq) distributed in time approximately as shown in figure 2b. Apart from a few points over west Yorkshire the model results and measurements are within a factor 3.Bearing in mind the sensitivity of the contamination at the measurement positions to small changes in the trajectories, and the uncertainty in the release pattern, it was felt that the results produced by the MESOS model were quite satisfactory, especially since the meteorological conditions were quite complex.

THE CHERNOBYL ACCIDENT

The analysis undertaken by ApSimon and Wilson and co-workers on the Chernobyl accident has been summarised in a recent paper (Ap Simon, Wilson and Simms1989), and also published in Russian (ApSimon 1990). The work will therefore only be briefly summarised here.

The early radiological data as the Chernobyl release spread across Europe were extremely varied, confused by a wide range of units, types of measurements and measurement techniques. Some countries reported peak activity, others indicated average levels and yet others provided little information at all. Although the MESOS model had

never been intended for application to a real accident situation, it was operational, only requiring meteorological data standardly available from the UK Meteorological Office. Consequently within 3 weeks of the accident we had undertaken an initial analysis of the pattern of dispersal, and deduced approximate source terms which were in good agreement with Soviet estimates subsequently presented in Vienna in August 1986. It was also apparent from observations over S Europe that there had been a continued release into the early days of May, although the reasons for this were not known at the time.

A detailed description of the spread of the release may be found in ApSimon, Wilson and Simms (1989). Sequential maps show the transport of the initial release to Scandinavia, where a weak frontal system led to some areas of high wet deposition. Subsequent material moved south across Poland; and then across central Europe, in a wedge shaped formation to the south of a ridge of high pressure which intruded from the Atlantic. Some material went east across the Soviet Union, and then trajectories turned westwards again across Europe. As a centre of high pressure moved across the north coast of Europe radioactive material circulated northwards behind it, reaching the UK on 2 May, and venting much of the material to the North Sea. During the early days of May trajectories moved down over Greece, and then turned north to give further enhanced levels over Scandinavia again.

As more extensive radiological measurements became available more detailed calculations were undertaken, and used to assess approximate daily releases of I-131, Cs137, and the less volatile Ru-103.These results are given in table 1. It should be noted that these are less than the Soviet estimates, which is to be expected since they exclude the coarser material which was deposited locally close to the reactor. The ratio of Ru to Cs increases in the later phases when temperatures in the core rose, blanketed by the loads of boron, lead, sand and dolomite dropped from above to cover the exposed core. The total I-131 released was two orders of magnitude greater than from the Windscale accident, but for Europe the main problems arose from Cs-137, which was patchily deposited according to rainfall patterns.

The calculated deposition of Cs, averaged over rather large grid squares is shown in figure 9. Based on these averaged levels of deposition the effective whole body collective dose commitment over 50 years was calculated for the European population outside the USSR. This allowed for exposure from both ingestion and external irradiation, and amounted to a total of 2.0. 10^5 man Sv., implying an average dose commitment to these

550 million people of 0.4 mSv; equivalent to about 2 months natural exposure to radiation. The variation about this was very large however, especially in areas where precipitation occurred.

It was concluded that the MESOS model had given a general indication of the pattern of dispersal, and provided a framework for interpretation of measurements. However it is a very simple model, essentially based on surface meteorological data, and its limitations were well recognised- for example in the frontal system over Scandinavia where the wind-field is very 3-dimensional in character, with ascending material following quite different trajectories than is indicated by surface winds.

New modelling tools for use in the event of a nuclear accident.

Since the Chernobyl accident there has been development of dispersion modelling tools in many countries.Some of these are based on trajectory calculations, but others are far more sophisticated requiring considerable computer resources. There are basically 2 approaches. The first integrates the advection-diffusion equations for a regular 3-dimensional grid, the so-called Eulerian approach. The second involves simulating the release as an assembly of particles followed across the domain- the Lagrangian approach. (There are also some hybrid models such as the particle -in cell approach of Lawrence Livermore Laboratory, which considers an assembly of particles but in conjunction with diffusion according to concentration distribution within a grid).

For our own improved model we chose a Monte-Carlo approach. There were several reasons for this; it avoids some numerical problems (numerical diffusion), it is easy to distinguish different sections of the release and revise results for different evolution of the source terms, and it gives some indication of probabilities of material deviating from the mean trajectory path. This model has been incorporated in an overall EC computer package (which is available from ENEA in Italy) for application in nuclear accident situations,together with models for closer-in assessment of contamination, and dose estimation and effectiveness of counter measures. Since it is quite likely that if depositing nuclides are released in an accident, they may become incorporated in frontal systems, we feel that it is especially important to study transport through such systems.
Accordingly studies are now in progress in collaboration with the UK Meteorological Office, together with more refined development of the overall 3DRAW model. Another aspect where technological capabilites can be improved is in the treatment of wet

deposition. In the UK, there is a good network of weather radars giving detailed observations of rain in space and time. Following Chernobyl we used this data to estimate small-scale maps of deposition (see ApSimon, Collier and Simms 1988), and successfully picked out the more contaminated areas. This approach could be very useful for direction of monitoring teams to the most affected spots in any future, and is feasible over increasing areas of Europe as weather radar coverage increases. Thus in any future accident far more sophisticated tools will potentially be available to assist in direction of emergency procedures.

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Crabtree J 1959 The travel and diffusion of the radioactive material emitted during the Windscale accident. Quart J Royal Met Soc. **85**, 362-370

Table 1:- Estimated daily releases of 3 nuclides based on MESOS model results and available measurements

Figure 1. Release patterns postulated for the release in 1957 of I-131 from the Windscale No I pile.

Figure 2. Surface synoptic charts 10-12 October 1957 (from Crabtree 1959)

Figure 3. Variation of air activity at a number of stations 11-13 October 1957(from Crabtree 1959)

Figure 4a Estimated trajectories initiated ahead of the front passing over Windscale

Figure 4b Estimated trajectories starting on evening of 10 October

Figure 4c Estimated trajectories starting on the morning of 11 October

Figure 5.Calculated time-integrated air concentrations (contours) compared with measured values (underlined) over N W England for release pattern (a)

Figure 6.Calculated time-integrated air concentrations (contours) compared with measured values (underlined) over N W Europe for release pattern (a)

Figure 7.Calculated time-integrated air concentrations (contours) compared with measured values (underlined) over N W England for release pattern (b)

Figure 8.Calculated time-integrated air concentrations (contours) compared with measured values (underlined) over N W Europe for release pattern (b)

Figure 9. Calculated deposition of Cs-137 from Chemobyl across Europe

Table 1:- Estimated daily releases of 3 nuclides based on MESOS model results and available measurements

day (from 21h00 to 21h00)	estimated release/(10^{15} Bq day^{-1}) ^{103}Ru	^{137}Cs	^{131}I
25/04 - 26/04	29	15	190
26/04 - 27/04	12	5.6	43
27/04 - 28/04	9.9	4.71	25
28/04 - 29/04	low[a]	low[a]	low[a]
29/04 - 30/04	0.76	0.38	3.8
30/04 - 01/05	3.6	0.76	1.1
01/05 - 02/05	6.3	1.1	4.9
02/05 - 03/05	3.5	1.9	15
03/05 - 04/05	4.9	2.7	19
04/05 - 05/05	5.9	3.0	21
05/05 - 06/05	low[a]	low[a]	low[a]
totals at 06.05	67	35	170

[a] Less than 1.0×10^{14}.

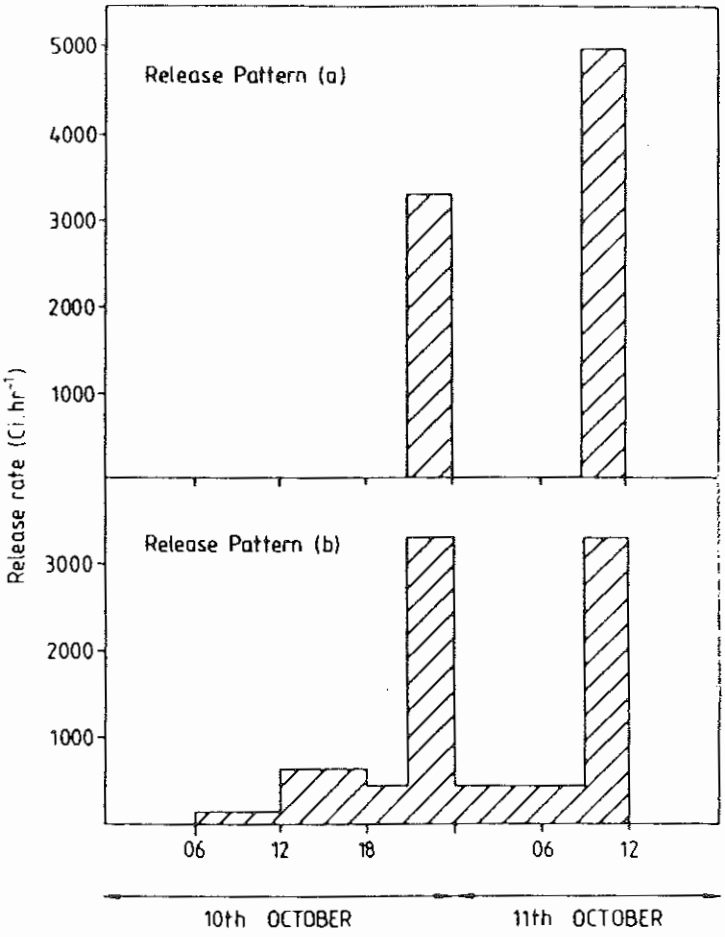
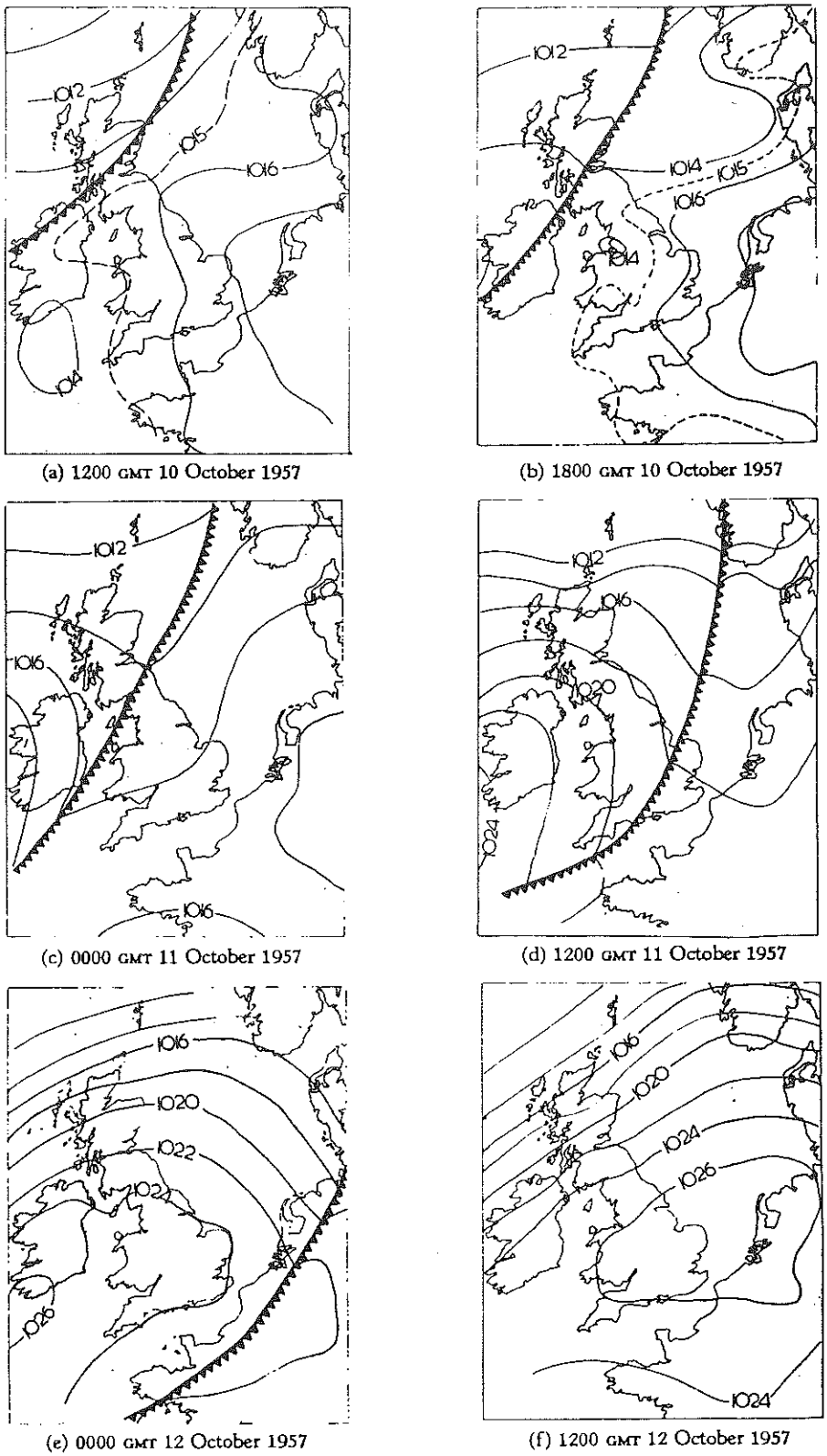


Figure 1. Release patterns postulated for the release in 1957 of I-131 from the Windscale No 1 pile.



(a) 1200 GMT 10 October 1957

(b) 1800 GMT 10 October 1957

(c) 0000 GMT 11 October 1957

(d) 1200 GMT 11 October 1957

(e) 0000 GMT 12 October 1957

(f) 1200 GMT 12 October 1957

Figure 2. Surface synoptic charts 10-12 October 1957 (from Crabtree 1959)

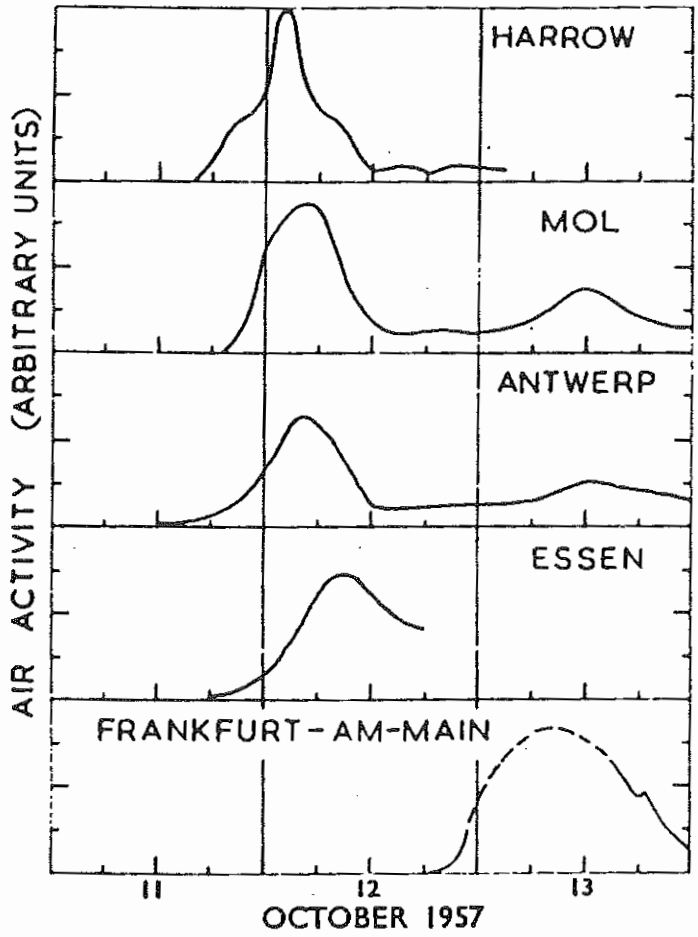


Figure 3. Variation of air activity at a number of stations 11-13 October 1957

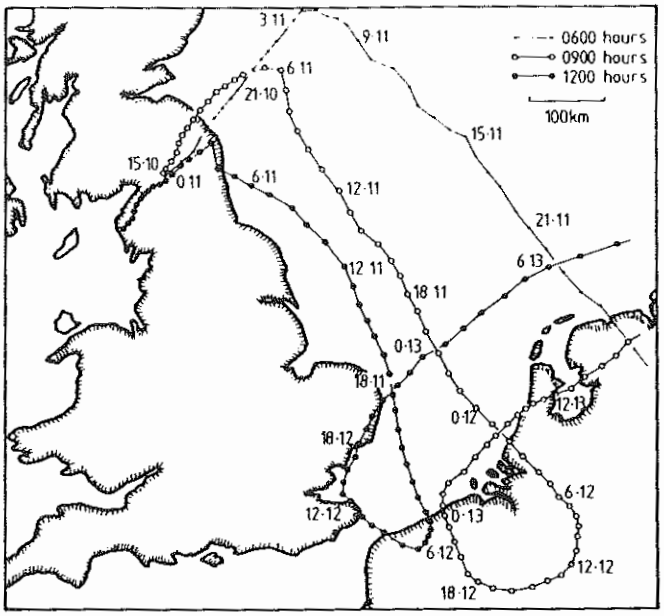


Figure 4a Estimated trajectories initiated ahead of the front passing over Windscale

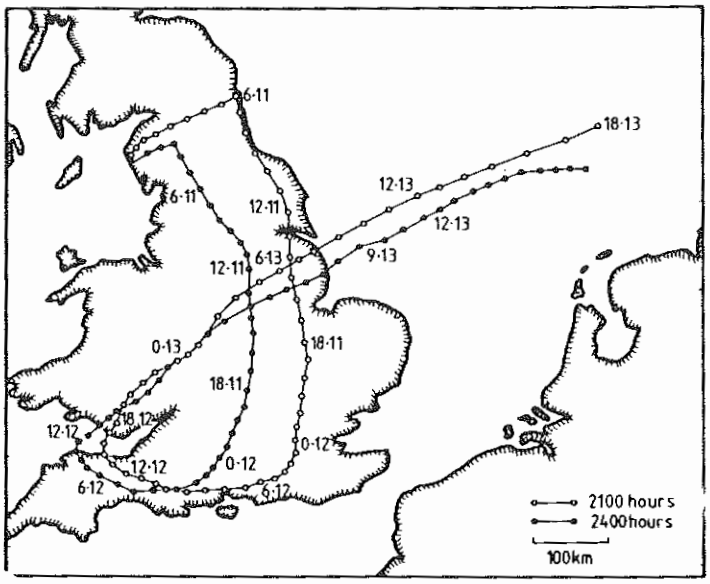


Figure 4b Estimated trajectories starting on evening of 10 October

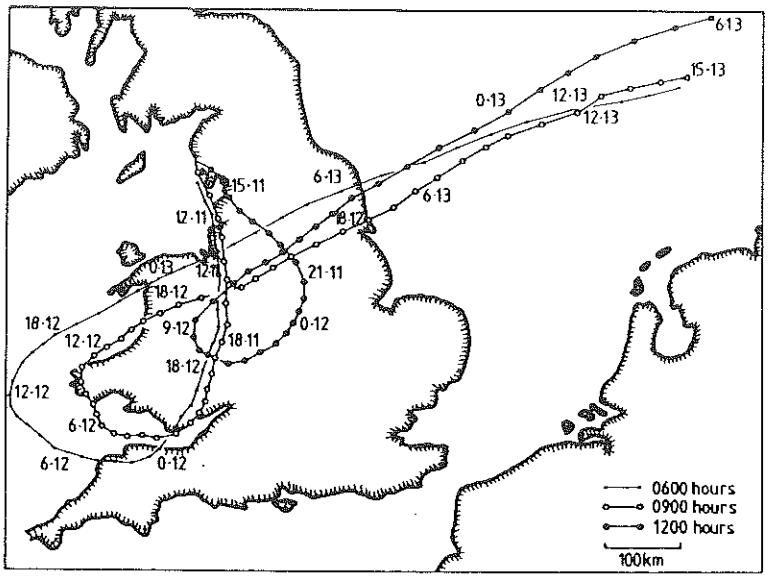


Figure 4c Estimated trajectories starting on the morning of 11 October

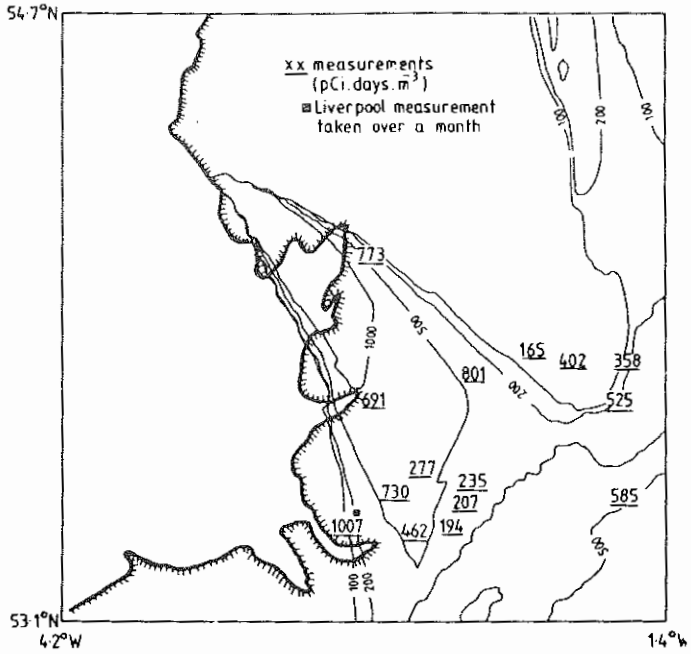


Figure 5.Calculated time-integrated air concentrations (contours) compared with measured values (underlined) over N W England for release pattern (a)

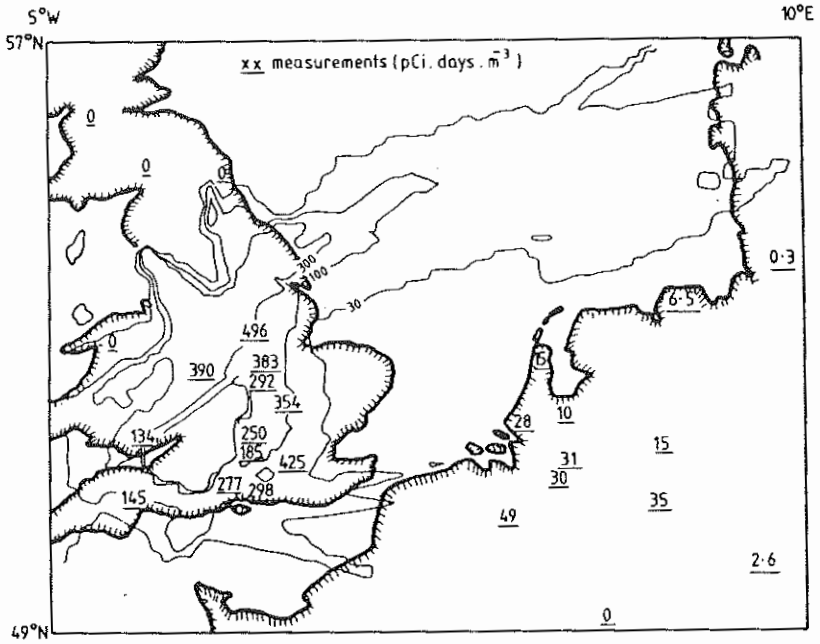


Figure 6.Calculated time-integrated air concentrations (contours) compared with measured values (underlined) over N W Europe for release pattern (a)

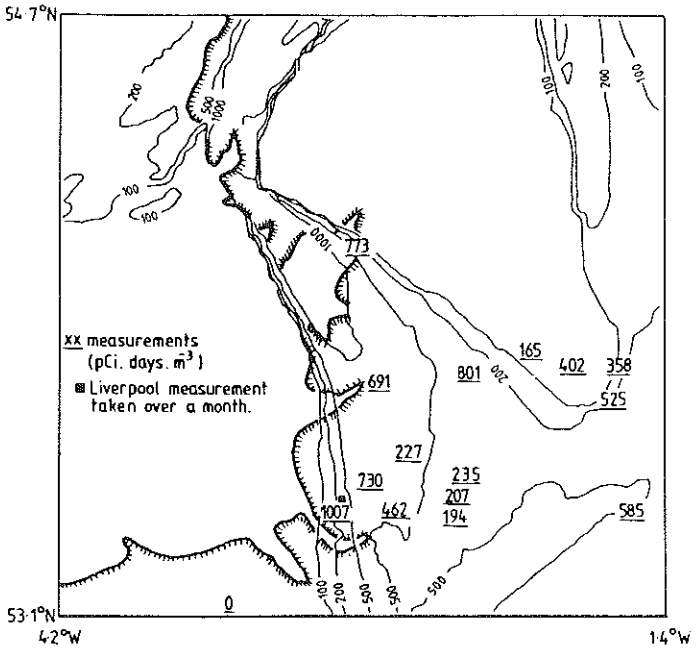


Figure 7.Calculated time-integrated air concentrations (contours) compared with measured values (underlined) over N W England for release pattern (b)

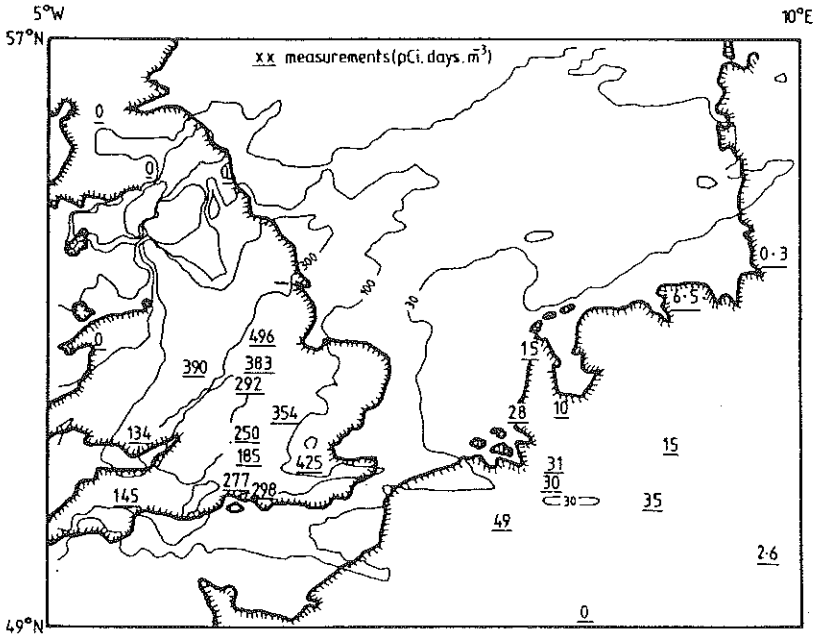


Figure 8.Calculated time-integrated air concentrations (contours) compared with measured values (underlined) over N W Europe for release pattern (b)

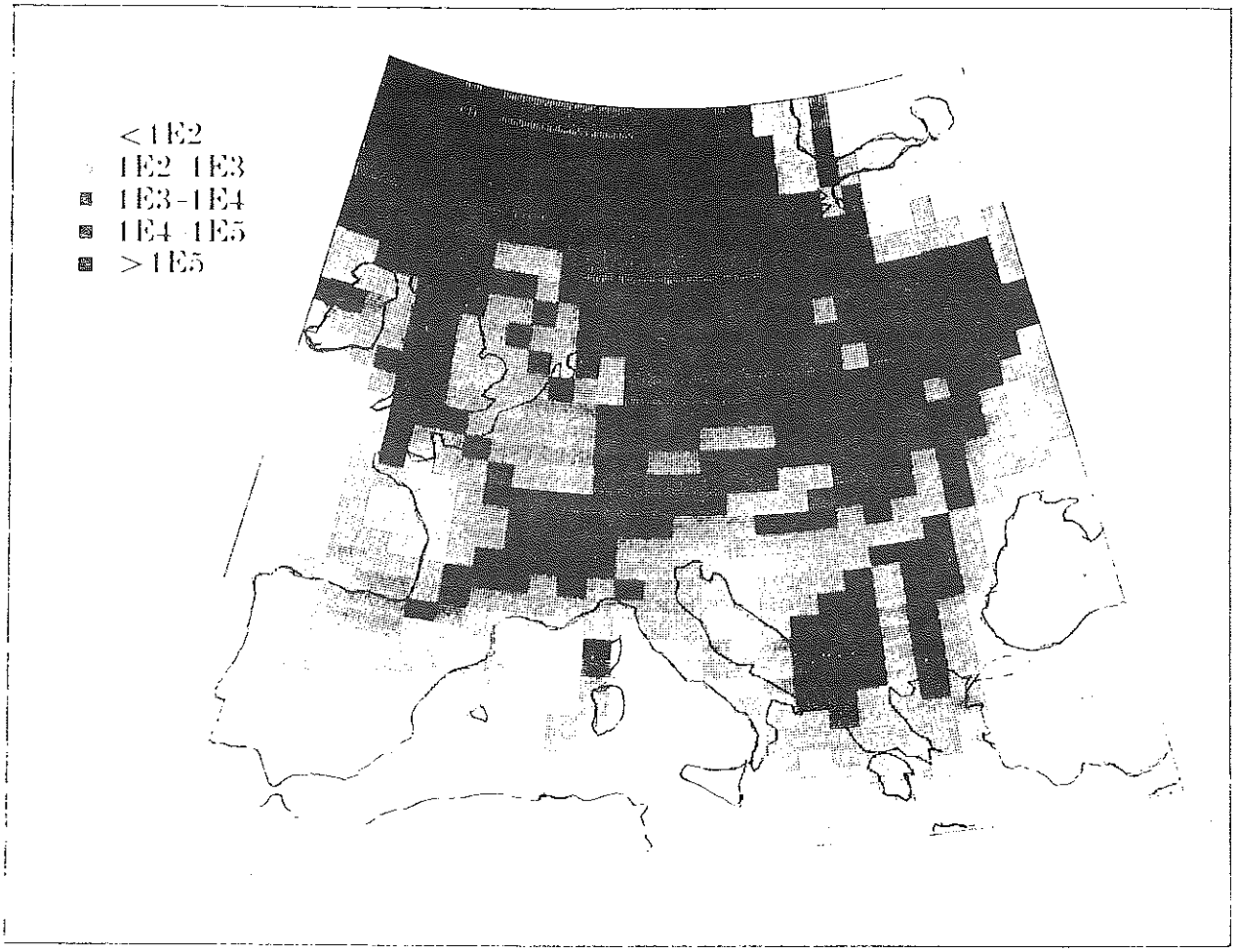


Figure 9. Calculated deposition of Cs-137 from Chernobyl across Europe

Some Common Features of the Formation and Conduct of the Products of Chernobyl Accident in the Environment

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INTRODUCTION

Permanent ingress of the long-lived artificial radionuclides into the environment due to global fall-out and wide use of the nuclear technologies contributes to transformation of the radioactive contamination into a new ecological factor of the environment. These radifonuclides are included into the natural biological cycle, penetrate into the man's organism over various biological chains, and are accumulated therein, causing a permanent radiation effect.

The relative annual effective equivalent irradiation dose resulting from global fall-out reached the maximum value of about 7% of the natural background in the early sixties, then it decreased to about 0.8% of the natural background in 1980, and now it equals 0.02 mSv.y ^{1} in absolute value.

On the contrary, the irradiation dose resulting from nuclear power engineering increased from 0.001% of the natural background in 1965 to 0.035% in 1980 and equals on average 0.001 $mSv.y^{-1}$ /1/. This is illustrated by the chart shown in Fig. 1.

Thus, the experience gained in the course of many years of service of the atomic power plants and enterprises of the nuclear power cycle under normal operating conditions proves that the radioactive contamination of the environment in the areas where these plants are located has no substantial effect on the irradiation doses of people nor of populations of organisms.

Thus, according to the data given in publication /2/, the individual effective equivalent dose of irradiation of the population living in the 35-km zone of the Chernobyl atomic power plant resulting from operation of the plant amounted to 0.0004 $mSv.y^{-1}$, the natural background being equal to 2.7 $mSv.y^{-1}$.

However, the atomic power plants and other installations of the nuclear power cycle which are radioecologically safe under normal operating conditions may become sources of large-scale contamination of the ecological systems in emergency situations associated with the escape of radioactivity into the environment, i.e. in case of a radiation accident.

1. Comparative Characteristics of Grave Radiation Accidents

Grave radiation accidents include first of all the accident at the plant of the Ministry of Defence on the Southern Urals (September 29, 1957), the accident at the plant in Windscale (October 8, 1957), and the accident at the Chernobyl atomic power plant (April 26, 1986). In the two latter cases, the accidents took place in industrial reactors, though the technological conditions of reactor operation at the time of the accidents and also nuclear fuel compositions were quite different. The accident on the Southern Urals occurred as a result of the explosion of the reservoir containing 70 to 80 tons of the high-activity liquid waste.

The cause of these accidents were analyzed in detail in a number of works /3,4,5/.

As a result of the accidents, a considerable amount of radioactivity got into the environment. The comparative evaluation of the total initial activity at the time of the accident and the share of the activity that escaped into the environment is given in Table 1 /3,6,7/.

Table 1

Accident	Total activity at time of accident, MCi	Activity which escaped into environment	
		MCi	%
Southern Urals	20	2	10
Windscale		0.038	
Chernobyl	1295 * (2015**)	50 ***	4 ***

* Disregarding inert gases and activation Np-239
** With regard for Np-239
*** Fission products ± 50%

Thus, with respect to the escape of activity into the environment, the accidents in the Southern Urals and at the Chernobyl atomic power plant are the largest accident that occurred throughout the use of the nuclear technologies. From the point of view of the radioecological effects, the radionuclide composition of the release into the environment is extremely important. These data in MCi are given in Table 2 /6,7,9,10/ and shown in Fig. 2 (Chernobyl accident).

Table 2

Radionuclide	Southern Urals	Windscale	Chernobyl
Np-239	-	-	29
Mo-99	-	-	3
Te-132	-	0.012	5.3
I-131	-	0.02	10 to 17
Ba-140	-	-	4.3
Ce-141	-	0.00019	2.8
Ru-103	-	-	3.2
Sr-89	-	0.00008	2.2
Zr-95	0.5	-	3.8
Nb-95			3.8
Ce-144	1.32	-	2.4
Pr-144		-	
Ru-106	0.074	-	1.6
Cs-134	-	-	0.5 to 1
Sr-90	0.11	0.000002	0.22
Cs-137	0.00072	0.0018	1 to 2
Pu-238	-	-	0.0008
Pu-239	-	-	0.0007
Pu-240	-	-	0.0001
Pu-241	-	-	0.14
Cm-242	-	-	0.021

It shall be mentioned that the data given in Tables 1 and 2 concerning the accident at the Chernobyl atomic power plant have been corrected according to the recent publications in the Soviet press /9/. By their evaluation, about 96% of the reactor full fuel loading is located in the reactor core and in the rooms of the 4th power unit. The estimated total release of activity (including the inert gases) equals 100 MCi ($3.7.10^{18}$ Bq), release of I-131 - 10 to 17 MCi (3.7 to 6.3) x 10^{17} Bq, release of Cs-137 - 2 MCi ($7.4.10^{16}$ Bq).

The gravity of the radioecological effects of the accidents was substantially different both from the point of view of the escape into the environment of the inert gases, radioiodine and long-lived products and with respect to the season of the year. These data are given in Table 3.

Table 3

Accident	Season of year	Activity of inert gases MCi	% of activity penetrating into environment (disregarding inert gases)			
			Sr-90	Cs-137	I-131	Pu
Southern Urals	Autumn	-	2.7	0.036	-	Traces
Windscale	Autumn	0.1	0.006	5	60	-
Chernobyl	Spring	50	0.5	4	20 to 34	0.3

Thus, the radioecological effects of the accident in the Southern Urals were caused mainly by the release of long-lived Sr-90 and Cs-137, the accident in Windscale - by the release of iodine and inert gases, and for the accident at the Chernobyl atomic power plant - by all these factors. The radioecological effects of the accident at the Chernobyl atomic power plant were aggravated by the season of the year when the accident took place. In this season of the year, the farming operations were coming to an end, the cattle was driven to the pastures, the natural and agricultural plants were at the beginning of the growing period but their vegetative mass was already sufficient.

The radioecological effects of the radiation accidents are closely connected with the area of the contamination zone which depends primarily on the height of the accident release and time of formation of its ground track. These data are given in Table 4.

Table 4

Characteristics of release	Accident		
	Southern Urals	Windscale	Chernobyl
Height of release, km	1	0.07	1 to 7
Time of formation of main track, hour	11	50	300
Power of release, Ci.h-1	"Salvo"	660	6700 to 40000
Accident release track area km2	15000 *	520 **	200000 *** 280000 ****
Number of evacuated population (thousand people)	10	–	115 *****

* With respect to 0.1 Ci.km^{-2} - double value of density of global Sr-90

** Area where limitations were introduced and content of I-131 in milk was checked

*** In the first days, with respect to isodose 0.2 mR.h^{-1}

**** Within the limits of Cs-137 fall-out density over 5 Ci.km^{-2}

***** In accordance with the decision taken in 1990, evacuation of the population will be continued

Characteristics of release	Accident		
	Southern Urals	Windscale	Chernobyl
Average doses for population living on contaminated territories, rem	1.2 *	0.02 **	5.3 ***
Alienated territories, km2	1000 ****	–	3100 *****

* The average effective equivalent dose of irradiation for non-evacuated population within 25 years

** The effective equivalent dose of irradiation (maximum dose for the thyroid gland : 16 rem for children and 9.5 rem for the adults)

*** The dose of the internal and external irradiation for the population living on the contaminated territories in 1988 (the second year after the accident)

**** By the present time - about 190 km^2

***** By the present time, the territories shall be equal to about 4800 km^2

It follows from Table 4 that the development of the emergency situations in time was quite different for all three accidents.

The accident in the Southern Urals was characterized by the explosive destruction of the technological equipment and "salvo" (instantaneous) release of radioactivity into the environment to a considerable height. The radioactive substance contained in the explosion cloud was deposited within a comparatively short period of time. This ensured formation of the track zone in the "classic" variant : of the extended form, with maximum density of the radioactive contamination on the track axis in the vicinity of the source of release of 15.10^3 $Ci.km^{-2}$ (4.10^3 $Ci.km^{-2}$ with respect to Sr-90), with monotonous decrease of the density of the radioactive contamination along the axis towards the periphery, and with sharp decrease of the density of contamination in directions perpendicular to the track axis /6/.

The accident in Windscale was more prolonged (because of the attemps at its localization) and from the radioecological point of view it consisted in the outflow of the gas component of the release through the ventilation pipe. The release was limited though the reactor was not provided with a containment.
The release into the atmosphere included about 100% of the inert gases, 12% or I-131 content in the reactor core, 7,5% of Te and about 0.03% of Sr /11/. The average level of air contamination in the course of the release was approximately equal to 0.2 $Bq.l^{-1}$ (the maximum level of contamination was about 1.1 $Bq.l^{-1}$ at a distance of about 3 km from the reactor). The contamination was of a "spot" pattern due to the effect of the atmospheric processes (Figs. 3 and 4).

The accident at the Chernobyl atomic power plant combined the "salvo" release (about 25%) of radioactivity to a height of approximately 7 km with explosive destruction of the reactor core and subsequent continuous outflow of the radioactive stream (about 75%) rising to a height of up to 1.5 km. For the estimation of the scale of this largest radiation accident, suffice it to say that only 5% of the fuel (about 7 tons) was involved in the accident in Windscale whereas approximately the same amount of the fuel was discharged from the reactor during the accident at the Chernobyl atomic power plant.

Recently, the scales and radioecological effects of the accidents in the USSR were revised and this accident was referred to the category of the ecological catastrophy /12/. In the course of development of the accident, 10 to 20% of the volatile radionuclides - fission products (I, Cs, Te, Ru) and 3 to 6% of the non-volatile radionuclides (Ce, Ba, Zr, Pu, Sr and others) were released into the environment.

The only thing which is common for the accidents at the Chernobyl atomic power plant and in Windscale is that both accidents took place in reactors and were similar in the first phase of their radioecological effects (escape into the environment of the inert gases and radioiodine with activity of 10 to 17 MCi and 0.02 MCi, respectively). However, the causes of the accidents and, which is most important, their development which determined the gravity of the radioecological effects were quite different.

In Windscale, two days passed from the moment of the error in the control of liberation of Wigner energy to the fire in the core. During this time period the personnel tried to correct the situation and managed to partially localize the accident. In this respect, the accident in Windscale is similar to the accident on Three Mile Island and sharply differs from the accident at the Chernobyl atomic power plant which is characterized first of all by the explosive initiation of the accident resulting in destruction of the reactor core and technological equipment of the power unit and by the long-time localization of the accident releases and by the considerable liberation of energy from the fuel remaining in the destroyed core. This determins the gravity of the radioecological effects of the accident at the Chernobyl atomic power plant - the greatest radiation accident since th time nuclear technologies are used.

/12/ Statement of Supreme Soviet of the USSR

Despite the differences in the effects of the three accidents which are under consideration, there are some common features. As it has been already mentioned, the accidents at the Chernobyl atomic power plant and in Windscale are characterized by the similar first phases of the accident effects - contamination of the environment with the inert gases and radioiodine. During the accident in the Southern Urals, like in the accident at the Chernobyl atomic power plant, the environment was contaminated with a considerable number of the long-lived radionuclides.

2. Some Common Peculiarities of Spatial-Nuclide Structure of Contamination Resulting from Accident at Chernobyl Atomic Power Plant

The radiation situation in the contamination zone, forecast of its variation in time, and gravity of the radioecological effects of the accident are connected in many respects with the pecularities of formation of the spatial-nuclide structure of contamination in the course of development of the accident.

Naturally, these pecularities will be primarily determined by the process of production of the released radioactive materials in the course of the accident, i.e. by the type and operation of the equipment as well as by the characteristics of the release, radionuclide composition and its peculiar features, physcial and chemical processess taking place in the course of development of the emergency situation, and the like, i.e. by the function of the source in a broad sense of this word.

Further on, formation of the spatial-nuclide structure of radioactive contamination will be affected by the mechanisms of transfer and deposition of the materials discharged during the accident which are primarily determined by the dispersal and phase of these materials as well as by the weather conditions in the area of the accident and in the remote areas in the direction of propagation of the contaminated air masses.

During the accident at the Chernobyl atomic power plant, formation of the release into the environment was determined by two processes :

- dispersion of the nuclear fuel as a result of the explosion of the reactor core on April 36, 1986 which led to the formation of an aerosol consisting of fuel matrix particles containing mainly the entire spectrum of radionuclides - fission products;

- outflow of the vapours (or gases) of the fragmentation radionuclides from the fuel remaining in the reactor core after is was heated to 1500 - 2000°C in the course of burning of the core graphite on April 27, 1986 to May 5, 1986 which resulted in formation of condensation aerosols which are mainly characterized by their monoradionuclide composition or a weakly developed nomenclature of radionuclides.

Along with the products of combustion, the release might contain small-divided fuel particles dispersed in the graphite (during the explosion) and the fission products sorbed by the graphite at the first stage. It was noticed /13/ that many of the aerosol particles were "hot" and differed from other radioactive particles by high individual total ($\alpha + \beta$) - activity exceeding 10 Bq (about 3.10^{-10} Ci) *.

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* The term "hot particle" shall be explained. From the point of view of the radiobiologists, it is a particle which produces the dose to the critical organ of the tissues of at least 1000 rem per year ("Biologcial effects of inhaled radionuclides", ICRP Publication, No.31, 1984). From the point of view accepted in the given publication, the "hot particle" is a particle of the micrometric (or submicrometric) size whose activity (absolute) exceeds by one or several orders the average activity of the given aerosol particles (see Bykhovskkiy A.V., Zarayev O.M.. Hot aerosol particles at technical use of atomic energy, 1974, Moscow, "Atomizdat" publishing house). It may be also a particle with high specific activity of about 310^{11} Bq.g^{-1} and consisting of non-volatile radioactive substances that are slightly soluble in water.

It may be also assumed that the complex processes of dispersion of the nuclear fuel, oxidation and reduction processes, phase transitions, sorption and desorption processes, processes of recrystallization of the nuclear fuel, and the like could result in a redistribution of the fission products in the nuclear fuel matrix.

A certain peculiarity of the characteristics of the released products could also be caused by the fact that fuel burn-up whose average value was 10.300 $\frac{MW.d}{kg\,U}$ might be different in various parts of the core of reactor P BMK-1000 in virtue of its technologinal peculiarities.

Thus, a mixture of the fuel matrix aerosol particles termed "fuel particles" and condensation aerosol particles termed "condensation particles" was released into the environment.

It was stated in the report of the Soviet experts in IAEA /8/ that owing to these processes the radionuclide composition of the release which is at all stages close to the composition of the irradiated fuel with a burn-up of 8 to 10 $\frac{MW.d}{kg\,U}$ was enriched with the volatile components (iodine, tellurium, caesium and partially ruthenium). In other words, the formation of the nuclide structure of contamination might be affected in the microscale by primary fractionation. As it is known, the process of fractionation of various fragmentation products leads to considerable deviation of the ratio of the activities of two radionuclides fractionating in a different manner from the theoretical values. Actually, if the fractionation standard is represented by the ratio of activity of the i-th radionuclide to the activity of refractory zirconium-95 (A_i / A_{95} = C_{N1}) or cerium-144 (A_i/ A_{144} = C_{N2}) by the time of the accident, then according to publications /4,12/ these ratios will correspond to the data given in Table 5.

Thus, if the fractionation coefficient equals :

$$F = \left(A_i / A_{95} \right)_{exp} \cdot \frac{1}{C_{N1}} > 1$$

this testifies to the prevailing release of the i-th radionuclide in comparsion with the refractory substances since the zirconium (and cerium) oxides are refractory.

Table 5

Radionuclide	I-131	Cs-137	Cs-134	Sr-90	Ru-103	Ru-106	Pu-239
CN1	0.66	0.66	0.03	0.05	1	0.46	$1.8.10^{-4}$
CN2	0.96	0.09	0.044	0.07	1.44	0.67	$2.6.10^{-4}$

Values of f = 1 ± 0.5 for all radionuclides (or non-volatile radionuclides) means that the radionuclides were transferred by the fuel particles.

By way of example, Fig. 5 illustrates the behaviour of $f_{131.95}$ and $f_{137.95}$ in the stream over the atomic power station. These data prove that enrichment with iodine and caesium was continued to greater or lesser extent practically in the course of the entire period of the accident release.

The complex nature of the processes of nuclear fuel dispersion and absorption of the radionuclides (or their oxides) possessing various degrees of volatility on the aerosol particles, has promoted their fractionation in the dispersed phase.

The results of the investigations /13,14/ show that distribution of the aerosols by size was mainly of the bimodal nature :

- fraction of the fine-divided aerosol with an active median diameter of 0.1 to 0.2 μ /3/;

- fraction of the coarse-divided aerosol with the active median diameter of 25 to 100 μ.

The results of our measurements show /13/ that -active nuclides are contained mainly in the coarse-divided aerosol and ß -active nuclides are contained in both fractions is evidently made up of the fuel matrix aerosol particles while the small-divided and fine-divided fraction includes mainly the condensation aerosol particles. The analysis of the content of the gamma-radiators in the fractions shows that the fine-divided fraction of the aerosols of ß -active nuclides is represented primarily by Ru-103, 106 (up to 90% of the content) while the coarse-divided fraction contains radionuclides Ce-144, 144 (up to 25%), (Zr+Nb)-95 (up to 45% of the content), Ru-103, 106 (up to 20%) and Cs-134, 137 (up to 8%). The isotopic composition of the coarse-divided fraction of α-radiators was determined mainly by Cm-242 (up to 85%), Pu-239 (up to 4%), Pu-238 (up to 3%) and Cm-244 (up to 3%).

As it is known (15/, fractionation in the dispersed phase has resulted in the fact that the fine-divided fraction of the condensation aerosol (and partially of the fuel matrix aerosol) was prevailing in the upper layers of the release cone and spreading cloud.

Three branches of the radioactive track of the accident release may be distinguished on the contaminated territory in the microscale : northern, southern and western branches whose formation was mainly completed on May 4 - 5, 1986 (Fig. 6). The map projection of the radioactive contmaination of the ground surface as a whole coincides with the map projection of air contamination /16/ and is indicative of the generic link between these two contaminations. During transport of the radionuclides fractionated in the dispersed phase, there occurred "potential" fractionation associated with different rates of dry deposition of the fuel matrix heavy particles and condensation aerosols, physical form of the inactive carrier, and the like.

The investigations of the isotopic composition in various branches of the radioactive track conducted by the authors of the present publication have shown that fractionation of the radionuclides is traced at distances over 15 to 20 km from the destroyed reactor. The contamination within this area is represented mainly by the fuel matrix aerosol which resulted in maximum contamination of these territories with the isotopes of plutonium and strontium. The contamination of the air with the two types of the aerosols explains abnormal fractionation of the fragmentation radionuclides on the soil cover : with the increase of the distance to the place of accident, the contribution of the condensation aerosols into general contamination grows, with simultaneous increase of the coefficients of fractionation of several isotopes, in particular, fractionation of radiocaesium.

Practically, all contaminations at distances over 100 to 150 km refer to a greatly fractionated kind of contamination. The contribution of the volatile radionuclides into the total radioactivity grows with the decrease of the exposure dose rate in the radioactive track (Fig. 6).

By virtue of these pecularities in the development of the accident, the spatial structure of any branch of the radioactive track produced by the accident at the Chernobyl atomic power plant differs from the "classic" structure formed as a result of the "salvo" release as it was on the Southern Urals.

A distinguishing feature of the contamination of the environment during the accident at the Chernobyl atomic power plant was that the contaminated areas were in the form of "spots" of various scales. Making no analysis of the causes of the mesoscale and macroscale "spots" whose formation was affected, apart from the fractionation processes, by the landscape and weather conditions, we shall state that the microstructure of contamination was to a great extent determined by the presence of "hot particles" in the fall-out. This peculiarity of the release during the accident at the Chernobyl atomic power plant is discussed below.

Nevertheless, on the southern branch of the accident release track characterized by negligible fractionation /17/, the change of the density of contamination with increasing the distance to the source is more regular. By way of example, Fig. 7 shows the data (as of 1986) relating to the measurement of the activity density of Cs-137 and ΣPu along the Pripyat - Kiev section. The pattern of contamination on the territory of Kiev was very complicated. Our measurements show that the activity density distribution was greatly affected by the extremely

broken relief of the city, by the microclimate depending on the temperature fields of the city, a great number of the natural water basins, forest parts, and the like. Thus, the range of change of the activity density of plutonium in 1988 varied from approximately 37 Bq/m^2 to 370 Bq/m^{2*} at an average value of about 100 Bq/m^2, caesium-137 - from 5600 Bq/m^2 to 25.10^4 Bq/m^2 at an average value of about 16.10^3 Bq/m^2. The contamination was mainly of the "fuel" (or close to it) type. The nature of the contamination was determined proceeding from the following condition : Ce-144/Cs-137 > 9 to 11 - "fuel" type, Ce-144/Cs-137 > 8 - "fractionated" type or intermediate type. The presence of several types of contamination resulted in that the correlations (Γij) between various radionuclides in the samples taken from various types were substantially different

For all types of contamination only $f_{134,137} = 1$. Fig. 8 shows the diagram of Γij for several radionuclides plotted by the results of measurements on the intermediate and fractionated types of contamination (dated April 26, 1986). Nevertheless, we managed to reveal the correlation between the content of plutonium and content of cerium-144 or ruthenium for each type of contamination.

Thus, according to publication /188/, the change of the plutonium content in the range of 0.005 to 15 $Ci.km^{-2}$ is described with an error of not more than 30% by the equation :

$$\log [A_{Pu}] = 0.88 \log [A_{Ru}] + \text{const.}$$

In the 60-km zone, the estimation given in publication /19/ seems to be more confident :

$$A_{Pu} \approx 6.10^{-4} A_{Ce-144}.$$

The complex radionuclide composition of the aerosol fall-out results in that the decrease of the dose rate (or activity) in the chernobyl

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* Subsequently such "spots" of plutonium were removed from the territory of the city.

fall-out zone is note approximated by a simple relationship of the C.t-1.2* type. This is characteristic of all three accidents considered above, though the reasons for it are different.

If we introduce the notion of the "apparent" decay-time for a certain time moment, it will be equal according to our estimation for t = 600 days :

for the "fuel" type of contamination - 180 ± 30 days;
for the "caesium-fractionated" type of contamination - 1020 ± 200 days.

A peculiar feature of the Chernobyl fall-outs consists in that they include, apart from Pu-238, 239 and 242, pure ß-radiator Pu-241 (E_{β}max 21 KeV, $T_{1/2}$ = 15, 16 years). Despite the fact that by the time of the accident its content in the reactor was about 0.1 of the content of Pu-240 (about 15 kg), its share equals about 98% of plutonium activity. In the results of ß-decay it transforms into Am-241 whose radiotoxicity is close to that of Pu-239.

In conclusion, we shall state that the results of our analyses of the soil cover samples show that the activity density of the long-lived radionuclides may be arranged in the following row, irrespective of the distance to the damaged reactor :

Cs-137 > Cs-134 > Sr-90 > Pu-239

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* Nevertheless, the average effective energy of γ-radiation of the accident products (0.6 to 0.8 MeV) is close to the energy of radiation of fresh products of the atomic blast.

3. "Hot Particles" in Accident Release at Chernobyl Atomic Power Plant

As it has been already stated, the peculiar feature of the accident at the Chernobyl atomic power plant consisted in the presence of "hot particles" in the aerosol composition. Proceeding from the results of the gamma-spectrometric analysis and determination of the content of strontium-90 and -active radionuclides, the "hot particles" were conventionally divided into two groups :

- fuel (or quasifuel) particles;
- condensation particles.

The first group included the particles containing the radionuclides (all of them or most of them) in the proportions corresponding to the average fuel burn-up of 8 to 10 $\frac{MW.d}{kg\ U}$.

We investigated the particles of this group measuring 20 to 100 μ which turned to be either fragments of the uranium oxide or the black amorphous conglomerates which were easily disintegrated into smaller fragments at a slight mechanical effect. Some of the particles are depleted with radiocaesium (as compared with its content in the fuel) which is evidently indicative of long exposure to high temperatures sufficient for diffusion and evaporation of the low-boiling elements. Other particles, on the contrary, are enriched with radionuclides Zr, Ce, Cs, etc.. In accordance with our measurement results, the content of plutonium in this type of the particles ranges from 0.02 to 6 Bq/particle (about 60% of Pu-239 and about 40% of Pu-238).

Some particles contain Cm-242 (its content is about $\lesssim$ 4%).** The total activity of "hot particles" of this size equals according to our measurements 250 to 1000 Bq/particle at an average activity of about 350 Bq/particle.

The smaller-divided particles of this group ($\lesssim$ 10 μ) have an even higher specific activity and are enriched with the volatile radionuclides (quasifuel composition). Their content in the soil cover depends on the azimuth and distance to the damaged reactor and equals 20 to 60% for the 30-km zone. The content of the radionuclides in these particles varies within relatively wide limits. According to our estimation, the average ß-activity equals 150 to 200 Bq/particle and the average α-activity amounts to approximately 0.5 Bq/particle.

The "hot particles" of the fuel group are present to a greater or lesser extent in all types of contamination in the near (up to 80 km) and intermediate (up to 150 km) zones of the track. Their presence was traced also in the far zone of the track /20, 21, 22/. Our estimation of the contribution of the "hard hot particles" (20 to 100 μ) to the irradiation dose shows that on the fuel type of contamination in March 1988 they could form 15 to 20% of the reference level for the dose rate*, i.e. 0.03 to 0.05 mR/h, and on the intermediate type of contamination - 5 to 10%.

The "hot particles" of the condensation group are more seldom found even on the highly-fractionated type of contamination. By our opinion their content on the fuel type of contamination does not exceed 5 - 7% of the total content of the "hot particles". Belonging to this group of the particles are first of all the "ruthenium" and "caesium" particles, i;e. particles containing the monoelements. Such particles, mainly "ruthenium" particles, were detected in the far zone of the track as well /23, 24/. The analysis of the inactive carrier of such particles has shown /25/ that they include ferrum, nickel, molybdenum, technetium, rhodium, palladium and do not include fixed oxygen. As a rule, the size of such particles is not over 1 μ.

The activity of these particles is extremely high. We have detected "ruthenium" particles with the activity of up to 6.10^5 Bq and "caesium" particles with the activity of up to 10^4 Bq. The content of plutonium in the "ruthenium" particles was not over 0.04 Bq and the content of strontium was not over 70 Bq. In the "caesium" particles, the content of plutonium and strontium was 0.004 and 10 Bq, respectively. It is worthwhile mentioning that ruthenium is contained in the particles evidently in the metal phase /23/.

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* The reference level of the exposure dose rate is 0.2 mR/h.
** December 1987

The condensation type of the "hot particles" is closely connected with the subgroup of the so-called "rarity hot particles" whose radionuclide composition is determined by ^{95}Zr + ^{103}, ^{106}Ru or by ^{141}, ^{144}Ce + ^{95}Zr. These particles were found also in the far zone of the accident release, in particular, in Finland /26/. Some particles of this subgroup which were at our disposal represented (by the data of the microprobe analysis) evidently dolomite with absorbed radionuclides, which was the result of the introduction of various fire extinguishers into the burning reactor.

The contribution of the condensation "hot particles" to the dose rate is small but these particles together with the small-divided fuel "hot particles" are referred to the fraction of the inhaled aerosols ($\leq 10\mu$) so that their radiation hazard in the first post-accident year was high. The "hot particles" are easily transportable which shall be primarily taken into account when estimating the biological hazard of these particles.

The investigation of the solubility of aerosols sampled in the area of the Chernobyl atomic power plant shows /14/ that some 25 to 50% of caesium in the atmosphere was in the non-soluble form, i.e. caesium was contained in the particles of the fuel matrix aerosol. This content complies with the values of the fractionation coefficient of 4 to 2 actually observed in the near zone (see Fig. 6).

Low transportation ability of the alpha-radiators /8/ confirms the assumption on their strong links with the fuel matrix. However, it is stated /27/ that some samples of the aerosols taken above the 4th power unit in May, 1986 were highly enriched with plutonium which testifies to the fact that conditions providing for production of the volatile forms of plutonium exist during burning and mixing of the fuel. Thus, the fragmentation radionuclides were contained in the atmospheric aersol in the soluble and non-soluble forms. The ratio betwenn these forms is evidently determined by the respective fragmentation coefficients. This effect should be most brightly displayed by the isotopes of caesium since all its compounds are absolutely soluble. The radionuclides contained in the particles of the fuel matrix aerosol are evidently not transportable. The particles of the condensation aerosol consisting of the metals, oxides or iodides of various isotopes of fragmentation and corrosion elements possess various degrees of transportation ability.

Thus, the peculiarities of development of the accident at the Chernobyl atomic power plant called for determination of the spatial-nuclide structure of the contamination zone by :

- spectrum of the fission and activation products of the nuclear fuel by the time of the accident;
- relative volatility of the radionuclides or their oxides at t>1500°C;
- amount of the released radioactivity;
- dispersal and relative content of the radionuclide in various dispersed phases;
- presence of "hot particles" in the accident release;
- meteorological conditions of particle transportation and type of particle deposition on the soil cover.

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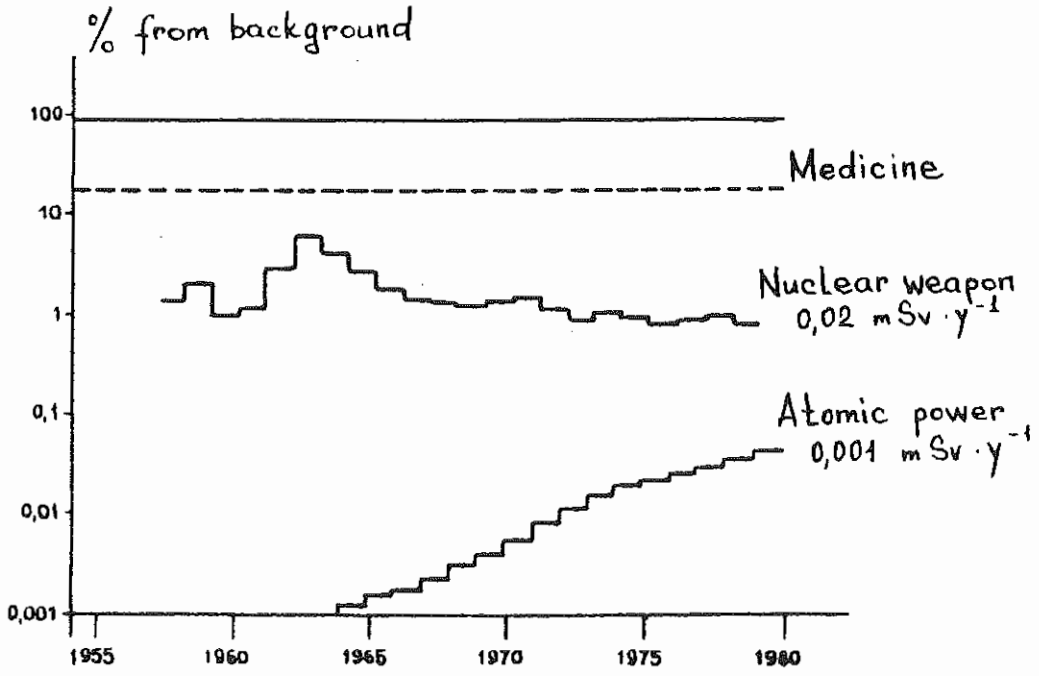

Fig.1 Average effective equivalent dose from power station, weapons fallout and medicine (in % of natural environmental dose)

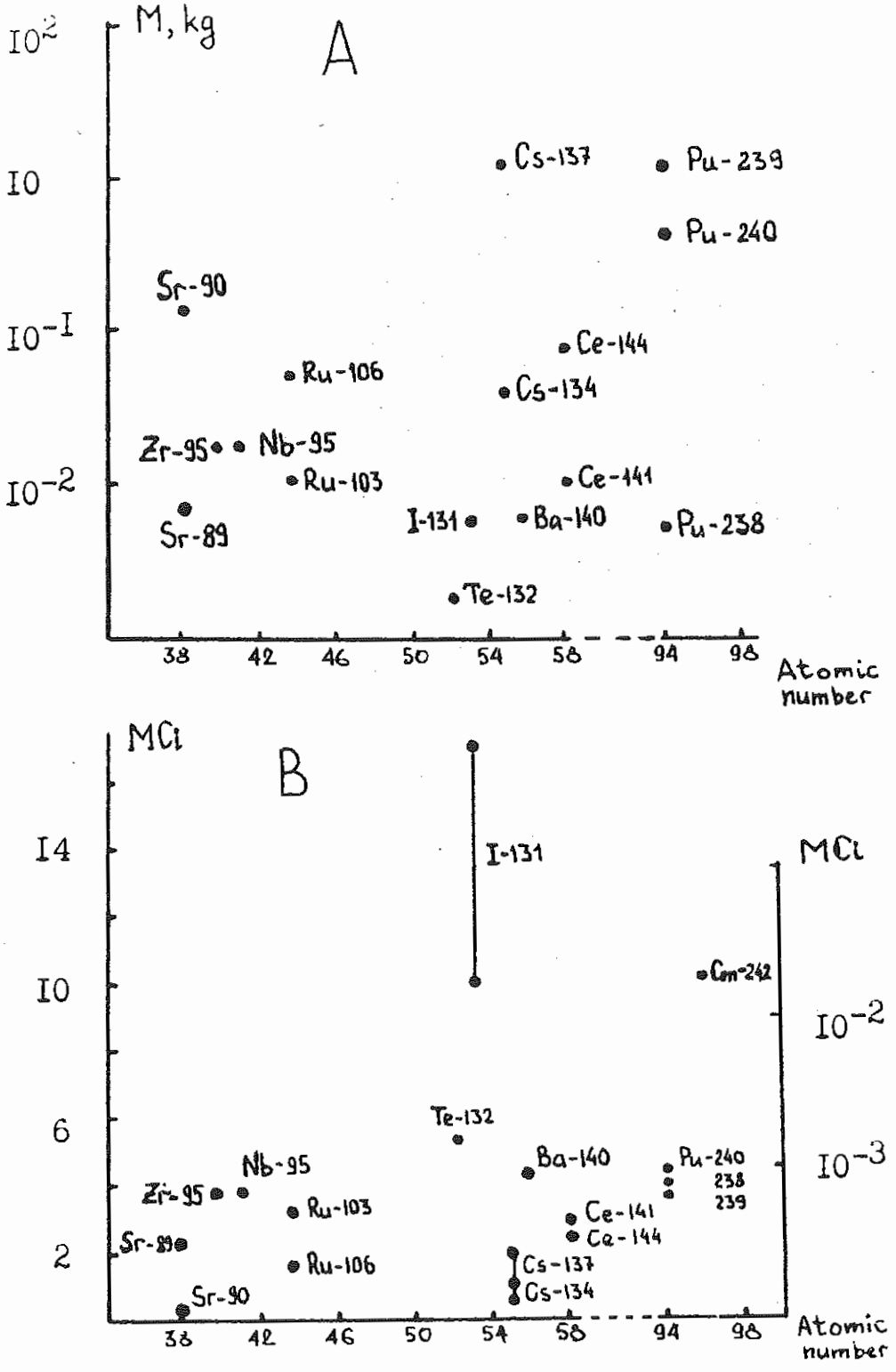

Fig.2 Chernobyl release (A-in mass unit, B-in activity uni

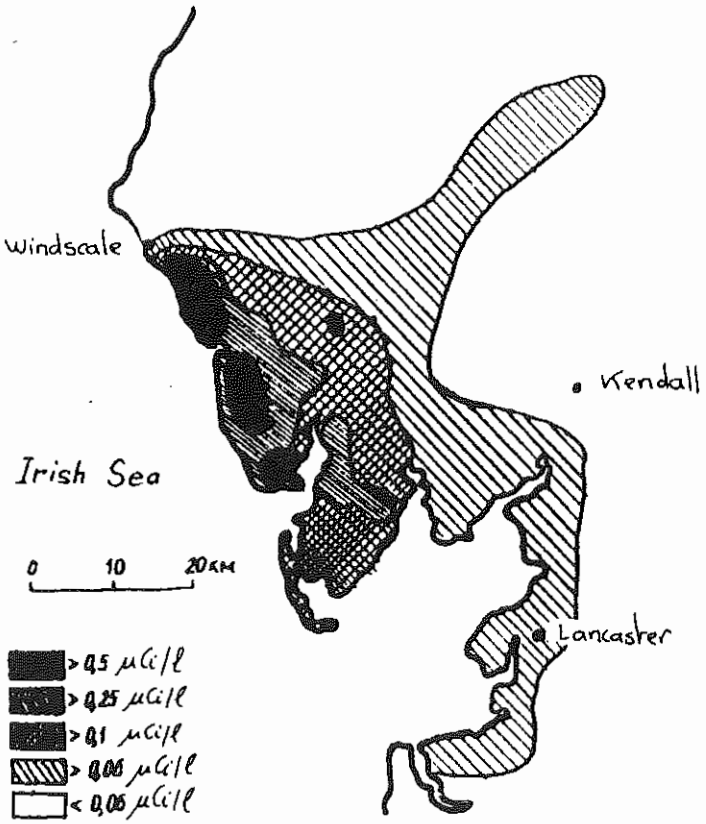

Fig.3 Levels of I-131 in milk after Windscale accident

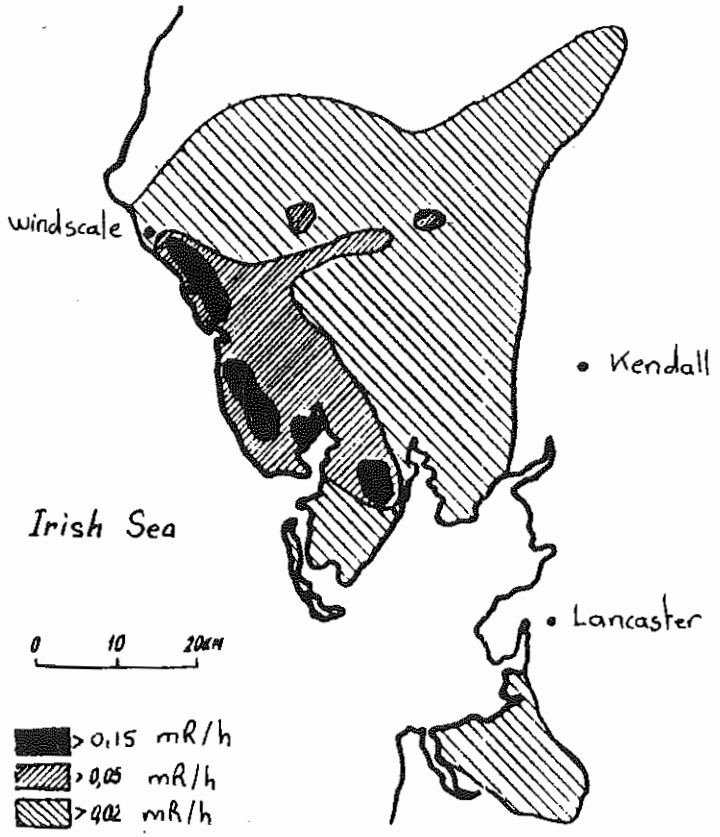

Fig.4 Levels of dose after Windscale accident

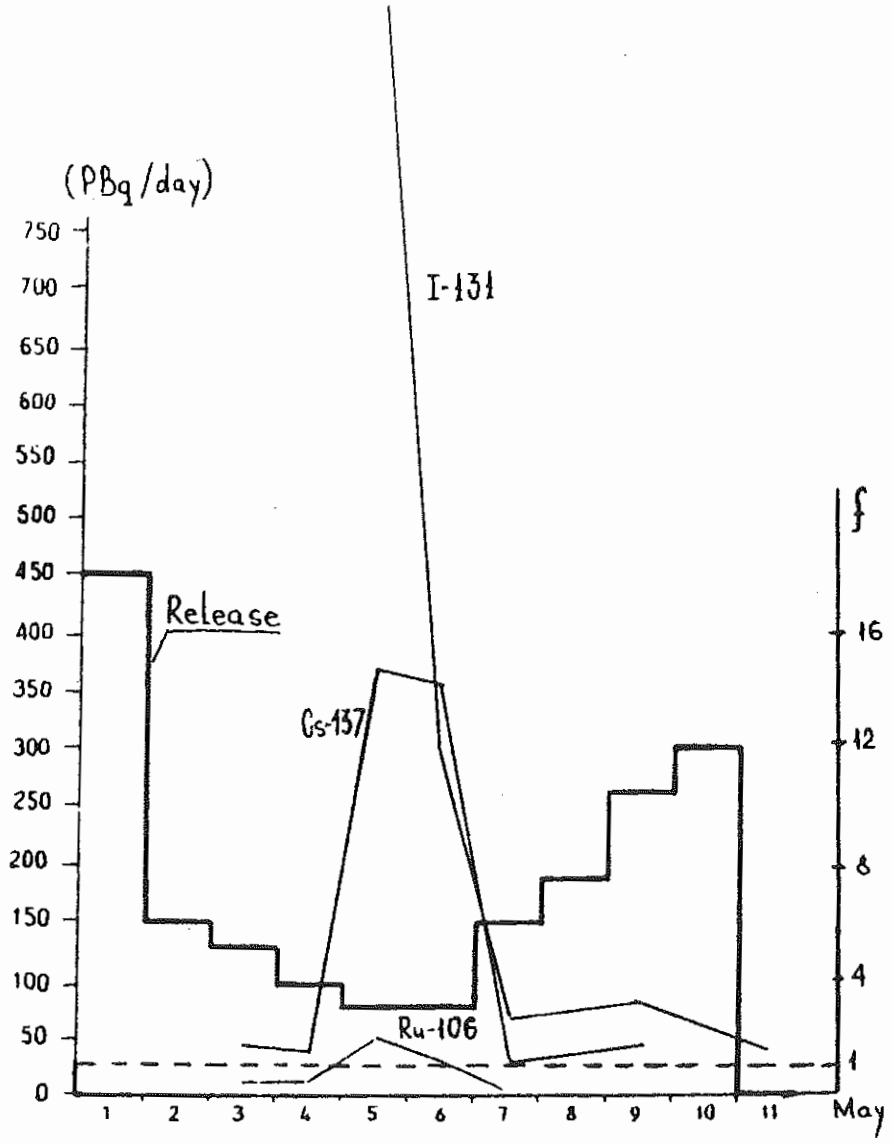

Fig.5 Fractionation of Cs-137, Ru-106, I-131 during Chernobyl accident

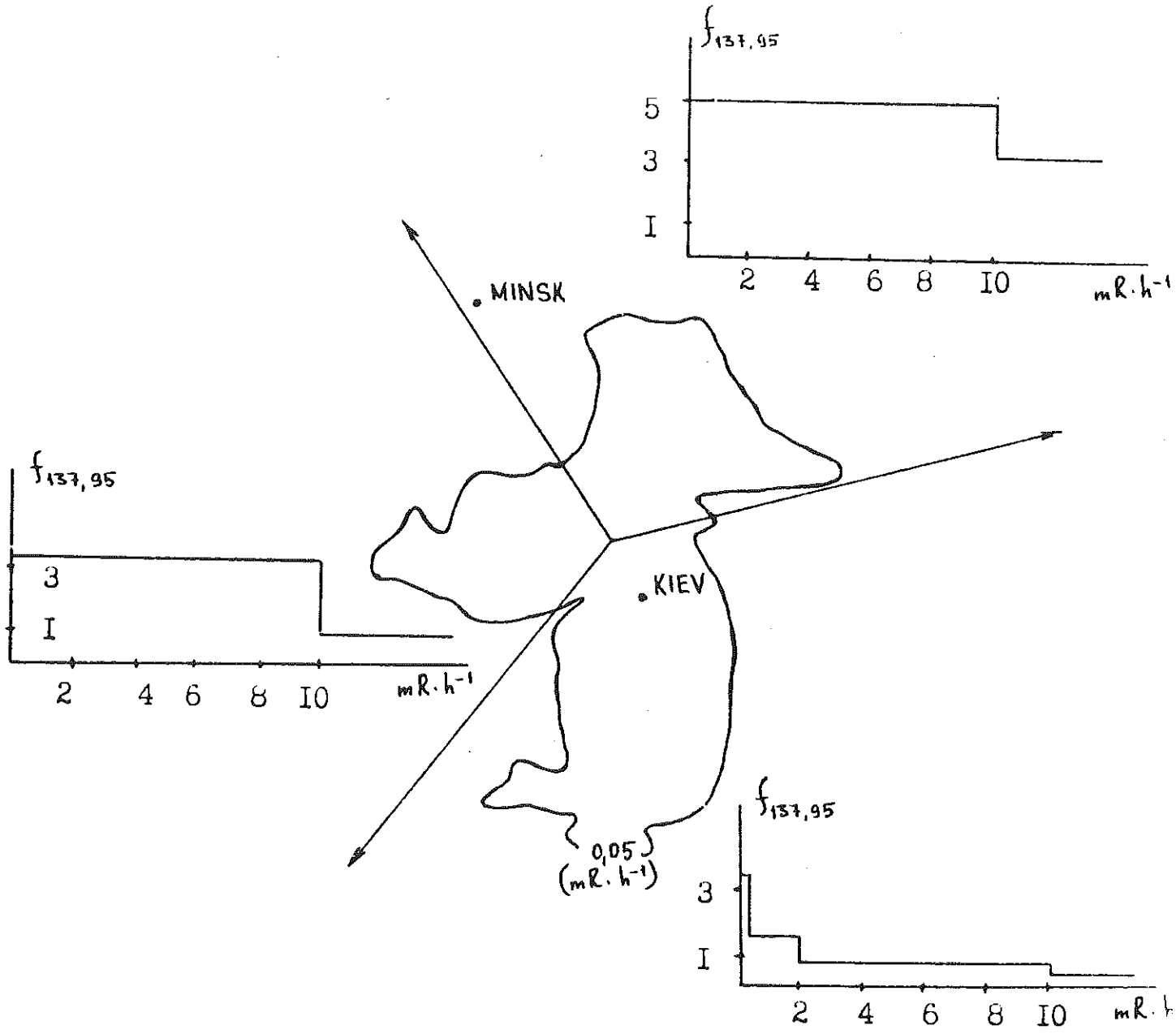

Fig.6 Behaviour of the coefficient of fractional $f_{137,95}$ for three directions of the compass at Chernobyl

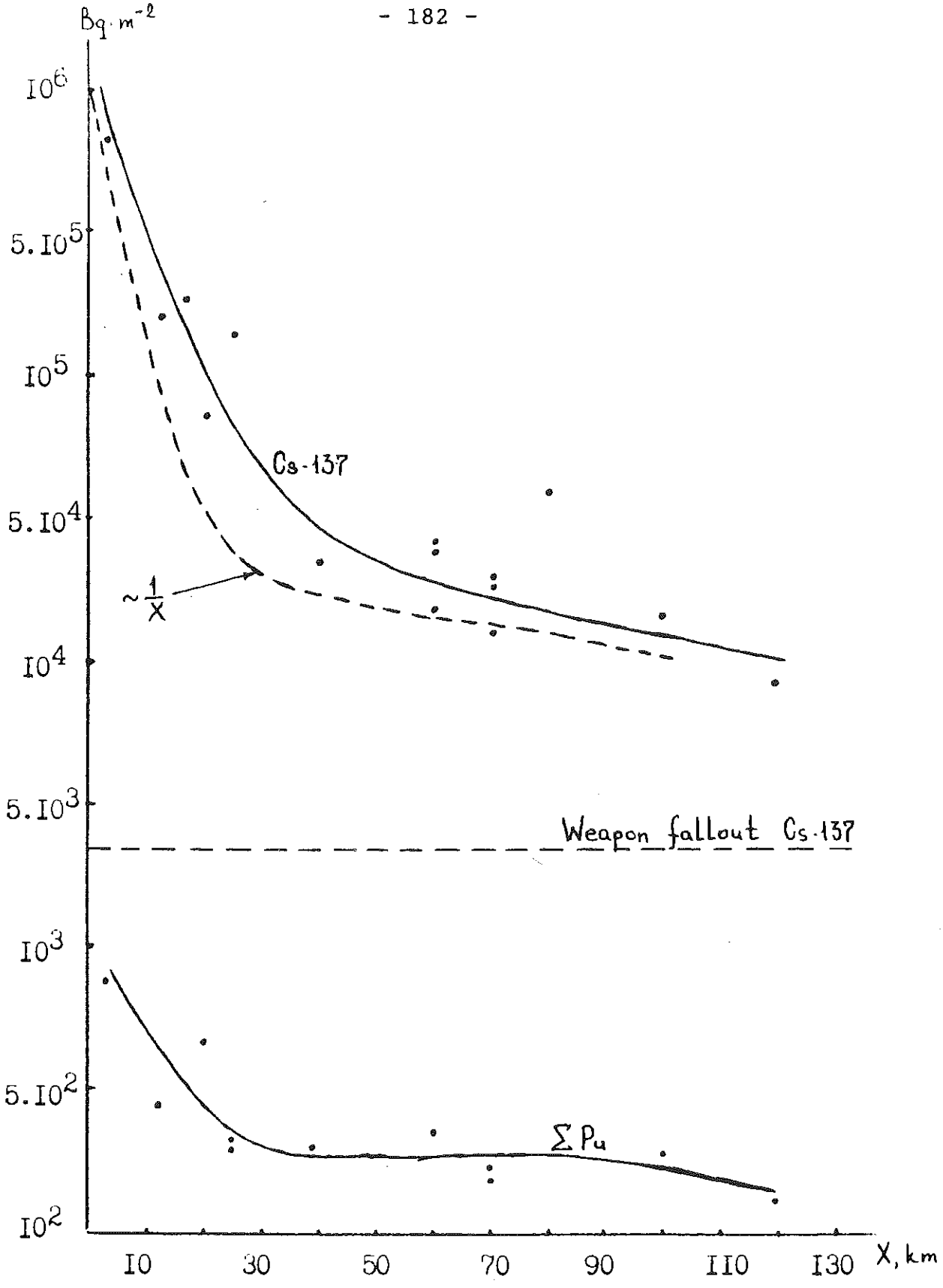

Fig.7 Distribution of Cs-137 and Σ Pu
(Pripiat-Kiev, south branch of the Chernobyl release)

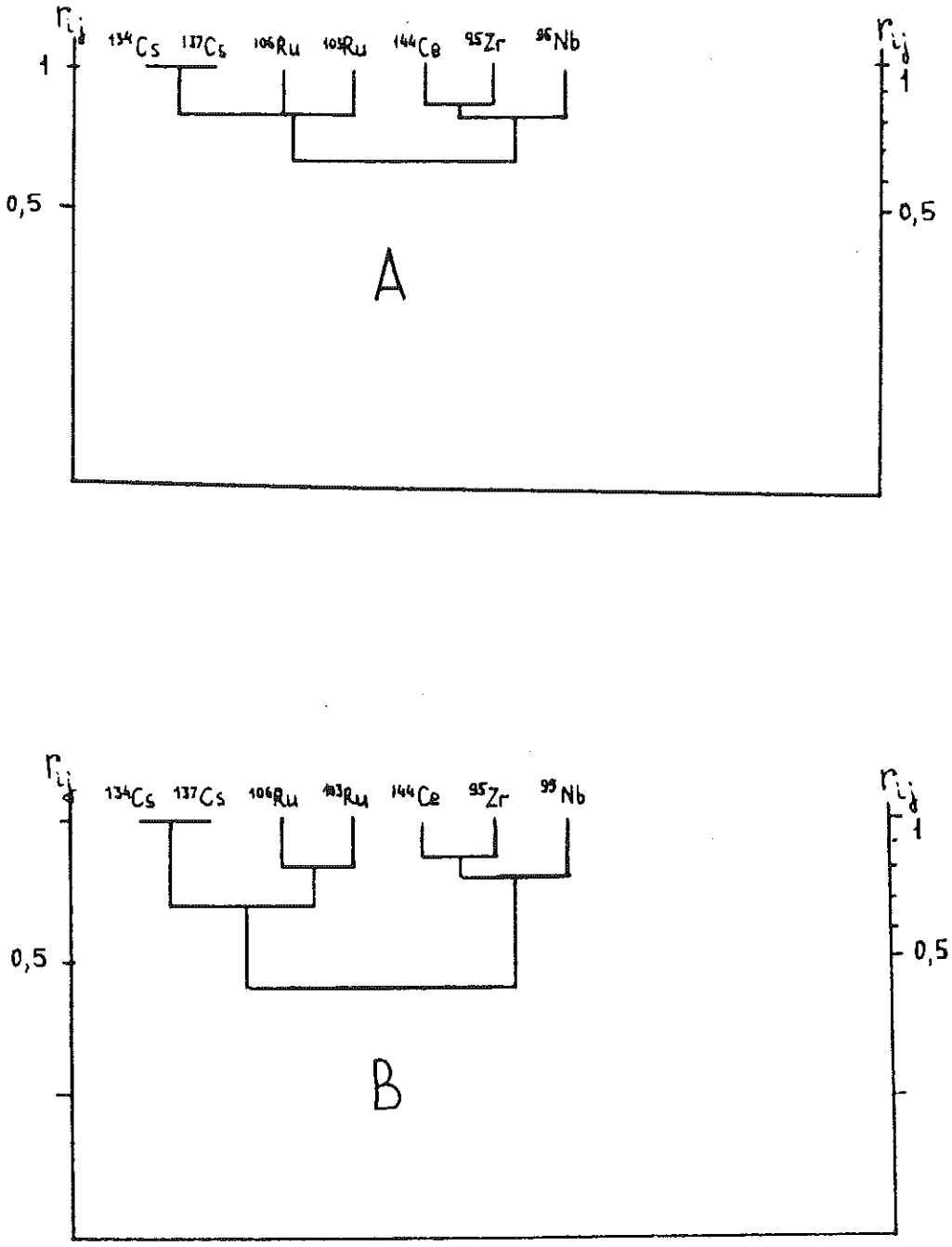

Fig.8 Data on the correlation coefficient r_{ij}
(A-fractionated type of contamination,
B-intermediate type)

Forms of Occurrence and Migration of Radionuclides from the Chernobyl NPP Accident in Typical Landscapes of Byelorussia

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ABSTRACT

In order to assess the radiation situation and predict how it will change we needed to establish the processes involved in the migration of radionuclides – especially in the topsoil as the main element in the radionuclide cycle in the biosphere.

Studies were carried out in three landscape-geochemical areas (typical for the affected Byelorussian territory in terms of radioactive contamination and the variety of soils/landscapes), the aim being to extrapolate the underlying processes found to the entire territory contaminated with radionuclides.

The following were studied:

1. Isotopic composition, degree of contamination, vertical distribution of radioactive fallout in the soil.

2. Ratio between the various forms of occurrence of the radionuclides – water-soluble, exchangeable, mobile and 'fixed'.

3. Abundance of active particles (including fuel particles) in fallout, their size, isotopic composition and geochemical stability.

4. Abundance of "hot" particles in the air and the likelihood of their being incorporated into the human body.

Studies of the vertical distribution of the radionuclides showed that four ?years after the accident the upper 0–1 cm layer of soil contains 50–97 % of the general content of radionuclides in the soil profile. Studies of the forms of occurrence of ^{137}Cs and ^{90}Sr revealed that in 1989 between 76 and 99 % of the caesium was in the 'fixed' state (does not separate from the soil when treated with 6N HCl). Specific to ^{90}Sr is the large proportion found in the water-soluble (up to 16 %), exchangeable (up to 88 %) and mobile states (up to 98 %). The amount of exchangeable strontium increases with distance from the Chernobyl NPP. Active particles of varying fineness are found in practically all the regions studied in concentrations of 10^3 – 10^5 particles per m^2. We also found particles enriched with ^{106}Ru, ^{144}Ce and ^{137}Cs. All isotopes were present in the fuel particles, including isotopes of plutonium, and we studied the content of active particles in human lungs and determined
their isotopic composition.

The complex radioecological situation arising after the Chernobyl accident meant that the contaminated area of Byelorussia needed to be monitored. This involved determining the level of radioactive contamination and forecasting how it would change over time, with a view to drawing up scientifically based recommendations for combating the negative effects of the accident. In order to assess and forecast the radiation situation, we needed to find out the main forms of occurrence of radionuclides and the main processes governing their migration, in particular in the topsoil, which is the main element in the cycle of radioactive substances in the biosphere.

In order to study the migration capacity of radionuclides, we selected three landscape-geochemical areas typical for the affected Byelorussian territory in terms of radioactive contamination, variety of soils and landscape forms (Fig. 1). The three monitoring sites - No. I: Khoyniki district (rayon), physico-geographical province of the Polesye (Pripyat Marshes); No. II: Cherikov district, province of the Orsha-Mogilev plain, and No. III: Korma district, straddling the boundary between provinces - are centred 40, 250 and 200 km respectively from the Chernobyl NPP. The following three main geomorphological levels are monitored at each site: floodplain, the terraces above the floodplain and the original plateau. The types of soil found in the selected areas are soddy-podzolic soil of light mechanical composition, soddy soil and peat-bog soil, which are most widespread in the Republic's southern areas. The fact that a selection of monitoring sites was used means that the data obtained on the migration of radionuclides can be extrapolated for the rest of the contaminated territory in Byelorussia, and that a long-term
forecast of changes in the radiation situation can be made.

This comprehensive study of the nature of the radioactive contamination was carried out at 12 reference points on the monitoring sites and focused on the following areas:

1) isotopic composition, degree of contamination and vertical distribution of radioactive fallout in the topsoil;
2) proportion of the various forms of occurrence of the radioactive isotopes in the soil (water-soluble, exchangeable, mobile and "fixed");
3) abundance of active particles in the fallout, their content, size, composition and geochemical stability;
4) analysis of the content of active particles in human lungs as an important integrating indicator of their migration.

To this end, soil samples in layers of 1 and 5 cm were taken every year at the reference points down to a depth of 0.5 m. The samples were then analysed to determine the content of the isotopes of caesium, ruthenium and cerium (using an ADKAM-300 gamma-spectrometer), and of ^{90}Sr and plutonium isotopes (using radiochemistry). To determine the forms of occurrence of the radionuclides in the soil, we used successive selective leaching [1, 2], a technique which is used in geochemistry and soil science to analyse the state of various elements, including radionuclides from global fallout. The presence of radioactive particles was established by means of radiography [3], with X-ray film as the light-sensitive layer.

It was established that the isotopic composition, amount and physico-chemical properties of the radioactive fallout depend on distance from the source of contamination. At all the reference points we identified the isotopes of ^{137}Cs, ^{134}Cs, ^{106}Ru, ^{144}Ce, ^{90}Sr and others, the relative content of each of which varied from one place to another. The greater the distance from the Chernobyl NPP, the greater the contribution of highly volatile radionuclides of caesium and the lower the proportion of ^{106}Ru and ^{144}Ce. The reference points were highly contaminated with gamma-emitting radionuclides. The level of fallout ranged from 20 to 100 Ci/km_2, the level of contamination at the distant sites (Nos. II and III) being approximately twice that of the near site (No. I). Most of the general gamma-activity was caused by ^{137}Cs (56-77%). The quantity of ^{90}Sr found in the soil varied from 0.3 to 3.7 Ci/km_2. The level of ^{90}Sr contamination at the distant sites was 4-5 times lower than that at the near site.

The dynamics of the behaviour of the main dose-forming isotopes (^{137}Cs and ^{90}Sr) were assessed using charts showing the distribution of the radionuclides in soil (in terms of the relative activity in layers of various depths) on the basis of observations made between 1987 and 1989. The results (Table 1 and Figs. 2 and 3) show that they migrate slowly through the soil profile. At most of the sites, three years after the accident the upper 0-1 cm soil layer was found to contain 50-97% of the general content of radionuclides in the soil profile. Moreover, a considerable proportion of the gamma- and beta-emitters was found in the surface layer (forest litter, moss, grass cover). In 1987 most of the radionuclides (70-90%) were in the forest litter and the surface layers of the soil. Between 1987 and 1989 the contamination level of the surface layer fell by 15-40%, the radionuclide content in the various layers in the upper 0-5 cm of the soil did not change by more than 15%, and the content in the lower layers did not exceed 2% of the total amount in the soil profile. As a rule, the specific activity of the soil layers at a depth of 20-25 cm is the same as background levels. Only in one case (site 20) was the contamination of the lower layers comparable with that of the surface layer. In this case, migration took place on soddy-podzolic soil with an alluvial sandy and well-aerated upper horizon. The reasons for the rapid migration in this case may be the intensive eluvial regime, the low physico-chemical absorptive capacity and the specific mechanical and mineralogical composition of the soil. The migration capacity of the various isotopes (^{90}Sr, ^{137}Cs and ^{106}Ru) did not differ significantly.

In order to determine the parameters influencing the speed of migration, the experimental data on the vertical distribution of radionuclides in soil were analysed using the following equation [4]:

$$q(x,t) = \frac{q_c a}{\sqrt{\pi M_1 \Delta t}} \exp\left(-\frac{x^2}{4 M_1 \Delta t}\right) + \frac{q_c b}{\sqrt{\pi \eta_1 \Delta t}} \exp\left(-\frac{x^2}{4 \eta_2 \Delta t}\right)$$

where $q(x,t)$ is the quantity of the radionuclide at depth x after time t from the moment of the accident, q_c is the quantity of the radionuclide on the surface of the soil at the time of the accident, M_1 and M_2 are the migration coefficients of slow and fast movement, and a and b their proportional shares.

We used this formula to produce a rough forecast of radionuclide movement deeper into the soil. The mathematical model in question was successfully used to forecast the dynamics of the migration of radionuclides in global fallout [4-6]. It made it possible to describe the main processes governing radionuclide displacement in the soil under natural conditions, viz. diffusion and convective migration. The migration coefficients express the overall speed of simultaneously occurring processes under the influence of factors such as the filtration of atmospheric precipitation, and transport on migrating colloidal and finely dispersed particles and through the root system of plants etc. It was established that the magnitude of the migration coefficients depends on the particular features of the soils and the physico-chemical state of the radionuclides, and that it ranges from 0.4 to 10 and from 10 to $90 \cdot 10^{-8}$ $cm^2 \cdot s^{-1}$ for the slow and fast types of movement.

The results of long-term forecasts for various periods of migration are shown in Table 2 on the basis of the distribution of ^{90}Sr and ^{137}Cs at reference point No. 17, which was typical for most cases. An analysis of the forecast shows that natural cleansing of the soil by means of vertical migration will proceed slowly. Most of the radionuclides will remain in the top soil horizon for many decades.

Since the egress of the radionuclides from the accumulative layer into other genetic horizons of the soil may lead to a considerable change in the migration coefficients, model calculations were carried out to yield the vertical distribution of isotopes which are known to have high coefficients:
$(100-300) \cdot 10^{-8}$ $cm^2 \cdot s^{-1}$. The variants for calculated soil profiles showed that in the next few decades there will be no danger of contamination of the groundwater in idiomorphic and semi-hydromorphic soddy-podzolic soils, but that there is likely to be contamination in floodplain soddy and peat-bog soils which have aquiferous horizons close to the surface and a high level of initial radionuclide contamination.

From the predictions of the changes in the spatial distribution of radionuclides in the topsoil over time we were able to estimate the effective period of semi-elimination (T_{eff}). The magnitude of T_{eff} is connected to two-half periods: physical decay ($T_{1/2\ phys.}$) and biological decay ($T_{1/2\ biol.}$), the latter being the time for semi-loss of the radionuclide as a result of vertical migration beyond the boundaries of the topsoil layer [7]. For the long-lived isotopes of ^{90}Sr and ^{137}Cs the values for $T_{eff.}$ ranged from 17 to 23 years and, in the case of intensive migration of radionuclides (for example, at reference point 20), the period of effective semi-removal was approximately 10 years.

It should be noted that the values for the period of semi-elimination are higher than the normal values for $T_{eff.}$ typical of radionuclides in global fallout [7,8]. This discrepancy can be explained by the particular features of the Chernobyl contamination, which include certain physico-chemical properties of the fallout (its fineness, solubility etc).

Tables 3 and 4 and Figs. 4-7 show the results of studying the forms of occurrence of caesium and strontium radioisotopes. It can be observed that ^{134}Cs and ^{137}Cs are present in all the soils analysed, mainly in the "fixed" state, which has hardly changed in the four years since the accident. In 1987 this form accounted for between 60 and 98% of the overall content of these radionuclides, and in 1989 between 49 and 98%, with part of them being in the firmly bound state (i.e. present in the soil residue after processing with 6 N HCl), possibly due to penetration of caesium isotopes into the crystal lattice of clayey minerals, such as vermiculite, montmorillonite and to the presence of low-solubility active particles in the composition. The quantity of water-soluble isotopes of caesium is not significant (max. 1%). The amount of caesium isotopes in exchangeable form does not exceed 22% and, in mobile form, 40%. The ratio between the various forms of occurrence of caesium radionuclides in the soil did not depend on the distance of the reference point from the source of contamination.

It should be noted that the caesium radionuclides from the Chernobyl emissions differ in their forms of occurrence from the isotopes of caesium in global fallout. The proportion of "fixed" caesium from Chernobyl is 2-5 times higher - and the relative quantity of exchangeable and mobile forms is significantly lower - than the proportion of these forms in global contamination.

In comparison with the isotopes of caesium, there is more ^{90}Sr in the water-soluble (up to 16%), exchangeable (up to 88%) and mobile (up to 98%) forms and, correspondingly, less in the "fixed" state (between 2 and 56%) (Table 4). The relative quantity of exchangeable ^{90}Sr rises with increasing distance from the Chernobyl NPP and hence, at a distance of 200 - 250 km from the reactor, ^{90}Sr is present in the soil mainly in the exchangeable form. In 1988 an increase in the content of ^{90}Sr in water-soluble form was observed in certain soils, in particular those with a high humus content - e.g. in peat-bog soil (reference points 4 and 11) and floodplain soddy soil (reference points 1 and 20) - which may be caused by the breakdown of the "fixed" form. There was a general trend towards an increase in the relative content of ^{90}Sr in exchangeable form in the lower soil layers (Table 5). Hence, vertical migration of ^{90}Sr isotopes takes place mainly in exchangeable form. In the case of ^{137}Cs, the ratio between the forms of occurrence hardly changes with depth. The data indicate that ^{90}Sr is more mobile than the isotopes of ^{134}Cs and ^{137}Cs.

Our study of the nature of radioactive fallout at the selected monitoring sites also showed that a considerable proportion of the radioisotopes is present in the form of active particles, which can be sub-divided into fuel particles ("hot" particles) and condensation particles. The content of active particles, their size and isotopic composition depend on the distance from the Chernobyl NPP.

Active particles of various sizes are present at almost all the reference points. Fig. 8 shows the radiographic traces of active particles detected at various sites. Fig. 9 shows an image of the largest particles detected at the site 40 km from the Chernobyl NPP. Within a radius of 40-250 km of the Chernobyl NPP, the greater the distance from the source of contamination, the lower the quantity of particles per m^2 (the figures ranging from $1.4 \cdot 10^3$ to $1.2 \cdot 10^5$), accompanied by a decrease in the proportion of active particles in overall specific gamma-activity.

There are significant differences in the isotopic composition of the active particles (Table 6). Some of the most frequently occurring particles at the sites (in all, about 100 particles were analysed) are rich in ruthenium (No. 7), cerium and ruthenium (Nos. 5 and 6), and caesium (Nos. 14 and 15). Particles rich in caesium are more common at the distant sites.

In order to study the chemical stability of active particles, soil samples containing such particles were treated with various acids, including those used for leaching during radiochemical analysis of samples to determine the amount of radioactive isotopes in them [9, 10]. Samples calcined at 650°C and non-calcined samples were processed. Concentrated solutions of the acids HCl, HNO_3, $HClO_4$ and the mixtures HNO_3 + H_2O_2 and $HClO_4$ + HF were used as solvents. The quantity of active particles, the total beta-activity and the part of the beta-activity which came from active particles were determined in each source sample and in each residue which had not dissolved in the above-mentioned reagents.

The data obtained (Table 7) show that it is much easier to dissolve the active particles in the calcined soil samples than in the non-calcined samples. Full dissolution of the active particles was observed only in the calcined samples when HNO_3 or a mixture of $HClO_4$ + HF were used.

A study of the correlation between the granulometric composition of the soils and the amount of isotopes in the various mechanical fractions, revealed that the distribution of radionuclides by fractions is virtually identical, irrespective of the type of soil and the distance of the reference point from the source of contamination. The highest specific gamma- and beta-activity was observed in the finest particles (< 10 μm), which account for no more than 20% of the mass. In this mechanical fraction we discovered some 50% of the ^{90}Sr, 30% of the ^{106}Ru, 40% of the ^{144}Ce, 25% of the plutonium isotopes, and the maximum content of active particles.

The results obtained indicate the capacity of active particles to play a role in the mechanical displacement of radionuclides via dust transport, surface runoff and penetration into the soil profile. Although it should be pointed out that currently the bulk of active particles (around 70%) is in the first surface layer, in peat-bog soils the active particles are fixed even down to a depth of 30 cm. When considering the surface distribution of active particles, it is important to take into account the possibility of contamination of ponded air and consequently the danger of particles being inhaled into the human body. Analysis of filters in filtration and ventilation units revealed up to several hundred active particles - many of which with an extremely high level of radioactivity - per million cubic metres of air in the populated areas studied. Active particles were also discovered on screens positioned 1 m above ground level in many populated areas of Byelorussia. It was revealed that the quantity of particles and their contribution to the total radioactivity was considerably higher in the south-eastern areas of the Republic than in the north-western areas. Thus, the distribution of active particles in the atmosphere was similar to their geographical distribution in the soil.

The hypothesis concerning the possibility of active particles being inhaled into the human body was also confirmed. In around 70% of the lung samples studied (the material being obtained from post-mortems on people from the contaminated area of the Gomel and Mogilev oblasts [regions]), active particles of various origins, including fuel particles, were detected with certainty. This last point must be taken into account in all recommendations on radiation safeguards, the performance of practical work (especially agricultural work) and when deciding on resettlement of people from contaminated areas.

In conclusion, the data in this paper on the nature of radioactive contamination in the southern areas of the Republic are useful for forecasting changes in the radiation situation. This thorough analysis of radionuclide migration, which takes into account the given landscape, soil characteristics, forms of occurrence of the radionuclides and physico-chemical properties of the active particles, has made it possible for us to explain the nature of the migratory processes in the topsoil, and to establish correlations between the relationships observed and those previously established for global fallout. This constitutes a basis for long-term forecasting of the migration of radionuclides in geochemical and biological chains.

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LAYER-BY-LAYER CONTENT OF THE ISOTOPES OF Cs-137 AND Sr-90 IN SOILS
(JULY 1988)

Number of ref. point	DESCRIPTION OF REFERENCE POINT	Cs-137 CONTENT				Sr-90 CONTENT			
		TOTAL Ci/km²	RELATIVE %			TOTAL Ci/km²	RELATIVE %		
		0-30 cm	0-1 cm	1-2 cm	4-5 cm	0-30 cm	0-1 cm	1-2 cm	4-5 cm
SITE No I LHOGNIKI DISTRICT (≈ 40 km from Chernobyl NPP)									
1	Low flood plain	23.4	64.2	24.1	0.7	2.8	50.0	30.8	2.6
3	Terrace above flood plain	28.5	91.9	3.6	0.4	3.7	83.5	9.6	0.6
4	Ameliorated meadow	17.6	86.2	3.0	0.4	3.0	79.4	3.2	1.7
6	Loessial plateau	11.8	55.7	34.9	0.9	2.3	48.6	44.4	0.4
SITE No III KORMA DISTRICT (≈ 200 km from Chernobyl NPP)									
20	Low flood plain	41.6	12.6	11.0	11.2	0.3	15.1	11.6	6.9
23	Terrace	82.9	65.1	32.6	0.3	1.5	97.0	2.6	0
27	Forest	46.5	62.0	9.6	2.7	0.4	42.7	11.9	4.6
SITE No II CHERIKOV DISTRICT (≈ 250 km from Chernobyl NPP)									
10	Low flood plain	59.7	94.8	1.8	0.2	0.4	75.4	14.0	0
11	Marshy flood plain	46.0	79.7	12.6	0.9	0.8	56.4	22.9	3.9
13	Terrace	47.8	61.1	18.9	2.2	0.8	50.0	26.1	2.1
17	Forest	32.9	56.1	13.3	2.5	0.4	68.3	15.8	1.2
19	Garden	67.0	64.5	17.1	1.6	0.9	53.2	20.0	2.6

TABLE 1

FORECAST OF THE DITRIBUTION OF RADIONUCLIDES BY DEPHT OF SOIL PROFILE

Depth of layer x (cm)	RELATIVE CONTENT OF ISOTOPES IN 1-cm LAYER (%)					
	Sr-90 ($cm^2 . s^{-1}$) $M_1 = 0,87 . 10^{-8}$ ($a = 0,87$) $M_2 = 28,2 . 10^{-8}$ ($b = 0,73$)			Cs-137 ($cm^2 . s^{-1}$) $M_1 = 0,50 . 10^{-8}$ ($a = 0,76$) $M_2 = 11,0 . 10^{-8}$ ($b = 0,24$)		
	5 years	10 years	50 years	5 years	10 years	50 years
0.5	41.1	29.7	13.5	48.0	35.2	16.2
1.5	28.9	24.9	13.1	26.8	26.2	15.3
5.0	1.4	3.8	8.8	2.3	2.6	7.9
10.0	0.6	0.7	2.5	0.8	1.1	1.5
20.0	0.1	0.3	0.3	0.01	0.1	0.6
30.0	0.01	0.1	0.2	traces	traces	0.3
40.0	traces	0.01	0.1	0	traces	0.1
60.0	0	0	0.1	0	0	traces

TABLE 2

FORMS OF OCCURENCE OF RADIOCAESIUM IN THE 0-1 CM SOIL LAYER
(1987-1989)

Table 3

Number of refer. point	H2O %			Exchangeable form (H2O + NH4 COOCH3 1H) %			HCl 1H %			"Fixed" form (GH HCl + soil residue) %		
	1987	1988	1989	1987	1988	1989	1987	1988	1989	1987	1988	1989
SITE No I LHOGNIKI DISTRICT (≈ 40 km from Chernobyl NPP)												
1	0.3 ± 0.1	< 0.1	0.1 ± 0.1	1.5 ± 0.2	0.6 ± 0.2	0.7 ± 0.2	3.1 ± 0.4	3.2 ± 0.2	1.0 ± 0.2	95 ± 8	96 ± 6	98 ± 13
3	0.3 ± 0.1	0.5 ± 0.1	0.3 ± 0.1	15 ± 2	1.8 ± 0.2	5.3 ± 0.9	5 ± 1	10 ± 1	12 ± 2	80 ± 8	88 ± 6	83 ± 8
4	0.2 ± 0.1	0.4 ± 0.1	0.1 ± 0.1	1.8 ± 0.2	3.4 ± 0.4	1.0 ± 0.3	6.1 ± 0.4	3.8 ± 0.5	4.5 ± 1.2	92 ± 7	93 ± 7	94 ± 12
6	0.5 ± 0.1	0.2 ± 0.1	0.3 ± 0.1	8 ± 1	6 ± 1	4 ± 1	7 ± 1	7 ± 1	10 ± 3	85 ± 8	97 ± 6	86 ± 12
SITE No III KORMA DISTRICT (≈ 200 km from Chernobyl NPP)												
20	0.5 ± 0.1	0.2 ± 0.1	0.2 ± 0.1	3.5 ± 0.6	1.9 ± 0.3	1.0 ± 0.2	7 ± 1	5 ± 1	1.8 ± 0.4	90 ± 10	93 ± 7	97 ± 7
23	0.9 ± 0.2	0.6 ± 0.1	1.3 ± 0.3	22 ± 3	6 ± 2	34 ± 4	18 ± 2	17 ± 3	17 ± 2	60 ± 5	77 ± 12	49 ± 7
27 ±	0.2 ± 0.1	< 0.1	0.1 ± 0.1	3.0 ± 0.4	2.6 ± 0.3	1.6 ± 0.3	20 ± 3	16 ± 2	10 ± 2	77 ± 8	81 ± 6	88 ± 11
SITE No II CHERIKOV DISTRICT (≈ 250 km from Chernobyl NPP)												
10	< 0.1	< 0.1	0.1 ± 0.1	0.3 ± 0.1	0.2 ± 0.1	0.7 ± 0.2	1.5 ± 0.5	0.8 ± 0.1	1.2 ± 0.3	98 ± 5	99 ± 6	98 ± 1
11	0.1 ± 0.1	0.1 ± 0.1	0.1 ± 0.1	1.2 ± 0.2	1.3 ± 0.2	1.2 ± 0.3	3.6 ± 0.6	3.4 ± 0.3	2.4 ± 0.7	95 ± 6	95 ± 6	96 ± 13
13	0.1 ± 0.1	0.1 ± 0.1	0.4 ± 0.1	4.3 ± 0.6	0.8 ± 0.1	2.3 ± 0.4	12 ± 2	2.1 ± 0.2	5.4 ± 1.2	84 ± 5	97 ± 6	92 ± 12
17	< 0.1	< 0.1	0.1 ± 0.1	2.0 ± 0.4	0.7 ± 0.1	8.8 ± 1.7	7 ± 1	3.4 ± 0.4	12.7± 2.1	91 ± 8	96 ± 5	78 ± 10
19	0.1 ± 0.1	0.8 ± 0.1	0.7 ± 0.1	5 ± 1	8.1 ± 0.4	18 ± 4	15 ± 2	16 ± 2	16 ± 4	80 ± 5	76 ± 5	66 ± 9

1, 20, 10 - Flood plain soils (sod meadow)
3, 23, 13, 17 - Soddy-podzolic sandy
4, 11 - Peat
6, 27, 19 - Soddy-podzolic sandy-loam soils

FORMS OF OCCURENCE OF STRONTIUM 90 IN SURFACE SOIL LAYERS (0-1 cm)
(1987-1989)

Table 4

Number of refer. point	H2O (%)			Exchangeable form (H2O + NH4 COOCH3 1H) (%)			HCl 1H (%)			"Fixed" form (%)		
	1987	1988	1989	1987	1988	1989	1987	1988	1989	1987	1988	1989
SITE No I LHOGNIKI DISTRICT (≈ 40 km from Chernobyl NPP)												
1	4 ± 2	7 ± 3	4 ± 2	37 ± 7	41 ± 6	49 ± 8	16 ± 5	14 ± 4	10 ± 3	47 ± 9	45 ± 5	41 ± 4
3	1 ± 1	6 ± 2	2 ± 1	35 ± 6	48 ± 8	77 ± 15	14 ± 4	8 ± 3	8 ± 3	51 ± 10	44 ± 8	15 ± 3
4	1 ± 1	5 ± 2	1 ± 1	14 ± 3	17 ± 4	20 ± 4	20 ± 2	21 ± 4	22 ± 4	76 ± 10	62 ± 5	58 ± 12
6	2 ± 1	3 ± 1	2 ± 1	28 ± 5	32 ± 5	41 ± 7	19 ± 4	17 ± 3	20 ± 4	53 ± 11	51 ± 10	39 ± 8
SITE No III KORMA DISTRICT (≈ 200 km from Chernobyl NPP)												
20	1 ± 1	16 ± 6	14 ± 5	87 ± 22	88 ± 22	93 ± 23	3 ± 2	4 ± 2	5 ± 2	10 ± 5	8 ± 4	2 ± 1
23	3 ± 1	3 ± 1	27 ± 11	64 ± 12	75 ± 12	96 ± 27	10 ± 3	14 ± 3	4 ± 2	26 ± 11	11 ± 5	1
27	1 ± 1	1 ± 1	1 ± 1	91 ± 18	88 ± 17	86 ± 15	5 ± 2	10 ± 4	11 ± 5	4 ± 2	2 ± 2	3 ± 1
SITE No II CHERIKOV DISTRICT (≈ 250 km from Chernobyl NPP)												
10	5 ± 3	5 ± 3	3 ± 1	76 ± 15	78 ± 15	83 ± 16	20 ± 10	20 ± 8	17 ± 7	4 ± 2	2 ± 1	1
11	1 ± 1	7 ± 3	5 ± 2	45 ± 9	62 ± 11	63 ± 9	25 ± 9	24 ± 5	26 ± 6	30 ± 6	14 ± 6	11 ± 4
17	4 ± 2	4 ± 2	7 ± 3	87 ± 17	84 ± 12	89 ± 12	9 ± 4	14 ± 5	10 ± 12	4 ± 2	6 ± 3	1 ± 1
13	11 ± 2	8 ± 3	14 ± 6	69 ± 9	69 ± 12	66 ± 12	24 ± 10	26 ± 10	24 ± 10	7 ± 4	5 ± 2	10 ± 5
19	4 ± 2	13 ± 5	19 ± 8	91 ± 17	84 ± 14	87 ± 16	7 ± 3	11 ± 4	12 ± 5	2 ± 2	5 ± 2	1 ± 1

Table 5: FORMS OF OCCURENCE OF RADIOACTIVE ISOTOPES OF CAESIUM AND STRONTIUM (July 1988)

Number of refer. point	Depth of layer cm	H2O (%)		Exchangeable form (H2O+NH4 COOCH3 1M) (%)		1N HCl (%)		"Fixed" form (%)	
		Total Cs	Sr-90	Total Cs	Sr-90	Total Cs	Sr-90	Total Cs	Sr-90
SITE No I LHOGNIKI DISTRICT (≈ 40 km from Chernobyl NPP)									
1 (a)	0 - 1	< 0.1	7 ± 3	0.6 ± 0.1	41 ± 6	3.2 ± 0.2	14 ± 4	96 ± 6 (30)*	45 ± 5
	2 - 3	< 0.1	5 ± 2	0.4 ± 0.1	72 ± 13	2.9 ± 0.4	18 ± 4	97 ± 6 (28)	10 ± 4
	5 - 6	-	-	2 ± 1	-	1 ± 1	-	97 ± 9 (30)	-
3 (b)	0 - 1	0.5 ± 0.1	6 ± 2	1.8 ± 0.2	48 ± 8	10 ± 1	8 ± 3	88 ± 6 (28)	44 ± 8
	2 - 3	< 0.2	5 ± 2	2.0 ± 0.6	95 ± 18	8 ± 2	4 ± 2	90 ± 7 (30)	1 ± 1
	4 - 5	-	-	2 ± 1	94 ± 35 ±	4 ± 2	2 ± 1	94 ± 7 (32)	4 ± 2
4 (c)	0 - 1	0.4 ± 0.1	5 ± 2	3.4 ± 0.4	17 ± 4	3.8 ± 0.5	21 ± 4	93 ± 7 (18)	62 ± 5
	5 - 6	-	-	4 ± 2	78 ± 20	4 ± 2	22 ± 6	92 ± 7 (16)	< 6
6 (d)	0 - 1	0.2 ± 0.1	3 ± 1	6 ± 1	32 ± 5	7 ± 1	17 ± 3	87 ± 6 (28)	51 ± 10
	2 - 3	< 0.2	3 ± 1	7 ± 1	77 ± 15	8 ± 2	18 ± 7	85 ± 6 (29)	5 ± 2
	4 - 5	-	-	-	77 ± 28	-	18 ± 9	- -	5 ± 3
SITE No III KORMA DISTRICT (≈ 200 km from Chernobyl NPP)									
20(a)	0 - 1	0.2 ± 0.1	16 ± 6	1.9 ± 0.3	88 ± 22	5.0 ± 0.6	4 ± 2	93 ± 7 (28)	8 ± 4
	2 - 3	0.1 ± 0.1	13 ± 5	2.6 ± 0.2	94 ± 30	5.0 ± 0.4	5 ± 3	92 ± 6 (30)	1 ± 1
	9 -10	-	-	15 ± 2	-	2 ± 2	-	83 ± 5 (29)	-

Table 5 (continued)

Number of refer. point	Depth of layer cm	H2O (%)		Exchangeable form (H2O+NH4 COOCH3 1M) (%)		1N HCl (%)		"Fixed" form (%)	
		Total Cs	Sr-90	Total Cs	Sr-90	Total Cs	Sr-90	Total Cs	Sr-90
23(b)	0 - 1	0.6 ± 0.1	3 ± 1	6 ± 2	75 ± 12	17 ± 2	14 ± 3	60 ± 5 (18)	11 ± 5
	2 - 3	0.4 ± 0.2	-	7 ± 1	91 ± 33	14 ± 2	8 ± 4	79 ± 5 (20)	1 ± 1
27(d)	0 - 1	< 0.1	1 ± 1	2.6 ± 0.3	88 ± 17	16 ± 2	10 ± 4	81 ± 6 (24)	2 ± 2
	2 - 3	0.5 ± 0.1	2 ± 1	14 - 1	97 ± 25	14 ± 1	3 ± 1	72 ± 5 (25)	< 1
	9 -10	-	-	17 ± 2	-	13 ± 2	-	70 ± 7 (23)	-
SITE No 11 CHERIKOV DISTRICT (≈ 250 km from Chernobyl NPP)									
10(a)	0 - 1	< 0.1	5 ± 3	0.2 ± 0.1	78 ± 15	0.8 ± 0.1	20 ± 8	99 ± 6 (41)	2 ± 1
	2 - 3	< 0.2	1 ± 1	0.2	83 ± 34	0.3 ± 0.1	17 ± 4	100 ± 5 (40)	< 1
11(c)	0 - 1	0.1 ± 0.1	7 ± 3	1.3 ± 0.2	62 ± 11	3.4 ± 0.3	24 ± 5	95 ± 6 (31)	14 ± 6
	2 - 3	< 0.1	1 ± 1	1.2 ± 0.2	76 ± 22	2.5 ± 0.6	24 ± 10	96 ± 7 (31)	< 1
13(b)	0 - 1	0.1 ± 0.1	8 ± 3	0.8 ± 0.1	69 ± 12	2.1 ± 0.2	26 ± 10	97 ± 6 (39)	5 ± 2
	2 - 3	< 0.1	3 ± 1	0.5 ± 0.1	71 ± 14	2.4 ± 0.3	27 ± 12	97 ± 6 (38)	2 ± 1
17(b)	0 - 1	< 0.1	4 ± 2	0.7 ± 0.1	84 ± 12	3.4 ± 0.4	14 ± 5	96 ± 5 (38)	6 ± 3
	2 - 3	0.1 ± 0.1	2 ± 2	0.6 ± 0.3	90 ± 22	4 ± 1	10 ± 4	95 ± 6 (36)	< 1
19(d)	0 - 1	0.8 ± 0.1	13 ± 5	8.1 ± 0.4	84 ± 14	16 ± 2	11 ± 4	76 ± 5 (25)	5 ± 2
	2 - 3	0.2 ± 0.1	2 ± 1	6 ± 2	93 ± 23	14 ± 3	7 ± 3	80 ± 7 (23)	< 2

(a) sod-meadow; (b) soddy-podzolic sandy; (c) peatbog; (d) soddy-podzolic sandy loam
* content of Caesium radioisotopes in soil residue after processing with 6N HCl

Table 6: ISOTOPIC COMPOSITION OF ACTIVE PARTICLES

Site index	Aγ (Bq)	Ce-144 (Bq)	Ru-106 (Bq)	Cs-134 (Bq)	Cs-137 (Bq)
1.	1.47 E 2	6.38 E 1	2.21 E 1	2.43	1.18 E 1
2.	1.69 E 2	6.73 E 1	3.26 E 1	3.95	1.89 E 1
3.	5.29 E 2	1.18 E 2	4.34 E 1	7.32	3.40 E 1
4.	2.54 E 2	1.00 E 2	5.48 E 1	1.08 E 1	4.20 E 1
5.	2.95 E 3	3.41 E 1	1.92 E 1	-	-
6.	1.72 E 2	6.39 E 1	4.71 E 1	-	-
7.	7.84 E 1	-	2.80 E 1	-	-
8.	5.92 E 1	-	1.24 E 1	1.48	4.47
9.	3.19 E 2	-	2.47 E 1	2.13	1.11 E 1
10.	3.32 E 2	-	2.73 E 2	-	3.15
11.	9.50 E 1	-	3.67 E 1	4.89	1.70 E 1
12.	1.97 E 2	1.30 E 2	-	1.34	2.19 E 1
13.	1.34 E 2	7.77 E 1	-	4.08	-
14.	5.39 E 1	-	-	2.67	1.40 E 1
15.	5.47 E 1	-	-	1.86	9.76

Table 7: PART OF THE β-ACTIVITY RESULTING FROM 'HOT' PARTICLES IN SOURCE SAMPLES, UNDISSOLVED RESIDUES AND LEACHATES

Acids	Processing time (hours)	Soil preparation C=calcined N=non-calcined	Part of β-activity which has gone into solution (%)	Part of activity resulting from 'hot' particles	
				Source samples (%)	Undissolved residues (%)
HCl	1.5	N	26	33	27
HCl	24	N	43	46	38
HCl	72	N	66	46	20
HCl	1.5	C	82	40	20
HNO_3	1.5	N	52	26	10
HNO_3	1.5	C	65	49	0
$HNO_3 + H_2O_2$	1.5	N	64	54	15
$HNO_3 + H_2O_2$	1.5	C	81	30	5.1
$HClO_4$	1.5	N	28	46	38
$HClO_4$	1.5	C	81	44	20
$HClO_4$ + HF	1.5	N	69	18	7.7
$HClO_4$ + HF	1.5	C	93	25	0

Fig.2: DIAGRAMM OF THE DISTRIBUTION OF CS-137 IN SOIL ON THE BASIS OF DATA FROM OBSERVATIONS IN 1987, 1988 AND 1989 (REFERENCE POINT 17)

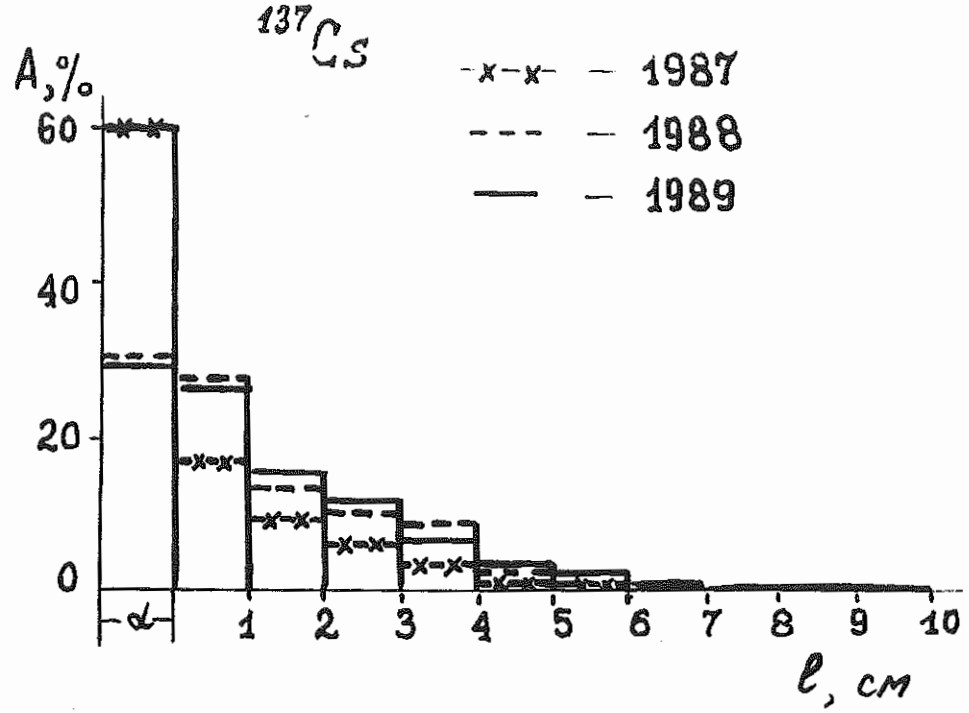


Fig.3: DIAGRAMM OF THE DISTRIBUTION OF Sr-90 IN SOIL ON THE BASIS OF DATA FROM OBSERVATIONS IN 1987, 1988 AND 1989 (REFERENCE POINT 17)

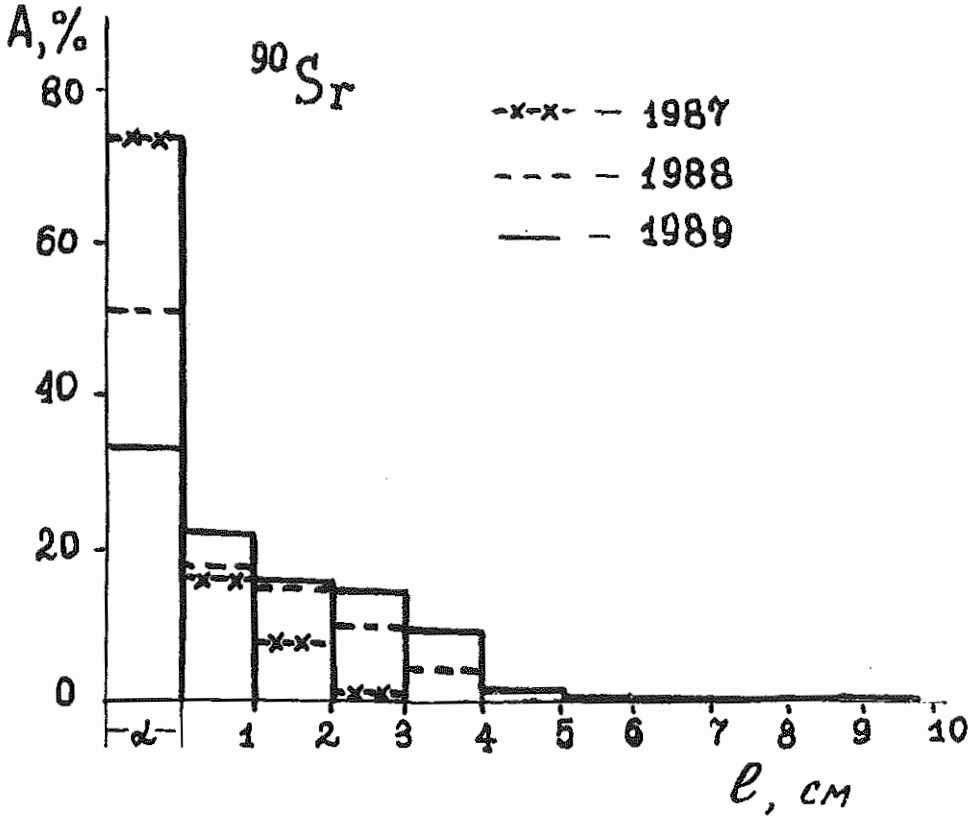


Fig.4: FORMS OF OCCURENCE OF THE RADIOACTIVE ISOTOPES OF CAESIUM AND STRONTIUM IN SANDY SOIL. SITE 3 (≈ 40 km FROM CHERNOBYL NPP)

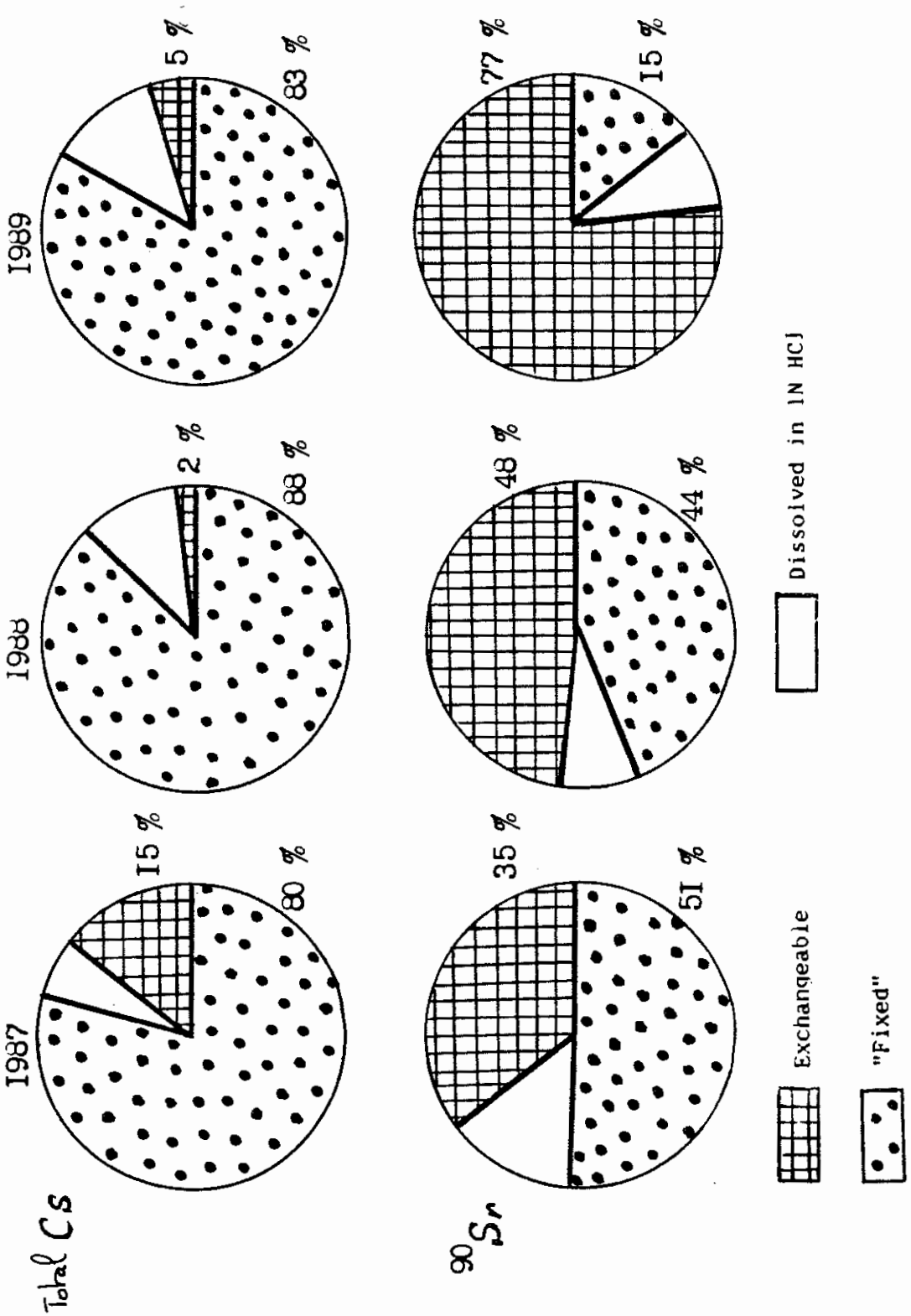


Fig.5: FORMS OF OCCURENCE OF THE RADIOACTIVE ISOTOPES OF CAESIUM AND STRONTIUM IN SANDY SOIL. SITE 17 (≈ 250 km FROM CHERNOBYL NPP)

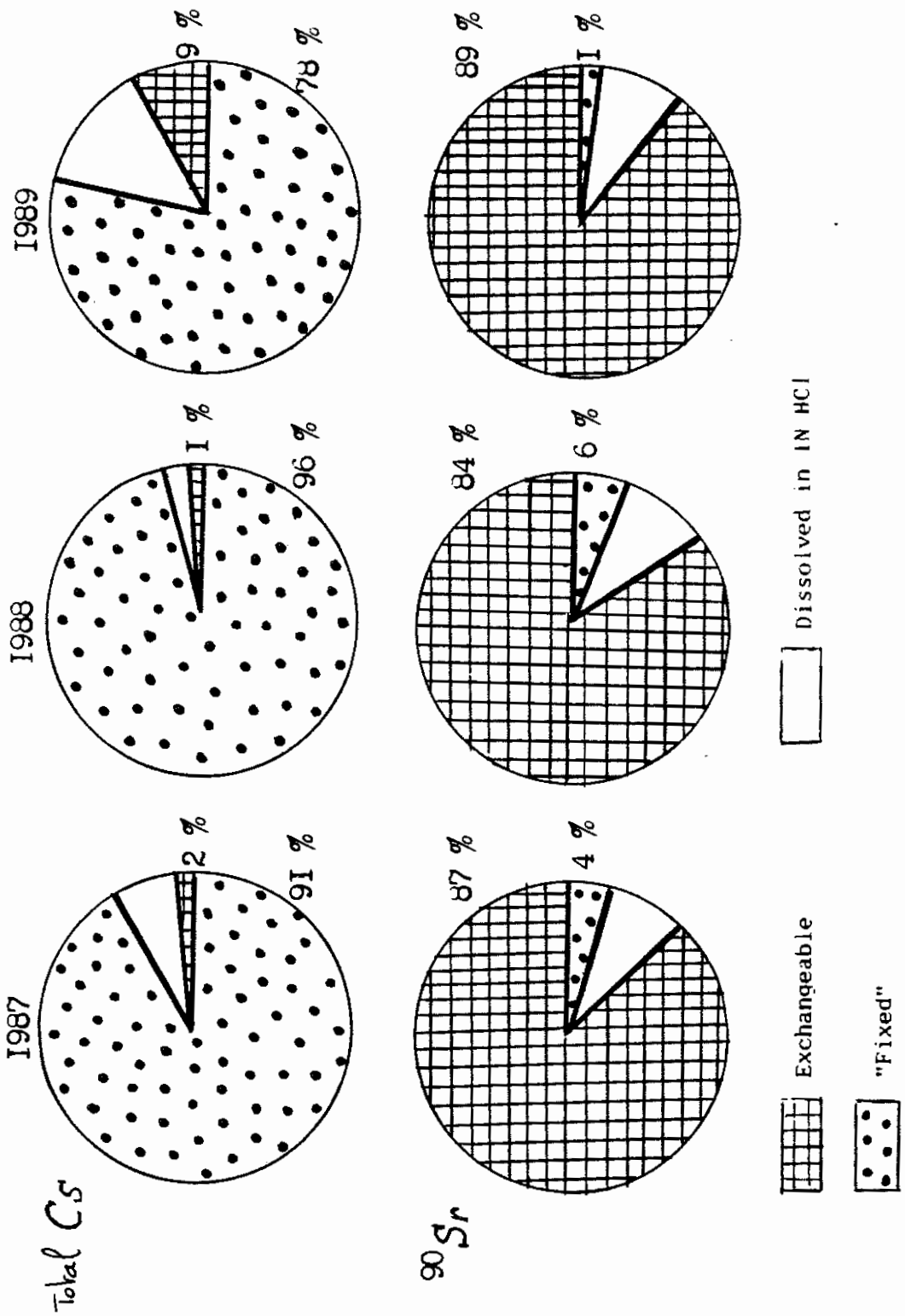


Fig.6: FORMS OF OCCURENCE OF THE RADIOACTIVE ISOTOPES OF CAESIUM IN SOD-MEADOW SOILS

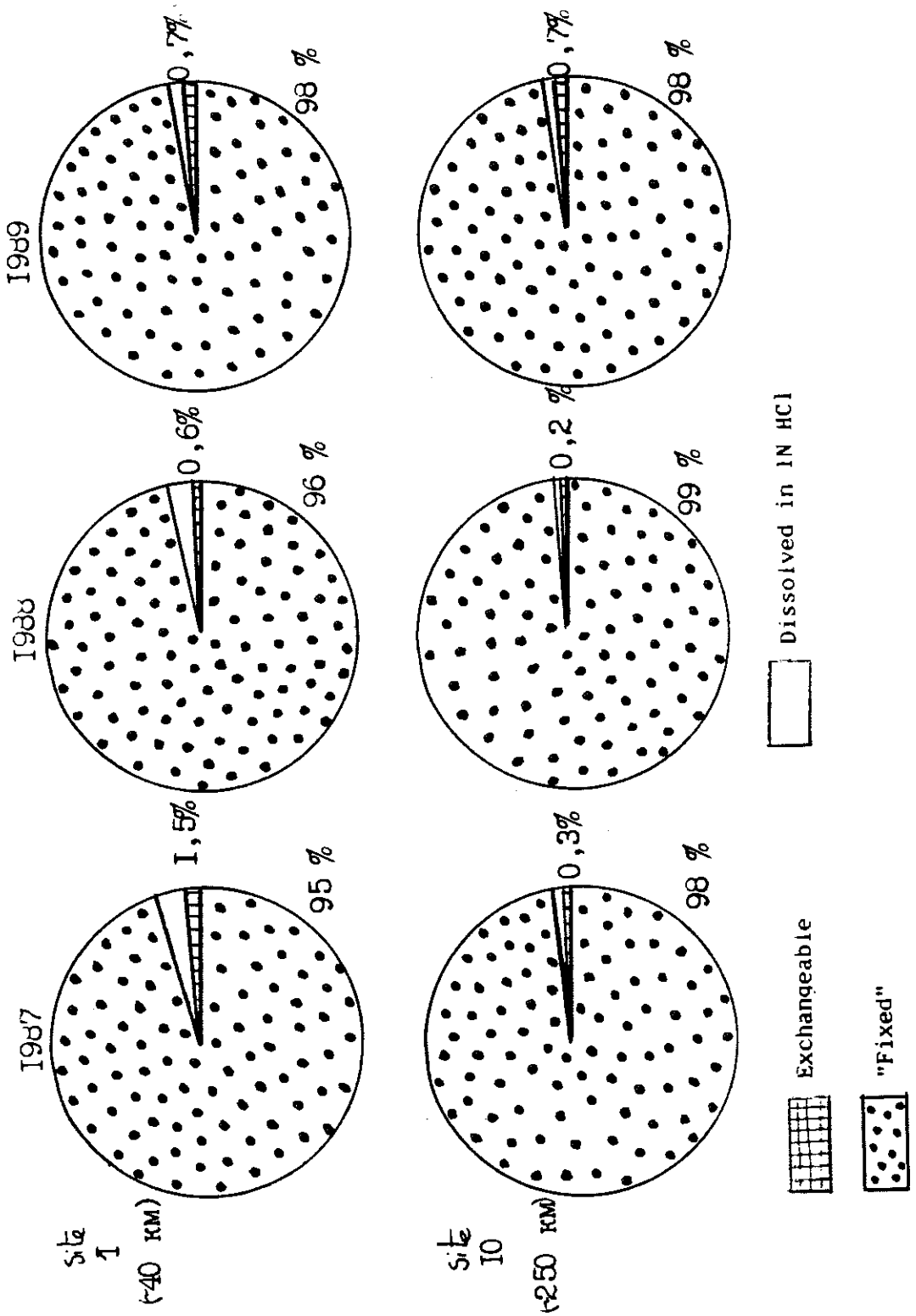
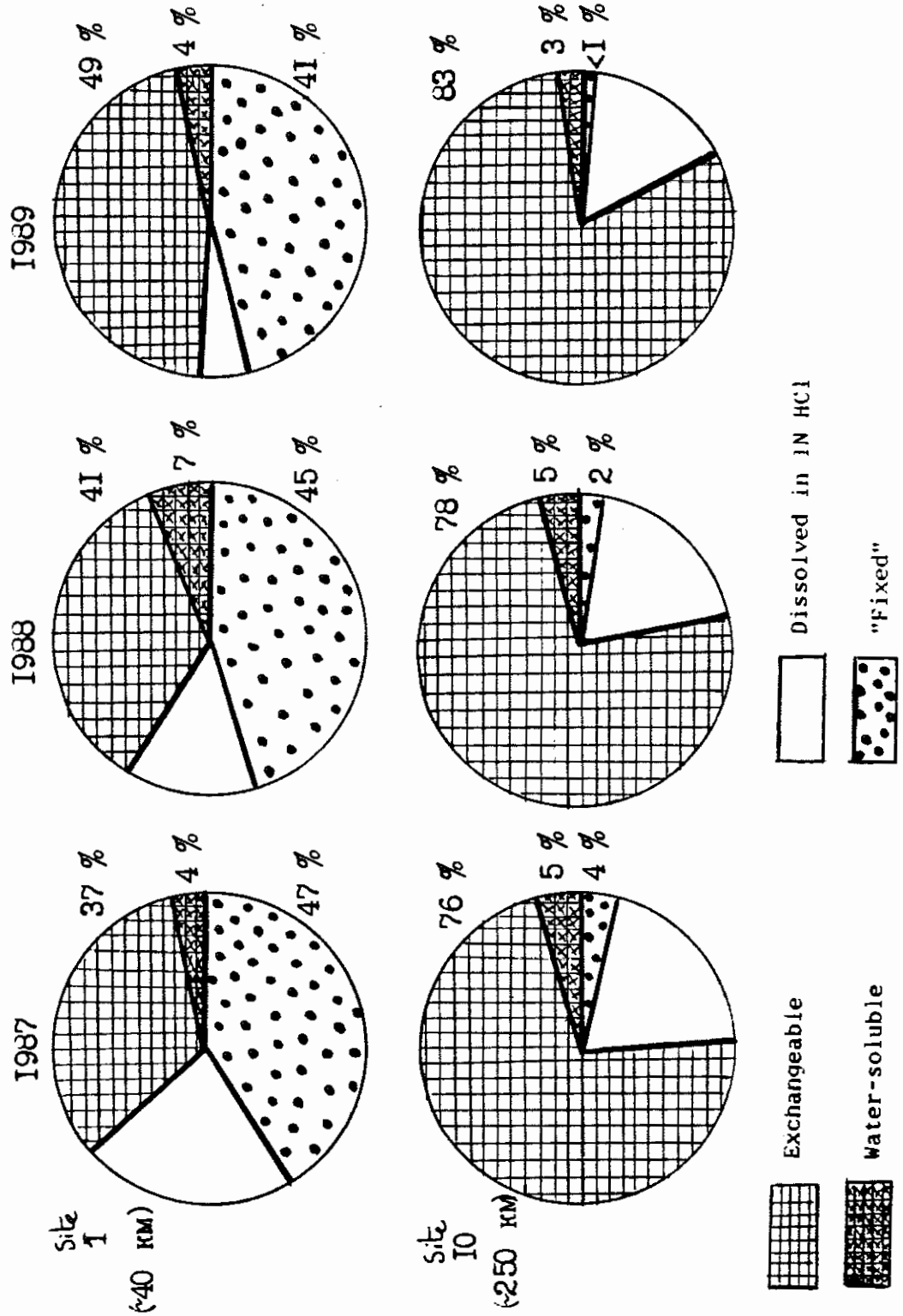
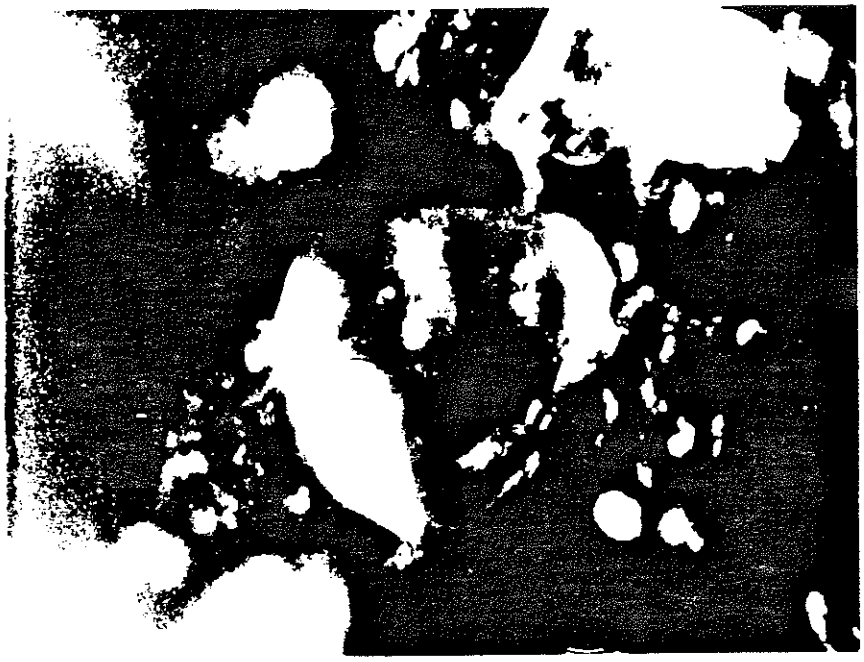


Fig.7: FORMS OF OCCURENCE OF THE RADIOSTRONTIUM IN SOD-MEADOW SOILS

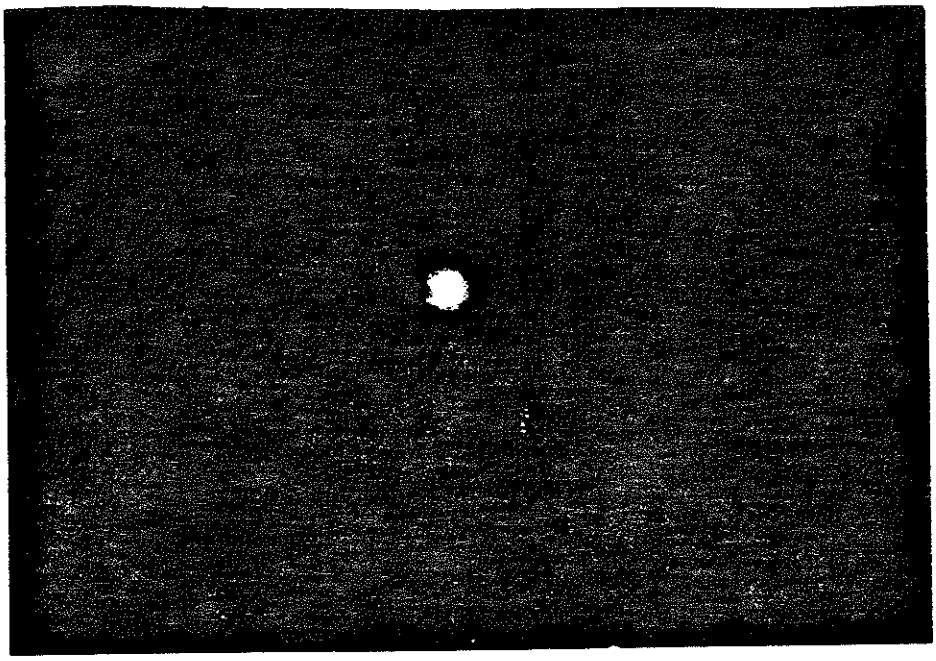




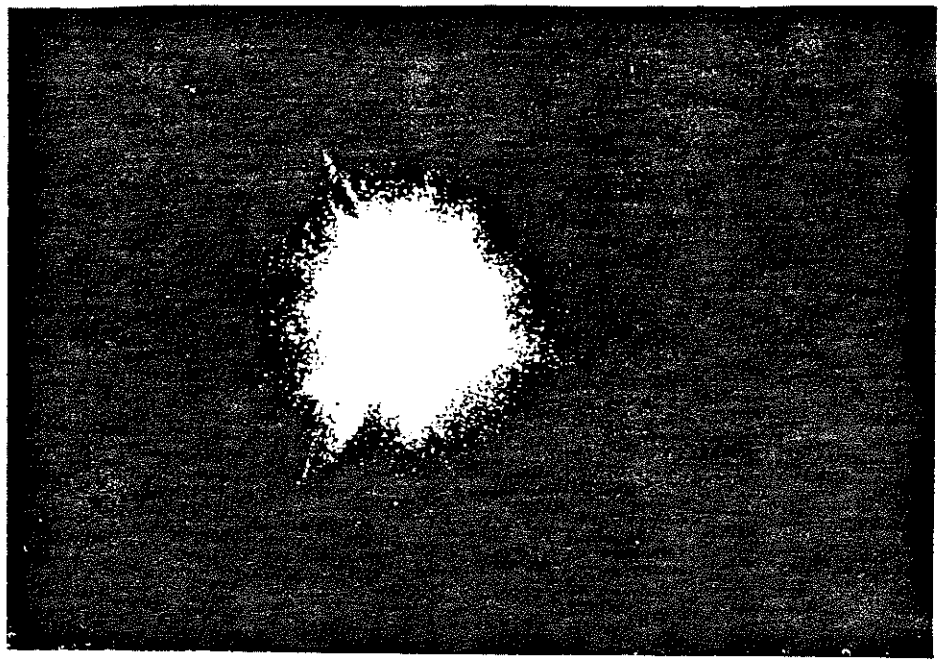
a)

b)

Fig.9: ACTIVE PARTICLES DETECTED AT THE MONITORING SITE 40 km FROM CHERNOBYL NPP. Photomicrograph
a) X 650
b) X 800



a)



b)

Fig.8: TRACES OF ACTIVE PARTICLES FOUND ON THE TERRITORY OF VARIOUS MONITORING SITES
RADIOGRAPHY X4 EXPOSURE TIME: 72 HOURS
a) 200 km from Chernobyl NPP
b) 40 km from Chernobyl NPP

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Hot Particles in Sweden One Year After Chernobyl

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ABSTRACT

Hot particles were collected on ground areas at three locations in Sweden one year after the Chernobyl accident. The aim was to determine the density of the particles on ground surface, the radionuclide and elementary composition, the structure, the size and other aerodynamic properties.
Altogether 36 particles were detected by an end-window GM-tube on the 476 m² total area investigated. The observed surface densities varied between Ø.Ø3 and Ø.Ø7 particles/m².
Based on gamma spectrometric investigations and gross alpha activity measurements the particles found could be classified into two groups.
One group of particles were fragments from the fuel elements, containing Zr/Nb-95, Ce-144, Ru-1Ø3/1Ø6, Sb-125 and alpha emitting nuclides. The highest total gamma activity was 8ØØ Bq with about Ø.7 Bq alpha activity after decay correction.
The Ru-1Ø3 and Ru/Rh-1Ø6 isotopes took almost 95 per cent of the activity in the second group without alpha emitters. The highest individual total gamma activity was over 2Ø kBq after decay correction. The scanning electron microscopic studies of these particles showed an elementary composition of Ru, Mo, Tc, Fe and Ni. The diameters of three of the particles were as high as 9, 11 and 12 microns.
The aerodynamic diameter and density of two of these particles were derived by their fall forced by gravity in air.

INTRODUCTION

The hot particles originated from the nuclear weapons' tests were studied intensively in the '60-s and '70-s [1-3]. These investigations were performed mainly on particles collected by air filters and fall-out samples. However very little is known about the hot particles' fate after their deposition into the environment.
Some of the hot particles found after the Chernobyl accident showed rather unusual properties [4-5]. These particles had very high gamma activity in a size of few micrometers without alpha radiation. The radionuclide composition and the lack of the alpha radiation have denied the possibility to originate this kind of particles from the fuel element material.
To answer these questions hot particles were collected and investigated one year after the Chernobyl accident in Sweden.

METHODS

An end-window type GM-tube (window thickness 2 mg/cm^2, surface 17 cm^2) was used to detect the particles on ground surface. The investigated areas were in grassy parks without trees and any cultivation. The grass was cut several times since the accident, but the first cut was about one and half month after the deposition in all cases.
The detector was kept to the ground surface as close as possible, but the average distance was between 1 and 2 cm because of the grass and ground's roughness.
The in-situ criteria to detect a hot particle was a doubled counting rate to the average background on the given place.
The collected soil samples took usually 100-200 g of mass.
We used a rather simple mechanical separation to prepare the samples for the gamma spectrometric measurements. We got finally soil fragments containing the hot particle with diameters of a few tenth of millimeter up to appr. 1 mm. In 9 cases of the total 45 ones it was not possible to identify a hot particle. Finally 36 particles were separated and fixed between two layers of adhesive paper.
The gamma spectrometric measurements were performed by a HpGe semiconductor detector.
For alpha measurements the samples were cleaned further, usually by the help of an optical microscope. The soil fragments

contained hard silicon crystallic structures, in which the particles were trapped. To remove the impurities and to destroy the silicon structures an organic solvent was used simultaneously with mechanical destruction between two glass plates. This method was repeated until we got the particle on a piece of paper of a few tenth of mm in size and without impurities visible under optical microscope.
The alpha intensity measurements were carried out by Si surface barrier type detectors.
For the scanning electron microscopic (SEM) studies further purification was made by ashing in several cases.
Two particles were prepared for alpha spectrometry by electrodeposition and Pu separation.

GAMMA SPECTROMETRIC INVESTIGATIONS

Based on our gamma spectrometric results the particles could be arranged into two groups (Table 1), in close agreement with the observations published earlier [5-8].
The medians and ranges of the total activities, corrected for the radioactive decay to 26 April 1986 can be seen in Table 1.
The ranges of the total activities illustrate, that the two groups differ considerably not only in the radionuclide composition, but in their total activities too.
The first type of particles, containing mainly Ru-103 and Rh-106 isotopes, by at least 95 per cent of the total activities, differ strongly from the hot particles found after the nuclear weapons' tests [1-3]. In most cases was found a small quantity of Co-60, Sb-125, Cs-134 and Cs-137 nuclides, but the last ones could from the soil impurities of the samples. In several cases we found Cd-109 too, but we could measure only very small activity or none of Zr-95/Nb-95 or Ce-144 isotopes.
The activity ratios of the Ru-103 and Rh-106 isotopes, ranged from 2.6 to 5.7, with the average of 3.7 . These figures correspond to 22500, 8000 and 13000 MWd/tU burn-up levels of fuel elements [6].
The more usual particles of the second type contained Zr-95, Nb-95 and Ce-144 radionuclides, by 60- 80 per cent of the total activity. About 10-20 % of the activity came from Ru-103 and Rh-106. Most of them also contained a small quantity of Cd-109, Sb-125, Cs-134 and Cs-137.

ALPHA MEASUREMENTS

Do not dissolve and loose the hot particles there was measured only the total alpha intensity for all of the samples.
The alpha intensity measurements have proved, that none of the Ru/Rh particles contained detectable alpha emitting radionuclides, i.e. fragments from the fuel element matrix.
This finding might be valuable to explain the origin of these particles. The Ru/Rh particles were claimed by several authors also to be fuel element fragments [5-6]. However our results support Chyssler's data [4] on the chemical composition of these particles and the explanation of Persson et al. [7] of their formation.
Fig. 1 shows the alpha spectrum of a Zr/Nb/Ce particle after electrodeposition. The measured and decay corrected alpha activities of two particles can be seen in Table 2.

STUDIES ON THE PARTICLES' SIZES AND COMPOSITION PERFORMED BY SEM AND AERODYNAMIC MEASUREMENTS

We concentrated on the rather peculiar Ru/Rh particles in this phase of our study.
We could localize, take photos and analyze the elemental composition of the hot particles in five cases. The presence of the particle was judged by X-ray analysis in all cases.
The SEM micrographs of three of the particles can be seen in Fig. 2-4. The surface of the particles was rough and the shape quite irregular, usually, although one of them showed rather regular shape and smooth surface (Fig. 2).
The data on the particles derived from the SEM photos and X-ray spectra are collected in Table 3.
The elemental composition supported the expectations referred in the previous paragraph. The only discrepancy was the presence of another metals, i.e. Fe,Ni and Al in the spectra, but they might come from the sample holder.
The aerodynamical properties of two particles were investigated by their fall forced by gravity in air. The falling time was determined by the help of two GM-tubes positioned in a known distance along a vertical tube. The aerodynamic diameter and the density of the particles could be derived because the geometrical diameters were known. These parameters are also presented in Table 3. The values of the specific activity of

Ru-103 (last line of Table 3) agree well, except of the two particles for which the diameter was derived only from back-scattered micrographs.

THE SURFACE DENSITIES ON GROUND OF THE PARTICLES

Table 4 summarizes the number of particles found together with the observed and corrected surface densities on ground.
It was observed under the preparation of samples, that the particles were found usually in the upper root zone of grass. However, because of the much higher efficiency of the GM-tube for betas we have to take into consideration the absorption of soil. It can be stated from these results, that we could probably detect a Zr/Nb/Ce type particle of the highest activity no deeper in soil than 3-6 mm, but we likely could not observe particles of activities lower than 90-220 Bq at all. For the Ru/Rh type particles the depth limit of the detection was between 7 and 9 mm for the upper end of the activity range, and 1-4 mm for the particles of the lowest activities.
These results give an acceptable explanation of the difference between the ratio of the numbers of particles of two types observed by us (Table 4) and published earlier. In Sweden a 5 to 41 [9], in Finland a 1 to 8 [6] ratio of the Ru/Rh and Zr/Nb/Ce particles were estimated. Our result (2 to 1 as an average) is much higher than these values.
The surface densities of the particles were corrected based on the detection limits and presumptions on the real activity distribution and initial densities on ground [10].
According to the corrected values (Table 4) a 15 per cent fraction of the Zr/Nb/Ce particles was still in a depth of up to 3-6 mm one year after the accident, indicating an environmental half-time of 130 days. 70 per cent of the Ru/Rh particles was in a depth of up to 3-9 mm, suggesting an environmental half-life of 700 days.
The ratio of the corrected surface densities of Ru/Rh particles in Gavle and Stockholm or Gotland (see Table 4) differs strongly from the ratios of the Cs-137 depositions on that areas. The last values were 2-3 kBq/m^2 in Gotland and Stockholm, but 100-120 kBq/m^2 in Gavle [7]. The most probably explanation of this discrepancy is, that the hot particles' deposition occurred mainly in the form of dry deposition, and was not affected considerably by the amount of precipitation.

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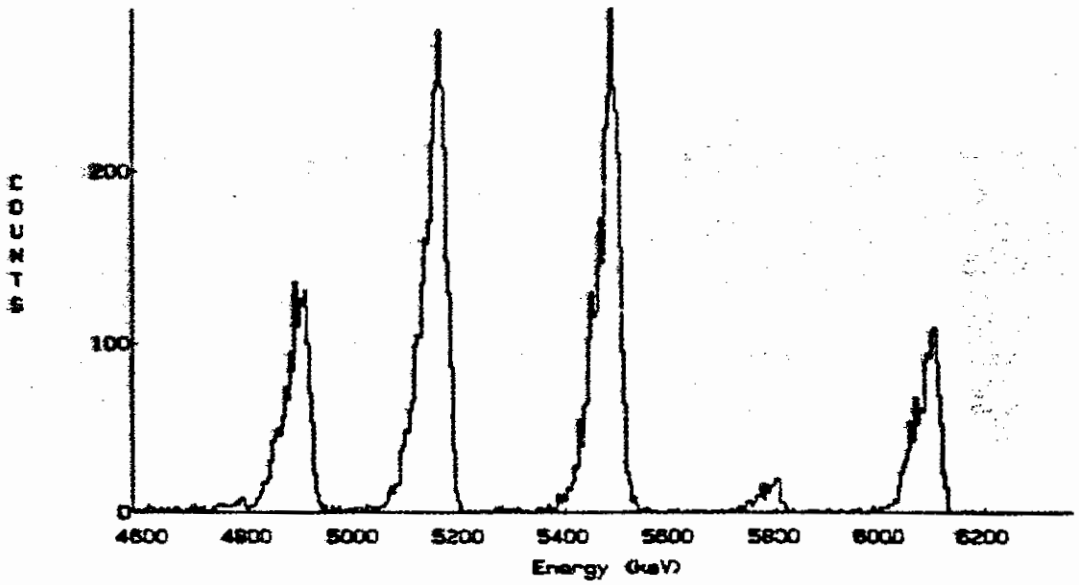

Fig. 1. The alpha spectrum of a Zr/Nb/Ce type particle after electrodeposition

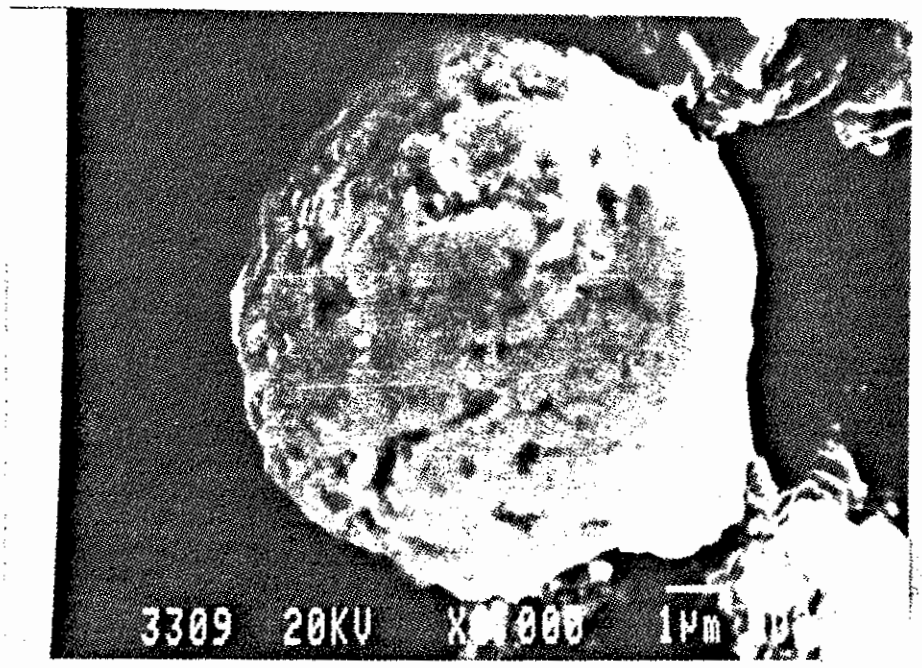

Fig. 2. The SEM micrograph of particle HAGA1

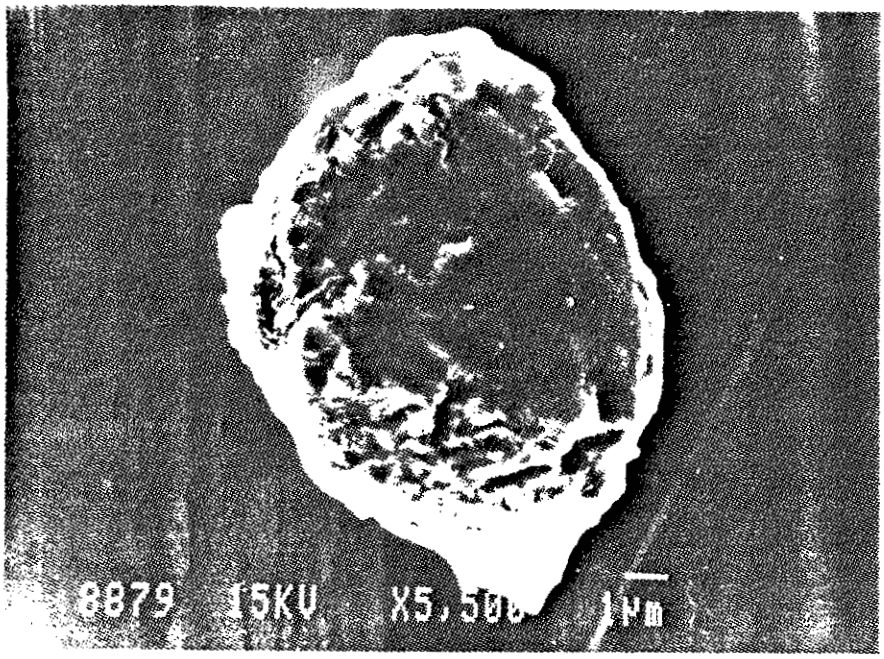

Fig. 3. The SEM micrograph of particle GUAN1

Fig. 4. The SEM-micrograph of particle GUAN2

Table 1. The gamma emitting radionuclide composition of the hot particles

	Total gamma activity* (Bq)		activity (per cent of the total)				
	median	range	Zr/ Nb-95	Ru-103/ Rh-106	Cd-109	Sb-125	Ce-144
1.	2000	320-22000	+	95	+	1	1
2.	400	25-770	60	20	1	1	15

* decay corrected for 26 April 1986

\+ detectable, but less than 1 %

Table 2. Alpha activities of two particles from the Zr/Nb/Ce group

Particle	Alpha activity in mBq (corrected for 26 April 1986)			
	Pu-239 /Pu-240	Pu-238 /Am-241	Cm-244	Cm-242
HAGA11	14.5	11.0	0.69	105
HAGA12*	53	50	3.6	580

* the particle was fragmented under preparation

Table 3. The information on the Ru/Rh particles derived from SEM investigations and aerodynamical experiencies

Particle	HAGA1	GUAN1 !	GUAN2 !	HAGA9	GAVLE3
total act.,kBq	22	49	41	4.4	9.8
composition*	Mo,Tc,Ru	Ru,Tc,Mo	Ru	Ru,Tc,Mo	Ru,Rh
diameter, μm	9	11.5	10.7	3.9+	2.4+
aerodyn. diam., μm	29	37			
density,g/cm³	10.3	10.7			
Ru-103 spec.act., Bq/μm³	49	50	46	116+	1097+

! particles found before the planned study
* Al,Ni,Fe are also present in most of the cases
+ uncertain, derived only from backscattered SEM photo

Table 4. The number and surface densities on ground of the hot particles

	surface density on ground, 1/m²		total	
	Ru/Rh group	Zr/Nb/Ce group	number	area, m²
		Observed		
Stockholm	0.034	0.040	13	177
Gotland	0.048	0.048	10	105
Gavle	0.068	-	13	190
		Corrected		
Stockholm	0.072	0.16		
Gotland	0.082	0.11		
Gavle	0.18	-		

Biological Monitoring of the Deposition and Transport of Radioactive Aerosol Particles in the Chernobyl NPP Zone of Influence

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It is well known by now that, as a result of the Chernobyl accident, a large quantity of finely dispersed radioactive aerosol (consisting of an extremely heterogeneous mixture of fuel and radionuclide particles of varying composition, sizes and physico-chemical properties) was deposited in the environment. Particles with high individual activity were earlier termed "hot" particles in the specialist literature. [1] According to some official data, the quantity of finely dispersed fuel ejected during the Chernobyl accident in the form of "hot particles" is estimated at 6-8 t, not including the "fuel dust" which remained within the enclosure or "sarcophagus".

The accident coincided with the active vegetation period of the higher plants in this region, and a considerable proportion of the radioactive particles settled on the leaves of trees, shrubs and herbaceous plants. Our initial research (one month after the accident) into the fallout of radioactive particles onto the leaves of plants in the Chernobyl NPP 30-km zone and in the city of Kiev, led us to draw certain conclusions as to the dynamics of behaviour and forms of the activity deposited, and demonstrated the expediency of using plants' leaf blades to solve geophysical problems and as the object of radiobiological experiments (in much the same way as traditional screens are used).

Plants are one of the main links in the trophic chains of radionuclide transport. The role of plants in such transport was studied mainly in relation to soluble compounds of radionuclides (as in the case of the Kyshtym accident, for example [2]), or to global fallout in which radionuclides were in soluble or exchangeable forms. The specifics of the Chernobyl accident led to the radioactivity occurring in particular forms, and the kinetics of radionuclide migration within trophic chains sometimes vary considerably from what was established in earlier experiments. It is important to study the interaction between plants and "hot particles", whose physico-chemical properties determine their non-solubility, which is characteristic, for example, of the carbides and oxides of some metals. When particles come into contact with plant surface tissues, "dissolving" factors come into play such as changes in the acidity of the solution or interaction with complex-forming compounds and organic materials exuded by the leaves of some plants. Thanks to these factors, many plants are capable of extracting compounds of low solubility from the soil minerals.

Making use of macro- and micro-radioautography, we set out to estimate the rate of conversion of low-solubility radionuclide particles into biologically mobile forms of radionuclides accessible to plants; to study the density of fuel particle fallout in the near-ground layer of the atmosphere and to assess how this varies at different distances from the fallout source over time (during the four years following the accident, 1986-1989); to study the size of the particles deposited on the leaves of plants at various strata, their activity, morphology and behaviour when kept in the form of herbarium exhibits; and to assess the contribution of alpha-active particles to the general amount of fallout and how it changes over time.

Methods and materials

From June 1986 to the present we collected leaves of plants at specific points in the Chernobyl NPP zone of influence (2, 5, 10, 15, 30 and 90 km from the wrecked unit), noting down every one or two months the stratum in which we found them, i.e. their distance from the ground surface, during the whole vegetative period (from April to October). In some experiments samples were collected more frequently.

The leaves thus gathered were dried in herbarium meshes and the part intended for radioautographic analysis processed as herbarium exhibits, which remain preserved in an undamaged state to this day. To assess their total activity the leaves were ground and, using a widely accepted method, prepared for gamma-spectrometric analysis of contamination.

"Hot" particles on the leaves were identified by bringing the leaves into contact with various radiographic films (RT-5 or RM-V) for a length of time specific to each experiment. We developed the films by a standard method, counted the number of dark blotches at the various particle locations, and determined the diameter of the blotches (this being known to correlate to particle activity).

To study individual particles we matched their autographs on the film against the specimen, cut out a section and isolated a given particle under an optical microscope; we then ascertained its size and, where necessary, photo-recorded it under the microscope. Gamma-spectral analysis of individual particles and beta-radiometry were carried out, and further radioautographs taken one or two years later for comparative analysis of their decay (if this formed part of the researcher's brief).

To identify the alpha-active particles we used solid-state track detectors (CN-85, CR-39 and LR-115) which, after being brought into contact with a specimen, were processed as specified in the accompanying instructions and analysed under an optical microscope. We used the same solid-state detectors to identify fission fragments following neutron activation of the specimens via a 10^{12} cm^{-2} neutron flux in the microtron-22 belonging to the Nuclear Research Laboratory of the Joint Institute for Nuclear Research (Dubna, Moscow region). The number of alpha-tracks was determined under a microscope to identify the uranium and plutonium nuclides.

An overall estimate of fallout over space and time was obtained via the horizontal gauze screen method involving gamma-spectrometric analysis of the screen contents.

Results

Since completion of the "sarcophagus" enclosing the wrecked reactor (November 1986), radioactive aerosol particles in the near-ground layer of the atmosphere have contributed to secondary contamination mainly through being resuspended from the ground by the wind. In order to learn more about this process we monitored the intensity and coefficient of wind-uplift throughout the year, drawing on gradient-related observations of the volumetric radionuclide concentration in the near-ground layer of the atmosphere, together with daily measurement of the volumetric concentration in the towns of Pripyat and Chernobyl and of radionuclide deposition across a network of points within the Chernobyl NPP 30-km zone.

Fig. 1 shows the change over space and time in the caesium-137 fallout value, based on screen measurement data obtained along the Pripyat-Zeleny Mys axis. A steady drop in the fallout value can be seen over time (this being particularly sharp in the first half of 1988 but slowing down in 1989). The values for "hot" particle deposition density evolve in a similar manner over distance and time (Fig. 2); the number of "hot" particles deposited on the leaves of plants in 1988 was five times lower than that deposited in 1986 in the area 5 km from the reactor (Kopachi, Novoshepelichi), and in the 30-km area (Otashev) 25 times lower.

Thus, radioactive aerosol emission into the atmosphere as a result of natural wind-uplift and other factors stabilises to a certain degree.

The data presented were obtained from radioautographic analysis of the activity deposited on plant foliage; some specimens are shown in Figs. 3-5. As we can see, in 1986 there was no difference in the fallout density of particles deposited on leaves in the various strata: it is the same for leaves growing 10-50 cm above soil level and for those 2 metres above it. However, the situation changes in 1987 and 1988, when the largest "hot" particles display a tendency to collect on the leaves of near-ground whorls. A decrease in the density of radioactive particle deposition on leaves observed at certain points in the area in 1988 and 1989, made it very difficult to carry out a representative autoradiographic analysis, thus leading to a 25-30% increase in measurement error. During these periods particle fallout density was assessed from leaves in the near-ground stratum (plantain, burdock, cinquefoil, etc.).

We assumed that the diameter of a dark blotch on the radiographic film was related to the activity producing the particle autograph [1], and - using this parameter - we plotted particle distribution at two points in the Chernobyl NPP zone - 5 km and 30 km from the reactor (Fig. 6). The main category at both points is that of fine, low-activity particles producing blotch aureoles smaller than 1 mm, but, as the histogram shows, at distances over 30 km there are no heavy and highly active particles (which produce dark aureoles larger than 10 mm). However, the complex character of the particles specific to Chernobyl prevented us from establishing the type of direct correlation between particle activity and size obtained via the well-known formula for global fallout. [1] Many of the particles consist of all possible combinations of an active nuclide with a non-active carrier. Therefore, to study the correlation between particle radioactivity and size we need to check the particles' size and morphology directly under the microscope, and an electron microscope is required to study the size and morphology of particles under 5 microns.

In view of the great complexity involved in singling out individual hot particles and determining their gamma, beta and alpha activity plus their microscopic parameters, we looked at a possible direct method of determining the size of particles from their alpha-activity track in thin layers of BYa-2 photoemulsion. Although this method can be used, it is more suited to studying a limited number of particles since it is very laborious and painstaking and ties up a large number of microscopists.

To assess the contribution of alpha-active particles to overall mass we used solid-state track detectors brought into contact with the particle at its location on the specimen (leaf) as indicated by the autograph on the RT-5 or RM-V type radiographic films after matching with the specimen. The alpha-activity tracks detected are shown in Fig. 7. The only drawback to this method is the difficulty of obtaining good makes of solid-state detectors from abroad.

As for the physico-chemical properties of the Chernobyl particles, a large number of reports - by researchers in various parts of the world - have appeared on this subject over the last two years.

An important feature of the "hot" particles studied by us is that since being collected they have been preserved in dry-air conditions, i.e. on dried leaves collected at various times since the accident, the earliest being 6 June 1986. This means we can see how the dynamics of change in the particles' activity and morphology relate to conditions at their location, given that most studies concern research into "hot" particles in the soil.

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Figure 1

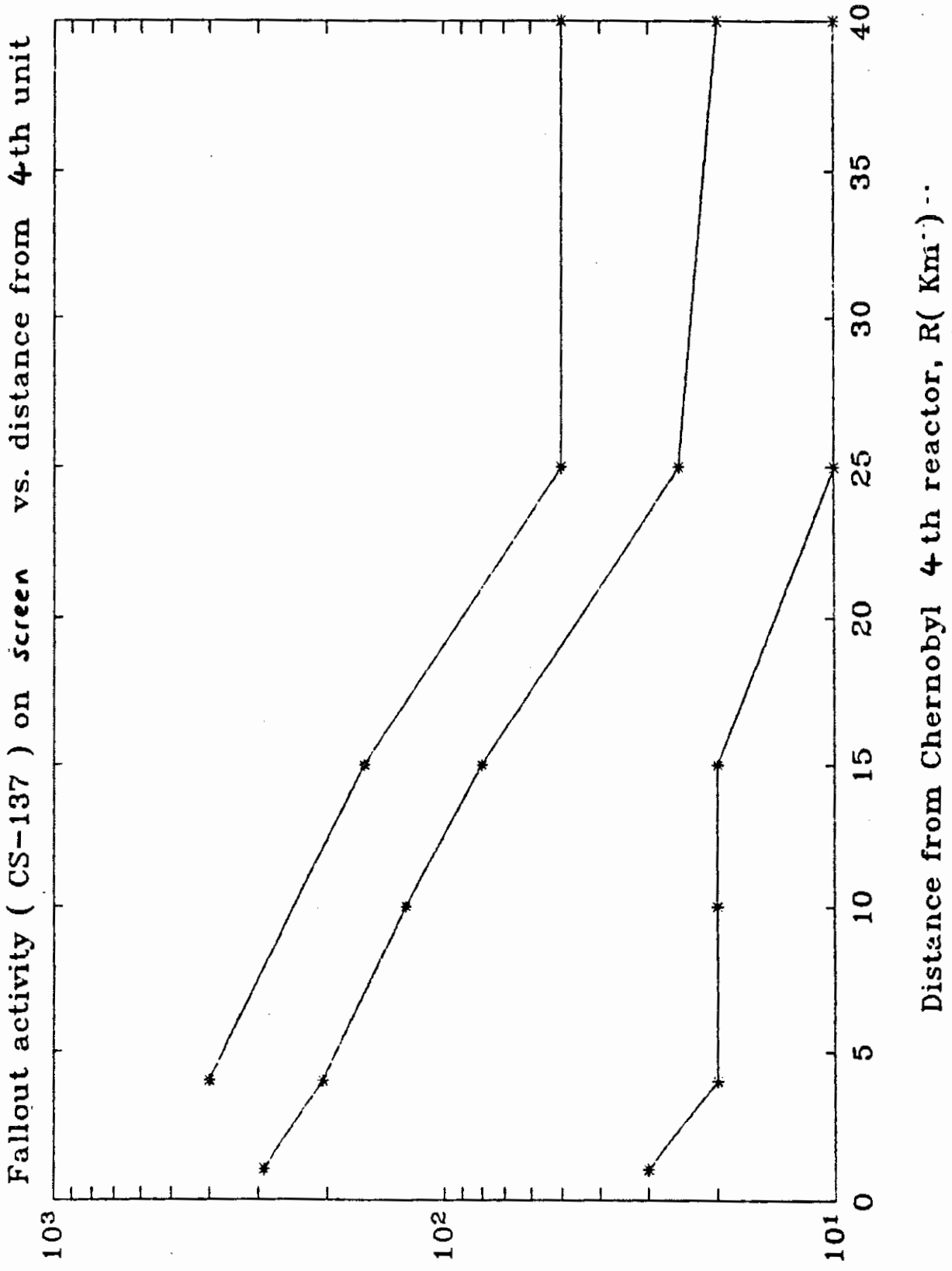
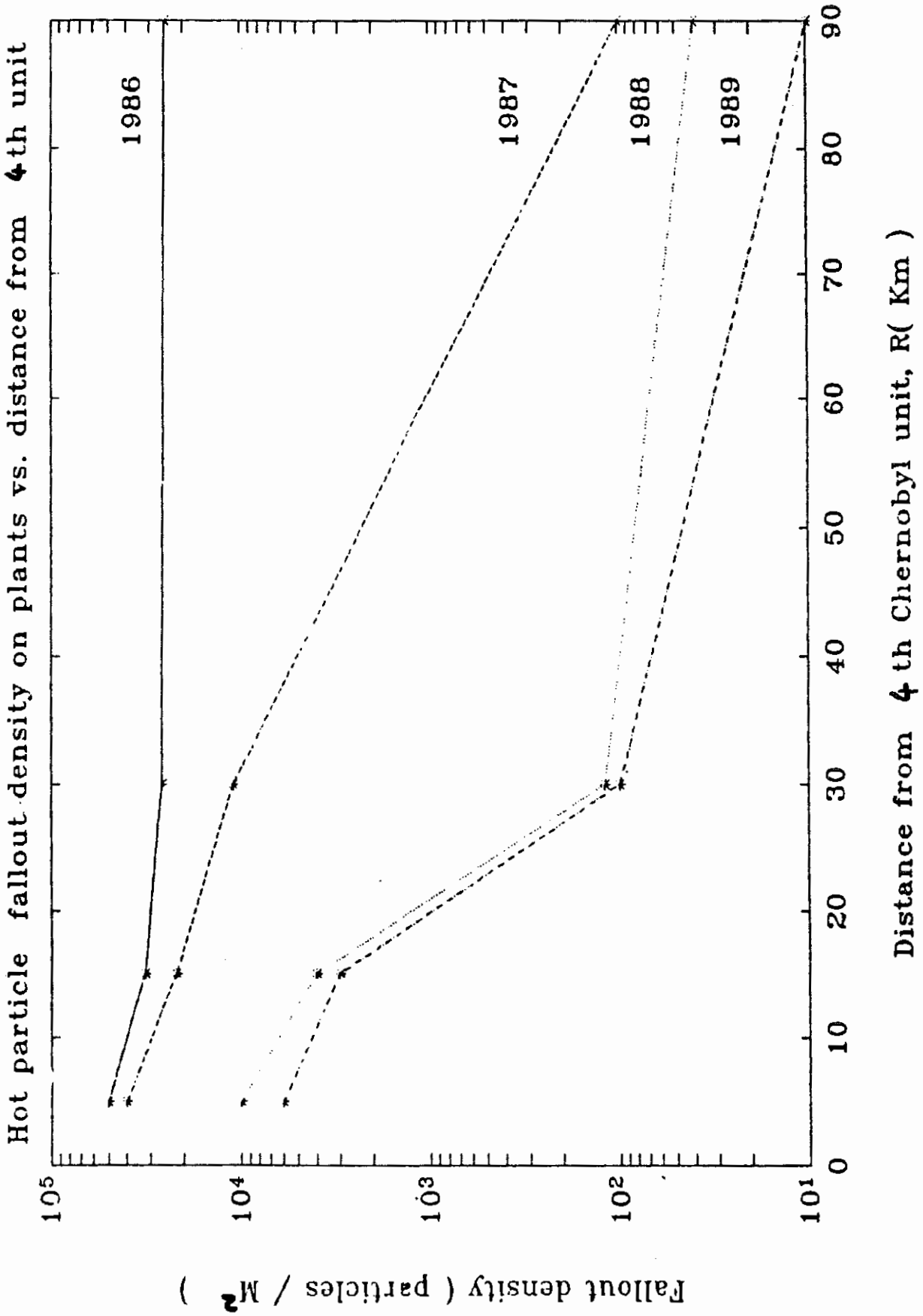

Fallout activity (CS-137) on screen vs. distance from 4th unit
Fallout activity (Ci / Km²)
10³
10²
10¹
0
5
10
15
20
25
30
35
40
Distance from Chernobyl 4th reactor, R(Km)

Figure 2


Hot particle fallout density on plants vs. distance from 4th unit
1986
1987
1988
1989
Fallout density (particles / M²)
Distance from 4th Chernobyl unit, R(Km)
10⁵
10⁴
10³
10²
10¹
0
10
20
30
40
50
60
70
80
90

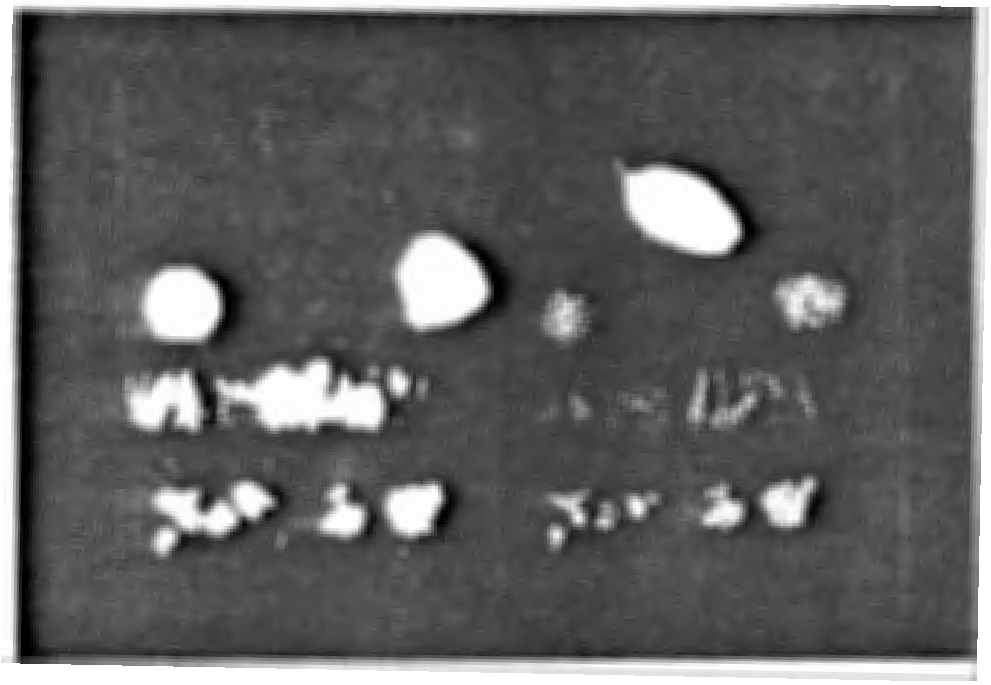
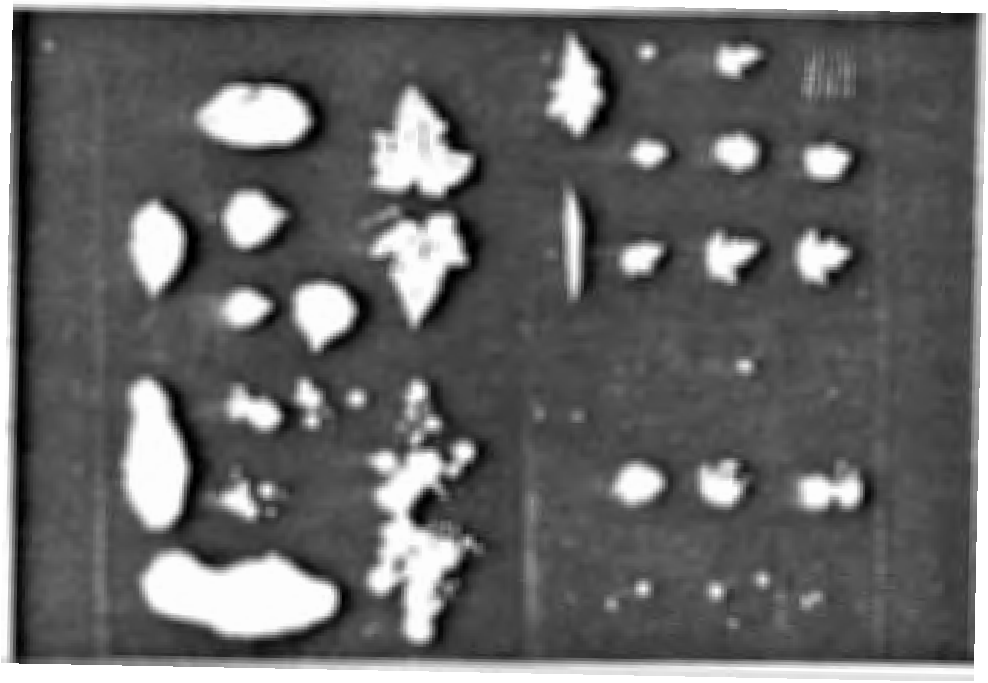


Fig.3, 4 Photos of leaves and their radioautographs; collected in 1986 in the Chernobyl NPP 30-km zone.



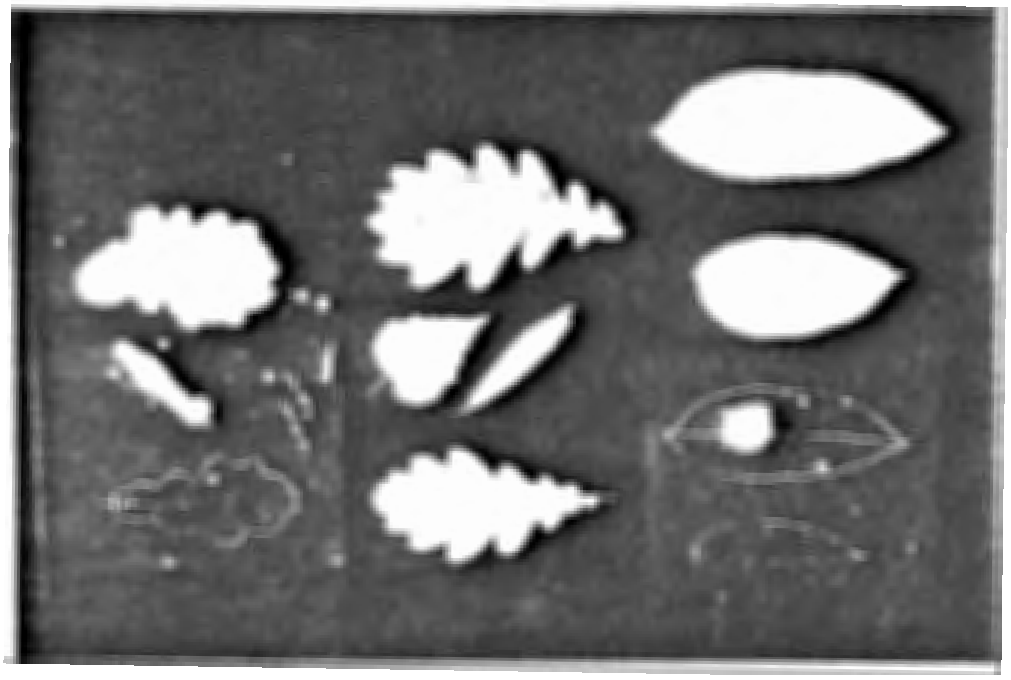
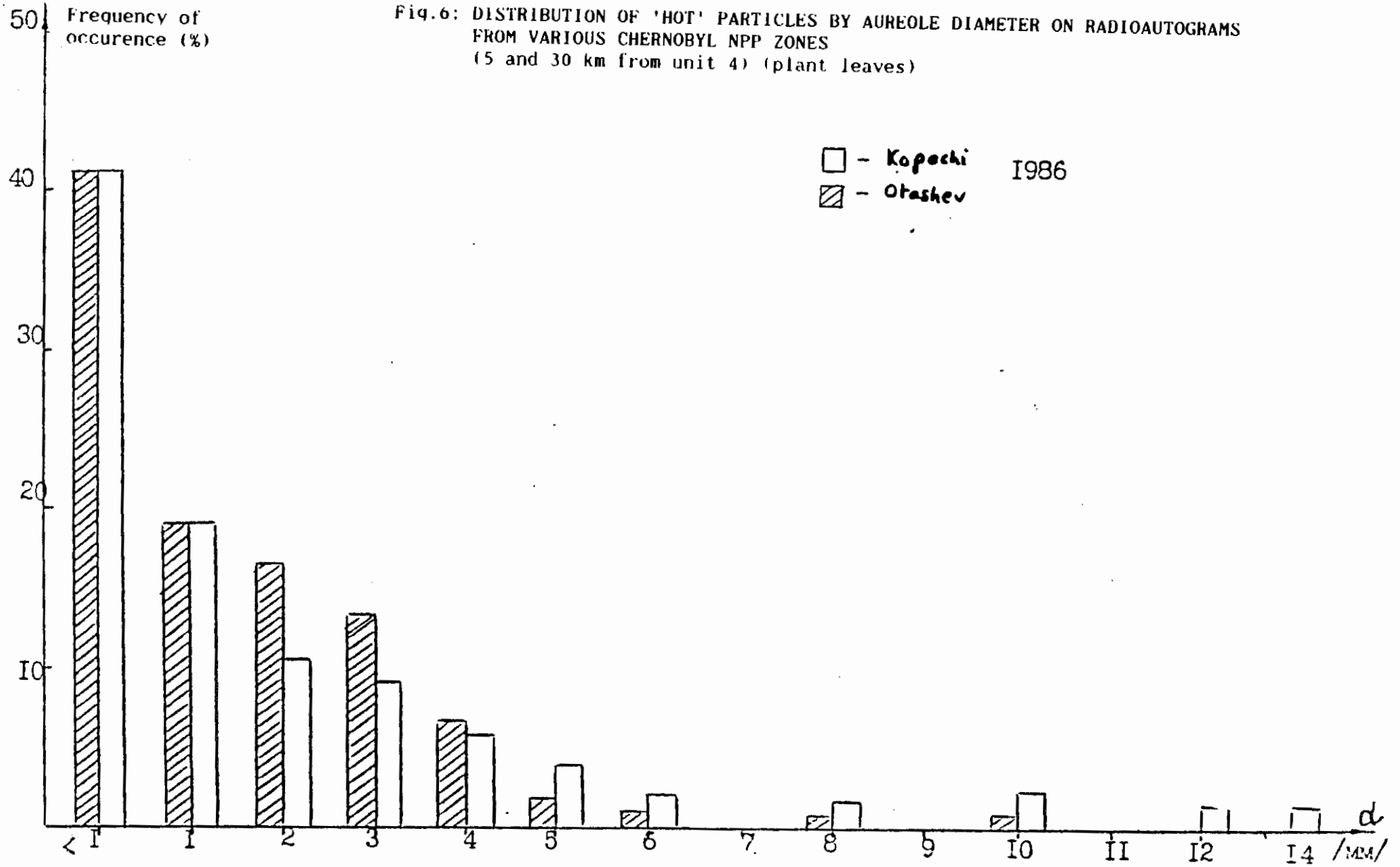


Fig.5 :Photo of leaves and their radioautographs; collected in 1986 in the Chernobyl NPP 30-km zone.

Fig.6: DISTRIBUTION OF 'HOT' PARTICLES BY AUREOLE DIAMETER ON RADIOAUTOGRAMS FROM VARIOUS CHERNOBYL NPP ZONES
(5 and 30 km from unit 4) (plant leaves)



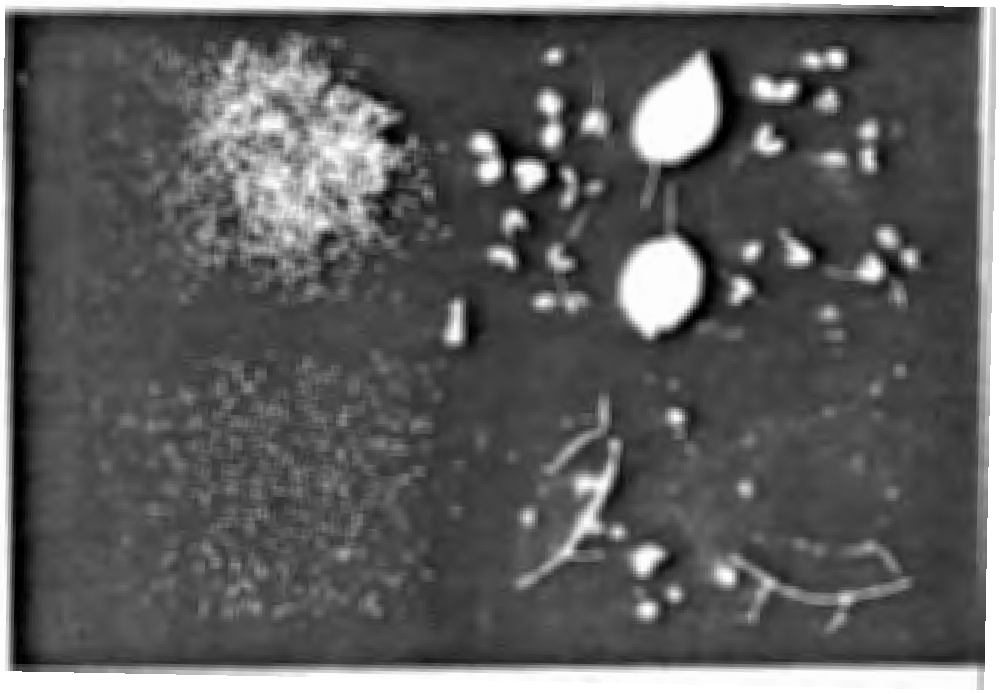


Fig.7 Tracks of alpha-activity (obtained using solid-state track detectors) on leaves collected 30 km from the reactor (edge of zone) in 1987.

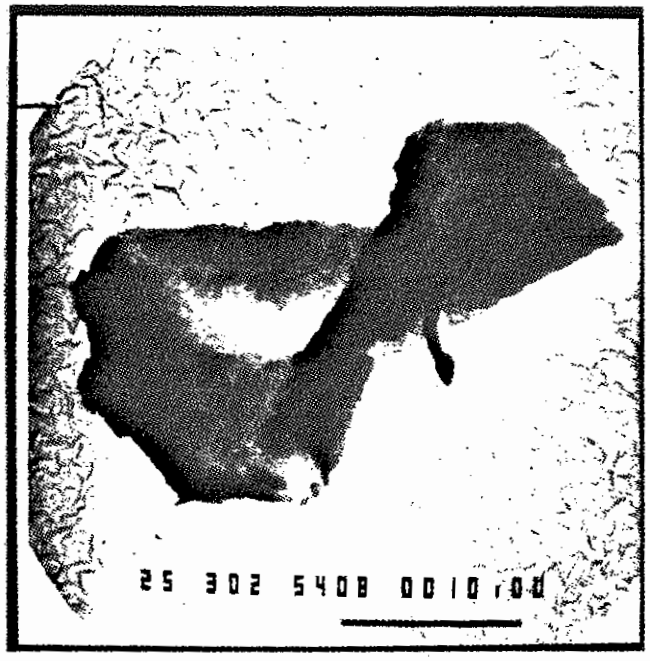
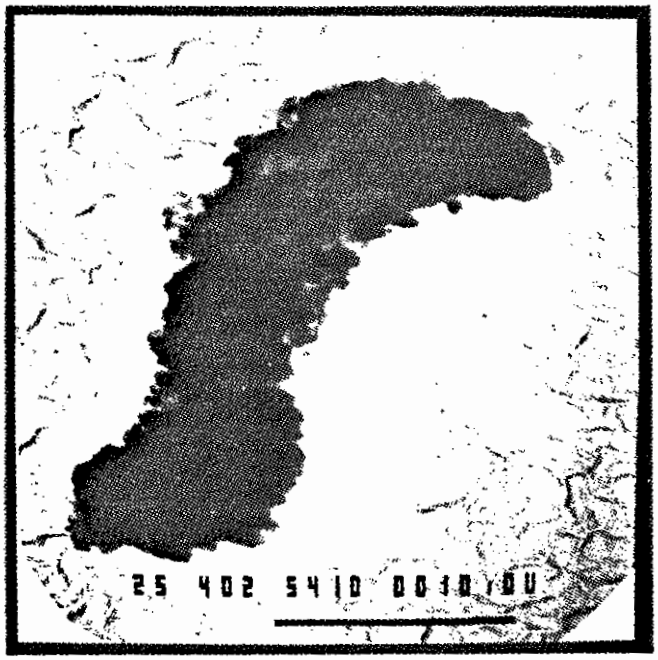


Fig.8 General view of "hot" particles on scanning electron microscope.



Deposition and Resuspension of Radiocaesium after Chernobyl

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ABSTRACT

An understanding of the processes of deposition and resuspension is required for evaluating the consequences of planned and accidental discharges of radionuclides. Opportunities for the observation of these processes following a widespread dispersal of material are rare. The Chernobyl accident provided an opportunity to measure deposition and resuspension of caesium isotopes, which were dispersed beyond the borders of the Soviet Union chiefly as sub-micron particles.

Direct observation confirmed that dry deposition of such particles is very slow, and allowed dry deposition velocities to be evaluated for rural and urban surfaces. For grass in Western Europe, the deposition velocity was about 0.5 mm s^{-1} although a few measurements gave higher results. Similar values were found for roofs.

Where rain occurred, wet deposition was dominant, but measurements of wet deposition parameters during individual rain events were very variable. A more useful estimate of the rate of removal by rain was derived from the variation of the concentration of radiocaesium in air over a period of several weeks following the accident. This approach indicated a mean of 330 for the washout ratio.

After a period of rapid removal from the atmosphere, concentrations of ^{137}Cs in air have remained measurable throughout Europe. The results of measurements indicate that resuspension is responsible, and that local variations in the amount deposited soon after Chernobyl are reflected in the resulting air concentrations. Traffic appears to be a significant mechanism at some locations. Other local effects are apparent as seasonal differences in concentration. However, resuspension appears to vary systematically with the amount deposited across Europe, despite the large range of climate. There is some indication of long range transport of resuspended caesium. The results may be used to assess the applicability of resuspension models in Europe. Models which indicate resuspension factors at the lower end of the range are consistent with the data. The rates of deposition of resuspended activity were extremely variable and unexpectedly large in some locations, suggesting that the contamination of crops and surfaces by resuspended material is potentially a significant process.

1. Introduction

Values of the parameters describing dispersion, deposition and resuspension are required for use in models for predicting the consequences of routine releases as well as accidents. The values used are based chiefly on laboratory or field experiments of limited scale, or on experience at nuclear facilities in North America. Data collected in Europe following the Chernobyl accident increase the available information, and extend it to include the moist temperate climate of Western Europe.

Several sampling networks operating in the countries of Europe have provided data that can be used to improve or confirm the parameter values in use. Here we summarise some of the available data, using ^{137}Cs as an example. Dry and wet deposition are discussed as well as resuspension.

2. Dry Deposition

Several authors in Europe determined the activity on grass and the soil surface after the peak concentration in air had occurred. Where rain did not fall, the deposition velocity, V_g, could be determined:

$$V_g \text{ (m s}^{-1}) = \frac{\text{activity deposited on grass, (Bq m}^{-2})}{\text{mean airborne concentration (Bq m}^{-3}) \text{ x duration (s)}}$$

In a few locations, direct measurements were made of activity on building surfaces and roads, either by sampling methods or by in situ gamma spectrometry.

Table 1 summarises several measurements. The data for grass provide valuable confirmation of the prior understanding of dry deposition. Many previous measurements and theoretical considerations indicated a deposition velocity of about 0.1 mm s^{-1} for particles in the range 0.1 to 1.0 μm, with a sharp rise for larger particles. Measurements of the size of airborne particles carrying ^{137}Cs from Chernobyl (12,13,14) showed that over Germany, Scandinavia and Switzerland the particles were principally 0.1 to 1 μm in diameter, with a small fraction up to 5 or 10 μm, and the contribution of these larger particles to deposition explains the observed deposition velocities, which generally cluster about 0.5 mm s^{-1}.

The results for building surfaces are also generally consistent with expectation, with very low deposition velocities for walls, and values for roofs and roads comparable with those for grass.

3. Wet Deposition

A number of direct observations of wet deposition have been reported. Generally, models require the fractional rate of removal by rain, λ, but the measurements have been used to determine the dimensionless washout ratio

$$W = \frac{\text{activity per unit mass of precipitation (Bq kg}^{-1})}{\text{activity per unit mass of air observed at ground level (Bq kg}^{-1})}$$

If the rate of rain is p and the vertical extent of the (assumed by well mixed) plume is H, λ can be related to W

$$\lambda = Wp/\rho H$$

where ρ is the density of air. In practice, the vertical profile of material in the atmosphere is rarely observed. Thus, λ can seldom be determined directly from measurements. Examination of a small number of data (Table 2) is sufficient to show that values of W vary widely. There are a number of possible explanations, but the most significant are probably (i) variations in the relationship between the concentration measured in air at ground level and that present at rain-forming altitudes, (ii) temporal variations in the airborne concentration so that the mean concentration during the air sampling period is not representative of that present during the (often much shorter) period of rain. These variations lead to difficulties in the collection of representative washout data.

Fortunately, another approach may be used. During the period from about 20 to 80 days after the Chernobyl accident, weekly air concentrations at several stations in the northern hemisphere were similar (Fig. 1), suggesting that on this averaging time the Chernobyl activity was fairly well mixed through the northern half of the troposphere. Moreover, during this period the concentration declined exponentially with a mean residence time of about 8.7 days. Given that the mass of the troposphere

is about 9000 kg m^{-2} and the precipitation rate for May to July averaged over the northern hemisphere is about 3.1 kg m^{-2} d^{-1}, and noting that wet deposition was far more important than dry deposition for Chernobyl caesium, the mean washout ratio can be deduced as

$$W = \frac{9000}{3.1 \times 8.8} = 330$$

This value is broadly consistent with values determined for the trace elements that occur in small particles (15). This value of W implies a mean value of 3×10^{-5} s^{-1} for λ, if a representative value of 3 mm h^{-1} is assumed for the precipitation rate. These parameter values are rather smaller than those used to model Chernobyl dispersion. For example ApSimon et al. (16) used $\lambda = 5 \times 10^{-5}\ q^{0.8}$ (where q is the precipitation rate in mm h^{-1}), equivalent to a mean effective value of about 8×10^{-5} s^{-1}.

4. Resuspension

4.1 Introductory remarks

By autumn 1986 it was obvious that the concentration of radiocaesium in the atmosphere was not continuing to fall at the rate observed from May to July. Possible explanations, including return of some material injected into the lower stratosphere and continuing dispersion of material from the vicinity of the damaged reactor, were considered. However these hypotheses could not account for the dependence of the air concentration on local deposition. It is now clear that resuspension of material deposited within the vicinity of each sampling point must explain most of the continuing air concentrations.

Previous studies of resuspension have included observations from nuclear weapon testing grounds, nuclear plant in America and the surrounding of Palomares in Spain. The only information relevant to the humid climate of North West Europe had been gleaned from wind tunnel investigations and small scale tracers experiments. To improve this data base, the IAEA has provided support to an assessment of the considerable amount of data available in environmental measurements during the years following Chernobyl.

Resuspension has the potential to cause persistent air concentrations and to redistribute material on the ground. Inhalation and contamination of crops may result in internal radiation exposures, and the contamination of previously clean areas causes the redistribution of external radiation doses. All three pathways need to be assessed in predicting accident consequences.

The resuspension factor

$$K(m^{-1}) = \frac{\text{Concentration in air, } C_a \text{ (Bq m}^{-3})}{\text{Surface deposit, d (Bq m}^{-2})}$$

has often been used to describe observations of resuspension. Both numerator and demoninator are directly measurable, but using K implies an equilibrium relationship between these two quantities which may be achieved only over an extensive area of uniform deposit. In principle, when the deposit varies spatially, concentrations would be better predicted using the resuspension rate,

$$\Lambda\ (s^{-1}) = \frac{\text{resuspension flux, R (Bq m}^{-2}\ s^{1})}{\text{d (Bq m}^{-2})} \ .$$

Thus Λ is the fraction removed per second by resuspension. Unfortunately this quantity cannot readily be deduced from field observations, but, if it were known, its use with a suitable dispersion and deposition model would enable the movement of contamination from place to place to be predicted. Such an approach is necessary for calculating air concentrations due to resuspension downwind of a contaminated area.

Whether Λ or K are used, it is clear that the value of the parameter must be expected to vary with many environmental variables. Perhaps the most important of these will be time after deposition, surface structure, nature of contaminant, wind speed, surface moisture and rate of mechanical disturbance of the surface. Measurements after Chernobyl must reflect all these, but it is unlikely that the data will contain sufficient detail to allow the influence of more than one or two to be distinguished.

4.2 Air concentrations

Sequences of two or three year's air concentration data are available for several stations across Europe. Where the deposition on the ground is known, K can be calculated. The results show a general decline of K with time, t, after deposition. A simple exponential function gave a reasonable description of the decrease with time for all stations (Table 3 and Fig. 2; see reference 17 for data sources). Results have been analysed for twenty stations in all. Some were unsuitable for the time trend analysis in Table 3. This data set includes a range of three orders of magnitude in initial deposit, d, and it is clearly apparent that the air concentration, averaged over suitable periods after deposition, does increase with d. However, the increase was less than proportional to d, so that K decreases as d increases (Fig. 3). This trend may reflect long range transport of airborne resuspended material from sites of high deposition to those of low deposition. Another explanation may relate to an effect of deposition mechanism. Areas of high deposition received their ^{137}Cs in rain, and this material may be less available for resuspension than that deposited in dry conditions.

Several models have been proposed for the resuspension factor. Some of the commonly used models are shown in Fig. 4, and the range occupied by the exponential functions fitted to the data is also shown. Clearly the results are more nearly consistent with the models that predict the lowest concentrations.

A number of details may be seen in the results. At some stations pronounced seasonal effects may be seen in the results, but it has not yet been possible to indicate the cause. At coastal sites on the island of Bornholm and at Risø (18), there is an effect of wind direction, which would be explained if most of the resuspended material measured derived from land areas within 20 km of each site. Finally, results at Risø (18) and also at Harwell (19) clearly indicate the effect of traffic.

4.3 Deposition of resuspended material

Several sites deploy deposition collectors. Often these are funnels which collect dry deposition as well as rain, and the results of measurements of deposition during the period influenced by resuspension are useful since

they give some information on the possible contamination of crops and other materials due to resuspension.

The results show some evidence of a relationship between the amount of resuspended material deposited during the year from August 1986 to July 1987 and the initial deposit (Fig. 5). However there is a broad variation in this relationship. At some sites the extent of deposition of resuspended material is quite minor, only a percent or so of the initial deposit, while at other locations it is as large as the initial deposit. This may reflect variations in the surroundings of the sites, but may also be influenced by sampling techniques. If the resuspended material were available for crop contamination, it might make a significant contribution to doses.

The resuspended material also exhibits a wide range of total deposition velocity (ie the long term average deposition flux including rain and dry deposition divided by the air concentration). At some stations this is of order metres per second indicating a contribution of very large particles, which can only be of very local origin, and local circumstances which favour or prevent the resuspension and sampling of particles several tens or hundreds of microns in diameter may be crucial in determining the result of the deposition measurement.

Conclusions

Results from measurements following Chernobyl have confirmed the expected dry deposition rate for small particles. The analysis of wet deposition rates has given rise to difficulties which were not completely unexpected. Wide variations in individual observations made the derivation of representative parameters difficult. The long term behaviour of the aerosol indicates a mean residence time which is in agreement with previous estimates of the residence time for the most persistent aerosols, but this leads to washout parameters somewhat smaller than the values commonly used in models.

More has been learned of resuspension. Previously, resuspension had never been observed from a widespread contamination incident in temperate, humid zones of Europe, but results have been obtained that demonstrate the significance of resuspension of radiocaesium in this area. Air

concentrations were modest, and caused only a minor increase in inhalation dose in comparison to the dose from the initial plume. However, the potential for contamination of crops and other surfaces by the deposition of resuspended material appears to be much more significant in some locations.

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Table 1. Dry deposition velocity for caesium isotopes measured following Chernobyl

Surface	Location	Deposition velocity mm s^{-1}	Reference
Grassland	S. England	0.44, 0.52	1
	S. Sweden	0.5 to 3.0	2
	Germany	0.5	3
	Denmark	0.2 - 1.0	4
	Italy	0.6	5
Roofs	Denmark	0.3	4
	S. England	0.3 - 0.9	6
Walls	S. England	<0.04, <0.005	6
	Denmark	0.01	4
Roads	Denmark	0.1	4

Table 2. Washout ratios measured after Chernobyl

Location	Washout ratio, W	Reference
Oxfordshire, England	230 - 6600	1
Lund Sweden	3000 ± 2700	7
Thessaloniki, Greece	700 - 830	8
Cumbria, England	490, 540	10
Munich, Germany	230 - 950	11

Table 3. The time dependence of the resuspension factor (K) for ^{137}Cs as expressed by the exponential function:

K (t) = A exp (-Bt) (t = time in months after mid-June 1986)

Location	Initial Deposit (Bq m^{-2})	Period of observation	A (m^{-1} x 10^{-9})	B ($month^{-1}$)
Klagenfurt, Austria	43700	1/87 - 12/89	3.6	0.049
Neuherberg, fRG	19700	7/86 - 12/88	7.4	0.090
Bregenz, Austria	13200	1/87 - 8/89	5.3	0.061
Ispra, Italy	11900	7/86 - 6/88	5.9	0.026
Nurmijarvi, Finland	7100	7/86 - 12/89	10.0	0.063
Wien, Austria	4000	1/87 - 8/89	12.8	0.053
Warsaw, Poland	3200	7/86 - 10/89	15.0	0.062
Braunschweig, FRG	2700	7/86 - 12/88	28	0.124
Eskmeals, UK	2500	7/86 - 6/89	16.0	0.082
West Berlin, FRG	2300	7/86 - 12/88	35	0.099
Riso, Denmark	800	7/86 - 12/88	31	0.106
Bornholm, Denmark	620	7/86 - 12/88	31	0.082
Skibotn, Norway	195	7/86 - 12/88	35	0.060
Chilton, UK	52	7/86 - 6/89	49	0.037

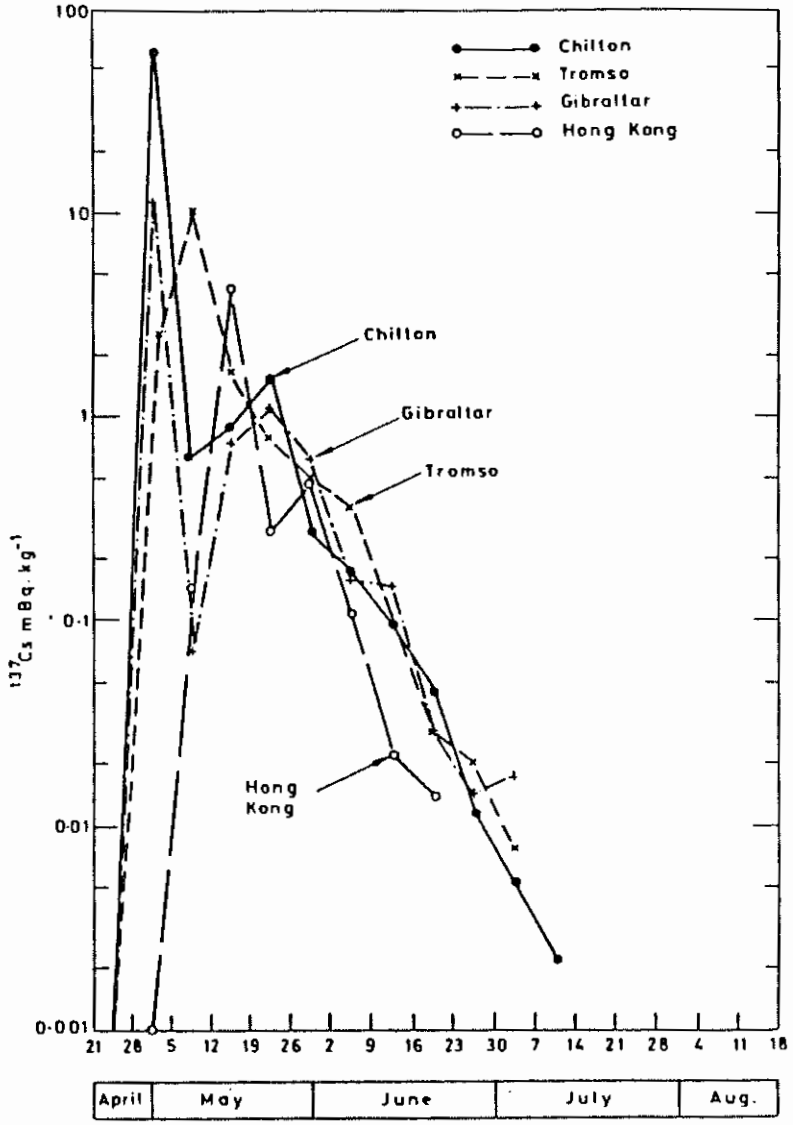

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FIG.1a CONCENTRATIONS OF ^{137}Cs IN AIR AT FOUR STATIONS IN THE NORTHERN HEMISPHERE.

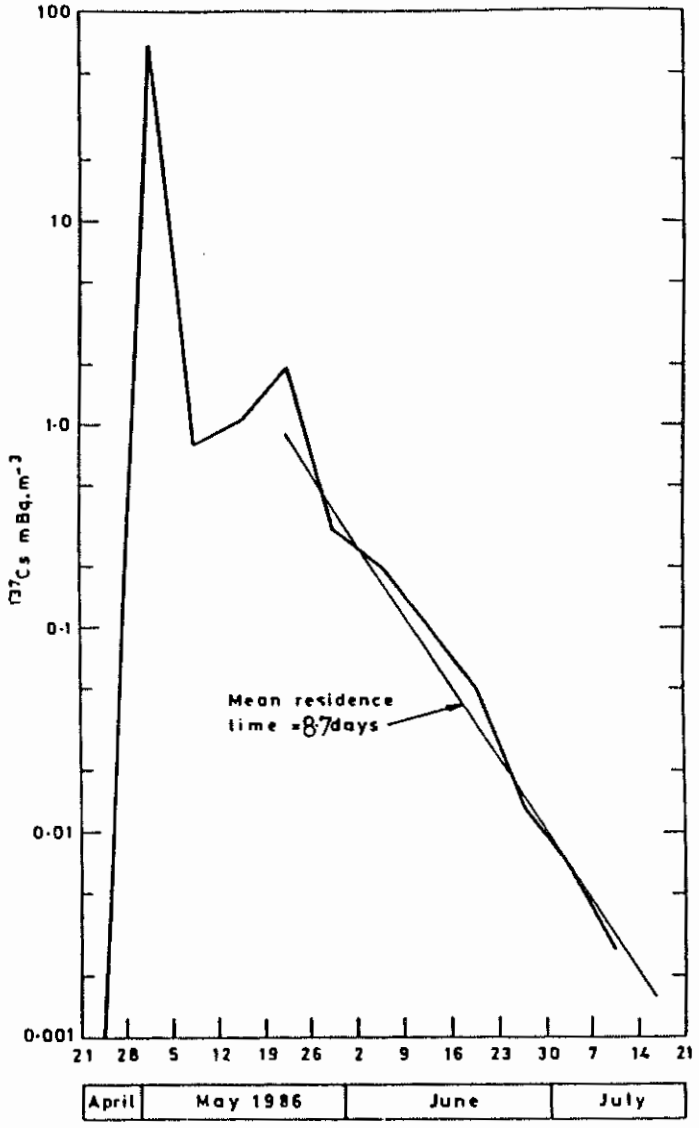

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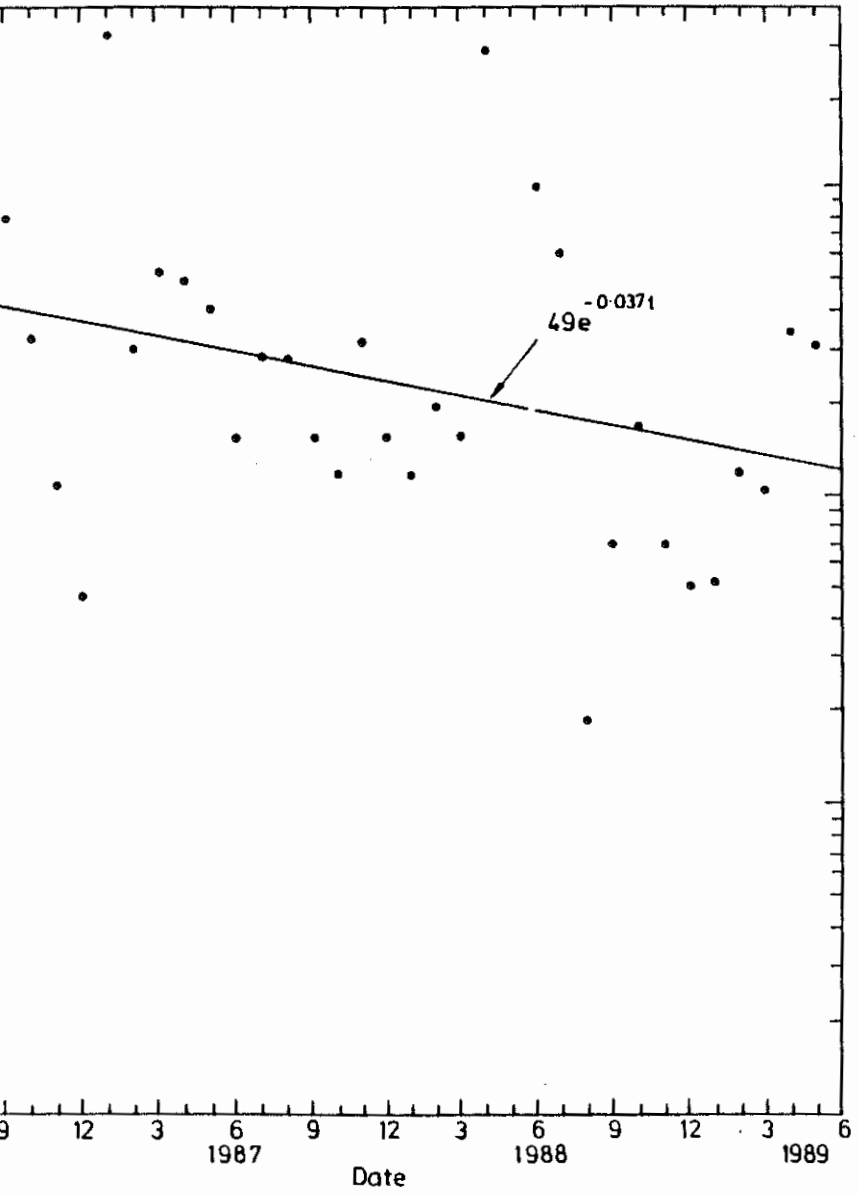
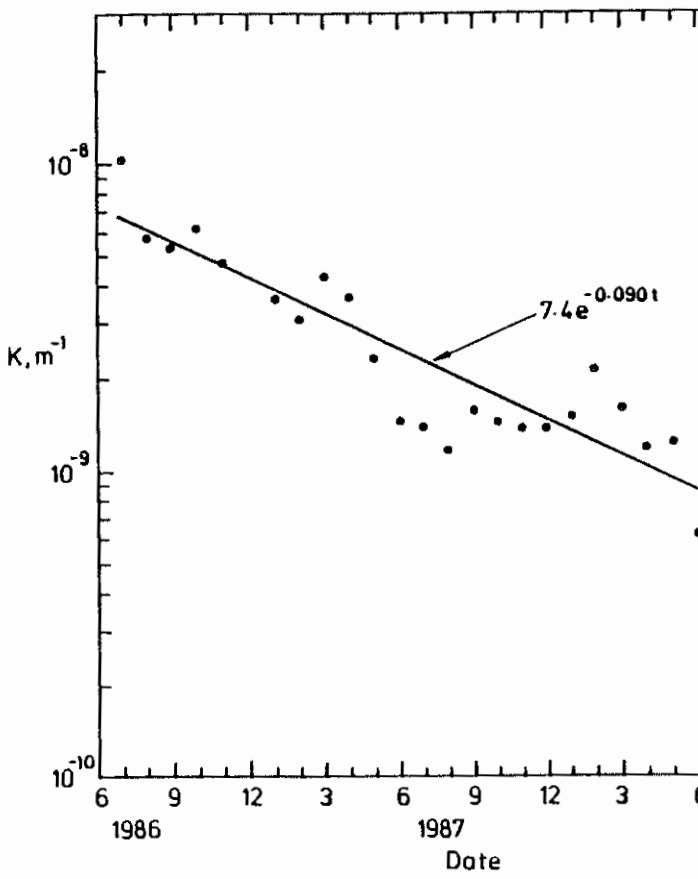
FIG.1b WEEKLY CONCENTRATIONS OF ^{137}Cs IN AIR AT CHILTON, MAY - JULY 1986.



ILTON : ^{137}Cs RESUSPENSION FACTOR (m^{-1})



NEUHERBERG : ^{137}Cs RESUSPENSION FACTOR

Fig. 2. Examples of data for resuspension of ^{137}Cs from Chernobyl

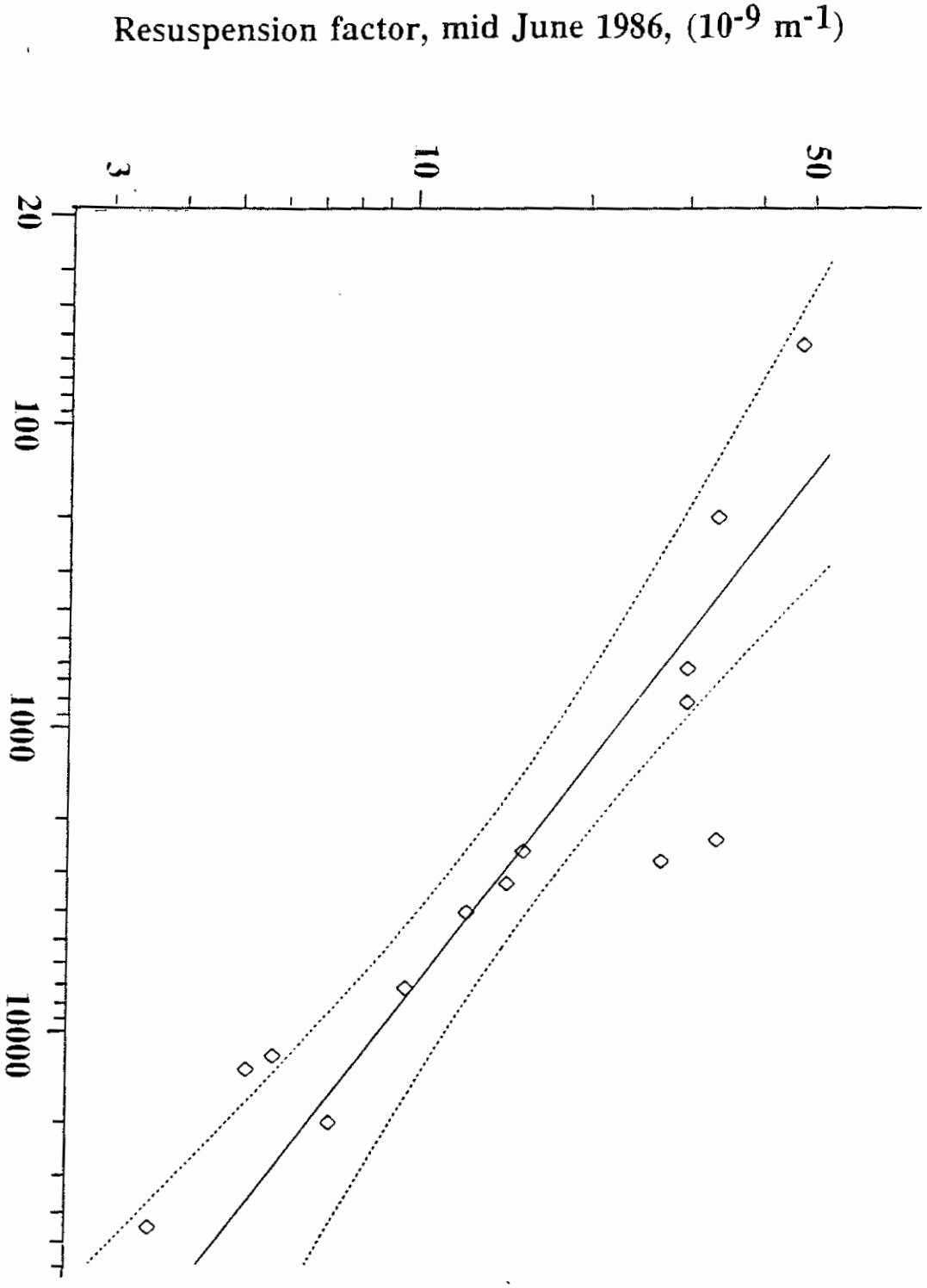


Fig. 3. Variation of the resuspension factor in mid-June 1986 with the amount deposited during April-June 1986.

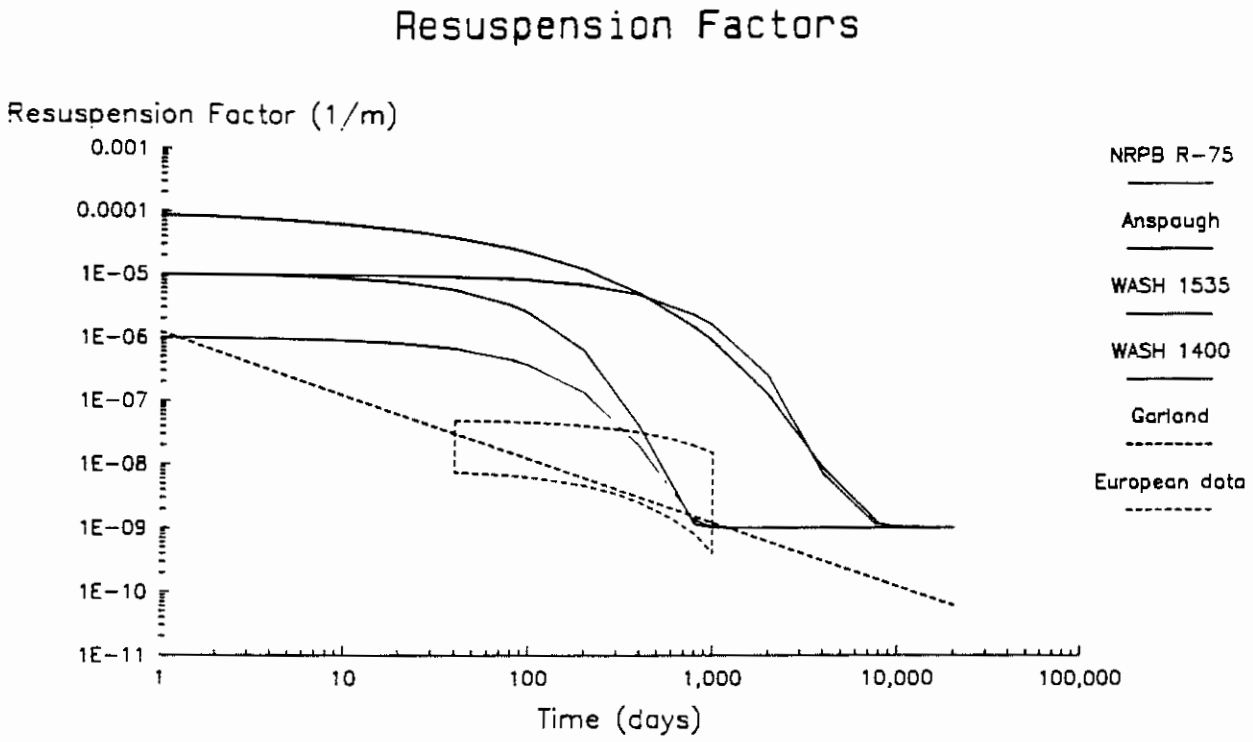


Fig.4. Empirical descriptions for the resuspension factor, used to predict air concentrations, showing the range occupied by the exponential functions fitted to European post-Chernobyl data.

Models:

USAEC (1974)

$$K = 10^{-5} \exp(-0.0139t) + 10^{-9} \ m^{-1}$$

USAEC (1975)

$$K = 10^{-5} \exp(-0.00185t) + 10^{-9} \ m^{-1}$$

Anspaugh (1975)

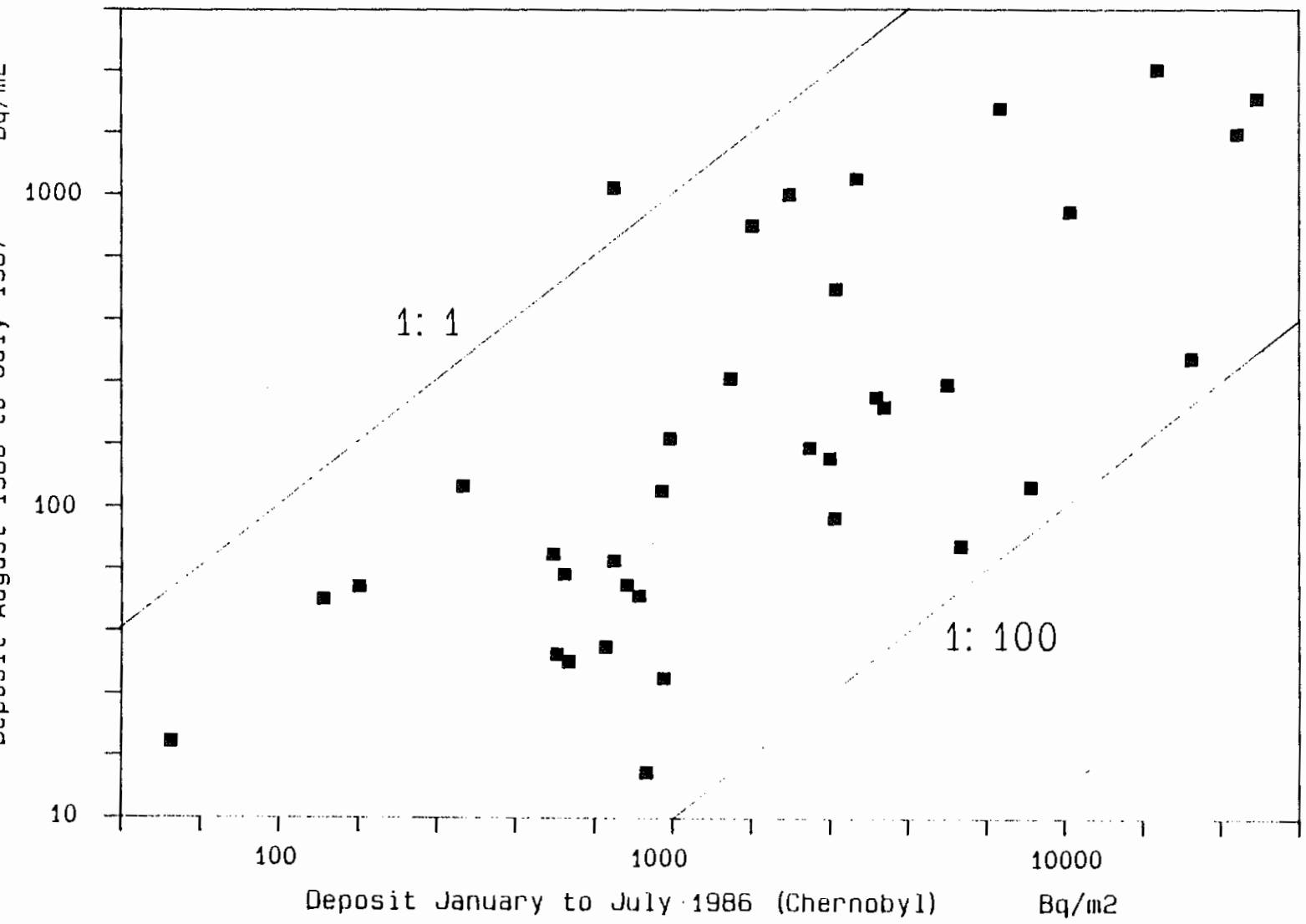
$$K = 10^{-4} \exp(-0.15\sqrt{t}) + 10^{-9} \ m^{-1}$$

Linsley (1978)

$$K = 10^{-6} \exp(-0.01t) + 10^{-9} \ m^{-1}$$

Garland (1982)

$$K = 1.2 \times 10^{-6} / t$$



5. Deposit during August 1986 to July 1987 plotted a
initial deposit from Chernobyl.

Characteristics of Primary and Secondary Caesium-Radionuclide Contamination of the Countryside Following the Chernobyl NPP Accident

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The accident at the Chernobyl NPP was the second serious accident involving a nuclear reactor (the first being at Windscale) and led to a wide area being contaminated with a large number of different radionuclides, including some elements with a high melting point.

From May 1986 onwards a team of researchers from the Soviet Goskomgidromet* and the Soviet Mingeo** carried out extensive studies around the Chernobyl NPP using aerial gamma and aerial gamma-spectrometric survey methods: these covered a 5-km zone around the Chernobyl NPP (78.5 km^2), a 60-km zone around the Chernobyl NPP (11 500 km^2) and five administrative regions bordering the accident zone (over 350 x 10^3 km^2). It was possible - via integration - to determine from these studies the absolute and relative contributions received by each zone from the total caesium-137 release into the atmosphere from the reactor. Comparison of a map plotting actual contamination of the countryside with estimates based on various mathematical models allows us to evaluate any shortcomings in the individual components of the models.

In order to obtain a quantitative assessment of possible redistribution of the original radioactive deposition, a recording network (with a radius of 60 km) was set up covering an area of 11 500 km^2 (geometrically centred on the fourth unit at the Chernobyl NPP). The network consists of over 400 recording points located at distances ranging from 1 to 60 km (36 radii, up to 19 points per radius). From 1987 to 1989 five series of soil samples were taken at nodal points in the network and analysed to establish their caesium-137 content. On two occasions (spring and autumn 1989) gauze screens were set up at various points throughout the network; after one month's exposure these screens were analysed for caesium-137 content.

Comparing the degree of caesium-137 primary contamination with the degree of resuspension for this radionuclide we were able to make quantitative assessments and predict windborne migration of primary contamination. The paper sets out the differences in regional contamination at micro-, meso- and macro-scale levels.

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* State Committee for Hydrometeorology

** Ministry for Geology

Since May 1986 a large group of scientists from various Soviet institutes and services, cooperating under a single programme entitled "Radiation monitoring of the Chernobyl NPP 60-km zone", has been working at the town of Chernobyl in the 30-km zone around the Chernobyl NPP.

The programme comprises the following main sections:

1. Radiation monitoring of the countryside using remote methods of aerial gamma-spectrometry.

2. Radiation monitoring of the countryside by taking and analysing soil samples from permanent and ad-hoc calibration networks.

3. Radiation monitoring of plutonium and strontium-90 radionuclide contamination of the countryside.

4. Radiation monitoring of fallout of radionuclides onto screens erected in the calibration network (area covered = 11.5 thousand km^2).

5. Radiation monitoring of "hot" particles deposited on the countryside after the Chernobyl accident and of fallout currently settling on screens as a result of wind resuspension of dust.

6. Radiation monitoring of contamination in populated areas, in both the 30-km zone and adjacent territories.

7. Initial evaluation of the impact of the accidental releases on people and the environment by a calculation of integrated indicators.

This paper covers some of the above.

The basic aims and tasks involved in monitoring radiation in the 60-km zone are as follows:

1. Collection, processing, rapid presentation and storage of information on radioactive contamination of the near zone affected by the accident.

2. Checking the certainty of the information obtained.

3. Devising ways of increasing the effectiveness both of the monitoring methods themselves and of data presentation.

4. Initial scientific interpretation and systematization of the unique material obtained.

I MAPPING AND DRAWING UP AN INVENTORY OF RADIONUCLIDE CONTAMINATION OF THE COUNTRYSIDE AND POPULATED AREAS USING THE MAKFAR-2 AERIAL GAMMA-SPECTROMETRIC SYSTEM

Since the Chernobyl accident many maps have been produced showing the different radiation contamination conditions, including for individual radionuclides. As a rule, such "radionuclide" maps, covering vast areas, were drawn up on the basis of the analysis of soil samples collected mainly in populated areas. Such samples were analysed for caesium-137, strontium-90 and total plutonium radionuclides. Although these maps were fully adequate for the operational tasks in the initial period following the accident, they do have a number of deficiencies.

First of all, a map drawn up as a result of sample analysis will always contain a degree of inaccuracy due to the small area represented by any given sample, and "Chernobyl" fallout is no exception here: after "envelope-type" collection of samples on an area of just 100 m^2, the readings from five samples sometimes differed by 3 to 10 times. But apart from such micro-scale differences in "Chernobyl" fallout, meso-scale differences also occurred, there being as many as 10 environmental contamination gradients over a single kilometre. Such differences could lead to fairly large patches of contamination being overlooked when drawing up maps, and make information on the shape of such patches and their internal structure highly uncertain.

An alternative to producing maps from analysis of samples is automated aerial gamma-spectrometric surveying of the ground coupled with automatic production of maps by computer. This method is based on installing a gamma-spectrometer, with a wide-range detector, on a helicopter (aeroplane) to survey a given area at a constant height along a predetermined set of flight paths, after which the information recorded on magnetic media is loaded into a computer, which produces maps (using a specific program) showing the ground contamination levels for any given radionuclide which is a gamma-emitter and whose gamma peak shows up on the gamma spectrum at flight height.

In the aerial gamma-spectrometric surveys undertaken after the Chernobyl accident we used various gamma-spectrometric systems of bothSoviet and foreign origin. Here we shall look at the results of the aerial gamma-spectrometric research carried out by a large group of scientists from the "Aerogeology" division of the Soviet Ministry for Geology and from the Chernobyl laboratory of the Institute of Applied Geophysics of the Soviet State Committee for Hydrometeorology, who used the "Makfar-2" aerial gamma-spectrometric system, consisting of:

- an on-board gamma-spectrometer with a system of detectors and primary information storage;
- an on-board navigation system locked in to the local area;
- a facility for in-situ information processing;
- a ground-based computer for processing the data and producing the maps.

The gamma-radiation detectors used consist of NaI(Tl) crystals produced by composite methods in the form of prisms 10x10x40 cm^3 and 5x10x25 cm^3 in size, have minimum and maximum overall volumes of 1.25 l and 48 l respectively, and a resolving power of 9.8% along the caesium-137 0.66 MeV line.

The on-board navigation facility consists of three main units:

A) a Doppler antenna for computing the flight speed and angle of drift of the aircraft;

B) a gyroscopic direction-finding system providing information on the aircraft's spatial orientation (course, bank, pitch);

C) an on-board navigational computer using data received from the two above-mentioned installations to plot the route covered using a geographical or quadrate system of coordinates.

Thanks to the above, the aircraft's current spatial coordinates can be calculated to within 35-45 m.

The "Makfar-2" aerial gamma-spectrometer is used to make surveys on scales of 1:10 000, 1:50 000 and 1:300 000 (i.e. flights spaced out every 100 m, 500 m and 3 km). The gamma-radiation spectra in the 0.38 -3.0 MeV range are recorded in steps of 10 keV on two channels simultaneously, using detectors with crystal volumes of 16 l and 1.25 l respectively. For further investigation of areas where caesium-137 surface contamination exceeds 15 Ci/km^2 we use the information recorded by the latter channel, whereas that recorded by the former channel is used for areas below 15 Ci/km^2. This means we can a) keep analogue-code converter "downtime" to a minimum in intensive gamma fields, thus allowing us to carry out quantitative measurements at high dose-rate levels (above 0.5-0.7 mR/h), and b) accurately measure areas where caesium-137 surface contamination is below 5 Ci/km^2.

The whole apparatus is installed on the aircraft, the survey height is between 140 and 160 m, the speed depends on the desired scale, varying from 70-110 km/h (scales 1:10 000 and 1:50 000) to 170-180 km/h (scale 1:300 000), and the measurement cycle is one second.

To convert the counting-rate unit (pulses/sec.) recorded by the "Makfar-2" into dose-rate units (mR/h) and surface contamination levels (Ci/km^2), we use readings from flights over calibration sites specially set up for this purpose at the end of 1987/beginning of 1988 (Table 1).

Four main criteria dictated our selection of sites: (1) mean contamination intensities on the various sites had to differ considerably from one site to another; (2) the sites had to contain areas with radionuclides of slightly fractionated composition and with clearly pronounced "caesium" contamination; (3) most of the sites had to have landscape features which clearly differed from one another (forest, arable land, meadow, water meadow, etc.); (4) the radionuclide contamination at each given site had to be evenly spread (as far as possible).

As Table 1 shows, despite the considerable time spent on choosing sites, it was not always possible to meet the last selection requirement. Therefore, aerial gamma-spectrometric surveys (scale 1:25 000) of all the sites are carried out each year to calibrate the gamma-spectrometer prior to starting (and also during) the work.

To check map accuracy we always compare the readings obtained with the findings for samples collected in situ throughout the survey area.

Table 1: DATA FROM CALIBRATION SITES USED TO CALIBRATE THE 'MAKFAR-2' SYSTEM

Site index	Area km²	Caesium-137 (Ci/km²)			Site radionuclide contamination ratios	
		min.	mean	max.	Cs-137/Ru-105	Cs-137/Ce-144
1	4.5	9.4	114	457	3.1 - 8.5	3.1 - 7.0
2	5.2	0.4	2.5	5.4	1.5 - 2.0	0.86 - 1.0
4	5	3.3	6.1	14	0.7 - 1.6	0.37 - 0.63
5	9	4.0	7.1	10.0	0.19 - 0.66	0.2 - 0.31
6	10	7.9	14.6	22	0.45 - 0.92	0.26 - 0.33
7	12	4.0	7.3	11.1	0.67 - 0.74	0.33
9	10	15	34	47	2 - 2.7	1.2 - 2.0
10	7	12	44	83	3 - 3.1	2.5 - 3.4
0.5T	1	62	148	208	3.8	2.6
1T	1	111	222	383	2.7	1.3
2T	1	101	333	742	2.9	2.04

Measurement accuracy (repeatability of results) was assessed by repeating flights (overlapping sections of 150 km). The resultant margin of error for the survey was ± 6.3% at a mean dose rate of 0.065 mR/h and a mean caesium-137 concentration of 7 Ci/km^2.

Due to practical economic considerations, the 15 Ci/km^2 and 40 Ci/km^2 isolines are the main points of interest on all the maps drawn up by us. In 1986 and 1987 5 Ci/km^2 was taken as the upper limit for carrying out farming and tending private holdings without decontamination being compulsory. The 15 Ci/km^2 isoline is the upper limit for areas where people are allowed to stay on without there being a need to bring in produce from outside the region. Therefore, it is essential that this isoline is defined precisely, not only so that important national economic decisions can be taken, but also to ensure that people can continue to live in safety in populated areas adjacent to contaminated patches with levels above 15 Ci/km^2.

Fig. 1 shows a typical result from aerial gamma-spectrometric surveying of a large populated area and its environs, with distances of 100 m (scale 1:10 000) and 250 m (scale 1:25 000) between the helicopter flight paths. As we can see, the town of Chernobyl, now unfortunately notorious throughout the world, lies in a zone where caesium-137 contamination is generally over 15 Ci/km^2 (but under 40 Ci/km^2). It should be noted for clarity's sake that the methodology involved in aerial gamma-spectrometric surveys of populated areas has not yet been fully perfected, although the results obtained now tally quite well with the readings from in-situ sampling.

As is known, the reactor ejected considerable amounts of radioactive products for more than 10 days. According to eye-witness accounts, the release initially took the form of an explosion, after which – as the fire developed and the temperatures at the source changed – it assumed the form of a jet dispersed by the wind. As this jet was blown in different directions the radionuclide composition within it changed, since the changes in the temperature and redox state of the source affected the molecular composition of the release and, consequently, its radionuclide composition. We now know that during the crucial period the release was rich in radionuclides of caesium-137,134 and iodine-131, and in order to determine which sections of the overall radioactive trail were rich in caesium-137, we used a computer to calculate the fields of concentration of the caesium-137/cerium-144 fractionation coefficient, using the formula

$$f_{144}^{137} = \frac{(\sigma_{137} / \sigma_{144})_E}{(A_{137} / A_{144})_T}$$

where (C_{137}/C_{144}) is the experimentally determined ratio of caesium-137 radioactive contamination ("volatile" component) to cerium-144 ("non-volatile" component), related to the time of the accident,

and

(A_{137}/A_{144}) is the ratio of the total activities of caesium-137 to cerium-144 stored in the reactor by the time the accident occurred.

It follows from this formula that there should be a link between a) the surveyed sites where and b) the processes involved in the "explosive" ejection of the slightly fractionated cocktail of radionuclides from the reactor, plus their subsequent additional removal during ventilation of the active zone when water was used to extinguish the fire.

Using the "Makfar-2" aerial gamma-spectrometer (supported by a powerful ground-based computer) meant we could not only produce maps showing contamination levels broken down into the various radionuclides, but also obtain schematic maps by combining the readings in various ways. This was how we drew up the cartogram showing the distribution, over the survey area, of the fractionation coefficient worked out using the above formula.

Fig. 2 contains part of the map (excluding the 30-km zone) showing the caesium-137 contamination level. The map was obtained on the scale of 1:200 000, whereas the fragment shown is on the scale of 1:500 000. It clearly shows six large patches containing significant amounts of caesium-137 in the soil, including what is known as the "western" trail (between the populated areas of Tolsty Les and Vilcha).

For the purposes of comparison with Fig. 3, Fig. 4 shows (with an unimportant change in scale for technical reasons) the -cartogram obtained from having the computer combine the caesium-137 and cerium-144 contamination levels. This shows clearly that the "western" and "southern" trails were caused by releases containing a slightly fractionated mix of radionuclides. It is also reasonably clear that the south-western and north-western patches are places where a large amount of caesium-137 fell during the second fallout phase, with some of this fallout being deposited in the northern sector as well.

Fig.1: CAESIUM-137 CONTAMINATION IN THE TOWN OF CHERNOBYL AND ENVIRONS
SURVEY SCALE 1 : 25 000

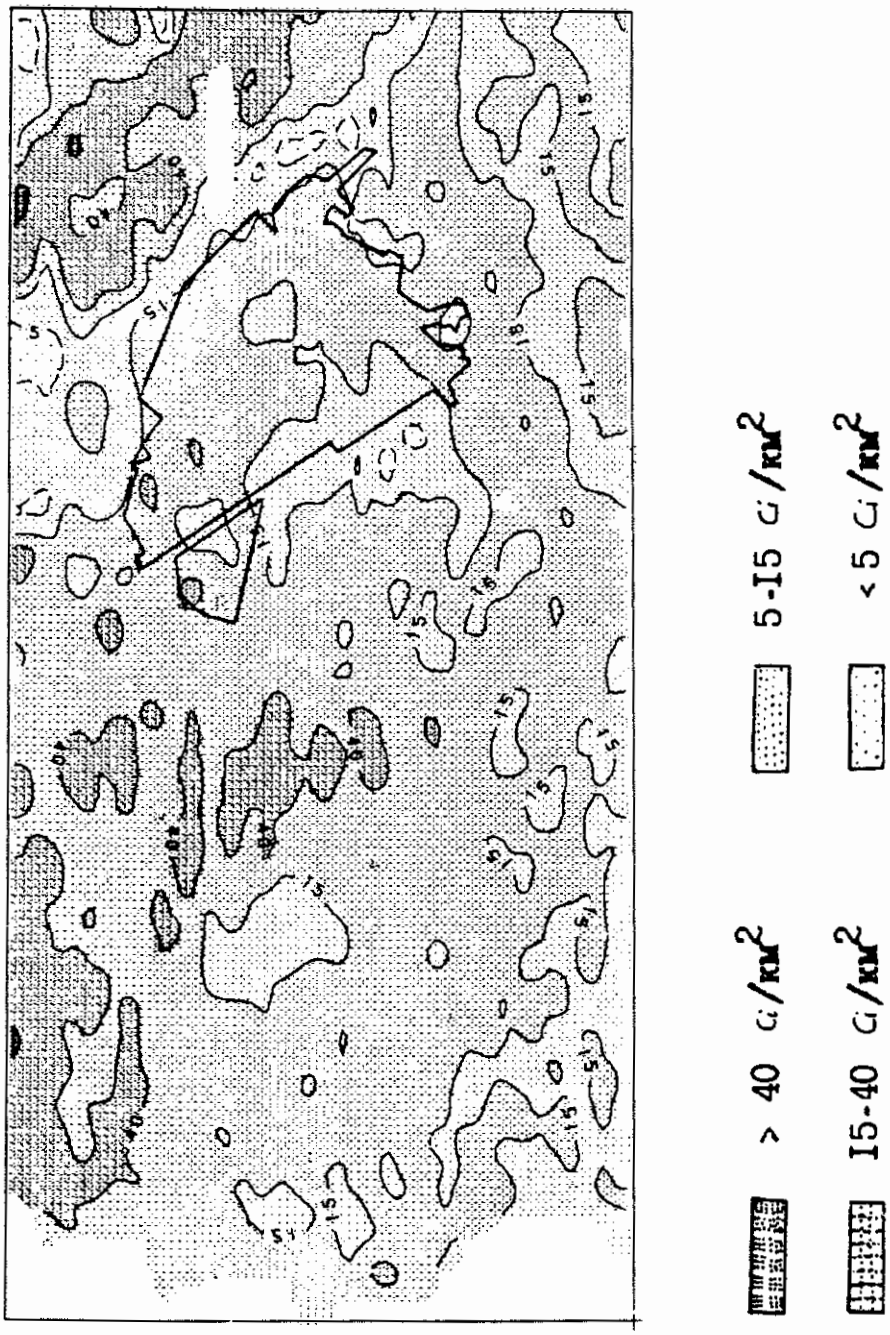

Fig.2: PART OF MAP SHOWING CAESIUM-137 CONTAMINATION EXCLUDING 30-km ZONE
SURVEY SCALE 1 : 200 000 - MAP SCALE 1 : 500 000

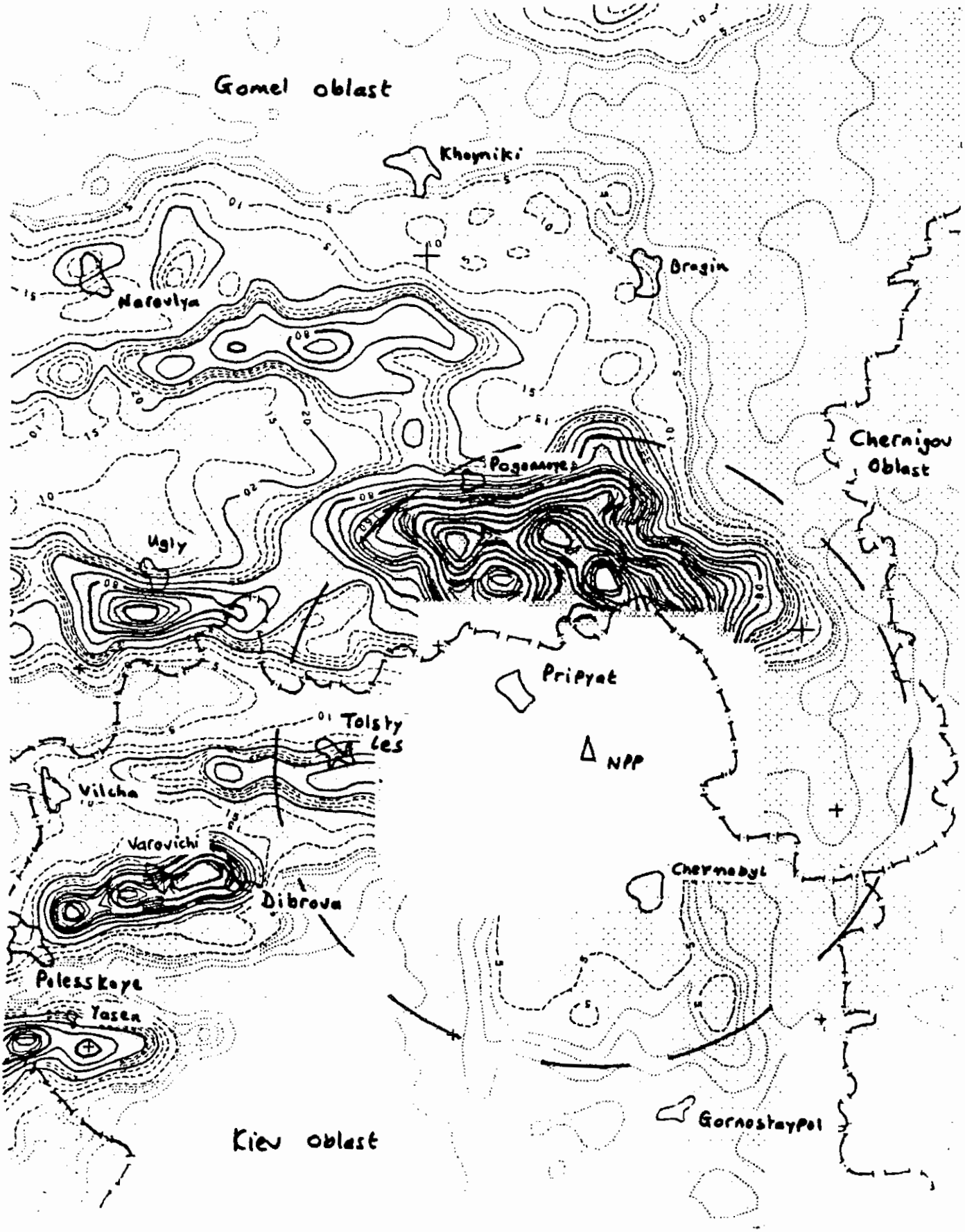

Fig.3: PART OF CARTOGRAM SHOWING DISTRIBUTION OF THE f_{144}^{137} COEFFICIENT, EXCLUDING THE 30-km ZONE

SURVEY SCALE 1 : 200 000

Aerial gamma-spectrometric surveying was an important phase in studying the consequences of the Chernobyl accident. It provided us with rapid information on the radioactive contamination over vast areas, by allowing us in the early stages to trace and pinpoint all the large "caesium patches" with severe contamination (above 5 Ci/km^2); then to plot in detail their shape and internal structure; to determine the areas of low contamination where residence and farming could continue; to pinpoint the populated areas with complex radiation conditions (situated on sites with sharply varying contamination levels) requiring examination of each individual farm or holding; to target areas for landscape and geochemical, radiobiological and geophysical scientific studies; and to harness the survey material in determining the radioecological characteristics of the areas studied.

Fig. 4 is a map obtained in 1987 using the "Makfar-2" survey system on flight paths spaced out every 3 km over an area larger than 10^5 km^2. It reveals the uneven spatial distribution of the contamination in both the near and distant zones affected by the Chernobyl accident. Thus, for example, the Chernigov oblast (region) of the Ukrainian SSR was in a healthier state than the Gomel and Mogilev oblasts of the Byelorussian SSR and the Bryansk oblast of the RSFSR.

It should be stressed that we never use the results of the aerial gamma-spectrometric survey on their own without comparing them with in-situ sampling findings. Table 2 shows (in concise form) the results of comparing the readings from aerial-gamma surveys of the Gomel and Mogilev oblasts with in-situ sampling data from a number of sites in the area. It sets out the differences between the in-situ and aerial data.

By making such comparisons we can draw up maps integrating both types of data, an example of which is the caesium-137 contamination map for the 60-km Chernobyl NPP zone shown in Fig. 5.

Fig.4: CAESIUM-137 CONTAMINATION WHERE CONTAMINATION LEVEL IS HIGHER THAN 15 Ci/km²

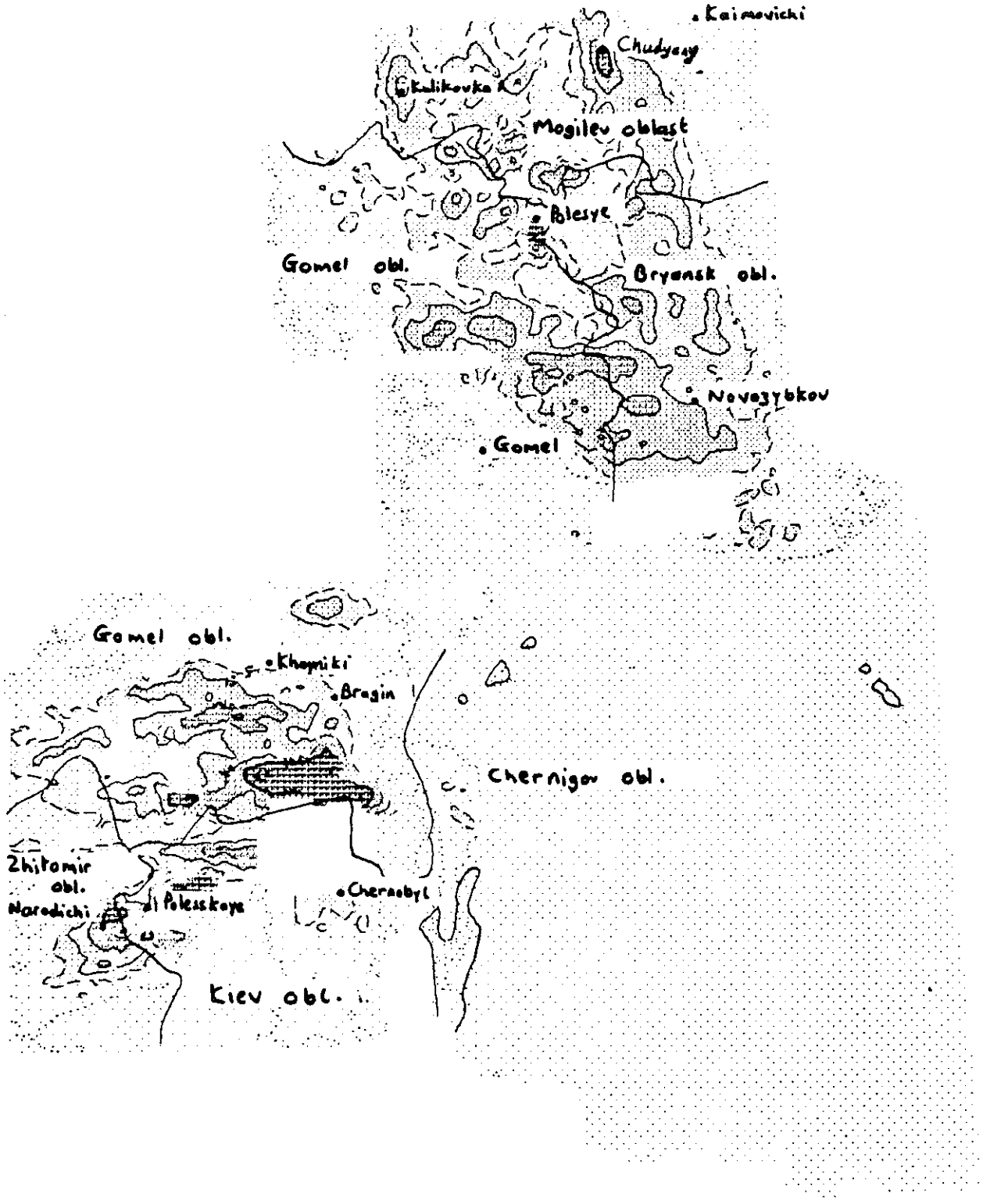

Table 2: COMPARISON OF DATA BRYANSK-MOGILEV OBLAST JULY 1989

Cs-137 (Ci/km²)		Difference
in-situ	aerial	
15	20	-5
89	100	-11
119	90	+29
38	48	-10
53	50	+3
60	60	0
52	70	-18
58	70	-12
63	80	-17
78	70	+8
33	45	-12
19	27	-8
25	32	-7
38	40	-2
26	40	-14
43	45	-2
28	36	-8
40	45	-5
35	45	-10
61	72	-11

$\Delta = \pm 10$ Ci/km²

$\sigma = \pm 24.5$ %
where mean = 48.65 Ci/km²

This figure is based on data collected up to January 1990, including:

1) the results from aerial gamma-spectrometric surveys of various parts of the 60-km zone on the scales 1:200 000, 1:100 000, 1:50 000 and 1:25 000;

2) information from the radiation monitoring network in the 60-km zone (results of collecting samples from the calibration network), and data pertaining to samples collected in-situ in populated areas.

Thus, to produce the map we used two types of information obtained by different methods, which is extremely important in ensuring that the maps give an objective picture, since each type of information verifies and supplements the other.

The map shows that the worst and most complex caesium-137 contamination occurring in the near zone after the accident is to the west and north of the Chernobyl NPP. The map clearly shows the narrow western trail from the Chernobyl NPP to Vilcha, the northern trails forming the contaminated sector of Chernobyl NPP-Narovlya-Khoyniki-Bragin, and the south-western trail from the Chernobyl NPP to Polesskoye. Contamination levels inside these trails exceed 100 Ci/km^2, both in the immediate vicinity of the Chernobyl NPP and in places much further afield.

The southern trail (Chernobyl NPP-Chernobyl) is also clearly visible on the map, but it quickly peters out; outside the 30-km zone levels exceed 5 Ci/km^2 at isolated points only.

The eastern trail (Chernobyl NPP-Nedanchichi) is not clearly pronounced outside the 30-km zone, and in the Slavutich zone occurs in the form of isolated patches only.

Paradoxically, the south-western and eastern parts of the 60-km zone suffered little from the accident; caesium-137 levels there do not exceed 1 Ci/km^2.

The figures on which this map is based are given in Table 3.

Fig.5: CAESIUM-137 CHERNOBYL NPP 60-km ZONE
(Ci/km²)

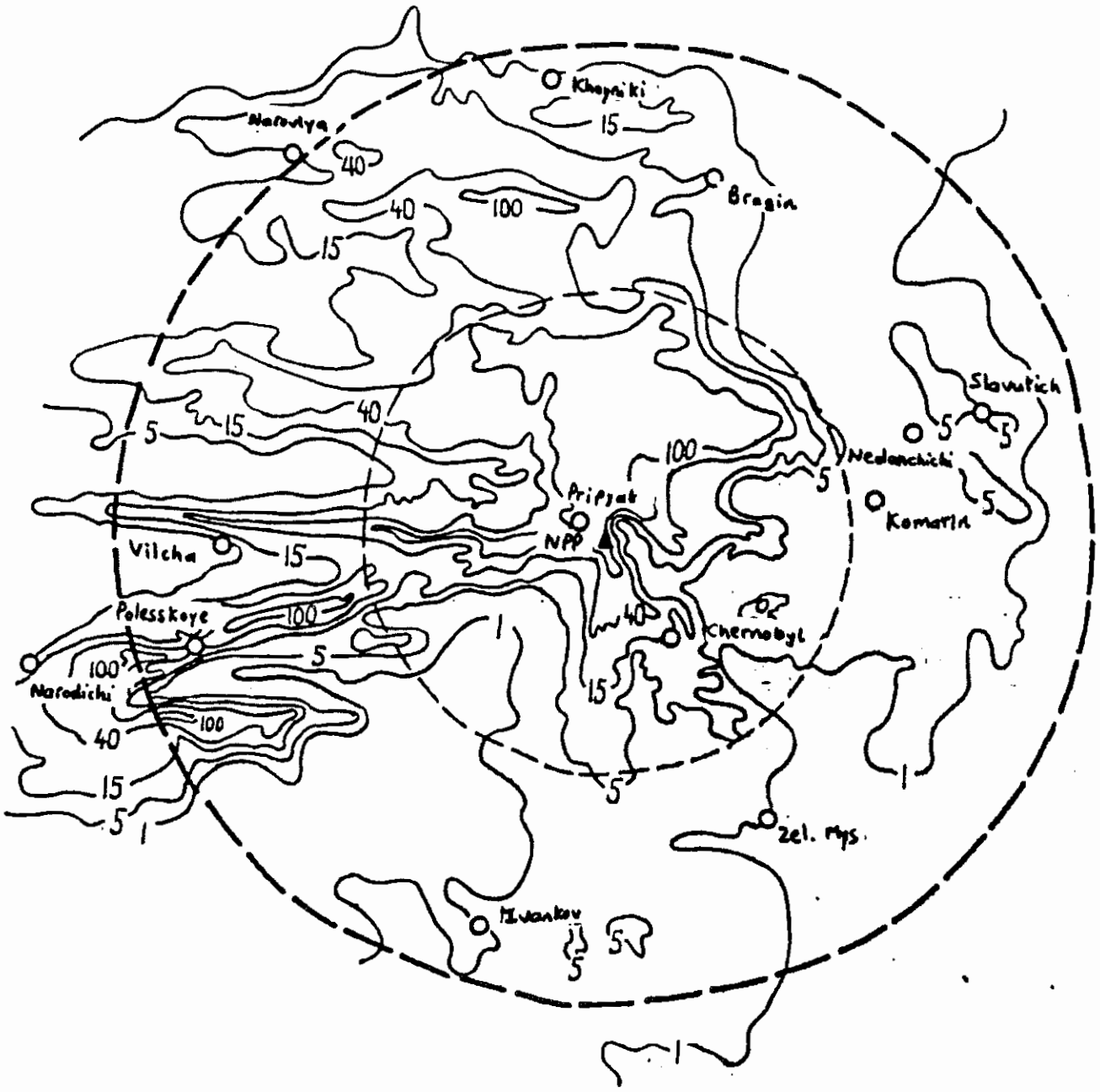

Table 3: DATA SERVING AS BASIS FOR MAP SHOWING CAESIUM-137 CONTAMINATION IN THE CHERNOBYL NPP 60-km ZONE

Levels Ci/km²	Area		Amount	
	km²	%	Ci	%
under 1	2445	22	1960	1
1 - 5	3805	34	11400	2
5 - 15	1585	14	15900	5
15 - 40	1505	13	40600	12
40 - 100	1280	11	89600	28
over 100	660	6	165000	51
Total	11300	100	324500	100

II DISTRIBUTION OF RADIONUCLIDES IN THE VERTICAL SOIL PROFILE AT THE CALIBRATION SITES

The purpose of having calibration sites in the Chernobyl NPP accident area was to intercalibrate the various methods of remote gamma-spectrometry under actual conditions, and to determine the dynamics of vertical migration of radionuclides in various landscape and geochemical contexts. Such geocomplex-related aspects dictated the choice of sites representing the main zones of the Polesye (Pripyat Marshes), taking into account landscape uniformity at any one site, the presence (absence) of farming activity, the size of the site and the degree of uniformity of radionuclide contamination and its isotopic composition. Our calibration sites are located on floodplains and terraces above them (ameliorated and natural), in watersheds under ploughing or used for ley, and in swamps and forest tracts (coniferous, broad-leaved and mixed), such places having a broad spectrum of soil types - from peaty-swampy to slightly podzolic forest and sandy soils with slight sod cover.

In the Polesye, soil formed mainly on sandy and sandy-loam substrates of morainal, fluvioglacial and alluvial origin, where the soil profile usually clearly separates (with the exception of individual super-aqueous phases) into an upper section rich in organic matter and loamy constituents, and a lower section usually of mainly mono-mineral siliceous composition. Given such a soil profile structure we expect to see a sharp contrast in radionuclide vertical-migration conditions.

Depending on the specific nature of the soil formation processes, the upper horizons are able to delay - for a certain length of time - the penetration of radiocaesium into the aeration zone's deep horizons and its ingress into groundwater. The evaluation of the duration of the presence of radiocaesium in the humic horizon (which depends on local landscape and geochemical conditions) is based not only on annual in-situ measurements of activity but also on the following experimentally determined parameters: forms of occurrence, degree of complexation with the individual fractions of organic matter, capacity of Quaternary sediments (of different origins and mineral composition) to sorb caesium and strontium, the calculation of the cleansing coefficients of solutions of soil composed of different minerals and the role of the latter as geochemical barriers, and plotting the curves describing the sorptive and desorptive processes involved in redistribution of activity in typical landscape and geochemical conditions in the Chernobyl NPP's zone of influence, etc.

In order to do the above, samples of soil and substrates are regularly

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The work for this section was carried out together with researchers at the Institute of Mineral Geochemistry and Physics, Ukrainian Academy of Sciences, under the direction of I. Sadolko.

Table 4: CHARACTERISTICS OF RADIONUCLIDE DISTRIBUTION IN THE PROFILE OF SOILS IN THE 30-km ZONE

Site index	Point of collection. type of soil	Year of measure-ment	Isotope	Soil layer with max. contam. level. cm	contribut. of isotope to total contamin. %	Location of centre of accum. cm	Content in 0-5 cm layer %	Minimum layer of soil where total A>>90% cm
1	Pl.4. T.1 (soddy. medium podzolic. peat cover) floodplain	1987	Cs-137	0 - 1		0.79	98.0	0 - 1
		1989	Cs-137	0 - 1		0.83	97.7	0 - 2
		1990	Cs-137	0 - 2	47.5	1.66	95.4	0 - 3
		1987	Ce-144	0 - 1		0.62	99.0	0 - 1
		1989	Ce-144	0 - 1		0.55	97.3	0 - 1
		1990	Ce-144	0 - 2	27.6	1.27	98.7	0 - 1
		1987	Ru-106	0 - 1		0.61	100	0 - 2
		1989	Ru-106	0 - 1		0.57	100	0 - 1
		1990	Ru-106	0 - 2	14.4	1.11	99.6	0 - 2
		1989	Sr-90	0 - 2		1.00	99.9	0 - 2
		1990	Sr-90	0 - 2		1.13	99.9-	0 - 2
2	Pl.5. T.3 (sod (soddy. medium podzolic. clayey -sandy-gleyey) meadow. floodplain	1987	Cs-137	0 - 1		0.67	99.0	0 - 1
		1989	Cs-137	0 - 1		0.65	98.2	0 - 2
		1990	Cs-137	0 - 1	52.1	0.68	98.5	0 - 2
		1987	Ce-144	0 - 1		0.59	100	0 - 1
		1989	Ce-144	0 - 1		0.70	97.8	0 - 3

Table 4: (continued)

Site index	Point of collection, type of soil	Year of measurement	Isotope	Soil layer with max. contam. level, cm	contribut. of isotope to total contamin. %	Location of centre of accum. cm	Content in 0-5 cm layer %	Minimum layer of soil where total A>>90% cm
2	Pl5., T.3	1989	Sr-90	0 - 1		0.63	99.9	0 - 2
3	Pl.5, T.4	1987	Cs-137	0 - 1		1.11	96.0	0 - 2
	(soddy, medium /	1990	Cs-137	0 - 2	46.7-	2.15	94.5	0 - 4
	slightly podzolic	1987	Ce-144					
	meadowy-swampy,	1990	Ce-144	0 - 2	23.5	1.77	98.4	0 - 4
	sandy) floodplain	1990	Ru-106	0 - 2	18.8	1.57	100	0 - 3
	waterlogged meadow	1990		0 - 1		2.04	95.8	0 - 6
4	Pl.7, T.3	1987	Cs-137	0 - 1		0.61	99.0	0 - 2
	(slightly podzolic	1990	Cs-137	0 - 1	44.8	0.91	99.7	0 - 2
	sandy, slight sod	1987	Ce-144	0 - 1		0.56	100	0 - 1
	cover, well-	1990	Ce-144	0 - 1	27.3	0.82	99.5	0 - 2
	developed litter	1987	Ru-106	0 - 1		0.55	100	0 - 2
	of 1-2 cm pine	1990	Ru-106	0 - 1	17.6	1.23	98.4	0 - 2
	forest	1989	Sr-90	0 - 1		1.30	98.3	0 - 5
		1990	Sr-90	1 - 2		1.15	98.1	0 - 5
5	Pl.4, M-12	1989	Cs-137	0 - 1		1.18	97.6	0 - 5
	(sand, no sod	1989	Ce-144	0 - 1		0.91	97.8	0 - 5
	cover)	1989	Ru-106	0 - 1		0.83	99.2	0 - 3
	floodplain	1989	Sr-90	0 - 1		2.30	93.1	0 - 7
6	Pl.1, T.5 (sand, no sod cover), sandy ridge	1989	Sr-90	0 - 1		3.08	80.1	0 - 13

collected from the calibration sites and subjected to gamma-spectrometric, radiochemical, physico-chemical, mineralogical and other types of examination.

The findings obtained in the past three years at a number of calibration sites are given in Table 4 and Figs. 6-10. Table 4 shows a) how the centre of radionuclide accumulation shifts over time, b) the soil layer with the highest radionuclide content and c) the amount (%) of a given radionuclide in the 0-5 cm layer for a number of typical soil types.

Figs. 6-10 show how increased moisture content affects radiocaesium penetration to a deeper level in identical or similar types of soil. Fig. 6 shows the comparative penetration of radiostrontium depending on the presence of organic substances in the soil, while Fig. 7 shows the redistribution of radiocaesium in the vertical soil profile at a number of calibration sites.

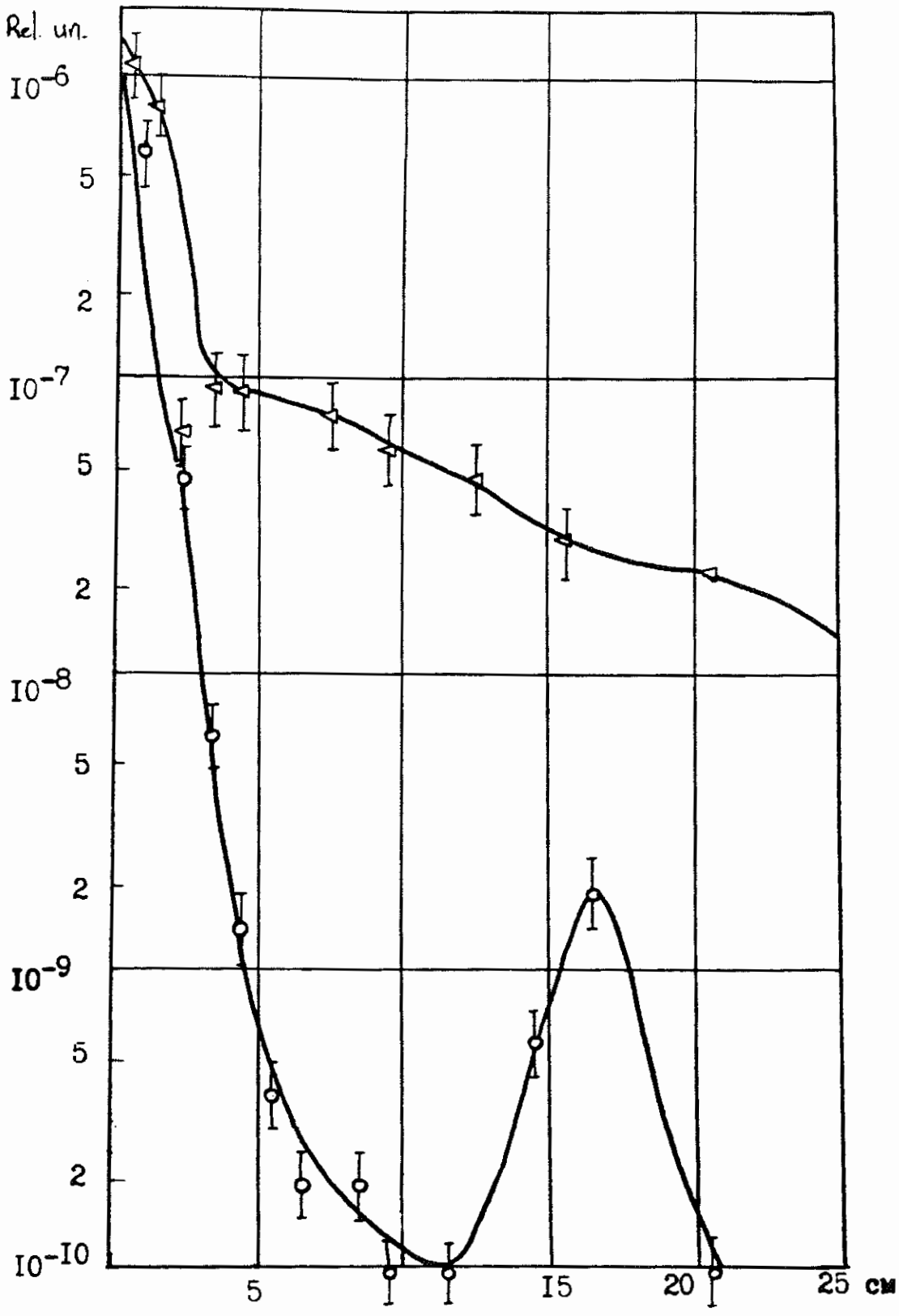


Fig.6: DISTRIBUTION OF RADIOSTRONTIUM IN THE VERTICAL PROFILE OF SOILS AT SITES 4 AND 1

-Pl.4. T.1 (peat cover, highly humic)
-Pl.1. 5T (sand, no sod cover)

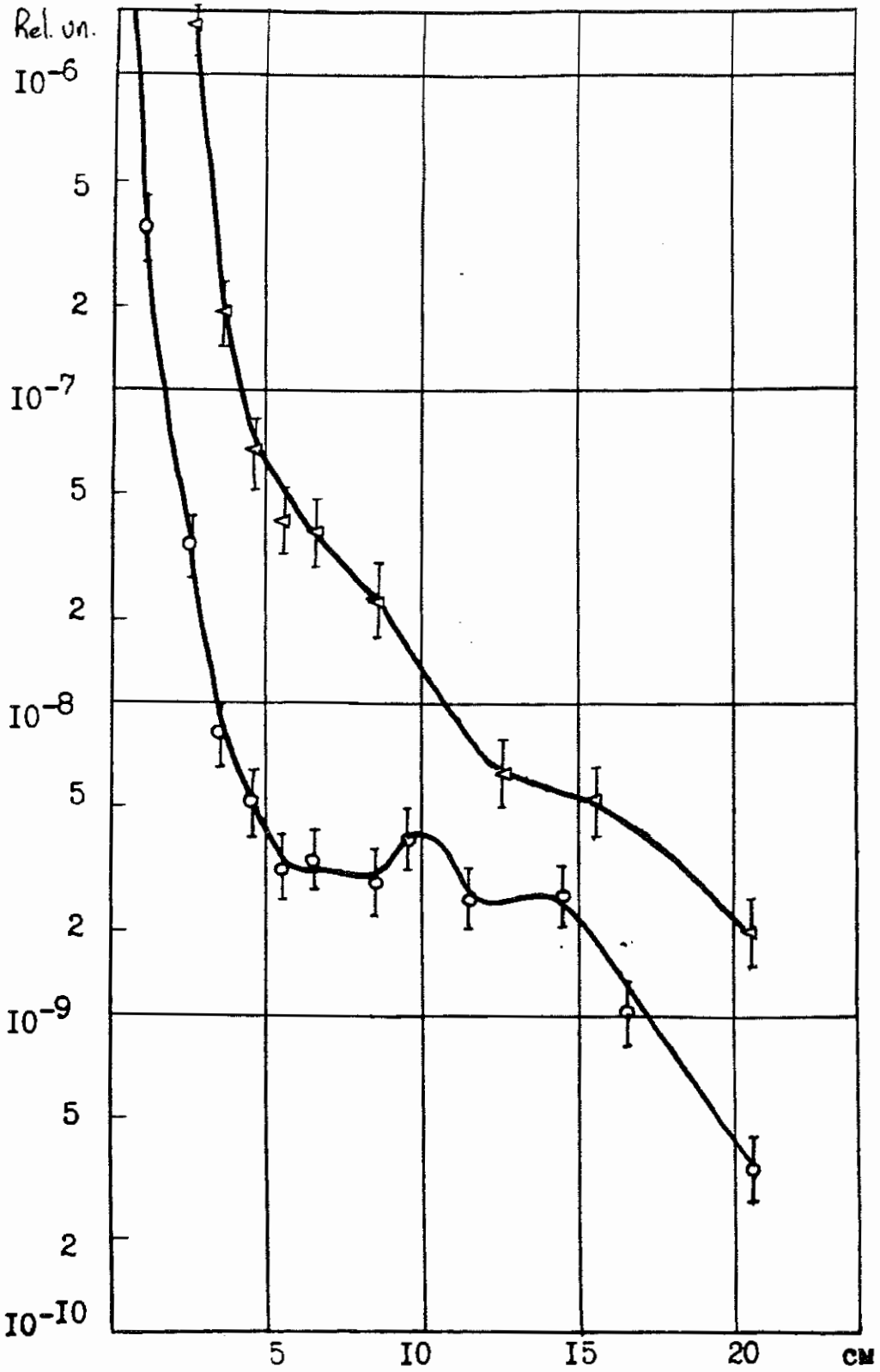


Fig.7: DISTRIBUTION OF RADIOCAESIUM IN THE VERTICAL PROFILE OF SOILS AT SITES 4 AND 1

-Pl.4, T.1 (soddy, slightly podzolic, silty-sandy soil with peat cover)

-Pl.1, M-42 (peaty-swampy-sandy soil, waterlogged)

Fig.8: DISTRIBUTION OF RADIOCAESIUM CONTAMINATION IN THE VERTICAL PROFILE OF SOIL AT SITE 4
(Soddy. slighly podzolic. silty-sandy soil with peat cover)

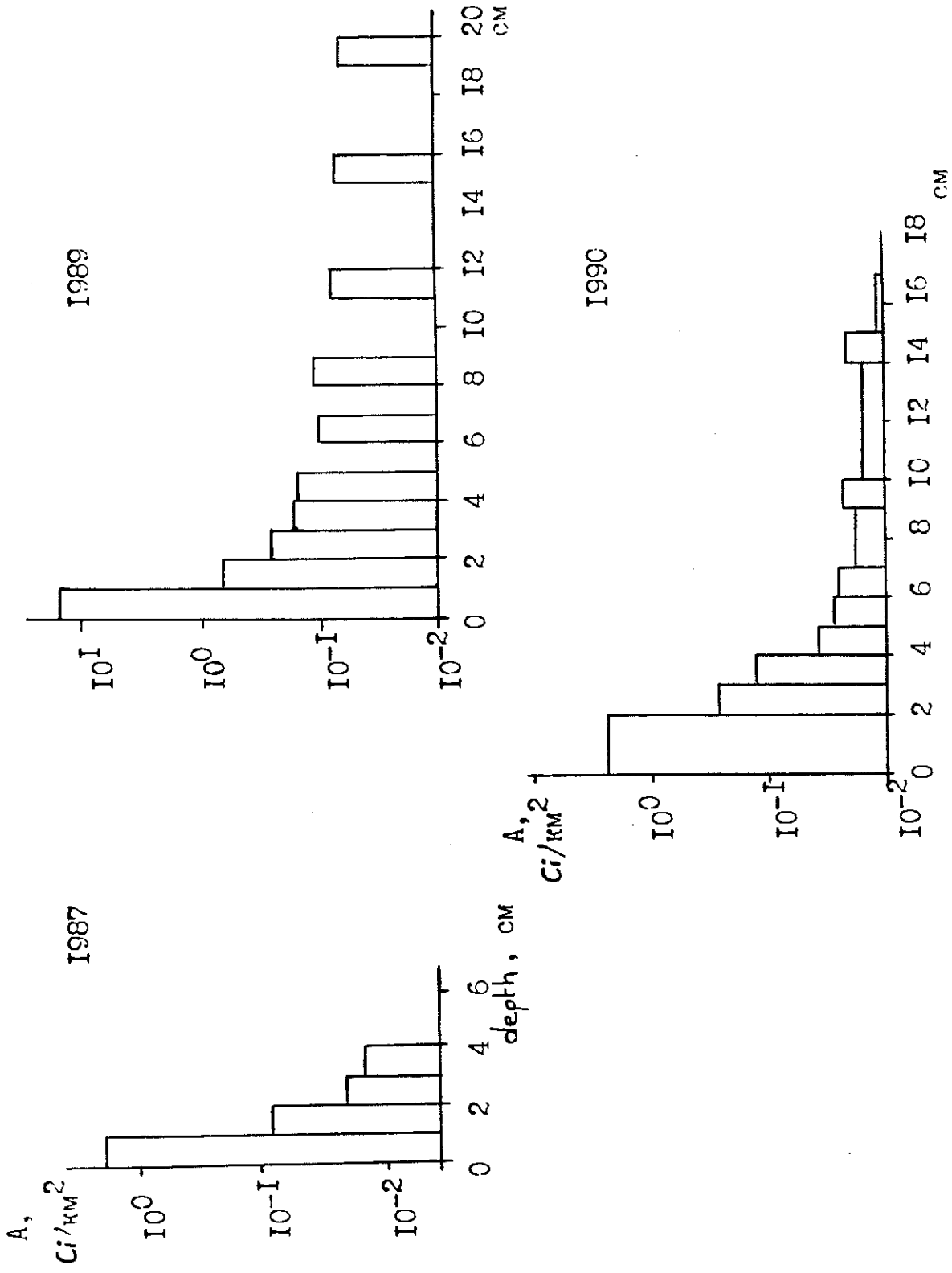


Fig.9: DISTRIBUTION OF RADIOCAESIUM CONTAMINATION IN THE VERTICAL PROFILE OF SOIL AT SITE 9
1987-1989

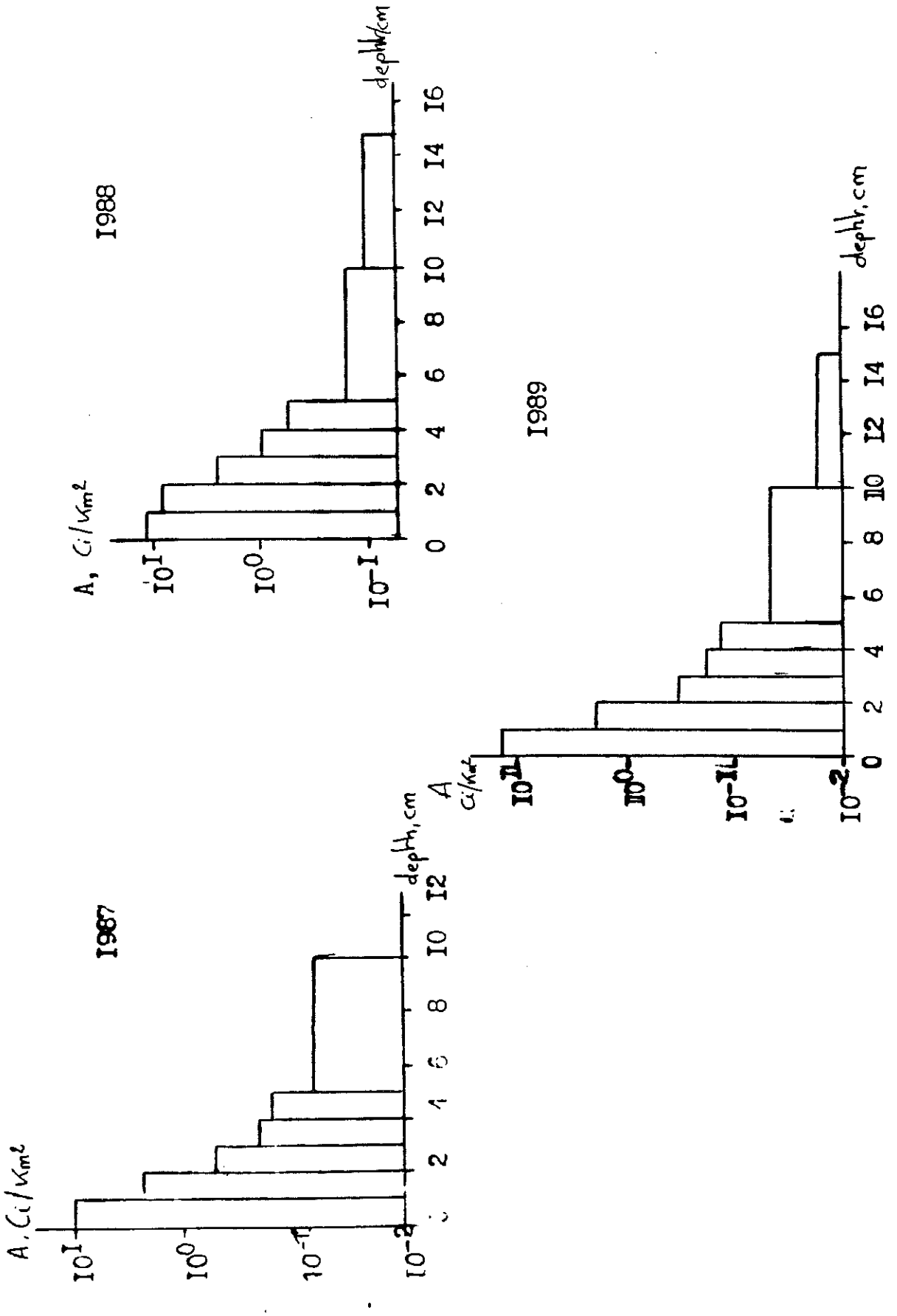


Fig.10: DISTRIBUTION OF RADIOCAESIUM CONTAMINATION IN THE VERTICAL PROFILE OF SOIL AT SITE 1
(arable land, ploughed in 1987)

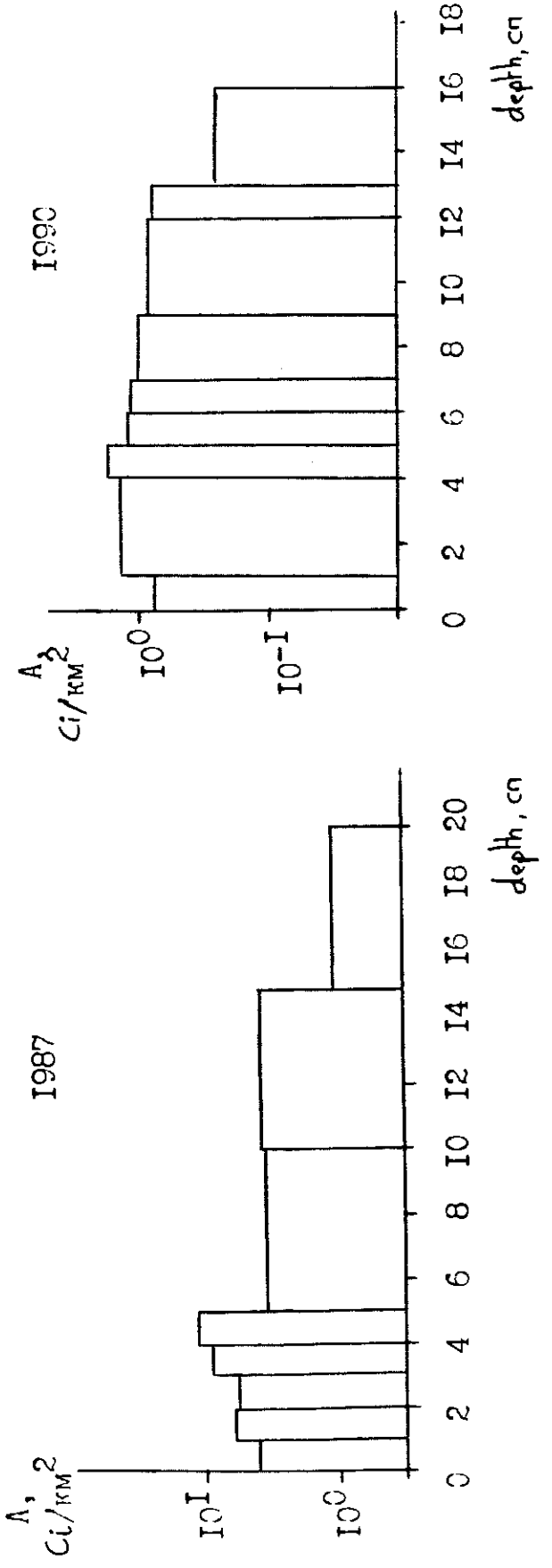
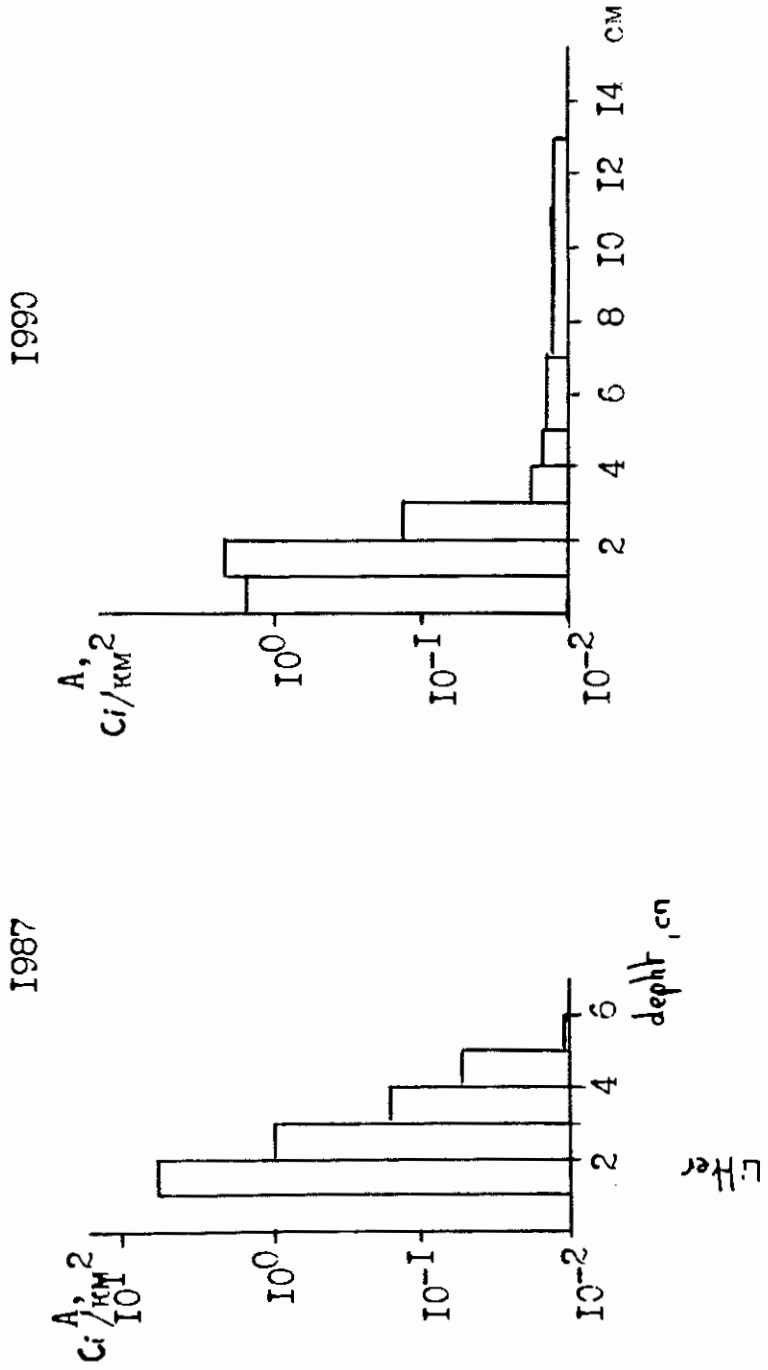


Fig.11: DISTRIBUTION OF RADIOCAESIUM CONTAMINATION IN THE VERTICAL PROFILE OF SOIL AT SITE 7 (pine forest) IN 1987-1990



The distribution of ^{137}Cs contamination was determined on the various sites at points in the same or adjacent locations.

Site 9 (Fig. 9) – Soddy, slightly podzolic, sandy-loam soil, hay meadow; the pattern of distribution hardly changed over three years – the bulk of the radionuclides (up to 97%) is found in the 0–5 cm layer, while in 1987 and 1989 the centre of accumulation was at a depth of 1.22 cm.

Site 1 (Fig. 10) – Soddy-podzolic, clayey-sandy soil, arable land, ploughed in 1987; the sampling points are located 5 m apart, but the overall contamination level varies considerably, while the pattern of radiocaesium vertical distribution is almost identical, the contamination it causes amounting to hundredths of a Ci/km^2 at a depth of 25–30 cm.

Site 4 (Fig. 8) – Soddy, slightly podzolic, silty-sandy soil, with thick peat cover; radiocaesium penetrates further (although not very rapidly) in this type of soil. Over four years the centre of accumulation shifted from a depth of 0.70 cm to 1.66 cm, while the amount in the 0–5 cm layer decreased from 98 to 95.4%.

Site 7 (Fig. 11) – Slightly and medium podzolic, sandy soil with thin sod cover and well-developed litter. The maximum amount of caesium (up to 99%) continues to be found in the litter and the top few centimetres, but it is now penetrating (in hundredths of a Ci/km^2) to a depth of 13–15 cm.

So far the research findings on vertical migration of Chernobyl accident radionuclides in the Chernobyl NPP region show the following:

1. The bulk of the radionuclides scattered over the surface of the contaminated areas is located in the 0-5 cm layer of soil (in the overwhelming majority of the local geocomplexes - by up to 90-98%).

2. In all the types of soil studied in the Chernobyl NPP 30-km zone the migrational capacity of radiostrontium is greater than that of other radionuclides.

3. The bulk of radiostrontium and radiocaesium binds with organic compounds having a molecular mass of (2.8-4.0) x 10_4 and (0.6-3.0) x 10_3. In grey-forest soddy-podzolic soil caesium-137 is mainly found in the form of highly insoluble compounds (up to 60-80%), with exchangeable forms accounting for 10-15%, those sorbed onto ferrous and aluminium hydroxide for 20-30%, and organic compounds 10-20%.

4. In most cases at the 30-km zone sites studied, 11-15% of the strontium-90 content is in water-soluble form, with 75-98% in the exchangeable form (60-65% in the near zone); this shows that practically all the strontium leaves the fuel matrix and other stable compounds, with the leaching processes occurring more quickly with increasing distance from the accident site.

5. Sandy soils (incorporating medium and large grains of sand) with no or only slight sod cover were the most macro-permeable in terms of radiostrontium vertical migration (floodplain sands, dunes, sandy ridges, eskers, etc.).

6. In descending order of sorptive capacity (or the capacity to retain radionuclides in the upper layer) the soils line up as follows: soddy-podzolic, loamy, clayey - soddy-podzolic, sandy-loam, with peat cover - meadow-bog, soddy-podzolic, sandy - sand without sod cover.

7. Radionuclides penetrate downwards more quickly in wetlands.

8. The forest soils displaying the greatest sorptive capacity were the soddy-podzolic and soddy-slightly podzolic types having a well-developed litter (the litter containing 70-80% of the radionuclides).

   Downward migration of radionuclides in wet or marshy forest soils is similar for non-listed areas. Soil density (porosity) plays a significant role in such downward penetration by radionuclides.

9. Over a three-year period the centre of radionuclide accumulation shifted downwards by no more than 0.5-1 cm on average as compared to 1987 (and by a little more in sandy soils with no sod cover, i.e. 1.5 to 2.5 cm for strontium). It should be noted that, whereas in 1986 and 1987 radiocaesium was found in several samples at depths of 10-15 cm in quantities of tenths and hundredths of a percent of overall column content, and in quantities of up to 0.5-1% in the 3-4 cm layer (in soils with slight sod cover, porous peat-covered and sandy water-logged soils in quantities of up to 2-2.5%), by May 1990 the radiocaesium content in these layers had increased by 1.5-3.0 times. Almost the same happens with strontium-90, except that in sandy soils with no sod cover it is found in

quantities of 2-5 x 10^{-9} Ci/kg at a depth of 40-41 cm and in quantities of 2-3 x 10^{-10} Ci/kg at a depth of 60-70 cm, while from the 3-5 cm to 30-40 cm levels its activity gradually decreases from the order of 10^{-8} to 5 x 10^{-9} Ci/kg.

10. The radionuclide migration rate greatly depends on how the radioactive fallout particles convert over time, i.e. on changes in the ratios between the nuclides' ionic, exchangeable and fixed forms.

11. The main factors determining radionuclide mobility in the top layers of the soil cover are the quantity and composition of the finely dispersed fraction and the organic matter present (the latter facilitating sorption of the radionuclides and accelerating their vertical migration).

12. Vertical migration of the radionuclides, although not very significant, is taking place, and study of subsoil geochemical barriers is therefore vital now.

III. "HOT" PARTICLES IN SECONDARY FALLOUT AFTER THE ACCIDENT

The problem of "hot" particles (and here we refer only to the geophysical aspects involved) became evident in the very early days after the accident. Radiographs of leaves from trees some 100 km south of the Chernobyl NPP revealed many large black blotches standing out clearly against a very weak fog. Sometime later, in mid-and late May 1986, we discovered - during radionuclide analysis of soil samples (collected using samplers with an area of some 150 cm^2) - a rather unexpected phenomenon, viz. that sometimes samples collected on a flat and evenly sod-covered area produced results differing by as much as tenfold from one another. We had never come across this phenomenon when studying trails of radioactive fallout from nuclear blasts, apart from in the zone nearest to ground zero. Subsequent autoradiography of the soil samples showed that sometimes several dozen highly active "hot" particles accounted for over 90% of the activity in the entire sample.

The problem of "hot" particles was first broached and worked on in the late fifties/early sixties, when highly active particles were discovered and studied which differed from the overall sample by their unusual radionuclide composition, since they mainly contained isotopes of refractory elements (or refractory compounds of elements). Subsequent studies extended and deepened our knowledge of the chemical and radionuclide composition of "hot" particles, the distribution of total radioactivity and individual radionuclides by particle size, and other characteristics and properties. General agreement was reached on what "hot" particles are exactly. In a fairly early piece of work based on an investigation of the relationship between particle size and radioactivity, Sisevsky showed that "hot" particles are not an exclusive group of particles but, in terms of size, essentially form part of the overall distribution of a radioactive aerosol.

The discovery of "hot" particles in the accidental releases from Unit IV of the Chernobyl NPP revived the old arguments on all aspects of this problem. This is not surprising, given that "hot" particles can - if we take nuclear power engineering as a whole - occur in the near-surface layer of the atmosphere in only two events: a) if a satellite fitted with a nuclear reactor fails to burn up completely in the Earth's atmosphere, and b) in the event of a serious reactor accident entailing the ejection of aerosols into the atmosphere. The second of these is involved here, of course, it being the second serious reactor accident (after Windscale) to leave a significant trail of radioactive fallout on the ground. The fact that "hot" particles formed in reactor accidents had not been fully studied, led to a revival of the old debates.

Nowadays, an accident involving active zone meltdown and complete (or partial) breaching of the vessel is classed as a serious reactor accident. In such incidents the quantity and radionuclide composition of the radioactivity released into the atmosphere depends on such a large number of factors that they cannot as yet be physically or mathematically modelled to any satisfactory degree (even when using highly powerful computers). The USA, Federal Republic of Germany, Japan and various other countries "armed" with nuclear power are carrying out intensive research into the chemistry of serious accidents and evaluating the associated releases of radioactivity. But each accident springs its own surprises. For example, studies of the accidental releases from the USA's Three Mile Island nuclear power plant revealed

that much less (by several orders of magnitude) iodine-131 and tellurium-132 was released than expected, while analysis of the Chernobyl releases shows that they contained highly significant amounts (also exceeding the computed expectations) of such low-volatility substances as barium, strontium and cerium radionuclides as compared to the amounts of the radionuclides of caesium, iodine and tellurium released.

Modelling of serious accidents also provides important (but not as yet extensive) information on the nature of the aerosol particles formed in accidental releases, "hot" particles included. Thus, for example, it has been established that intense fuel burn-up facilitates the formation of the smallest particles by size and the greatest release of radioactivity, that radionuclide fractionation is related to fuel porosity, that the overall mass of aerosols rises with increasing temperature and scale of oxidation processes, and that much of the substances released in accidents is transported in the form of aerosols ranging from 0.01 to 10 microns in size.

Since we do not wish to become involved in, or continue, the controversy (against the backcloth of Sisevsky's findings) about what a "hot" particle is, people should decide for themselves what they understand by this designation for the highly active particles which show up clearly when exposed to an X-ray plate while the fog from the fine particles goes practically unnoticed.

In our view, the above-mentioned geophysical aspects of the "hot" particle problem embrace all issues relating to their distribution inthe various parts of the environment (soil, air, bed sediments, leaves and needles of trees, etc.), particle distribution by size (insofar as this is linked to fallout characteristics), radionuclide distribution by various particle types and, of course, secondary contamination of the atmosphere by "hot" particles due to wind resuspension of dust.

Aerosol contamination of the atmospheric air occurs over time in two stages, the first (a relatively short one) encompassing release of radioactivity from the reactor, transport of radioactive clouds in the atmosphere and settling of aerosols onto ground and water surfaces, while the second (which is continuous) involves secondary contamination of the atmosphere via wind resuspension of dust.

On the basis of this breakdown we also proceeded to collect samples of radioactive aerosols and the "hot" particles contained therein.

In order to obtain samples of such particles from the soil, we took the thin upper layer (less than 1 cm thick), and then carefully dried and intermixed it prior to radiography, doing the same for samples of bed sediments taken from rivers and other water bodies.

To collect samples of atmospheric fallout deriving from resuspension of dust we used the calibration network mentioned earlier, cutting out rectangles (15 x 20 cm) from the centre of screens in order to study the hot particles.

Figs. 12 and 13 are radiograms of pieces of gauze cut out of screens; they show "hot" particles which were lifted into the air by wind resuspension of dust and settled on the screens. Fig. 12 (screen 8 km from the Chernobyl NPP) shows four large (in terms of specific

Fig.12: AUTORADIOGRAPH OF CENTRAL SECTION OF A GANZE SCREEN EXPOSED FOR A MONTH (2.09.88 - 5.10.88) AT A CALIBRATION POINT WITH A BEARING OF 250°. 8 km FROM UNIT IV OF THE CHERNOBYL NPP

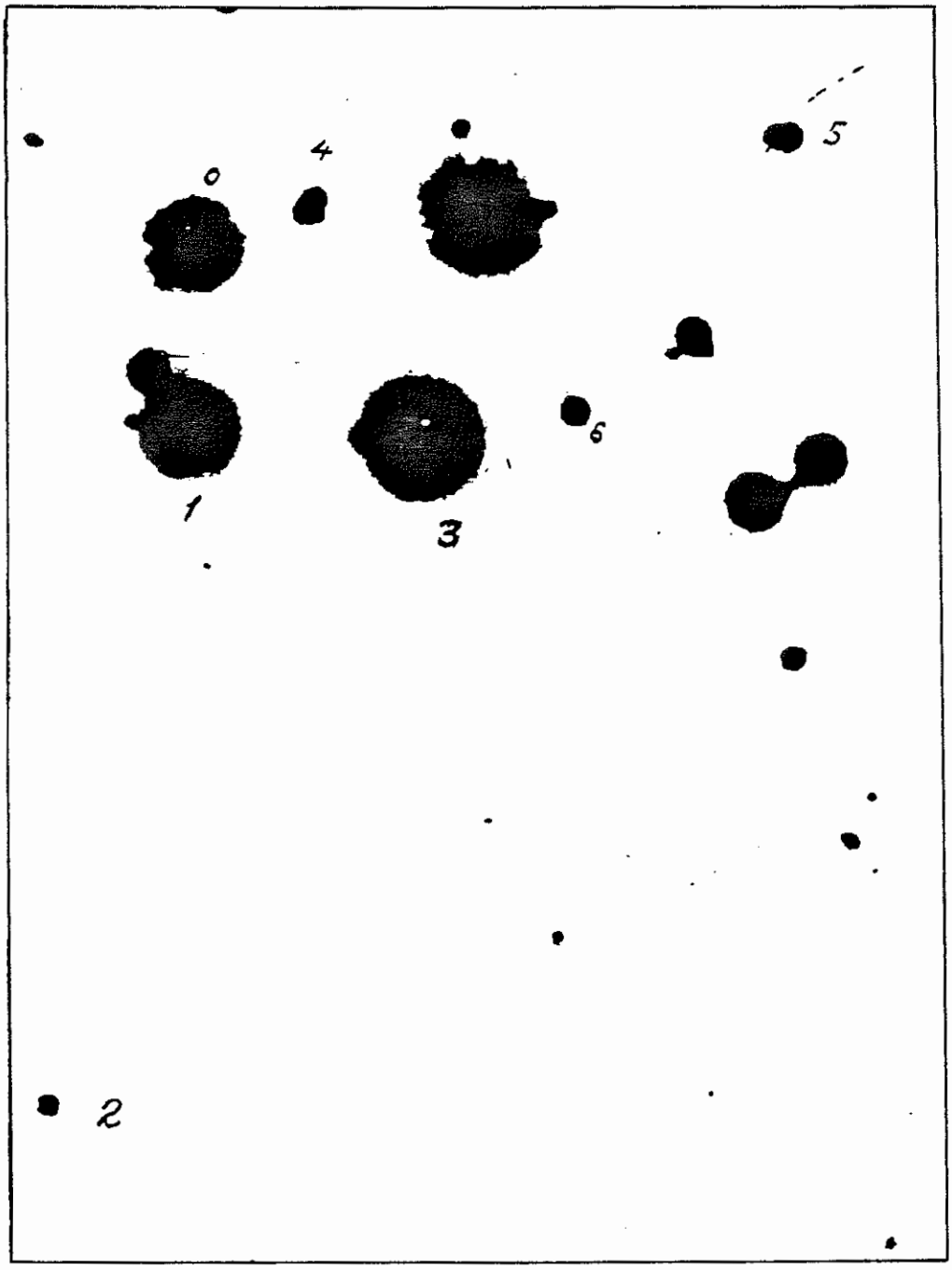
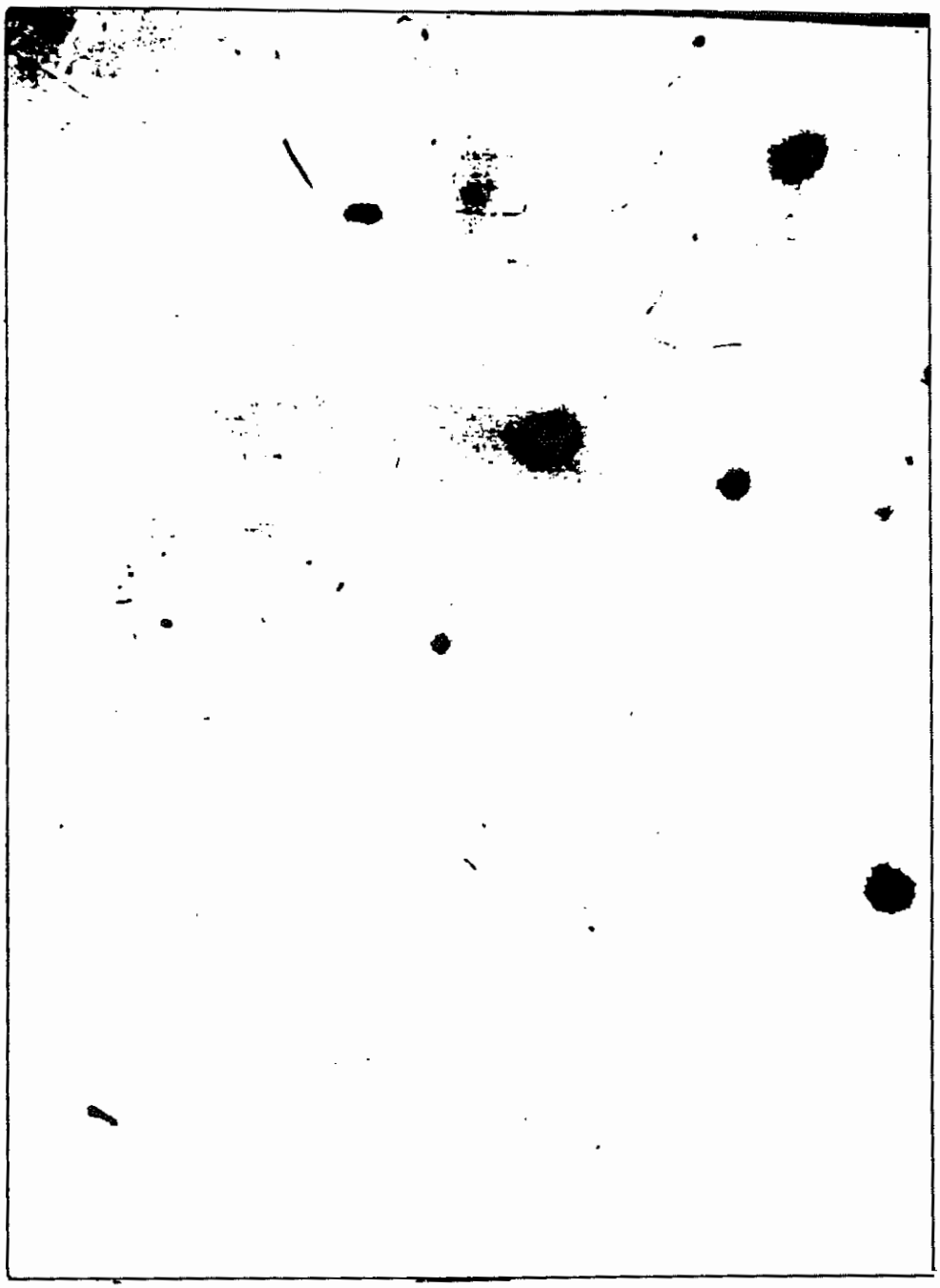


Fig.13: AUTORADIOGRAPH OF CENTRAL SECTION OF A GANZE SCREEN EXPOSED FOR A MONTH (9.09.88 - 10.10.88) AT A CALIBRATION POINT WITH A BEARING OF 20°, 30 km FROM UNIT IV OF THE CHERNOBYL NPP



activity) particles and eight "medium-sized" particles represented by black blotches which are clearly smaller. Fig. 13 (further out from the Chernobyl NPP at 30 km) has only four particles, and these are medium-sized.

Autoradiography was the main method of pinpointing "hot" particles for subsequent removal from the samples collected.

Our investigations involved several hundred autoradiographs of various samples, mostly of soil, bed sediments and dust which had settled on the gauze screens.

The only main alternative to autoradiography is selection of particles using a tightly collimated detector pointed at the surface under study, and which is moved automatically or manually to scan the surface successively and continuously. In our experience (we used this back in the early sixties) this method is very laborious and does not guarantee that researchers will pinpoint even all the highly active particles.

The autoradiography method we use allows us to a) establish unequivocally the presence and number of "hot" particles on the surface of the sample studied, b) classify them initially by degree of activity (by blotch size and intensity), c) pick out individual particles (if they are not in the conglomerate) from the whole sample for further gamma-spectrometric and radiochemical analysis, d) detect particles of fairly low-level activity by exposing the sample for a long time, and e) set up a documentation bank on "hot" particles.

Following autoradiography, all parts of the exposed samples on which any significant dark blotches were detected were subjected to visual checking under a powerful microscope. It should be noted that during the whole time we worked with particles we very rarely saw any individual "hot" particles; they were either fixed as "riders" on larger particles, or were bound up in the particle conglomerate, from which it was quite difficult to pick out the "hot" particle proper. We put this down to the fact that all the particles we collected are what one might call "secondary" particles, a term explained below.

Given the accident phenomena involved here, it can be said that the aerosol component in the release must, of necessity, have included the following two basic types of particle (by structure and composition):

- Type 1 - dispersed substances stemming from the fuel elements and ejected into the atmosphere, including both radionuclides from the fuel as well as shattered and induced radionuclides which had accumulated during the reactor's operational life, and

- Type 2 - particles formed via the condensation on various germs (condensation nuclei) of substances which evaporated during the explosive and high-temperature processes.

The radionuclide composition of Type-1 particles should typically be reasonably stable, whereas the composition of Type-2 particles greatly depends on the condensation nuclei. Obviously, combinations of (highly variegated) particles from these two types also exist, for example when the condensation nucleus of a Type-2 particle is a dispersed substance, i.e. a Type-1 particle.

The phase involving formation of primary particles was followed by a phase in which they changed their form and composition during atmospheric transport due to their interaction with non-radioactive atmospheric dust, a time when many of the primary particles formed conglomerates. The formation of particles with radioactive carriers (which we designated "secondary" particles) ended as the radioactive atmospheric dust fell onto the earth, water or anthropogenic surface or as it interacted with vegetation, the particles deposited in this manner being richer in non-radioactive components.

We traced this process - from the primary particle through to the dust destined to be resuspended in the air by the wind - in order to back up the following two assumptions: a) the dispersive size of the "hot" particles appearing in the air in the course of wind transport should not differ from the dispersive size of the non-radioactive dust in each specific area, and b) when evaluating the medico-biological aspect of the "hot" particle hazard, we should not be guided by the size-related distribution of the primary "hot" particles, since this might lead to large errors in our evaluations.

Fig. 14 is a photograph (taken under a microscope) of a typical conglomerate of radioactive and non-radioactive particles (extracted from a dried sample of bed sediment taken from the cooling pond). On the particle about one millimetre in size we can see smaller fused "riders", which are also primary "hot" articles deposited near the accident site.

Fig. 15 shows the result of interaction between fine radioactive dust and an "item of vegetation". This secondary "hot" particle fell onto a screen in the summer of 1989 (bearing 250°, 8 km from the stricken unit).

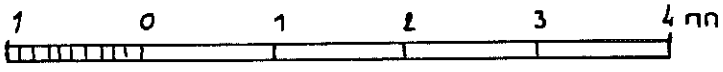
Fig. 16 shows a "hot" particle of particular interest since it represents a special class of "hot" particles in the "Chernobyl" releases. It, too, was taken from a screen in 1989 (bearing 250°, 8 km away). Structurally speaking, it is a fragment of graphite ash, one of whose formation sources was the black jet of ash ejected (mainly southwards and south-westwards) in the second half of 26 April 1986, as confirmed by precise eye-witness accounts. These particles crumble easily when touched lightly by a needle, and this is clearly visible under the microscope.

After being checked visually and selectively photographed, all parts of the sample showing up on the autoradiographs as clearly identified black blotches were cut out, tagged and then subjected to gamma-spectrometric analysis.

Fig.14: PHOTO (under microscope) OF A TYPICAL CONGLOMERATE OF RADIOACTIVE AND NON-RADIOACTIVE PARTICLES
(bed sediment from cooling pond)

Fig.15: FINE PARTICLES ATTACHED TO AN "ITEM OF VEGETATION"

Fig.16: 'HOT' PARTICLE COMPOSED OF GRAPHITE ASH: SETTLED IN 1989 ON SCREEN 8 km FROM THE CHERNOBYL NPP



So far we have subjected several hundred particles (or particle conglomerates) to radionuclide analysis using semiconductor gamma-spectrometers. Without going into too much detail, we can divide all the particles into two types:

- particles containing cerium-144 and zirconium-95 with one or other collection of radionuclides;

- particles in which these radionuclides are either completely absent or are present in hardly noticeable quantities only.

The fact that there are two groups is quite logical, given that radioactivity was released from the stricken reactor in two phases. The release phase relating to the series of explosions (in the early hours of 26 April 1986) involved the formation of aerosols with a radionuclide composition similar to the non-fractionated type, reflecting the composition of the fuel and fission fragments prior to the accident. The phase of releases during the reactor fire was accompanied by high temperatures and, as already known, involved large amounts of radionuclides of caesium-137, caesium-134, iodine-131 and other volatile products.

Table 5 gives the findings from radionuclide analysis of "hot" particles taken from fallout landing on screens in spring 1989. As we can see, the "hot" particles greatly differed in their radionuclide composition and were found in fallout throughout the 60-km zone.

Some of the particles stand out on account of their being very rich in ruthenium-106 (as Table 5 shows). Fig. 17 shows the gamma spectrum of one such particle collected in the "red" forest (measured on 1 March 1989). The caesium-137 peak on the gamma spectrum of the overall soil sample collected at the same site is several times higher than the ruthenium-106 gamma peak, while in Fig. 17 it is the other way around.

It is interesting to compare the fractionation characteristics of the radionuclides in the "hot" particles in the manner the authors did earlier for samples of atmospheric aerosols and fallout from nuclear explosions. Figs. 18-20 give the results of just such an analysis for four pairs of radionuclides: the reference pair caesium-137/cerium-144 and the pairs under scrutiny - zirconium-95/cerium-144, ruthenium-106/cerium-144, and antimony-125/cerium-144.

The fractionation coefficients were calculated using the usual formulae. As Figs. 18-20 show, the correlational dependencies of the fractionation coefficients studied are similar to those in nuclear blasts: there is hardly any fractionation of zirconium-95 from cerium-144, while the volatility of ruthenium-106 is somewhat lower than that of caesium-137, and that of antimony-125 is, in contrast, greater than that of caesium-137.

Table 5: CONTENT OF RADIONUCLIDES IN 'HOT' PARTICLES SETTLING ON SCREENS IN THE CALIBRATION NETWORK

Site index	Coordinates of place at which screen erected		Content of radionuclides in 'hot' particles in relation to date of accident (Bq/preparation)					Number of particles setting on centre of screen
	bearing degrees	distance (km)	106	125	134	137	144	
1.	10	5.0	-	-	0.50	0.53	-	1
2.	230	6.0	-	-	0.29	0.42	-	6
3.	340	7.0	-	-	0.70	1.6	-	11
4.	240	10	-	-	1.8	2.4	-	5
5.	40	14.5	-	-	7.2	13	-	6
6.	10	17.5	-	-	5.1	11	-	8
7.	20	20	-	-	0.37	0.96	-	3
8.	50	25	-	-	2.3	8.5	-	3
9.	20	30	-	-	6.9	15	-	7
10.	40	45	-	-	0.24	0.33	-	1
11.	260	60	-	-	0.89	1.3	-	4
12.	310	5	-	-	0.99	2.6	44	4
13.	250	8.3	-	-	3.3	6.2	280	16
14.	120	10	-	-	3.0	5.2	350	5
15.	190	12	-	-	3.3	8.9	430	6
16.	360	14.5	-	-	1.5	6.2	120	7
17.	10	17.5	-	-	3.7	12	210	8
18.	60	25	-	-	3.0	5.4	270	2
19.	200	37.5	-	-	0.51	0.54	46	2
20.	270	52.5	-	-	2.7	7.6	240	1
21.	250	60	-	-	4.9	11	100	5
22.	340	5	44	1.0	2.8	5.7	110	6
23.	250	8.3	200	-	9.0	21	2700	16
24.	60	10	23	0.59	0.96	4.2	150	2
25.	340	12.5	120	2.6	1.3	2.5	330	2
26.	320	20	19	-	1.3	2.4	41	3
27.	60	25	26	-	1.4	2.5	200	2
28.	270	30	100	-	2.3	3.9	220	4
29.	20	37.5	210	3.9	28	56	660	1

Fig.17: GAMMA SPECTRUM OF A HOT PARTICLE RICH IN RUTHENIUM-106
'RED' FOREST, BEARING 210°, 1500 m FROM UNIT IV
OF THE CHERNOBYL NPP

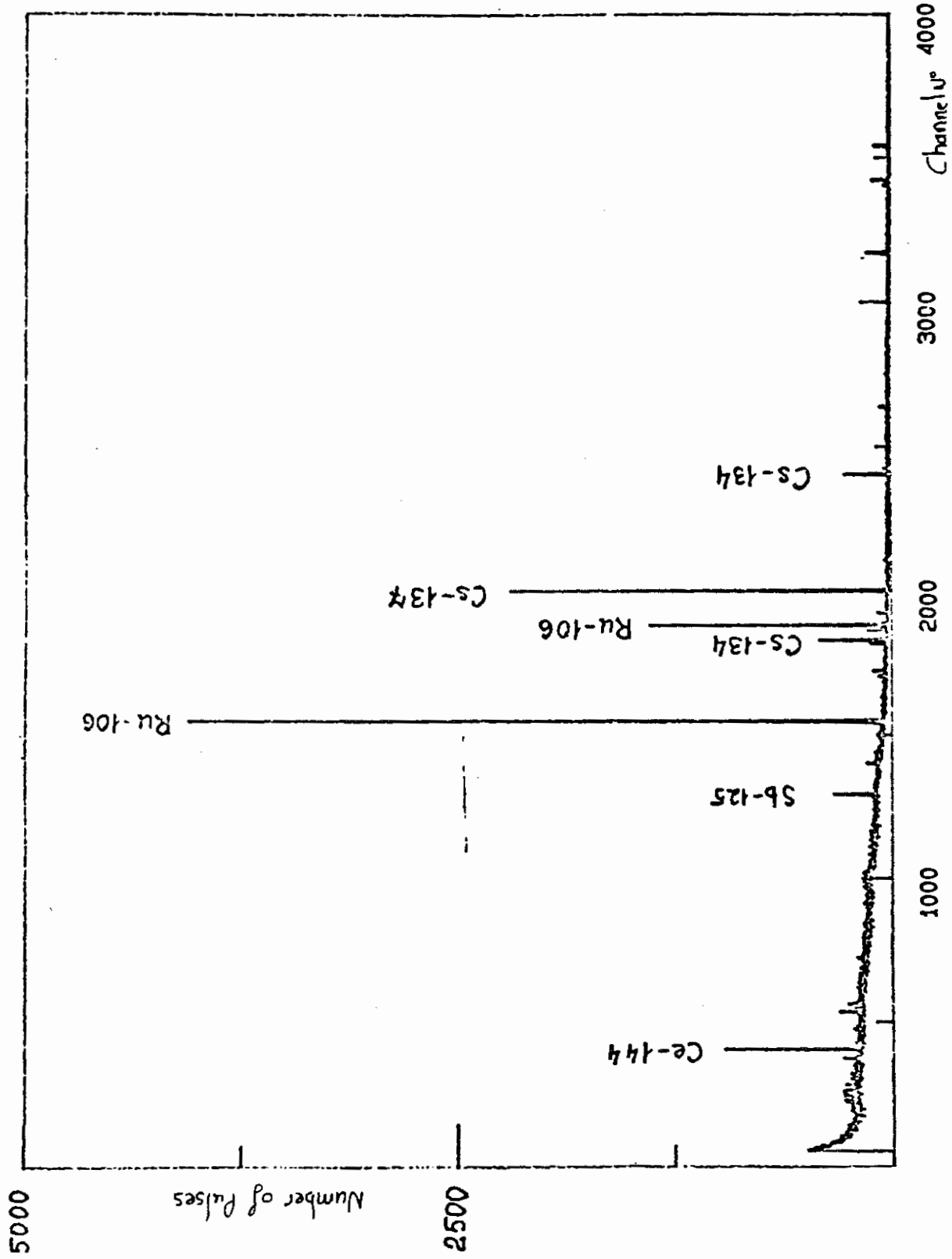


Fig.18: CORRELATIONAL DEPENDENCE OF THE FRACTIONATION COEFFICIENT f^{95}_{144} ON THE FRACTIONATION COEFFICIENT f^{137}_{144} 'HOT' PARTICLES TAKEN FROM SAMPLES OF SOIL. 1987-1988

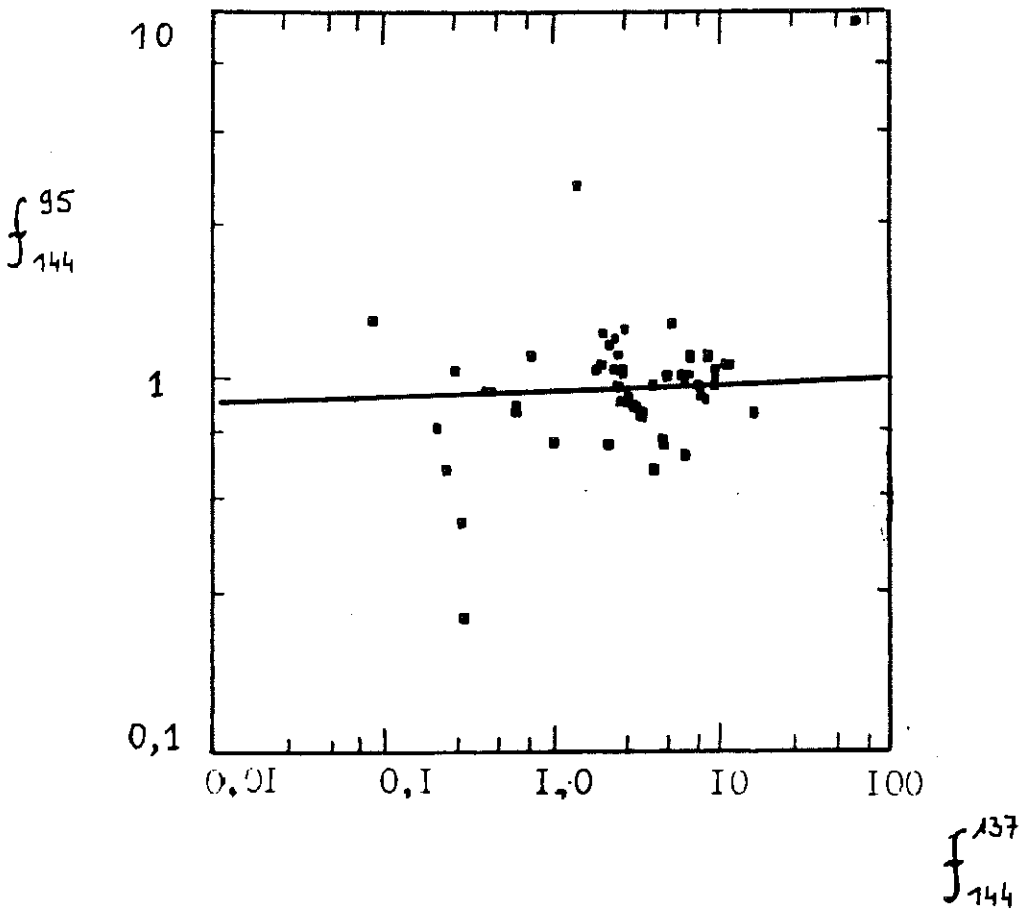


Fig.19: CORRELATIONAL DEPENDENCE OF THE FRACTIONATION COEFFICIENT f_{144}^{106} ON THE FRACTIONATION COEFFICIENT f_{144}^{137}. 'HOT' PARTICLES TAKEN FROM SAMPLES OF SOIL. 1987-1988

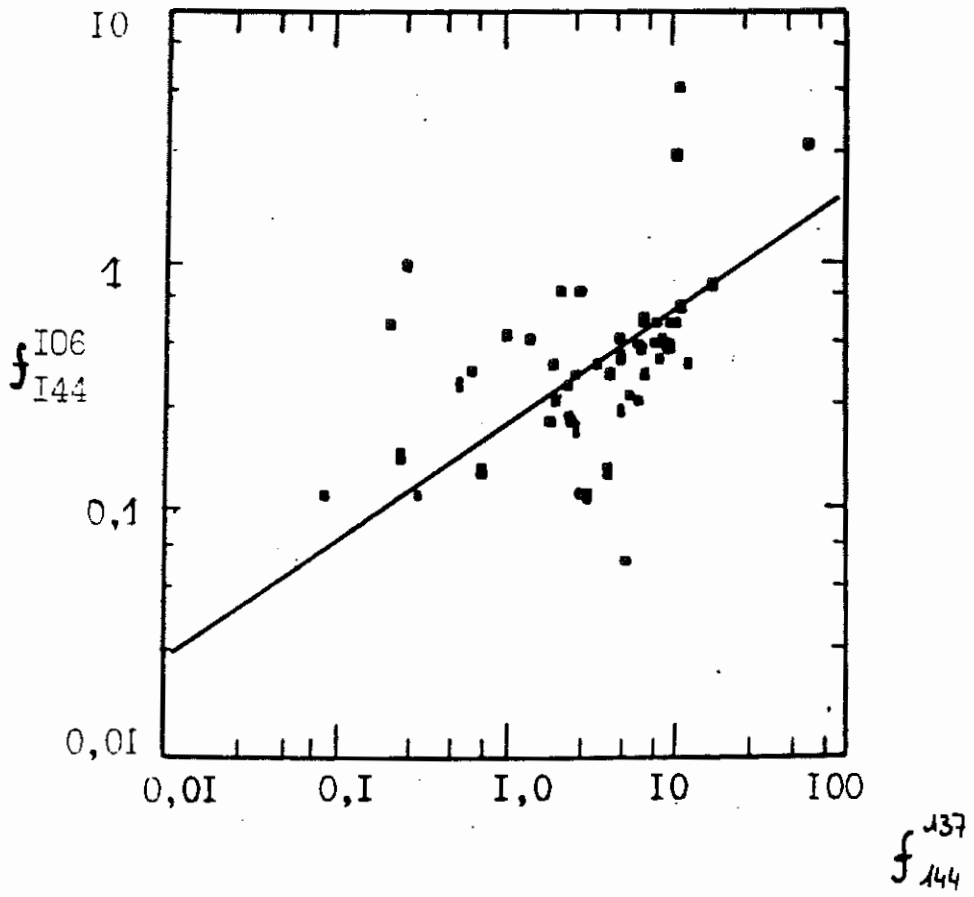
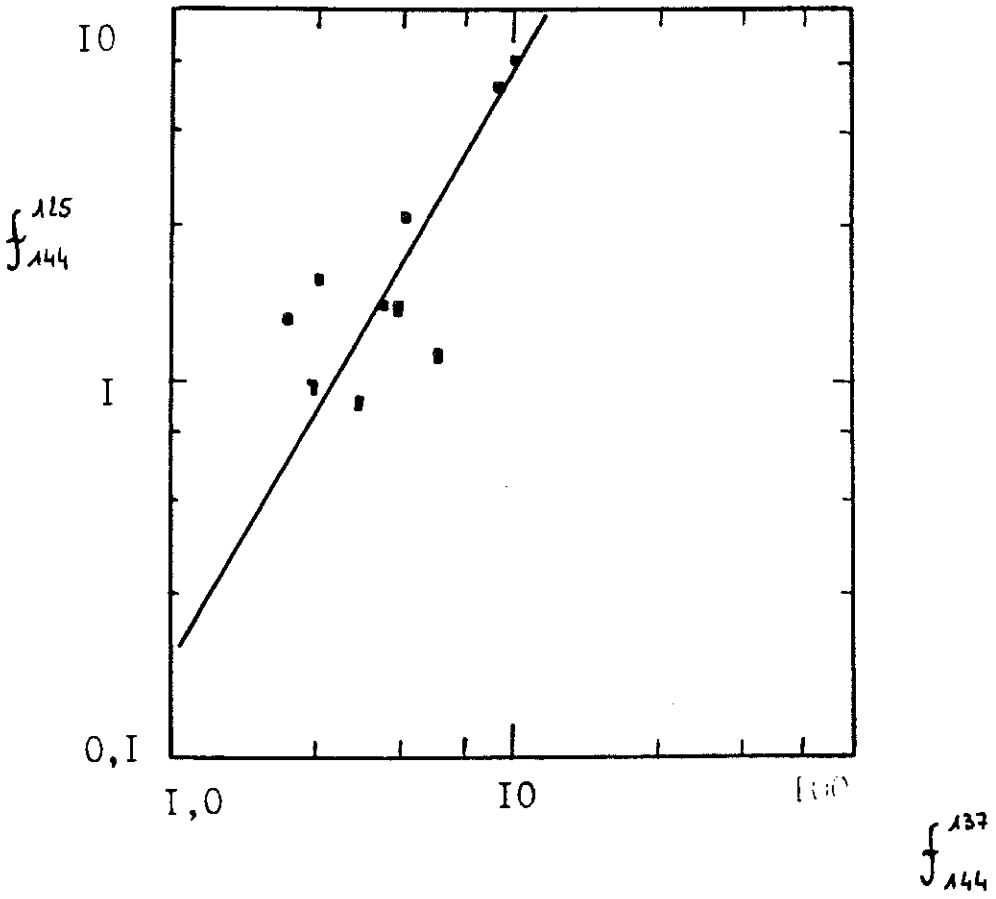


Fig.20: CORRELATIONAL DEPENDENCE OF THE FRACTIONATION COEFFICIENT f_{144}^{125} ON THE FRACTIONATION COEFFICIENT f_{144}^{137}. 'HOT' PARTICLES TAKEN FROM SAMPLES OF SOIL, 1987-1988



Although four years have elapsed since the Chernobyl NPP accident, our knowledge of the intensity of radiation, and the forms, structures and chemical composition of "hot" particles still leaves a lot to be desired. And we should not forget that each individual "hot" particle contains a great deal of information about the physico-chemical processes occurring at the time of the accident, in particular at the time any given particle was formed and ejected into the atmosphere.

Session IV

ENVIRONMENTAL CONTAMINATION AND TRANSFER

Part I

Deposition of Activity from the Windscale Accident

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ABSTRACT

The deposition pattern was determined by measuring activity in grass and soil, gamma dose rate near the ground and gamma dose at 150 m (aerial survey). The deposited activity was mainly ^{132}Te + ^{132}I and ^{131}I. The $^{137}Cs/^{131}I$ ratio in deposition was 0.02, compared with about 0.1 in air filters. Relatively small activities of Sr and refractory fission products were deposited.

The effective half life of ^{131}I in herbage was 4.9 days, and in milk 4.7 days. Assuming a herbage consumption of 30 kg wet weight per day, the transfer factor herbage/milk was $3x10^{-3}$ d l^{-1}.

A ban on milk consumption was enforced where the peak concentration of ^{131}I exceeded 0.1 μ9∪⊠$^{-2}$, corresponding approximately to the 1 μCi m^{-2} isopleth of deposition.

In the area of maximum deposition, near Seascale, ^{137}Cs in milk reached about 1500 Bq l^{-1}. Outside the restricted area, concentrations of ^{137}Cs in milk were of the same order as those due to weapons fallout.

1. Pattern of deposition

The deposition from the Windscale accident was described in a paper by Dunster et al to the 1958 Geneva Conference[1] and in a series of published papers and reports, copies of which have been lodged with IAEA. It is not proposed to attempt to reproduce all this data in the present paper, but to bring out some points for comparison with the Kyshtym and Chernobyl accidents.

After the accident, the first measurements of deposited activity were made with ionisation chambers mounted on vehicles, but the vehicles picked up activity from the roads, raising the background, and it was found better to use hand-held geological survey instruments. These were of the gamma scintillator type, calibrated in mR/hr against radium sources, but with an energy-dependent response. The gamma dose was due mainly to ^{132}Te + ^{132}I, ^{131}I and ^{137}Cs. In the period 4 to 20 days from the accident, the dose from ^{131}I was dominant, and this enabled a simple conversion from recorded dose rate to deposited activity of ^{131}I to be calculated[3].

Beginning 8 days after the accident, an aerial survey of the district was done, with an aircraft flying at 150 m height[4]. By this time, the pattern of deposition had been established from ground measurements, but the aerial survey was useful in confirming it. Subsequently, the results of the gamma surveys and measurements of ^{131}I in grass and soil were correlated to establish the isopleths of deposition more accurately[5].

Emissions from Windscale lasted about 18 hours, from 1600 on 10th October to 1000 on 11th October 1957, at which time the fire was put out with water[2]. On 10th October, the wind was light from the SW, but at 0100 on the 11th a weak cold front passed over, giving a change in wind direction to NW[6]. The activity emitted before the passage of the front was carried to the NE, then swept down to the SW.

The release peaked about 0900 on the 11th, and gave a plume of deposition running SSE. At 100 km from Windscale, the half value width of this plume was 8 km[5], similar to the width of the Kyshtym plume at this distance[7]. The maximum dkeposition of ^{131}I, 30 $\mu Ci/m^2$ (1.1×10^5 Bq/m^2) was in this plume at 7 km from Windscale.

2. Nuclides deposited

With the gamma spectrometry available in 1957, the deposition of ^{137}Cs could not be assessed until the ^{131}I had decayed. A survey of ^{137}Cs in

soil, carried out in 1958[8] showed a pattern of deposition similar to that of ^{131}I, the $^{131}I/^{137}Cs$ ratio (as at 10.10.57) being 50:1 (Table 1). This compares with 20:1 in the fuel and 11:1 on air filters[9,10].
Despite the use of water to extinguish a graphite/uranium fire which had reached 1300°C, there was no explosion at Windscale, and very little dissemination of uranium or refractory fission products. The release of ^{90}Sr was less than 1% of that of ^{137}Cs, and the deposition of ^{90}Sr near Windscale could not be readily distinguished from that deposited locally in previous emissions of uranium oxide particles[11], or from distant weapon tests. ^{210}Po behaved similarly to ^{137}Cs in respect of fractional release from the fire zone and deposition to grassland (Table 1).

3. Retention on herbage

During the month after accident, repeated samples of grass were taken from a field at Seascale, 4 km from Windscale, which was not being grazed by cattle, and these showed activity of ^{131}I, on grass, per m^2 of soil surface, diminishing with a 4.9 day half life[12]. The corresponding field loss half life is 13 days. The $^{131}I/^{137}Cs$ ratio in the grass decayed with an apparent half life of 9.5 days, slightly longer than the radioactive half life of ^{131}I. This implies that field loss of ^{137}Cs was slightly faster than that of ^{131}I. Re-volatilization of ^{131}I from grass or soil was not observed, and there was no evidence of re-suspension of activity in dust. Most of west Cumberland is grassland, the climate is moist, and soil erosion is minimal

4. Velocity of deposition

There was no measurement of air dosage in the area immediately downwind of Windscale. In Lancashire and Yorkshire, cellulose filters were operated by local authorisities for measuring smoke in the atmosphere. These were collected and analysed at Harwell[10]. Relative to the dosage deduced, the local deposition of ^{131}I gave a velocity of deposition of 4 mm/s[5]. The filters would not have trapped organic iodine vapour, but they were probably fairly efficient for inorganic iodine. The velocity of deposition of ^{137}Cs can be estimated to havge been about 0.7 mm/s, similar to values noted in Britain and Denmark for ^{137}Cs from Chernobyl.

5. Transfer of activity to milk

West Cumberland is a milk producing area, and, at the time of the accident, the cows were grazing the fields. It was decided to sequester milk in areas where the ^{131}I activity exceeded 0.1 μCi/l (3700 Bq/l) and the measures taken to do this were fully described in ref (1).
At a farm in Seascale, where extensive measurements were made, the transfer factor grass/milk defined by

$$\text{Transfer factor} = \frac{^{131}I \text{ per litre milk}}{^{131}I \text{ per kg fresh wt of grass}}$$

was 0.1[1,12]. Assuming a daily consumption of 30 kg wet weight (allowing for some supplementary feed in autumn)

$$F_m = \frac{^{131}I \text{ per litre milk}}{\text{Daily intake by cow}} = 3\times10^{-3} \text{ d/l}$$

^{137}Cs in milk in the same area was not measured until 28th October, because of limited facilities for gamma spectrometry. By extrapolation, the peak activity was about 40 nCi/l (1500 Bq/l). Measurements of ^{137}Cs in dried milk from several areas of England and South Wales showed transient peaks in October/November 1957, compared with varying levels due to weapons testing[13]. There was no comparable increase in ^{90}Sr in milk, even at farms in the zone of maximum fallout[1].

6. References

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Table 1

Activities relative to ^{137}Cs = 1.00

	^{131}I	^{90}Sr	^{210}Po
Reactor(1)	20	1	0.13
Air filters	11	0.004	0.20
Grass/soil	50	-	0.16

(1) Inventory in zone of melting

Landscape and Geochemical Approach to Drawing up a Soil Distribution Profile for Chernobyl Radionuclides in Distant Areas

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ABSTRACT

A study (between 1987 and 1989) of the distribution of radionuclides in the 0-5 cm soil layer in various typical landscapes 60-200 km from Chernobyl revealed a link with landscape structures. One year after the accident, concentrations of caesium-137/134, ruthenium-106 and cerium-144 were found at depths of 1.5-5 cm in the soil in forests and in meadowy regions of a hydromorphic nature. In subsequent years distribution of the radionuclides within the soil varied in line with the biological cycles and typomorphic component of the soil (organic and inorganic, such as iron and manganese hydroxides and calcium carbonate forms).

While most of the radionuclides accumulated in the 0-5 cm layer, some penetrated to depths of 10-15 cm and deeper in sandy hydromorphic soils and swampy soils, with ruthenium-106 proving most and cerium-144 least mobile.

The study findings show how vital it is to analyse the distribution of Chernobyl radionuclides and their secondary migration in the environment, and to draw conclusions on the role played by spatial effects.

The landscape and geochemical structure of the region contaminated by technogenic radionuclides is of considerable significance both in the initial local distribution of radionuclide fallout and the secondary redistribution of chemical elements in soil, vegetation and subsoil water. This is because the landcape-geochemical structure reflects the variable physical and chemical properties of soil-forming rocks, soils, water, biocenoses and their generic composition, typomorphic elements and compounds and geochemical barriers, which define the character and scope of the aerial, aquatic, mechanical and biogenic migration of chemical elements, including radioactive ones [1-10].

General landscape and geochemical characteristics of the contaminated area

The Pripyat-Dnieper region where the accident took place is characterized by slightly fragmented type-1 geochemical landscapes (polesye = Pripyat marshes) on ancient alluvial and sand deposits. They are a combination of agricultural land and pine forests on podzolic and soddy-podzolic soils on watersheds and terraces (eluvial landscapes), on the one hand, and herbaceous communities on alluvial stratified, acidic, meadow and marshy soils (superaqueous landscapes) on the other. Distinctive features of the "polesye" are more intensive elimination of chemical elements under autonomous conditions and the active migration of chemical elements (acid, acid-gley class) in water, inter alia in mobile soluble organic complexes. The elements are concentrated in the soils at biogeochemical, oxygenous, gley and sorptional barriers. The migration of cationogenic elements in landscapes of the H-Ca water migration class is normally slower; in the region in question, these landscapes are represented by meadows and coniferous and mixed forests on glacial and topsoil deposits of varying composition. In addition to the geochemical barriers mentioned above, the alkaline barrier also has a definite role to play here. The Mozyr heights (formed by aeolian and eluvial sand and loess deposits on an underlying moraine) and the Bryansk and Chernigov opolye (eluvial landscapes comprising agrocenoses with fragments of oak forest on grey forest soils, soil-forming rocks and loessial and covering loams) stand out as islands on the low plains. They are characterised by their highly fragmented nature (type-II geochemical landscapes), the combination of chemical and mechanical denudation and improved fixation of many cationogenic elements in biogeochemical, sorptive and alkaline barriers. Water migration of elements in automonous Ca-class forest-steppe and steppe landscapes (the less fragmented ones in the southern part of the region) is the least significant. This does not relate to elements capable of forming mobile carbonate complexes, for example plutonium isotopes (see report by F. I. Pavlotskaya, et al.). In addition, where watersheds and slopes are fragmented and more intensely cultivated, mechanical migration of elements plays a more significant role.

Distribution of the main gamma-emitters in the soils of some connected landscapes in the outer zone affected by the Chernobyl accident

Apart from revealing the main geochemical landscapes, landscape and geochemical investigations presuppose a study of the catenary interlinking of elementary landscapes which reflect a combination of elements of the relief, types of plant communities and soils on identical source rocks[7,11] using test sections.

The authors investigated such sections between 1987 and 1989 in various directions from Chernobyl. On the whole, the concentrations and ratios of the main gamma-emitters corresponded to the estimates of contamination[12, 13] Fig. 2). With a similar ratio of main gamma-emitters in the upper 5 cm of eluvial and superaqueous soils, in 1987 their absolute content in the catenae differed by between 2 and 5 times (Tables 1, 2). In contrast to watershed superaqueous meadows, watershed forest landscapes were characterized by their relative accumulation of all radionuclides, most nuclides being concentrated in the forest litter. Examination of the terrace-edge zones - which are the accumulators of global fallout of artificial radionuclides - did not show a different relative accumulation pattern for Chernobyl radioisotopes. This leads us to assume that the contrasting nature of elementary landscapes was a significant primary factor and was linked to the influence of relief and vegetation on the character of the fallout and its retention. In addition, it could not be ruled out that some of the nuclide fallout was lost from meadow plant communities in 1986 due to removal of biomass through haymaking and grazing.

In undisturbed soils, most of the radionuclides are concentrated in the upper 5 cm. In the Khocheva River valley (30 km south of Chernobyl) in 1988, for example, this layer contained virtually 100% of the ^{144}Ce, ^{125}Sb and ^{155}Eu, 92-100% of the ^{134}Cs, 75-99% of the ^{137}Cs and 61-97% of the ^{106}Ru[10]. Maximum accumulation of all radionuclides was noted in soils in forest belts situated in watershed areas (the forest-edge effect, Fig. 3). Arable soils were found to have the lowest levels of radioisotopes, which were more evenly distributed, sometimes inversely. This inverse distribution was also seen in undisturbed fragmented loessial landscapes at the bottoms of gullies (Fig. 4). This indicates that secondary radionuclide redistribution in the catenae had begun and that it is possible to use them as reference points in tracing mass transfer in landscape and geochemical systems.

Trends in the dynamics of radionuclide distribution in the soils of connected landscapes

Re-examination of a number of test sites in the Khocheva River valley and on the Ovruch plateau (one year later) showed no significant lateral or vertical migration during this period of the main artificial gamma-emitters beyond the top 5 cm of ground cover and soil (comparison of concentration intervals with the concentrations of elements, account being taken of natural decay; Fig. 5). At the same time, mention may be made of a number of trends towards vertical and lateral redistribution of radionuclides, for instance:

1. A relative reduction in radionuclide concentrations in the upper 5 cm of soil, in some cases up to 10% by comparison with the amount calculated for this horizon, with a simultaneous increase in the proportions contained in the 5-15 cm layer both in eluvial landscapes under pine forest on sandy soils and in superaqueous grassy landscapes on alluvial soddy sandy-loam soils (an increase in this layer of up to 50%);
2. A lateral relative increase in radionuclide concentration in certain transitional grassy elementary landscapes linked with eluvial forest soils, the overall level of radionuclide contamination being maintained throughout, cannot be excluded (Figs. 5 and 6). The inverse distribution of radionuclides at the bottom of gorges in fragmented loessial landscapes referred to above (Fig. 4) may also be the result of secondary lateral redistribution.

Conclusions

The data given here underline how important it is to take account of the landscape and geochemical structure of a geographical area when studying and forecasting radioisotope migration and distribution in the environment at all stages of the investigation into the post-accident contamination profile. This allows the spatial distribution of Chernobyl radionuclides to be classified and a forecast to be made of changes in distribution due to mass transfer in the landscape. It is advisable to study the dynamics of radionuclide distribution and migration in natural conditions in geochemically contrasting landscapes, represented by connected structures, with repeat sampling at sufficiently long intervals.

Table 1

The contrast between connected landscapes in the upper and central reaches of the Vit river (60 km south-south west of Chernobyl) in 1987

Elementary landscape	Soil/ vegetation cover	Contrasting radionuclide concentrations (0–5 cm)						
		^{95}Zr	^{95}Nb	^{106}Ru	^{143}Cs	^{137}Cs	^{144}Ce	
				Upper reaches				
Neo-eluvial	Grass meadow with couch grass and euphorbia on soddy-podzolic cultivated sandy-loam soil	I	I	I	I	I	I	I
		2.5	1.9	21.0	9.3	25.6	39.7	100.0
Superaqueous	Grass-sedge meadow on alluvial humus gley loamy soil	1.7	3.5	1.6	1.9	1.7	2.0	1.9
		2.2	3.5	18.4	9.4	24.0	42.4	100.0
Superaqueous	Mixed-grass meadow on alluvial soddy layered sandy-loam soil	1.7	5.4	1.9	1.5	1.6	2.0	1.9
		2.2	5.5	21.2	7.5	20.7	42.9	100.0
				Central reaches				
Neo-eluvial	Pine forests: Moss and lichen on soddy-podzolic cultivated sandy soil	2.2	1.2	1.3	1.1	1.1	1.5	1.3
		1.5	4.6	20.0	10.7	28.8	34.1	100.0
Eluvial - accumulative	Blue-grass meadow on loamy sandy soil	1.04	0.9	0.8	0.9	0.9	1.1	0.9
		1.7	4.8	17.7	11.5	30.7	33.6	100.0
Superaqueous	Cinquefoil-blue-grass meadow on illuvial ferriferous sandy-loam soil	I	I	I	I	I	I	I
		1.5	5.1	19.5	11.9	32.4	29.6	100.0

Comments: Upper line - lateral contrast of connected landscapes, the concentration of nuclides in one of which being taken as 1; Lower line - contribution of separate nuclides to the total gamma-emitter contamination in separate landscapes (as a percentage of total contamination).

Table 2

Contrasting concentrations of the main gamma-emitters in the soils of connected landscapes in the Sozh river valley (250 km north of Chernobyl)

Elementary land-scape	Vegetation	Soil cover	Contrast in radionuclide concentrations			
			^{106}Ru	^{134}Cs	^{137}Cs	(Ru + Cs)
Superaqueous	Cinquefoil -grass meadow	Humic peat gley	1.7	1.4	1.5	1.6
			9.9	24.3	63.9	98.1
Superaqueous (terrace-side)	Horsetail/ grass meadow	Humic gley	1.9	1.3	1.4	1.4
			11.5	23.2	65.3	100.0
Neo-eluvial	Blue grass/ grass meadow	Humic podzolic	2.3	2.0	2.1	2.1
			9.8	24.7	65.5	100.0
Eluvial	Spruce grove/ mountain sorrel	Podzolic sandy loam	I	I	I	I
			8.8	26.0	64.6	99.4
Eluvial	Apple orchard with mixed legume -grass cover	soddy podzolic sandy loam	1.5	1.3	1.4	1.4
			9.2	24.2	66.6	100.0

Notes: 1. See comments on Table 1.

2 The final column gives the proportion of total gamma activity in the soil (between 0 and 5 cm) accounted for by the three main gamma-emitters.

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Additional comments

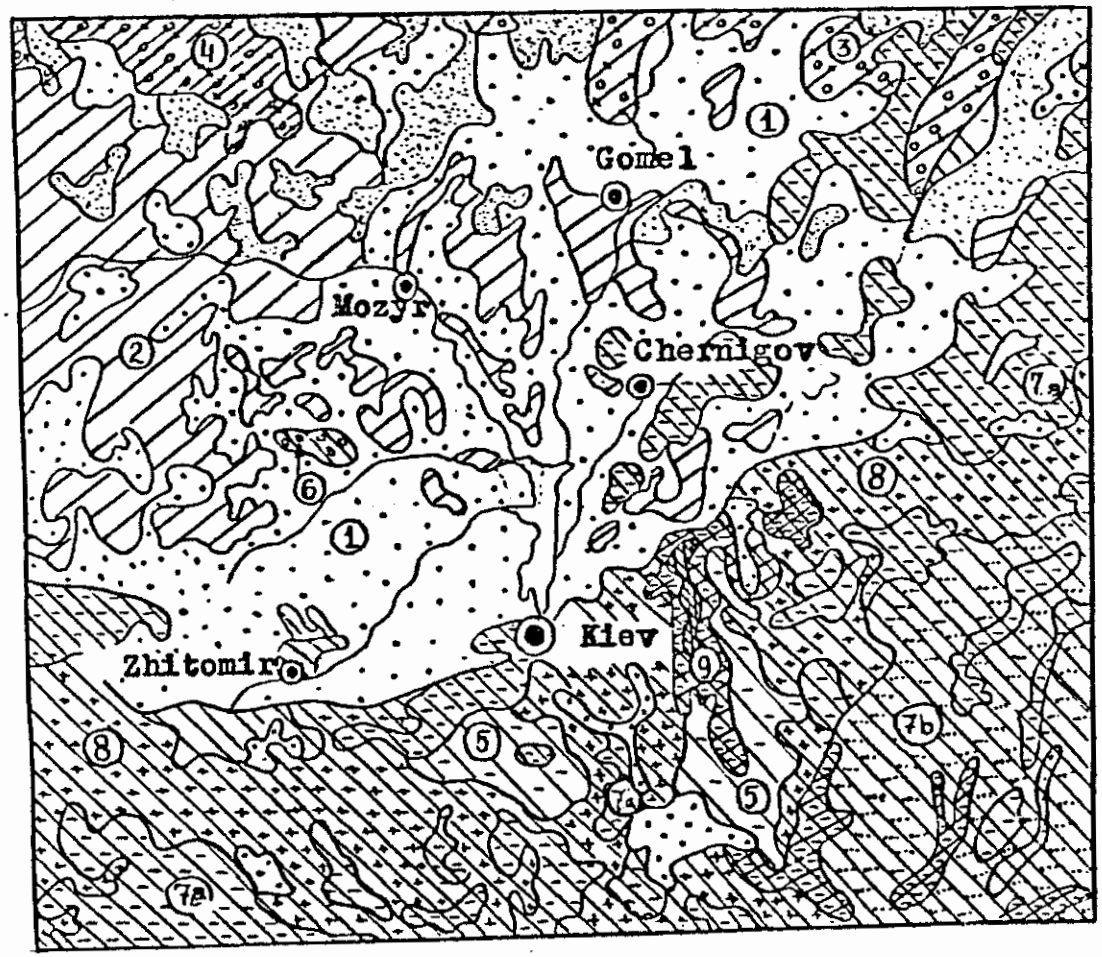
Fig. 1 Legend to survey map of geochemical landscapes:
Genuses: I - flat plain relief; slow water exchange, chemical denudation often predominates over mechanical denudation, constant boundaries between autonomous and subordinated landscapes, insignificant differences (where they exist); II - hilly relief, broken heights; mean water exchange rate; varying proportions of mechanical and chemical denudation; distinct boundary between autonomous and subordinated landscapes.
Types: a) on glacial deposits of varying composition (boulder loams, sands and combinations thereof); b) alluvial and fluvioglacial sands, mainly quartz; c) loess and loess-type rocks; d) combination of loess-type rocks with platform sedimentary formations (limestones and clays), including those near a related seam of shield formations.
Classes (by water migration type): A) slightly mineralized neutral or slightly acid waters, sometimes rich in organic matter: H) acidic, H-Fe) acid-gley; B) hydrocarbonate/calcium slightly and moderately mineralized waters, often hard: Ca) carbonate, Ca-Fe) carbonate-gley; C) moderately and heavily mineralized waters, hydrocarbonate and chloride-sulphate: H-Ca) transitional from acid to calcium, sometimes combined with acid and calcium, Ca-Na) calcium-sodium.
1a) polesye landscapes with predominantly podzolic soils in autonomous conditions, 1) soddy podzolic. The division in contour 7 is due to the greater role played by water migration in Starodub landscapes (7a; A. I. Perelman) compared with Poltava landscapes (7b).

Fig. 3 Description of vegetation and soil cover at the test sites: 159) oak and pine forest with birch on slightly soddy, medium podzolic sandy soil;

160) arable soddy slightly podzolic soil sown with fodder grass; 161) grass meadow on soddy gleyey sandy soil; 162) mixed-grass meadow on soddy gleyey sandy loam soil on banded alluvium; 164) mixed-grass meadow on soddy gleyey cultivated soil; 165) grass/leguminous crops on soddy slightly podzolic sandy loam soil; 166) field-protecting belt of pine forest with forest litter on soddy slightly podzolic cultivated sandy soil; 167) grass meadow on soddy slightly podzolic sandy cultivated soil; 168) sparse pine stands on similar soil.

Fig. 4 Soil and vegetation cover: 120, 129 - slightly loamy leached chernozem under crops; 121-123, 126-128, 131 - natural grass/herbaceous vegetation on loamy chernozems, partly podzolized, sometimes with buried humic horizons; 124, 125, 130, 132 - sedge/herbaceous/grass meadow on meadow chernozems, partly washed or silty.

Figs. 5, 6 Soil/vegetation cover on test sampling sections: 158) pine forest on formerly arable soddy, slightly podzolic sandy soil on alluvial sand; 157) herbaceous/grass meadow on soddy sandy cultivated soil on alluvial sand; 156) herbaceous/grass pasture on soddy gleyey moderately loamy silty soil on sandy alluvium; 153) grass/leguminous/herbaceous meadow with sedge on alluvial stratified moderately loamy soil with buried humic horizon; 152) herbaceous/leguminous/grass meadow on soddy gleyey sandy soil on stratified alluvium; 151) herbaceous/grass meadow on soddy podzolized sandy soil on stratified sandy alluvium; 150 - pine forest with green moss and forest litter on slightly podzolic cultivated sandy soil.



Genus	Type	Class A: H	Class A: H-Fe	Class B: H-Ca	Class B: Ca	Class C: Ca-Fe	Class C: Ca-Na
I	a			3			
I	b	1a, 1b	2				
I	c				5	9	10
II	a			4	6		
II	c				7a, 7b		
II	d				8		

Fig. 1 Map of natural geochemical landscapes in the Pripyat-Dnieper region (A.I. Perelman, 1962,1975; with additions).

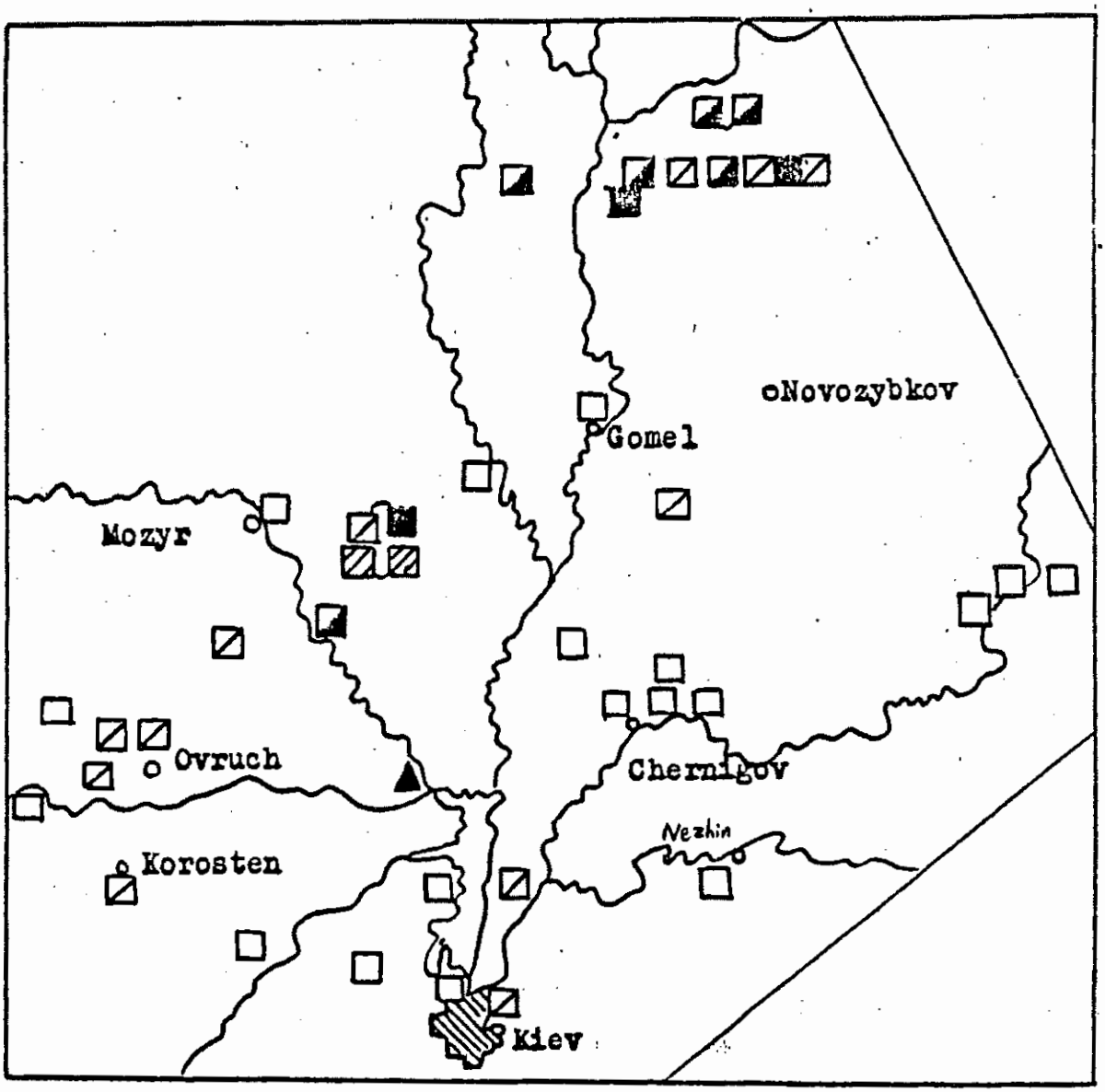
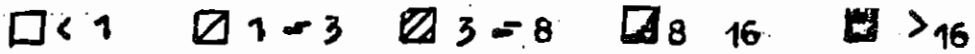


Fig. 2 Density of topsoil contamination by gamma-emitters (resp. units) at sampling sites on 28.07.1987.



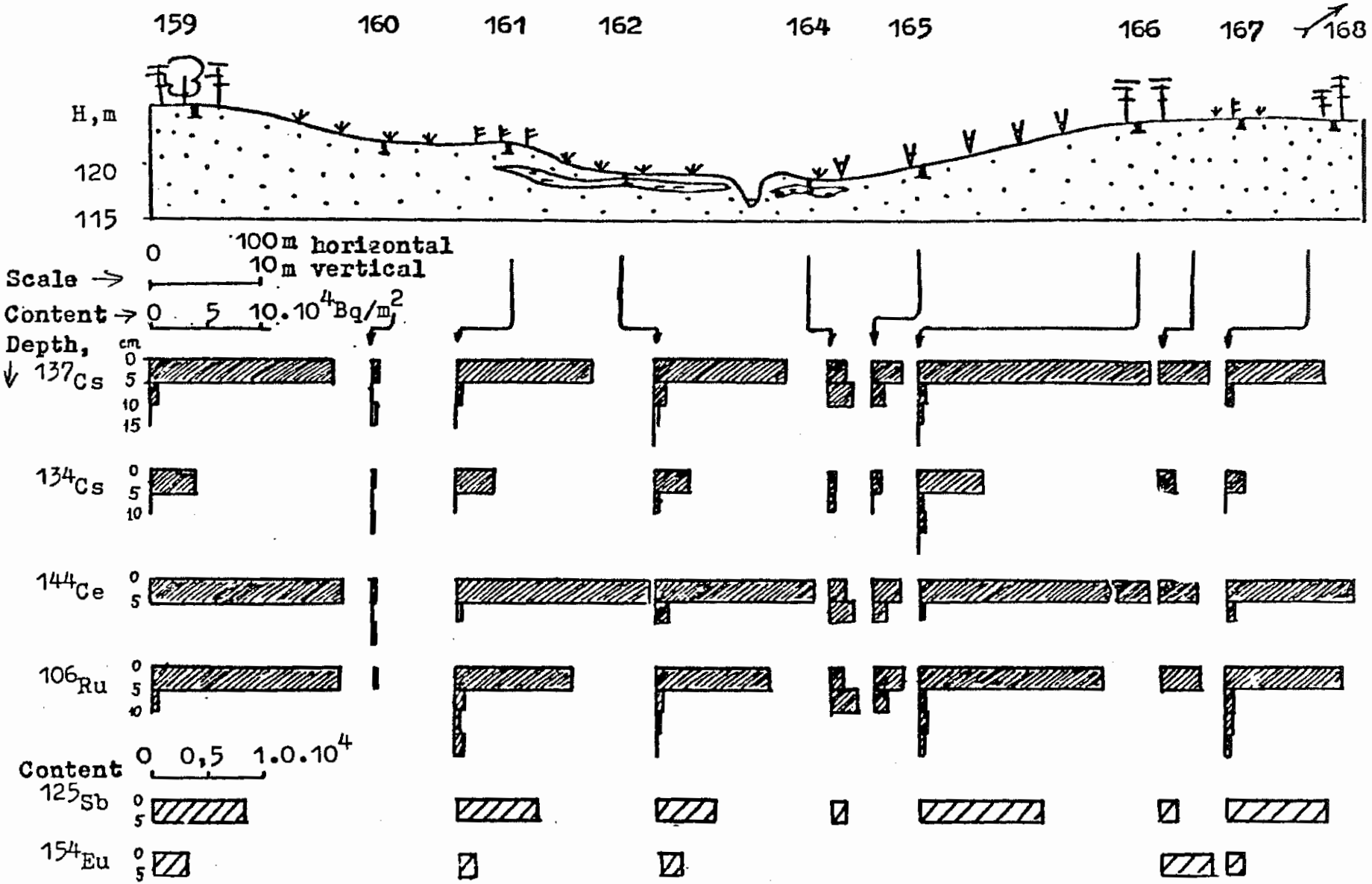


Fig.3 Distribution of gamma-emitters in the topsoil of connected landscapes in the middle reaches of the Khocheva river (polesye landscape, 30 km south of Chernobyl), 1.08.1988. (Light shading means that the element was found in one out of three samples).

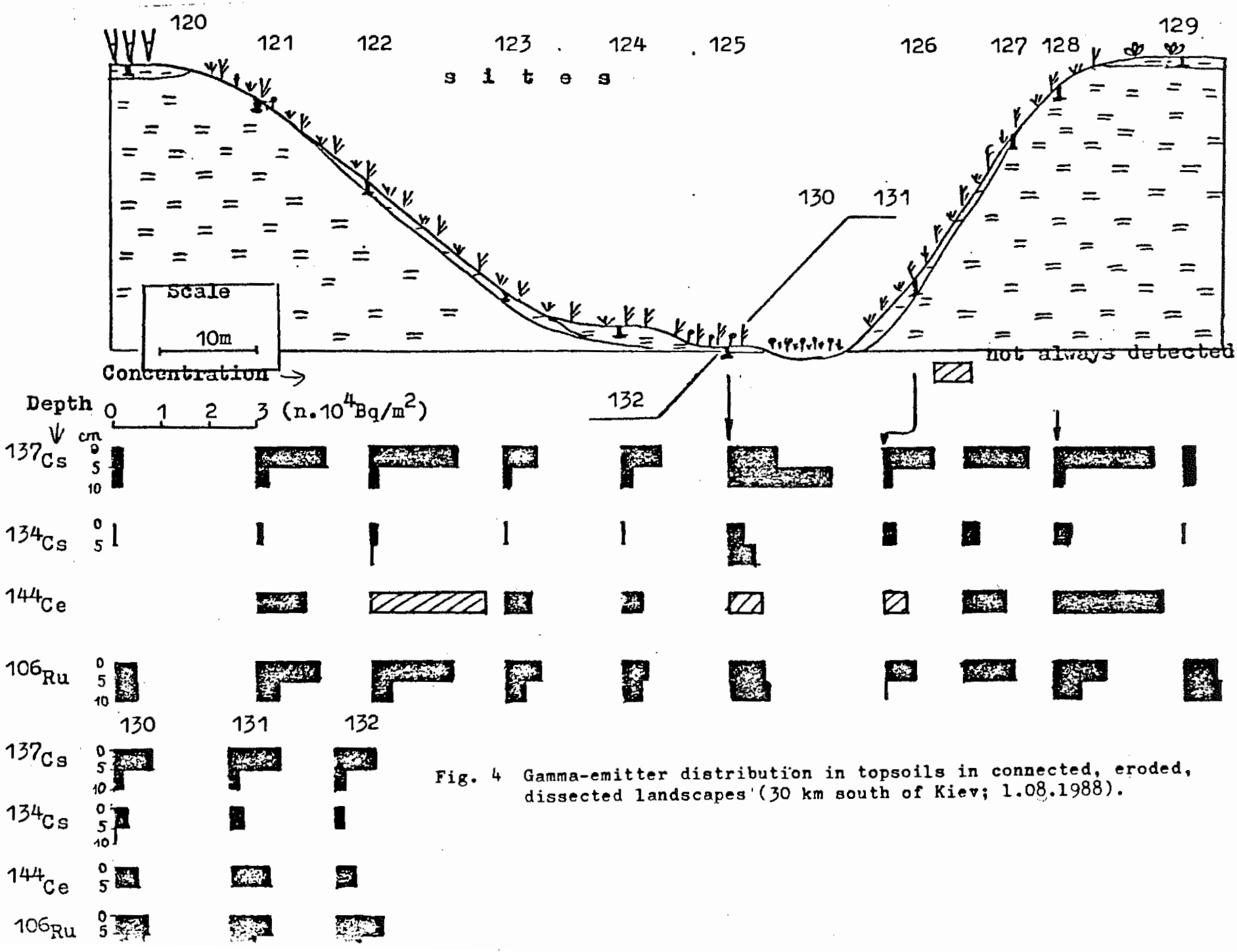


Fig. 4 Gamma-emitter distribution in topsoils in connected, eroded, dissected landscapes (30 km south of Kiev; 1.08.1988).

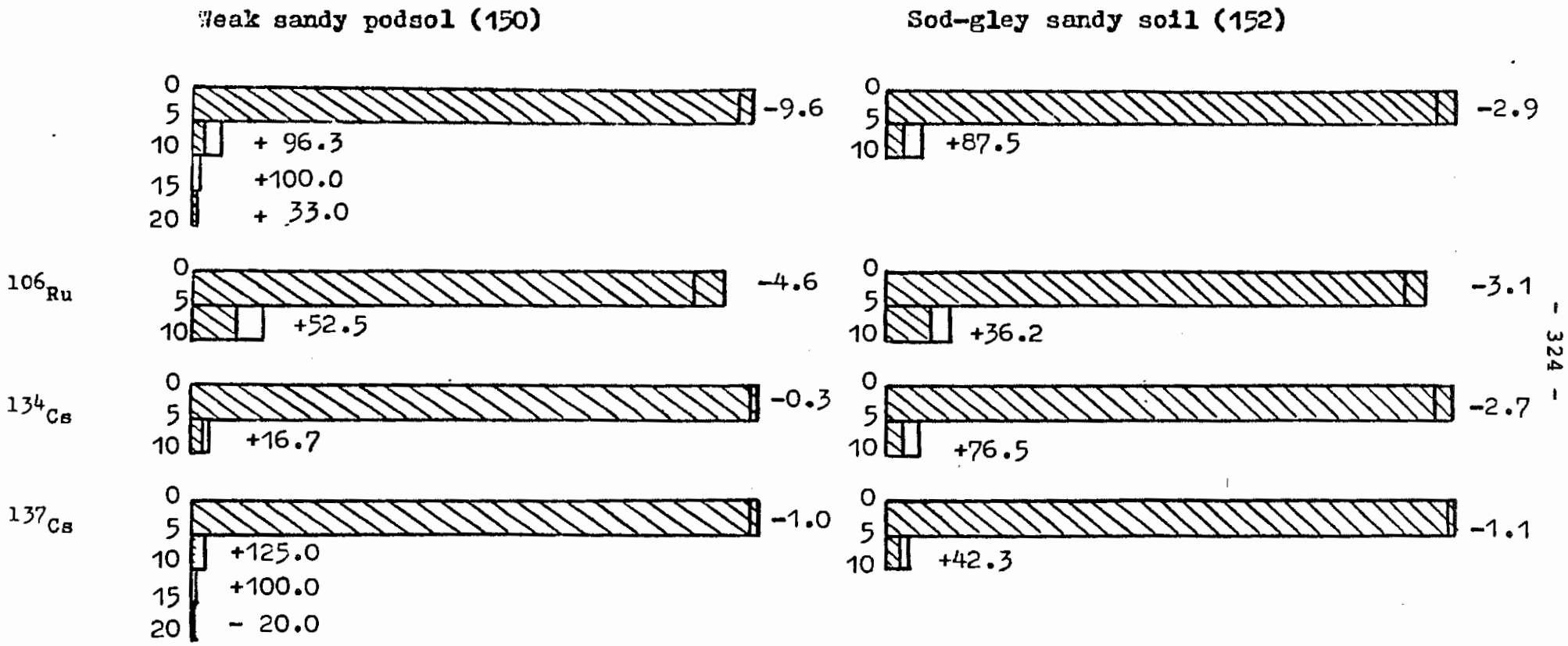


Fig. 5 Prognosis based on 1988 data (assuming no radionuclide migration) and actual result of repeat sampling in 1989 (unshaded); expressed as a percentage of radioisotope concentration in separate soil horizons (where 100% is the general concentration of elements in a section to the corresponding depth).

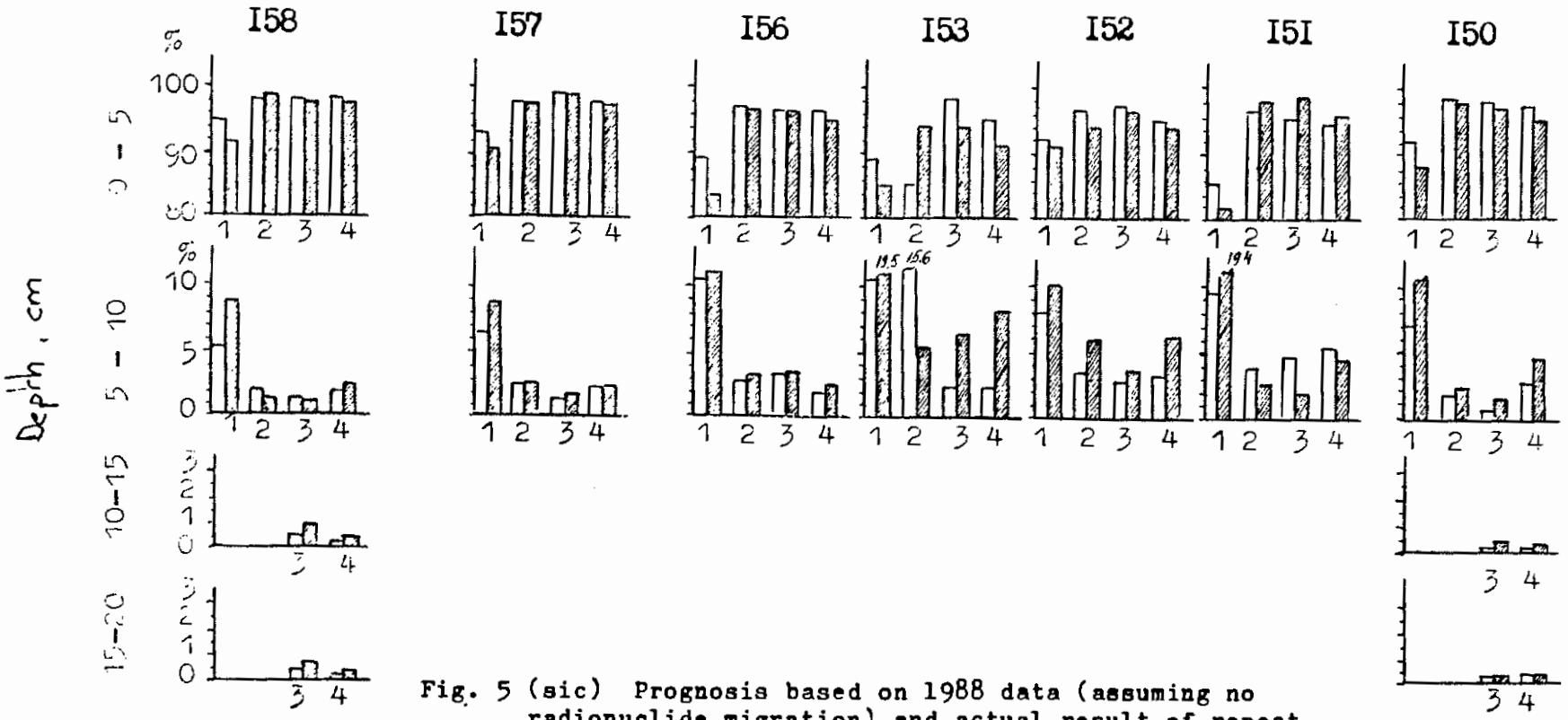


Fig. 5 (sic) Prognosis based on 1988 data (assuming no radionuclide migration) and actual result of repeat sampling in 1989 (shaded); expressed as a percentage of radioisotope concentration in separate soil horizons (where 100% is the general concentration of elements in a section down to the corresponding depth).

1 - ^{106}Ru; 2 - ^{134}Cs; 3 - ^{137}Cs; 4 - total gamma activity.

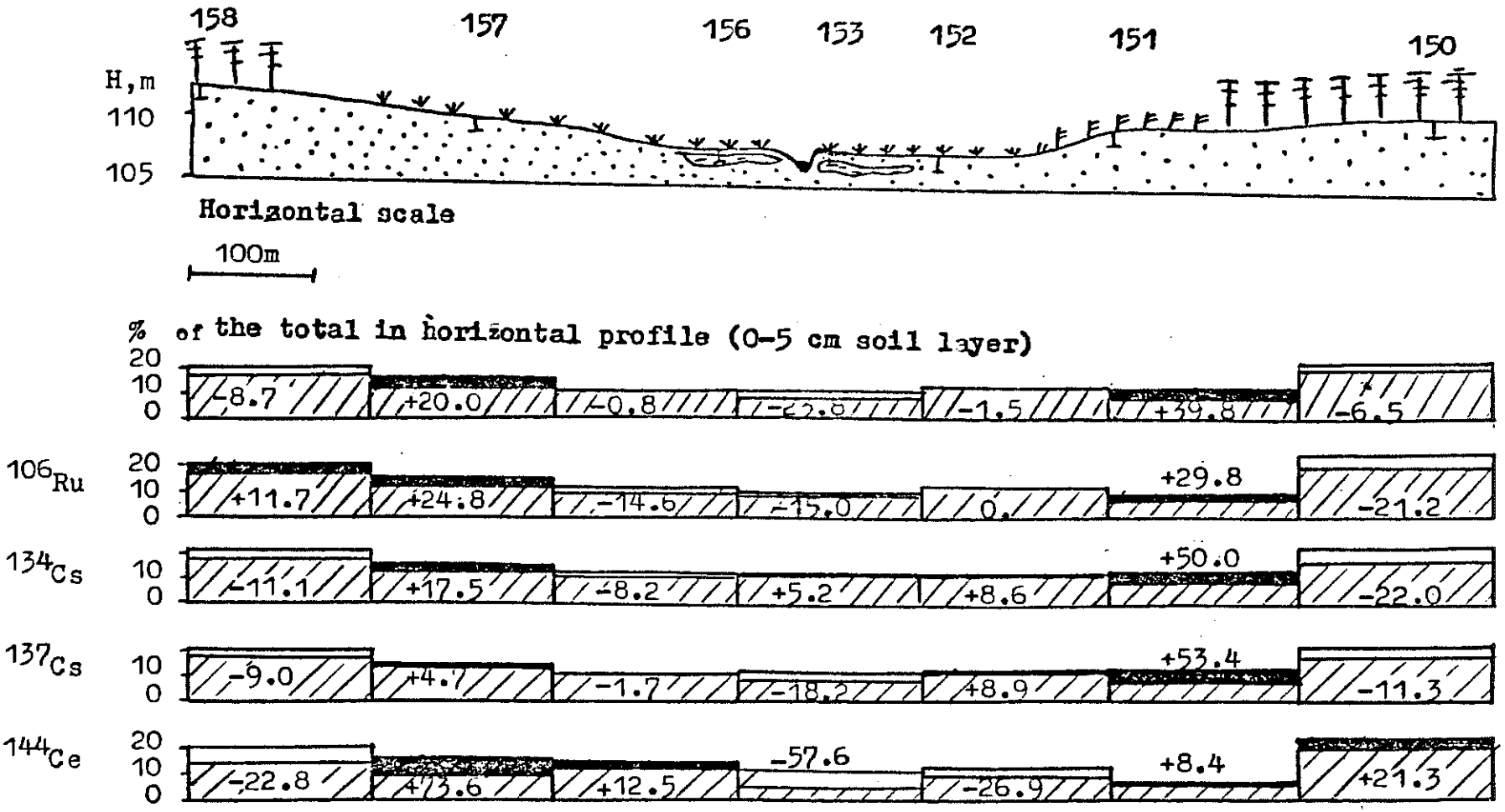


Fig. 6 Prognosis based on 1988 data (assuming no radionuclide migration) and actual result of repeat sampling in 1989, expressed as a percentage of radioisotope concentration in the 0-5 cm topsoil layer in connected landscapes in the lower reaches of the Khocheva river on 1.08.1989 (where 100% is the general concentration of elements in the catena soils; figures show the deviation (in %) from the calculated data).

Radionuclides in the Soil and Vegetation in the 30 km Accident Zone Around the Chernobyl NPP

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ABSTRACT

Under the programme of the USSR Academy of Sciences to combat the consequences of the Chernobyl reactor accident, radioecological research was carried out in interconnected landscape components affected by runoff at various distances from the damaged unit inside the 30-km accident zone.

The results showed that the radionuclides studied could be arranged in the order of ^{137}Cs > ^{134}Cs > ^{90}Sr in terms of decreasing content in the soil and vegetation, with a higher contamination density occurring in the immediate vicinity of the nuclear power plant. No geochemical trend was observed in the migration processes in interconnected landscape components affected by runoff. Displacement of the maximum content of ^{90}Sr and $^{134,137}Cs$ to a depth of 10-15 cm was observed only in the most dynamic geochemically linked components (valleys and floodplains). In forest and open areas radionuclides were concentrated in the forest litter and the upper 0-5 cm soil layer. Of all the wild plant species studied, mosses had the greatest concentration capacity; their ^{90}Sr and ^{137}Cs content and concentration coefficients were 5-10 times higher than those for the overground mass of members of herbaceous plant communities.

In the immediate vicinity of the damaged reactor ^{90}Sr is found in the soil in the firmly fixed state, but a substantial proportion of it converts into ion-exchangeable forms with increasing distance from the reactor. Work carried out by the Experimental Scientific Research Station along the radiation trail of the 1957 accident also showed that 72-95% of this radionuclide is in the ion-exchangeable form. The period of semi-elimination of ^{90}Sr from the 0-5 cm layer of contaminated soil is 9 years. Cs radioisotopes in the soil are mainly in the form of firmly fixed compounds.

Under the programme of the USSR Academy of Sciences to combat the consequences of the Chernobyl reactor accident, radioecological research was carried out in interconnected landscape components affected by runoff at various distances from the damaged unit inside the 30-km accident zone. The first area was 6 km south-east of the NPP and covered the top, central section and foot of the slope leading to the River Pripyat floodplain. The second area was 18 km south of the NPP and also covered the top, central section and foot of the slope forming the swampy bayou of the River Uzh.

On each of the main relief elements of the geochemical profiles mentioned we exposed soil cross-sections and from these took samples in layers of 0.5-1.0 cm down to a depth of 5-10 cm, recording the collection site and the genetic horizons. In the areas around the exposed soil cross-sections, we took samples of herbage, forest litter and other dead plant matter. The presence of ^{90}Sr was determined by means of the daughter radionuclide ^{90}Y, which was measured using a low-background UMF-1500 unit with an SBT-16 end-window counter with a margin of error of up to 15%. The content of the radionuclides $^{134,137}Cs$ was determined on an AM-A-02FI multi-channel analyser with a DGDK-50-B semiconductor detector. The areas of the photopeaks were calculated using equipment with a measurement error of no more than 3%.

The results obtained show that the radionuclides studied could be arranged in the order $^{137}Cs > ^{134}Cs > ^{90}Sr$ in terms of decreasing content in the soil and vegetation, with a higher contamination density occurring in the area in the immediate vicinity of the destroyed NPP unit. Thus, the level of radionuclides in the upper 0-2 cm soil layer amounted to hundreds of kBq/m^2 (for ^{90}Sr and ^{134}Cs) and thousands of kBq/m^2 (for ^{137}Cs) (Fig. 1). In the areas 18 km from the NPP the amount of radionuclides was about one order of magnitude lower. The maximum concentrations of radionuclides were confined to the upper 0-2 cm soil layer in open areas and to the layer of forest litter in forest stands. Of all the types of wild plants studied, mosses were found to have the greatest capacity for accumulation, the concentration of nuclides therein being 5-10 times higher than that in other types of plant. Spatial differentiation in radionuclide content in the various relief elements was not yet evident because of the short space of time which had elapsed since the accident.

In the period after the accident the involvement of radionuclides in the biogeochemical migration cycles depended on their capacity to form compounds with varying degrees of mobility, as well as on their general content and distribution in the soil. The physico-chemical forms of ^{90}Sr and ^{137}Cs in slightly humic sandy soils at various distances from the damaged reactor were studied in this connection.

The Table shows that ^{90}Sr is found primarily in the fixed state (up to 74%) near the damaged unit (3 km), while no more than 1% is in the water-soluble compound form, and the exchangeable and acid-soluble forms account for 17.7% and 7.7% respectively.

Table 1

Relative distribution of the physico-chemical forms of ^{90}Sr and ^{137}Cs with varying degrees of mobility in soils (%)

(Data for 1987)

Distance from NPP (km)	Physico-chemical forms of radionuclides in soils			
	Water-soluble	Exchangeable	Acid-soluble	Fixed
		^{90}Sr		
3	0.6±0.3	17.7±2.9	7.7±2.1	74.1±1.7
18	1.8±0.4	48.4±1.2	11.7±1.0	38.2±2.6
		^{137}Cs		
3	3.1±0.4	6.6±0.9	13.1±1.6	77.2±1.7
18	1.5±1.2	8.3±3.0	8.5±1.7	81.8±2.6

The proportion of water-soluble and exchangeable radiostrontium rises with increasing distance from the accident site, whereas that of fixed radiostrontium falls to 38.2%. This has been confirmed by the research work carried out in the zone of the Kyshtym accident in the Urals in 1957, which showed that 72-95% of this nuclide occurs in ion-exchangeable form and that the period of semi-elimination from the contaminated soil is approximately 9 years[1].

The Table shows that ^{137}Cs is present in the soil primarily in the form of firmly fixed compounds, irrespective of the places where the soil samples were taken.

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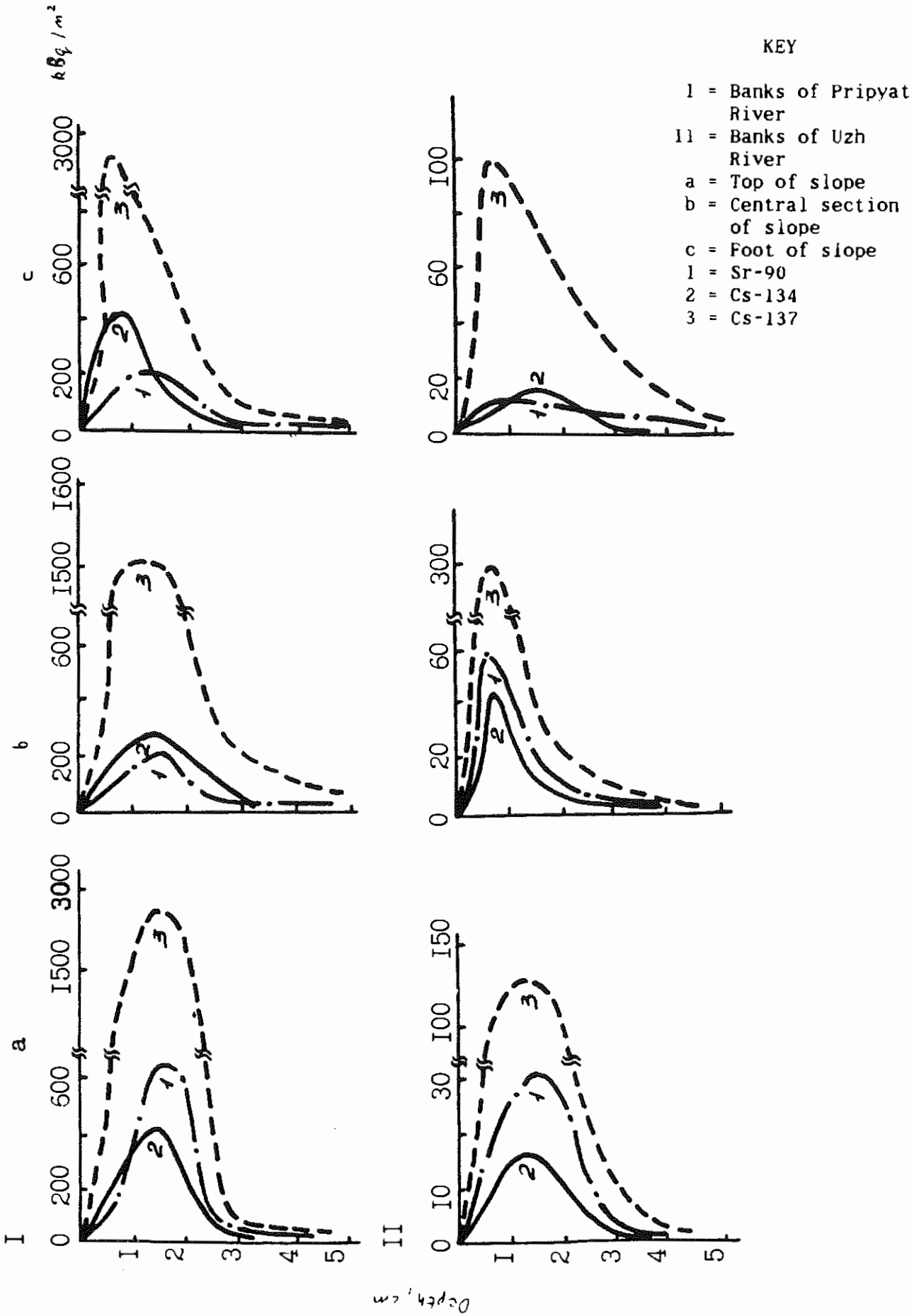
Fig.1: RADIONUCLIDE CONTAMINATION LEVEL IN THE SOIL AND VEGETATION OF THE ACCIDENT ZONE AROUND THE CHERNOBYL NPP



Content of Cs-134, Cs-137 and Sr-90 in Birch Crowns in Woods Within 30 km of the Chernobyl Power Plant During the Initial Years Following the Accident

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ABSTRACT

Between 1987 and 1989, as part of the programme of the Soviet Academy of Sciences, we studied the effect of the radioactive releases from the damaged Chernobyl reactor on Betula verrucosa Ehrh. stands located 1.5, 6 and 18 km from the power plant.

It was established that the content of $^{134,137}Cs$ and ^{90}Sr in crowns decreased significantly with increasing distance from the power plant, with major differences being observed even within each stand. Thus, the concentrations of $^{134,137}Cs$ in seeds produced in 1987 varied by a factor of 14 near the NPP, and by a factor of 30 at the most distant site. The concentration of total $^{134,137}Cs$ in catkins in 1988 was lower than in 1987 by a factor of 1.7, whereas in 1989 it was, on average, 17 times higher than in 1987. The concentration of ^{90}Sr in catkins rose over this three-year period. Evidently, the temporary decrease in the amount of radiocaesium in the catkins was caused in part by decay of ^{134}Cs atoms, while the increase in the radionuclide content in crowns in 1989 reflected greater ingress of radionuclides into the trees through the roots.

The concentration of $^{134,137}Cs$ in seeds in the birch stands studied was similar to that in the leaves, but lower by a factor of 2.2 than that in the fruit scales of the catkins. The concentration of ^{90}Sr in seeds was 2.1 times higher than in the fruit scales, while the radionuclide content in leaves was higher than – or hardly differed from – that in seeds. The concentration of $^{134,137}Cs$ in seeds, fruit scales and leaves was higher than the ^{90}Sr concentration.

The research also showed that the birch seeds produced in 1987 in the stand close to the NPP had better sowing qualities and greater resistance to gamma irradiation challenge than seeds from the two other stands.

A number of authors in the USSR and other countries have shown that in the event of airborne contamination of forest areas by radioactive substances as much as 40 - 90% of such substances are retained by the tree crowns.[1-4] The structural elements in the crown play varying roles in the radionuclide cycle in forest biogeocenoses. Thus, the skeletal parts of the crown - the trunk, branches - act as a store and as transport routes for the radionuclides. The parts of the crown which are shed each year - leaves, needles, reproductive organs - can be considered as donors of radionuclides to axillary organs and the soil but also as recipients of radionuclides which enter trees via the roots. Although of all the elements which are shed by the crown the leaves play the most important part in the radionuclide cycle, one should not ignore the contribution made by the reproductive organs which, in the case of some trees such as willow and birch, can be quite considerable in fruitful years. In addition, the uptake of radionuclides by fruit and seeds leads to a build-up in their internal irradiation. In this connection our study of the influence of high background radiation on the biological properties of birch seeds within 30 km of Chernobyl NPP includes investigations of the mechanisms involved in the uptake of $^{134,137}Cs$ and ^{90}Sr in the catkins and leaves of this species of tree. In the literature we found no references to the accumulation of the radionuclides in question by reproductive organs of birch.

The present report contains data on the accumulation of $^{134,137}Cs$ and ^{90}Sr in reproductive organs and leaves of the Betula verrucosa Ehrh. birch tree within a 30-km radius of the Chernobyl NPP in the first three years after the accident there.

The investigative method used was as follows. In three stands of mixed pine and birch at distances of 1.5, 6 and 18 km from the demolished NPP unit, work in the first year centred in each area on 15-20 heavily fruiting young birch trees; in August 1987, 1988 and 1989 catkins were collected, and leaves as well in the next two years.

The radionuclide content in catkins and leaves was determined separately for each tree and also for the structural elements of the catkins - seeds, seed wings and fruit scales. The concentration of ^{90}Sr was measured by its daughter ^{90}Y, using the low-background UMF-1500 apparatus with an
SBT-16 end-window counter. The concentration of radiocaesium was determined by means of an AM-A-02FI multichannel analyser with a DGDK-50-B semiconductor detector.

Because of the very poor fruiting of birch trees in 1988 and 1989 in stands further away from the NPP no count was made in those years of the concentration of radionuclides in reproductive organs and in leaves.

Fig. 1 shows that the concentration of radiocaesium in birch catkins produced in 1987 diminished with increasing distance of the stand from the power plant. Thus, the concentration of this radionuclide in seeds from the first stand 1.5 km from the NPP was 113 kBq/kg of dry material and from the second and third stands, at distances of 6 and 18 km from the plant, the concentrations were 65 and 25 kBq/kg of dry material respectively. Seeds and seed wings had practically the same concentration but this was 1.6 to 2.2 times greater than in fruit scales.

In the next two years there was an increase in the content of radiocaesium in both seeds and leaves, with the most noticeable increase - by a factor of 9 to 10 - in trees near the NPP. It should be noted that the concentration of $^{134,137}Cs$ in seeds and leaves was similar in 1988 and 1989.

Table

Amount of $^{134,137}Cs$ in seeds and leaves of Betula verrucosa growing at different distances from Chernobyl NPP during the initial years following the accident, measured in kBq/kg of dry material

Growth element	Year	Distance from NPP in km	
		1.5	6
Seeds	1988	420 ± 115.5	45 ± 18.5
	1989	4 187 ± 535.5	164 ± 36.6
Leaves	1988	407 ± 121.2	38 ± 12.0
	1989	3 628 ± 861.4	149 ± 26.3

Since there was not a sufficient volume of catkin samples to allow the radiochemical analysis of radiostrontium, material from five trees in each of the stands investigated was used. The amount of ^{90}Sr in structural elements of birch catkins and leaves decreased with increasing distance from the demolished unit (Fig. 2). Thus, in the first stand, the amount of radionuclides per kilogram of dry material in seeds produced in 1987 was 120 kBq and in the second and third stands it was respectively 25 and 0.75 kBq. In 1988 and 1989 the increase over the 1987 concentration of radiostrontium in structural elements of catkins and leaves was significantly lower than the increase referred to above in the concentration of radiocaesium. In the second stand this did not emerge so clearly. Within the catkins the concentration of ^{90}Sr was higher in the seeds than in the fruit scales. However, these differences were statistically significant only in seeds produced in 1989 from the first stand (t_{st} = 4.0 at $t_{0.05}$ = 2.3). It will also be seen from Fig. 2 that the concentration of this radionuclide in seeds and leaves is, as a rule, identical. An exception to this is found in specimens of such seeds and leaves gathered in 1989, in which the amount of ^{90}Sr is clearly higher in the leaves (t_{st} = 2.8 at $t_{0.05}$ = 2.3). From the above it is clear that in the three years after the Chernobyl accident there has been a build-up of an ever increasing amount of caesium and strontium radionuclides in the reproductive organs (catkins) and leaves of Betula verrucosa growing within a 30-km radius of the power plant. The uptake of radionuclides in tree crowns was more marked in 1989 in the stand 1.5 km away from the demolished unit. This was due to some extent to repeated

contamination of the trees by radioactive dust raised into the air as a result of decontamination operations in the territory adjoining the stand. However, since similar decontamination operations were also carried out in this zone in earlier years, the main reason for the phenomenon referred to is evidently the increased uptake of $^{134,137}Cs$ and ^{90}Sr by the trees through the soil in 1988 and 1989. This in turn led to an increase in the return of radionuclides into the soil through the shedding of leaves and, in the final analysis, to an extension of the scale of their incorporation in the cycle of matter in the biogeocenosis.

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Fig. 1. Content of $^{134,137}Cs$ in 1987 in crowns of Betula verrucosa growing within 30 km of Chernobyl NPP. 1 - seeds; 2 - seed wings; 3 - fruit scales.

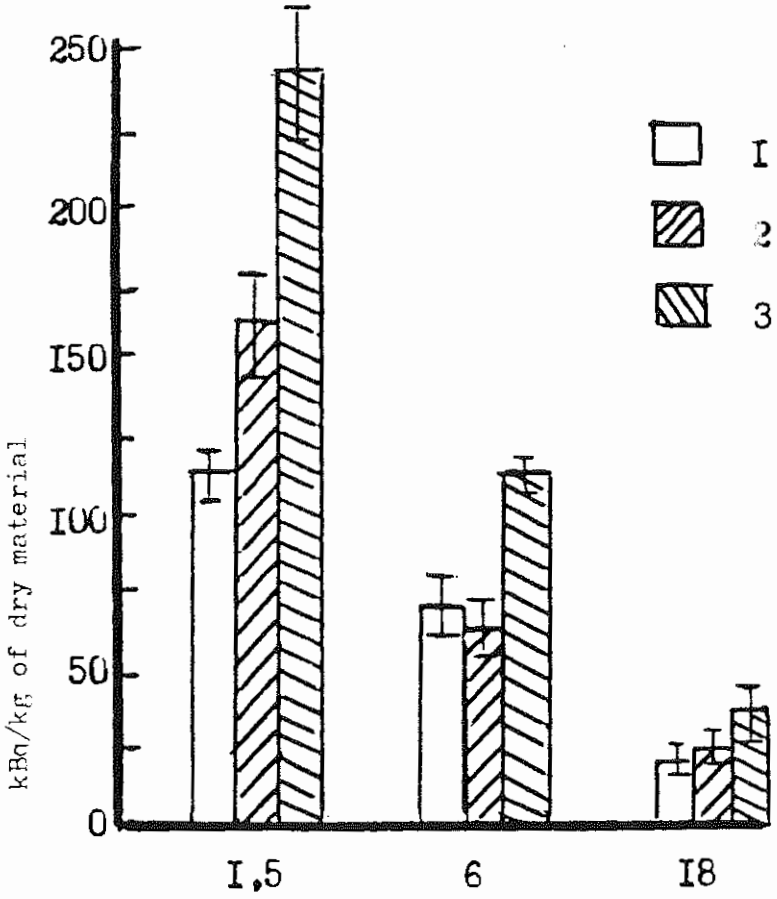
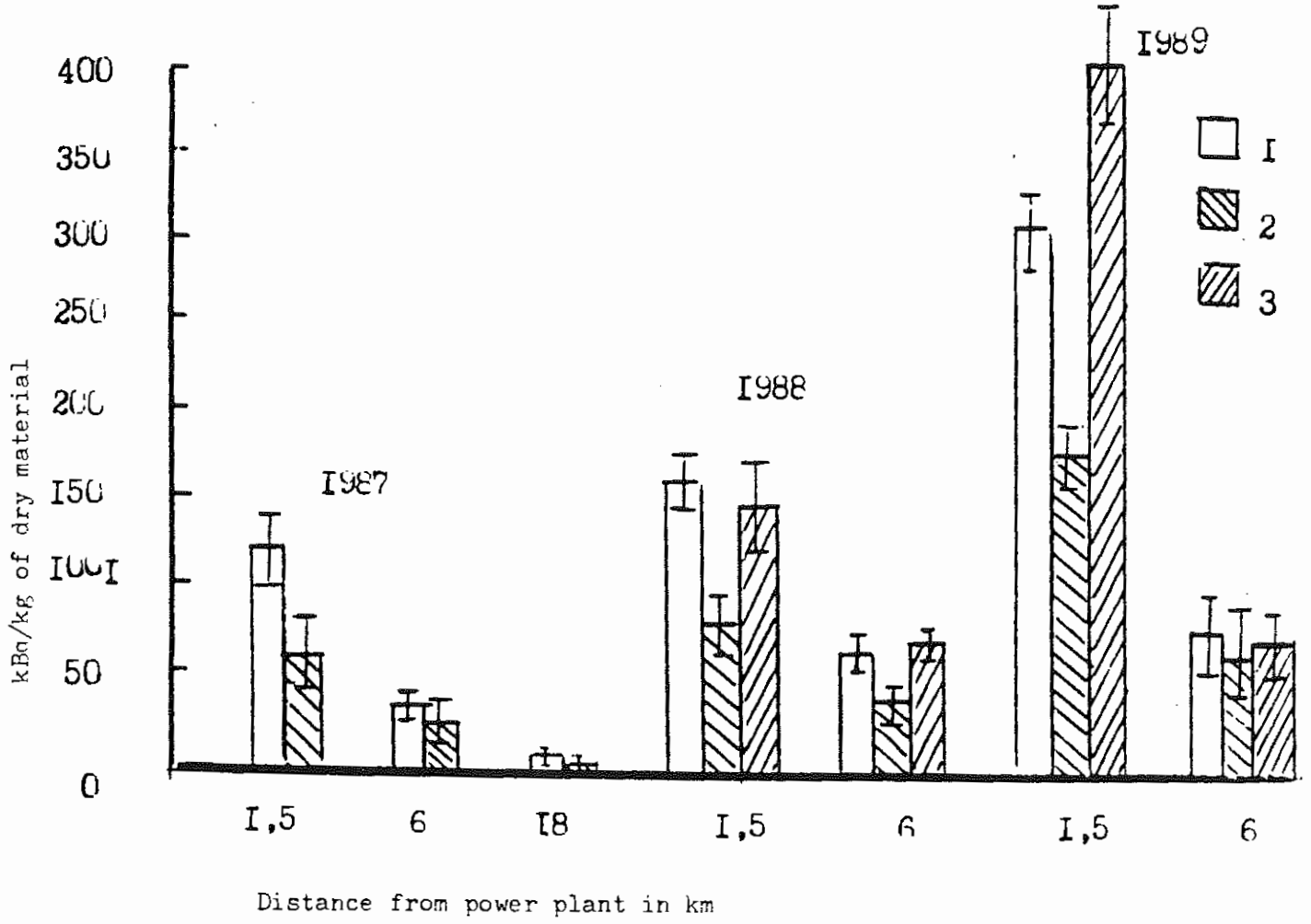


Fig. 2. Changes in the content of ^{90}Sr in crowns of Betula verrucosa, growing within 30 km of Chernobyl NPP, in the first three years following the accident. 1 - seeds; 2 - fruit scales; 3 - leaves.



Forms of Radionuclide Occurence in Soil

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ABSTRACT

This paper covers research into the forms of the ^{137}Cs and ^{144}Ce radionuclides having entered the soil as a result of the Chernobyl accident. The difference between the sorptive behaviour of these radionuclides and that of the same radionuclides introduced into the same soil in ionic form may be useful in drawing conclusions about the forms of radionuclides in the soil.

86 soil samples were taken at the following distances to the north of the reactor: 6, 80 and 150 km (Gomel region) and 260 km (Mogilev region) of Byelorussia.

In addition to sorption and desorption experiments using solutions of different composition, we also carried out experiments involving ultrafiltration of solutions (using membranes with a pore diameter of 0.5, 0.19, 0.1 and 0.06 microns) produced by desorption of Cs and Ce in real and model experiments.

From the overall results we conclude that Cs and Ce are present in soils of widely differing composition, partly in ionic forms and partly in the form of particles, the matrices of which are either uranium dioxide or graphite.

In order to solve the technical problems involved in overcoming the aftermath of the Chernobyl accident, it is essential to know in which form the radionuclides occur in the environment.

What we mainly need to know is whether a radionuclide occurs in the form of particles or in dissolved, ionic form. After the Chernobyl accident two conflicting views were advanced. The first was that all the radionuclides in the environment were in the form of particles, whose matrices were either uranium dioxide or graphite. The second view was that all the radionuclides were present in dissolved, ionic form. It should be pointed out that identifying the form of a radionuclide in the environment is a formidable task. Although there is a substantial body of literature on the state of radionuclides in solutions and in soils, little is known about the forms of occurrence of the radionuclides deposited on the soil as a result of the Chernobyl accident. The point is that the overwhelming majority of investigations were conducted with model systems even if real soils and real river and sea water were used.[1-3] The main characteristic of such models is that dissolved, ionic forms of radionuclides are introduced into the soil or water, after which the researchers examine what form a radionuclide will assume in a solution of given composition, how it is distributed between water and soil, etc. In our case the original form of the radionuclide is not clear, being determined by the circumstances of the accident. However, it is precisely this form of the radionuclide that has to be determined.

Changing conditions in the reactor, changing meteorological conditions and uninterrupted ejections over a ten-day period may have led to the formation of very different forms of radionuclides on the territory of Byelorussia.[4,5]

In this study conclusions on the form of the radionuclides were based primarily on data on the sorptive and desorptive behaviour of the radionuclides Cs-137 and Ce-144 deposited in soils as a result of the accident, and the behaviour of these radionuclides when introduced into the same soils in ionic form. The difference in the sorptive behaviour of radionuclides in real and model systems was the basis for conclusions on the forms of radionuclides in the soil. In addition we used ultrafiltration with the aid of nucleopore filters (pores 1 to 0.05 μm in diameter) and the ion exchange method.

The soil samples used were collected in October 1986 along the "northern track" at a distance of 6 km from the reactor (at Masany), and at a distance of 40 and 150 km in the Khoyniki and Narovlya rayons (districts) in Gomel oblast (region), and at a distance of 260 km in Mogilev oblast (Chudyany).

The standard soil core was cut into two layers. Since over 95% of the radioactivity was concentrated in the top layer of soil, this layer was used for the real experiments and the bottom part for the model experiments. For the model experiments the soil samples were immersed in an aqueous solution (pH 4) containing the radionuclides Cs-137 and Ce-144 in ionic form. The mixture was stirred for 7 hours and left to stand overnight. After this the solution was filtered off through a paper filter (pore diameter 2.5 μm) and the soil was dried in the air. In these conditions the sorption rate was 97-99% for Cs-137 and 80-95% for Ce-144.

For desorption, the following solutions were used: H_2O; 0.1 mole HCl; 0.1 mole HCl + 0.1 mole KCl + 0.1 mole $FeCl_3$; and 0.1 mole NaOH + 0.1 mole $Na_4P_2O_7$. The 0.1 mole N_aOH + 0.1 mole $Na_4P_2O_7$ solution is used to extract humic acids from the soil[6], the data on desorption of radionuclides by this solution possibly giving some indication of the role of humic acids in binding radionuclides in the soil.

The desorptive solution was poured onto real and model soil samples (weighed amounts) and stirred for 7 hours. The solution was separated from the soil through a "blue band" paper filter. The results shown in the tables and figures are average values from 5 or 6 experiments.

Table 1

Extraction of Cs-137 from soil

Soil	Amount extracted, % Real experiments				Amount extracted, % Model experiments		
	H_2O	0.1M HCl	0.1M HCl, KCl, $FeCl_3$	0.1M NaOH, $Na_4P_2O_7$	H_2O	0.1M HCl	0.1M HCl, KCl $FeCl_3$
Sandy	3±1	3±1	38±8	12±2	1.6±0.3	1.3±0.4	19±1
Grey meadow	5±2	6±2	28±14	30±9	1.4±0.2	0.9±0.2	22±4
Loamy	8±2	6±1	24±15	6±1	1.0±0.1	1.2±0.8	10±2
Soddy-podzolic	2±1	3±1	19±4	30±12	0.3±0.1	2.2±0.6	22±2
Peaty	4±1	7±3	7±5	–	1.4±0.8	–	28±5

As the results show (Table 1), water and 0.1 mol/litre HCl solutions extract only a small fraction of Cs-137 and the quantity does not vary much with soil type.

The ultrafiltration data reproduced in Figures 1 and 2 show that in the real 0.1 mole HCl and H_2O solutions, Cs-137 partly occurs in particle form (50%). But this also applies to the model solutions. What is the nature of these particles?

The data reproduced in Table 2 show that Cs-137 is well sorbed by all soils examined from the 0.1 mole HCl solution and even from the HCl + KCl + $FeCl_3$ solution.

Table 2

Sorption of Cs-137 by soils

Soil	0.1M HCl			0.1M HCl+0.1M KCl+0.1M $FeCl_3$		
	Soil amount (g)	Sorbed quantity		Soil amount (g)	Sorbed quantity	
		Absolute	%		Absolute	%
Loamy	11	517	97	7	57	55
	27	440	98	14	13	52
	46	300	95	23	12	50
Sandy	1	132	34	15	6	32
	12	170	88	17	5	31
	26	204	93	19	5	32
Soddy-podzolic	1	168	43	4	10	18
	9	91	80	12	7	30
	20	81	88	21	26	74
Peaty-swampy	2	990	84	-	-	-
	13	760	91	-	-	-
	30	250	95	-	-	-

This may serve as an indication that the Cs-137 particles in the solution are particles of soil with adsorbed ions of caesium, i.e. Cs-137 occurs in the solution in the form of pseudocolloids. The fact that a solution containing the ions H^+, K^+ and Fe^{3+} increases caesium desorption from the soils evidently indicates that these ions displace caesium. The conclusion to be drawn from the data as a whole is that Cs-137 is mainly found in the soils in ionic form.

The data in Table 1 show that the greater the quantity of humic acids the more Cs-137 is extracted by the 0.1 mole NaOH + 0.1 mole $Na_4P_2O_7$ solution. At the same time, the data in Figure 4 show that the presence of dissolved forms of humates in the solution has no significant effect on the sorption of Cs-137 by sandy soil.

This suggests that Cs-137 interacts weakly with humates. It is not impossible that the sorptive properties of humic substances in the soil differ from their properties when in pure form.

The data presented in Figure 3 show that independently of the distance from the reactor, Cs-137 occurs in the soil mainly in ionic forms, i.e. no significant change in the forms of occurrence of Cs-137 is observed as a function of distance.

Similar investigations were conducted with Ce-144. The data reproduced in Table 3 show that there is a substantial difference between the sorptive behaviour of ionic forms of Ce-144 in the model experiments and that of Ce-144 entering the soil as a result of the accident. While in the model experiments 80% of the ionic forms of Ce-144 are extracted from the soils (irrespective of type), in the real experiments the quantity extracted was between 15% and 30%.

Table 3

Extraction of Ce-144 from soils (%)

	Real experiments				Model experiments		
0.1M Soil KCl,	H_2O	0.1M HCl	0.1M HCl, KCl, $FeCl_3$	0.1M NaOH $Na_4P_2O_7$	H_2O	0.1M HCl	HCl, $FeCl_3$
Sandy	1±1	1±1	49±7	23±7	1±1	14±1	82±2
Grey meadow	2±1	4±1	38±6	67±12	10±1	30±4	86±2
Loamy	2±1	1±1	13±1	-	10±1	34±8	79±4
Soddy-podzolic	3±1	2±1	28±12	75±10	1±1	27±3	81±3

The ultrafiltration data (Figures 5 and 6) show how the forms of occurrence of cerium differ in the real and model solutions. In the real solutions of 0.1M HCl (Figure 5) approximately 70% of the Ce-144 occurs in the form of particles, while in the model solutions the figure is approximately 40%. What is the nature of these particles? In such low concentrations (10^{-10} to 10^{-12} mol/litre) cerium cannot itself form particles of colloidal dimensions. In the 0.1M HCl solution, prepared with twice-distilled water, at all concentrations cerium occurs only in ionic form and only as Ce^{3+} cations.[7] In this case the cerium is evidently not in the form of particles whose matrix is uranium dioxide. In point of fact, the ultrafiltration experiments with solutions containing the ions H^+, K^+ and Fe^{3+} (curves 2,4) show that - as in the model experiments - cerium occurs only in ionic form, i.e. in these solutions Ce is not sorbed by soil particles nor do pseudocolloids of cerium form in the solution as a result.

The data in Table 4 show that there are differences in the ratio of Ce-144 to Cs-137 in the soil (prior to contact with solutions) and in solution.

Table 4

Ratio of Ce-144 to Cs-137 in soil and in a 0.1M HCl + 0.1 M KCl + 0.1M $FeCl_3$ solution

Distance from reactor in km	Ratio of Ce-144 to Cs-137	
	Soil	Solution
6	2.0 ±0.2	11.6±2.7
30	0.4 ±0.2	2.4±0.4
40	0.3 ±0.1	4.0±0.4
70	0.6 ±0.3	1.5±0.4
260	0.01±0.01	0.2±0.1

This suggests that the process is one of desorption or leaching, and not the dissolving of particles. Moreover, these processes differ for caesium and cerium.

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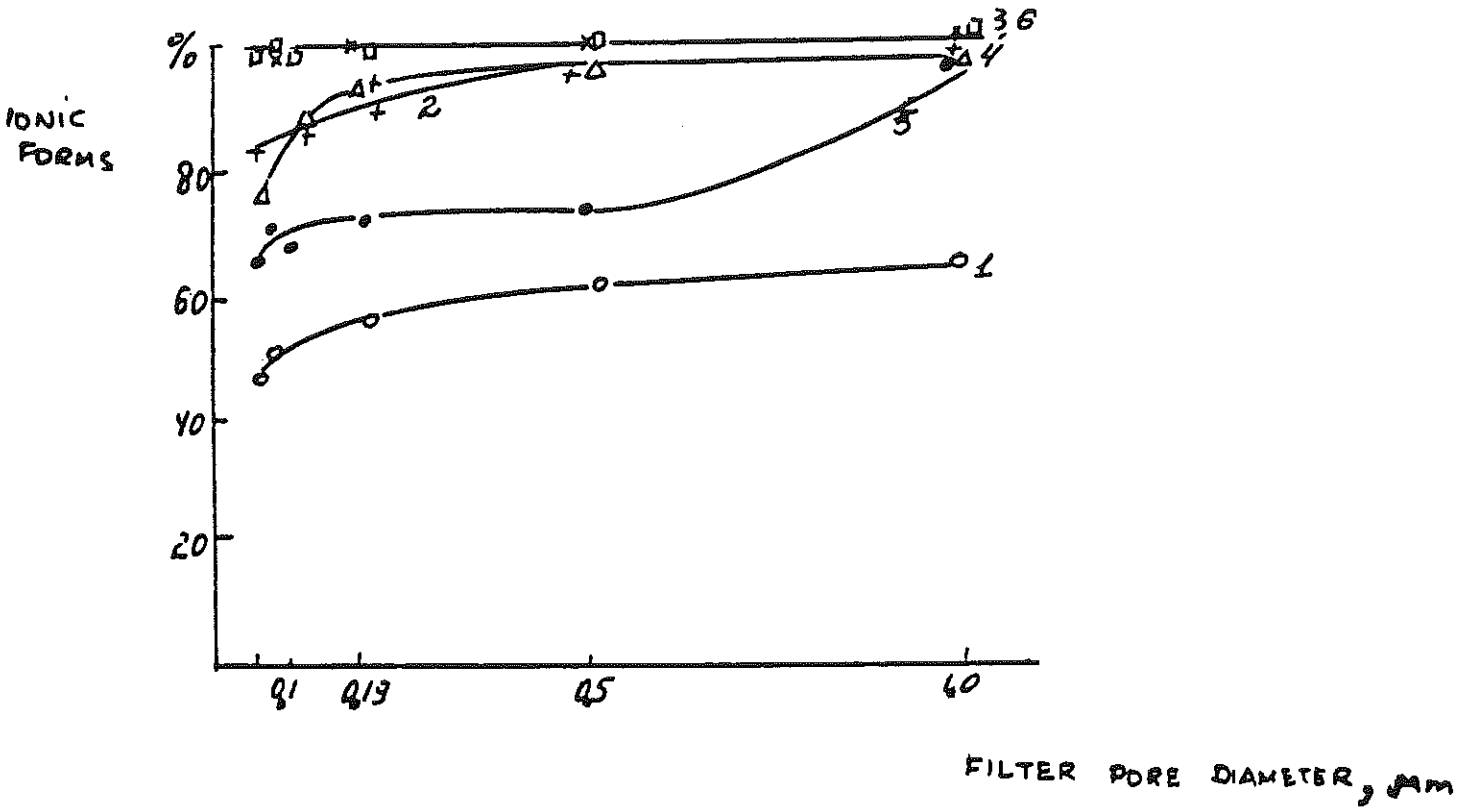

Figure 1: Cs-137 ultrafiltration. Sandy soil.

Real solutions:
1) 0.1 mole HCl
2) H_2O
3) 0.1 mole each of: HCl, KCl, $FeCl_3$

Model solutions:
4) 0.1 mole HCl
5) H_2O
6) 0.1 mole each of: HCl, KCl, $FeCl_3$

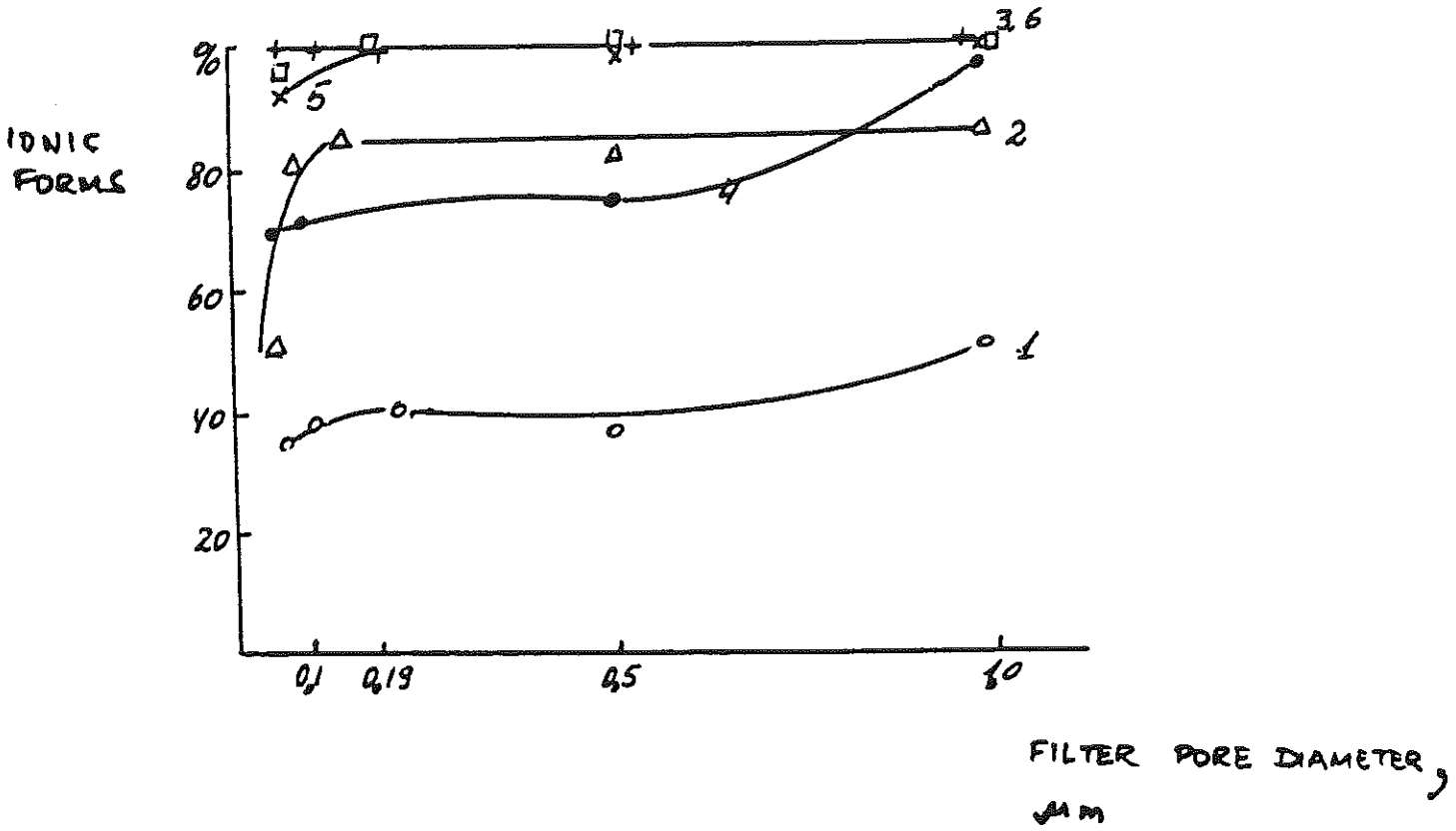

Figure 2: Cs-137 ultrafiltration. Soddy-podzolic soil.

Real solutions:
1) 0.1 mole HCl
2) H_2O
3) 0.1 mole each of: HCl, KCl, $FeCl_3$

Model solutions:
4) H_2O
5) 0.1 mole HCl
6) 0.1 mole each of: HCl, KCl, $FeCl_3$

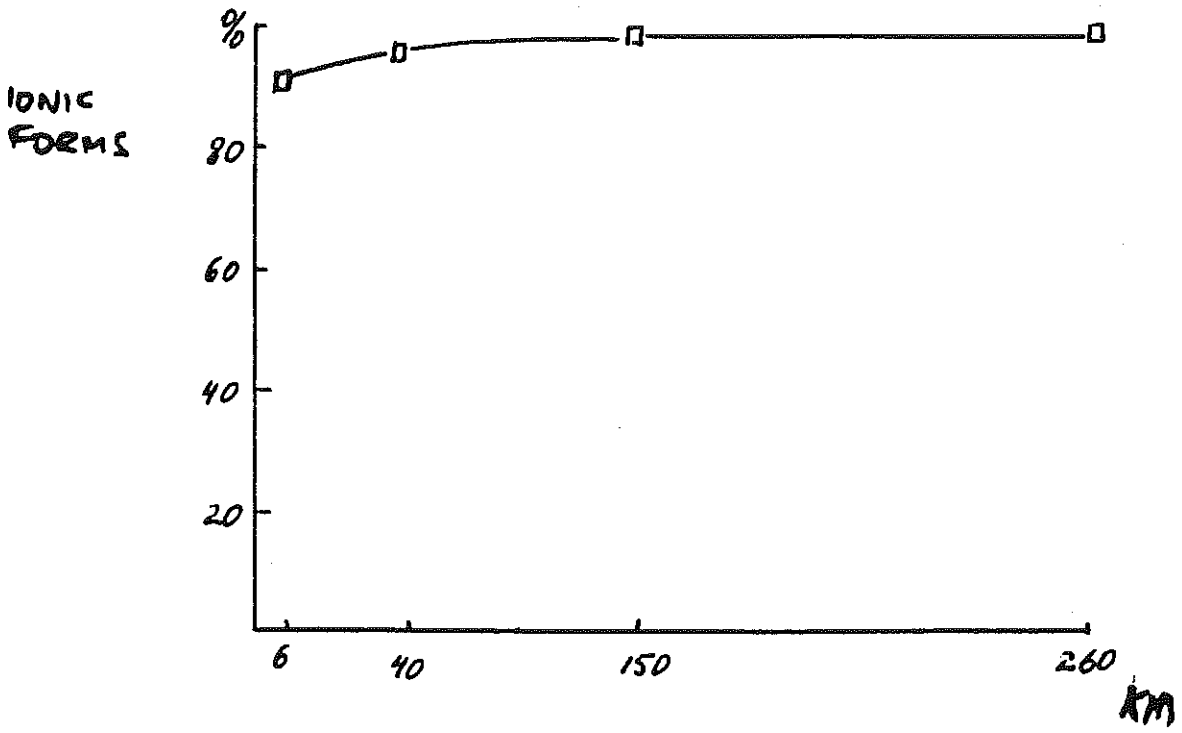

Figure 3: Content of ionic forms of Cs-137 in a 0.1 mole HCl + 0.1 mole KCl + 0.1 mole $FeCl_3$ solution as a function of distance from reactor.

Filter pore diameter: 0.1 micron

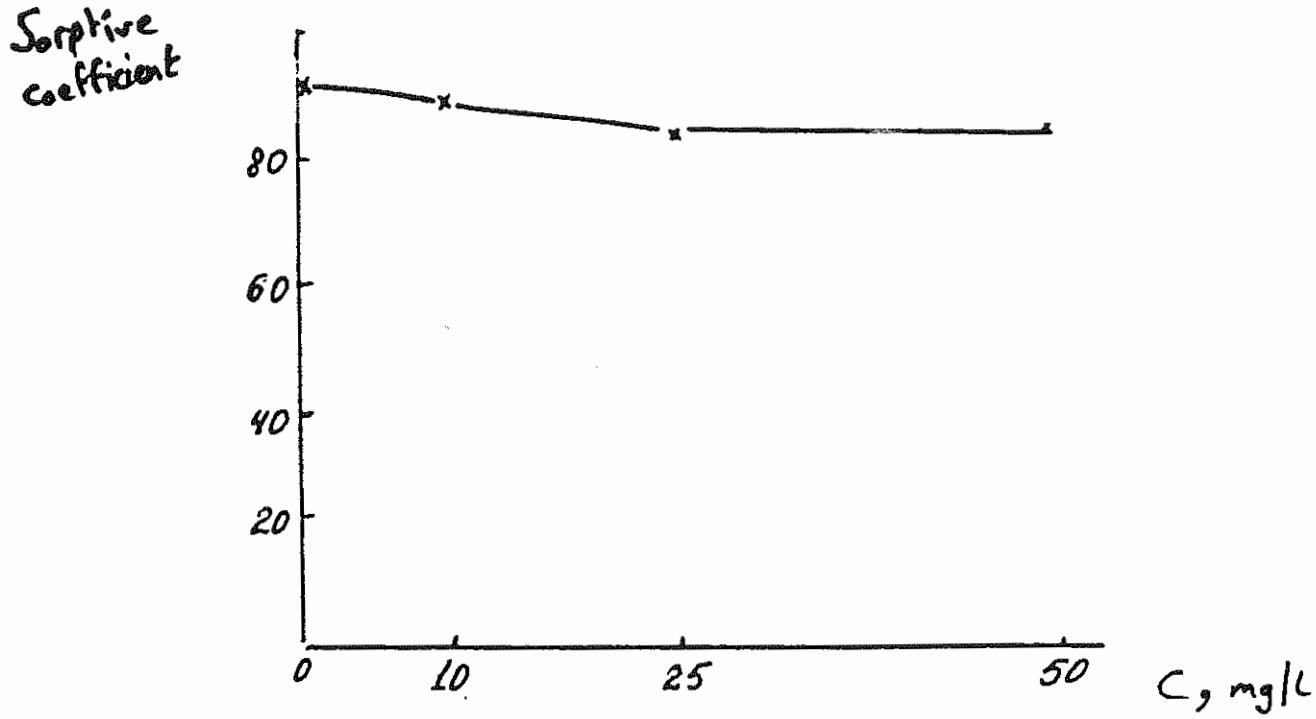

Figure 4: Sorption of Cs-137 by sandy soil as a function of the concentration of humic acids in a solution of pH = 8.1.

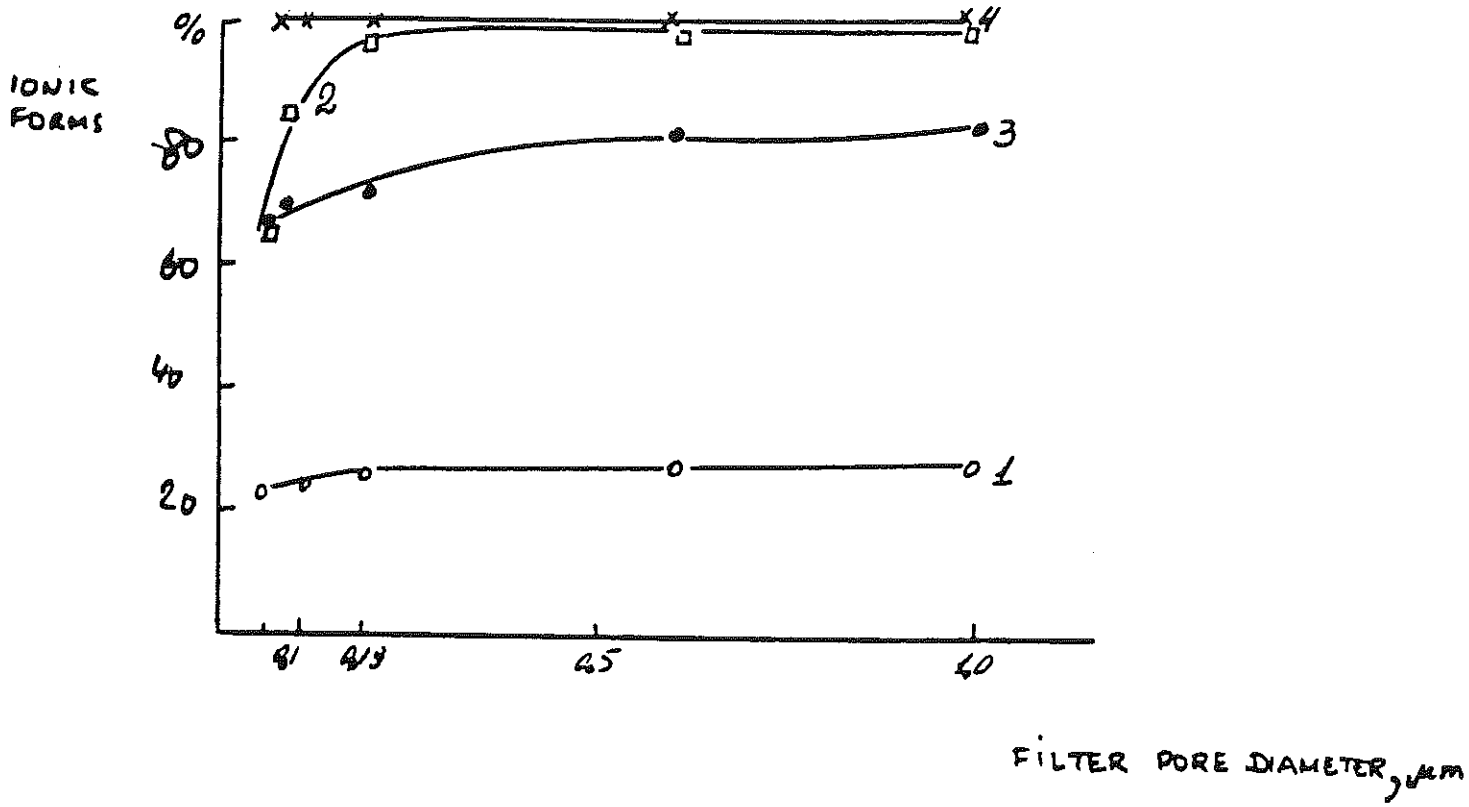

Figure 5: Ce-144 ultrafiltration. Soddy-podzolic soil.

Real solutions: 1) 0.1 mole HCl
2) 0.1 mole each of: HCl, KCl, $FeCl_3$

Model solutions: 3) 0.1 mole HCl
4) 0.1 mole each of: HCl, KCl, $FeCl_3$

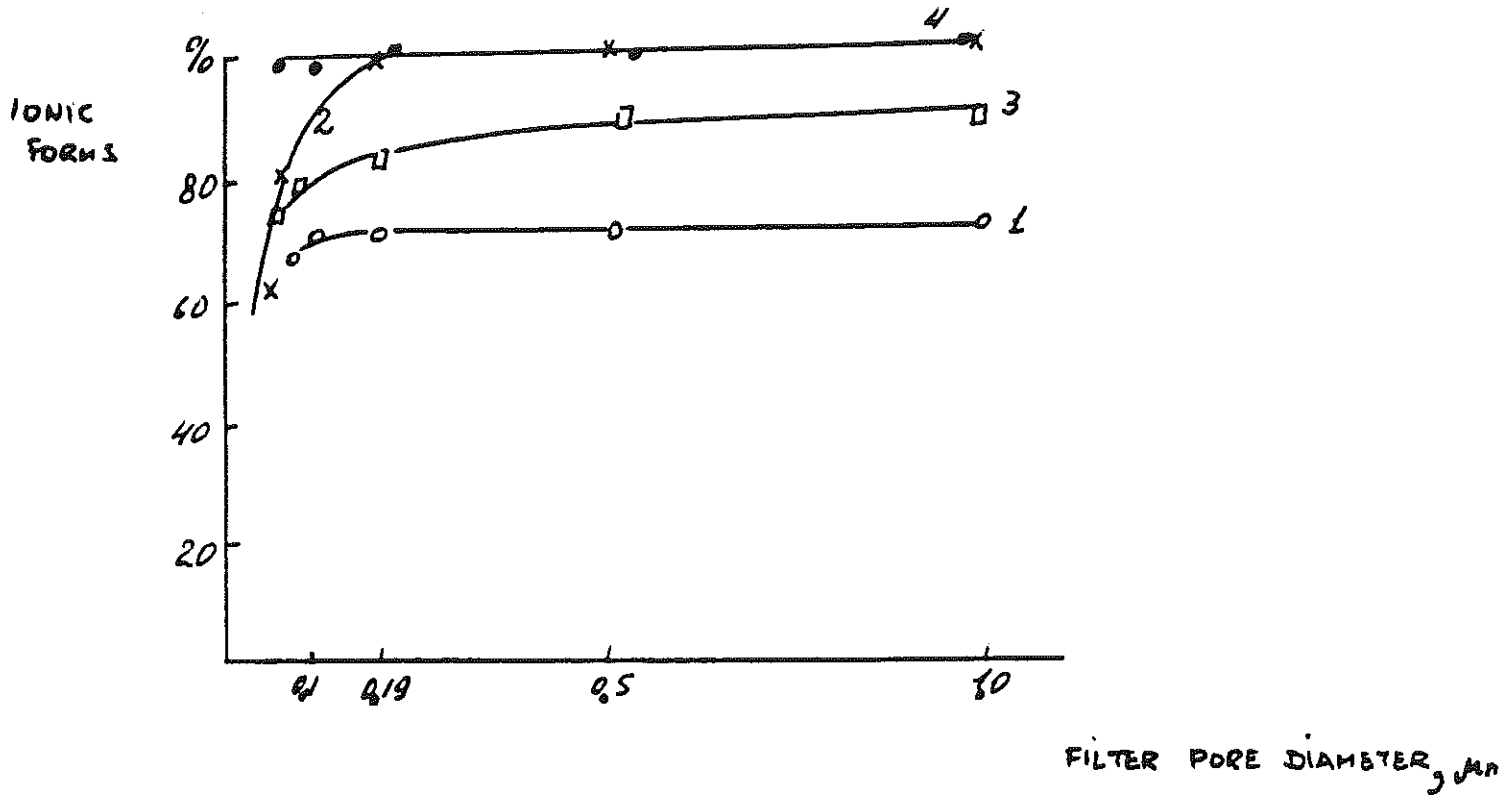

Figure 6: Ce-144 ultrafiltration. Sandy soil.

Real solutions: 1) 0.1 mole HCl
2) 0.1 mole each of: HCl, KCl, $FeCl_3$

Model solutions: 3) 0.1 mole HCl
4) 0.1 mole each of: HCl, KCl, $FeCl_3$

Migration Peculiarities of Long-Lived Radionuclides in Soils in Areas Contaminated by the Chernobyl NPP Accident

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ABSTRACT

We studied migration of long-lived radionuclides of caesium-134/137, cerium-144 and ruthenium-106 in soil samples from regions contaminated by the Chernobyl accident. Samples of three types of contamination - fuel (A), quasi-fuel (B) and aerosol (C) - were taken at calibration sites in August 1988 down to a depth of 5 cm using conventional core samplers. The radioactivity of the samples was measured at intervals of 0.5 cm, as was the activity of three mechanical fractions of the samples: 1-2 (>2) mm; 0.25-1.0 mm; <0.25 mm.

The equipment used to measure the radioactivity included the AFOPA LP-4900B multichannel programming analyser (Finland) and the DGDK-100V-3 semiconductor (germanium) diffusion drift detector (USSR), the results being processed by the ASPRO automatic programming facility (Vernadsky Institute, USSR).

Regardless of the type of soil and landscape most of the caesium-134/137, cerium-144 and ruthenium-106 is concentrated in the upper 0-2 (to 5) cm in the form of strongly bonded compounds. Migration of caesium and ruthenium radionuclides down to a depth of 3.5-4.5 cm was observed in humus-deficient, soddy-podzolic sandy soils (found in watersheds) and in soils of a hydromorphic nature.

The maximum concentrations of caesium, cerium and ruthenium radionuclides (30-50 % of total activity) were found in the 1-2 (>2) mm fractions (grass, roots and tillering nodes) in the 0-1 (to 2) cm layer, which reflects the increasing role of biogenic migration processes in mass transfer of radionuclides despite their apparent lack of mobility in biocenoses. The soil's dust and clay fraction (<0.25 mm) helps to fix the radionuclides in the crystal structures of soil minerals and organic matter. The ratio of caesium-134/caesium-137 in soils exhibiting the quasi-fuel type of contamination varies from 0.17-0.55 in the 1-2 mm and <0.25 mm sample fractions, evidently reflecting the various radionuclide occurrence forms and topographies in the fallout matrix, isotope separation included.

Introduction

One of the most serious consequences of the accident at the Chernobyl NPP was the greatest radioactive contamination of the biosphere, including the soil cover, that has ever taken place in one go.

The need for in-situ studies, spread over many years, of the vast area contaminated made it necessary to set up special calibration sites - areas typical both as regards the various types of contamination and the physico-geographic areas affected.

The radiochemical studies of radionuclides carried out at these sites form one of the main bases for radioecological monitoring of the contaminated areas of the USSR.

This report covers specific studies into the dynamics and forms of occurrence of long-lived "Chernobyl" radionuclides in samples of soil collected in August 1988.

Methods

Collection of samples *

The soil samples were collected from virgin unused land chosen on the basis of landscape and geochemical features and the various types of radioactive contamination discovered, and systematic observations were carried out from 1986 onwards. At the calibration sites we selected a 5 x 7 m area with an incline of no more than 10°, without any hollows, with vegetation no higher than 5-10 cm and located no more than 150 m away from the nearest forest.

The basic types of contamination specified in 1986 by B. F. Myasoyedov, I. A. Lebedev et al. (Vernadsky Institute) were classed according to the differing radionuclide composition of the fallout discharged from the damaged unit over the protracted period and the changes occurring during this time:

- Type A: ratios of all the radionuclides close to those calculated for fuel with a burn-up intensity of 10.3 GWd/t;
- Type B: disruption of the radionuclide fuel ratios, with an increase in the relative amounts of plutonium, ruthenium, barium and caesium as compared to type A;
- Type C: samples containing a large amount of volatile components from the fuel due to graphite burn-up: up to 14 times more plutonium, up to 30 times more barium, up to 80 times more ruthenium and up to 900 times more caesium than in type A;
- Type D: even greater anomalies in radionuclide composition, with up to 200 times more plutonium and caesium than the calculated levels, but with a low ruthenium content.

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* O.V. Rumyantsev and E. M. Korobova were responsible for the collection and description of the samples.

Type A occurred at the following calibration sites:

No 5, situated south of the settlement of Zalesye and bounded by the Chernobyl-Kiev road and the River Uzh (30-km zone), where the soil is of the floodplain, soddy, sandy type on layered sandy alluvium;

No 7, situated west of the Cherevach-Zeleny Mys road between the settlements of Opachichi and Kupovatoye, where the soil is of the soddy-podzolic sandy type on alluvial sandy deposits.

Type B: this type of contamination was found at the following sites:

R-Sh-9, in the Khocheva River valley near the settlement of Stepanovka, on tussocky herbaceous meadowland, directly to the south of the town of Pripyat on the 30-km zone boundary, where the soil is of the soddy-gleyey, sandy type on alluvial sandy deposits.

R-Sh-8, at the mouth of the Khocheva River directly south of the town of Pripyat on the 30-km zone boundary, in an area of oak and pine forest with birch, where the soil is of a medium podzolic sandy type on alluvial loam.

Type C: this type was found in soils of a soddy-meadowy sandy-loam type on alluvial loam collected near the settlement of Malye Kleshchi 100 km south-west of the town of Pripyat.

The soil samples were collected using standard core samplers (15 cm in diameter, 5 cm long), then the total surface gamma activity and that of the individual radionuclides were measured (Table 1), after which the samples were measured layer by layer (in steps of 0.5-1 cm), as was the activity of the following three mechanical fractions in each sample: 1-2 (> 2) mm; 0.25-1.0 mm and < 0.25 mm.

The radioactivity of all the samples was measured on integrated gamma spectrometric equipment, including the AFOPA LP-4900B multi-channel programming analyser (Finland) and the DGDK-100V-3 semi-conductor (germanium) diffusion drift detector (USSR), and the data processed by the ASPRO automatic programming facility (Vernadsky Institute).

Model experiments to study the form of occurrence of the individual long-lived radionuclides/gamma-emitters were carried out on the above-mentioned samples of soil. We looked at desorption of the radionuclides in static conditions by means of consecutive elutriation using the following solutions: distilled water, sodium acetate (pH 7) and hydrochloric acid 1M. The radionuclide concentrations in the eluate and samples were determined via gamma spectrometry.

Discussion of results

The studies of the distribution peculiarities of the long-lived radionuclides still responsible for the gamma-radioactive contamination in the soils up to this day, show that - regardless of the form of contamination, the type of soil and landscape - they are mainly concentrated in the top 0-2 (to 5) cm (Table 2).

RELATIVE GAMMA-ACTIVITY AT LONG-LIVED RADIONUCLIDES ON THE SURFACE AT THE CALIBRATION SITES ON 15.08.88 - TOTAL ACTIVITY TAKEN AS 1*

Contamination Types / Site	Soil type	RELATIVE ACTIVITY OF RADIONUCLIDES			
		Ru-106	Ce-144	Cs-137	Cs-134
A					
A Site No 5	Floodplain soddy sandy	0.16	0.27	0.45	0.10
Site No 7	Soddy-podzolic sandy	0.22	0.30	0.36	0.10
B					
R-Sh-9 Stepanovka	Soddy-gleyey sandy	0.33	0.23	0.34	0.10
R-Sh-8 Khocheva	Medium podzolic sandy	0.17	0.34	0.38	0.10
C Moly Kleshchi	Soddy-meadony sandy-loam	0.02	0.00	0.76	0.29

* Gamma-activity of samples measured by AFORA-ASPRO facility

Table 2

Comparative content of long-lived radionuclides in the 0-1 cm layer as against content in the sample as a whole (0-5 cm)

Collection site, contamination type	Radionuclide content, as a % of total in 0-5 cm layer			
	cerium-144	caesium-134	caesium-137	ruthenium-106
Site No 7, A	67	62	59	65
Site No 5, A	98	98	97	97
R-Sh-9, B	76	63	69	42
R-Sh-8, B	68	81	76	79
M. Kieshchi, C	-	69	60	-

By the end of 1988 the radionuclides deposited on the surface of the soil had entered (in varying degrees according to their form of uptake, physico-chemical form in the fallout and their particular characteristics) the geochemical migration cycle, as demonstrated by the data given in Table 3.

DISTRIBUTION OF LONG-LIVED RADIONUCLIDES BY DEPTH
(Total activity of sample taken as 1)

Contamination Type / Site	Depth cm	RELATIVE ACTIVITY OF RADIONUCLIDES AT TIME SAMPLE COLLECTED			
		Ce-144	Cs-134	Cs-137	Ru-106
A	0-1	0.98	0.97	0.96	0.98
Site No 5	1-1.5	0.02	0.02	0.03	0.01
	1.5-2.0		0.001	0.008	0.01
	2-3				
	3-4				
	4-5				
Site No 7	0-1	0.61	0.62	0.63	0.64
	1-5	0.39	0.38	0.47	0.26
B	0-1	0.76	0.66	0.62	0.42
R-Sh-8	1-1.5	0.10	0.12	0.15	0.20
Stepanovka	1.5-2.0	0.16	0.21	0.22	0.38
	2-3				
	3-4	0.003		0.001	
	4-5				
R-Sh-9	1-0	0.68	0.81	0.75	0.20
Khocheva	1-1.5	0.32	0.19	0.22	0.80
	1.5-2.0		0.002	0.03	0.01
	2-3-4-5				
C	0-1		0.68	0.62	
Malye Kleshchi	1-2		0.24	0.22	
	2-3		0.03	0.12	
	3-4		0.02	0.01	
	4-5		0.02	0.02	

No particular differences were noted in the distribution of the long-lived radionuclides in the 2 cm layer in the soils with different types of contamination, and no major change in the nuclide composition of the fallout had been noted by the end of 1988.

Furthermore, we fully established that there was a tendency for the general patterns of radionuclide migration in soils to be followed, regardless of uptake source, i.e. accumulation in local geochemical barriers, concentration in the sod, litter and humic horizon, migration to 3-5 cm in sandy soils (Site 7) and of a hydromorphic nature during periodic flooding (M. Kleshchi).

Determination of the concentrations of radionuclides in the mechanical fractions showed that a substantial amount was concentrated in the roots, in the earth around the roots and in the litter: up to 70-80% of the total activity in the 0-1 cm layer in the case of type A contamination, up to 40-50% in the case of type B and ~ 30% in the case of type C. This testifies to the increasing role of uptake of radionuclides into plants via the roots, and to their increasing mobility, i.e. to biogenic migration in the mass transfer of radionuclides despite their apparent poor mobility in biogeocenoses.

The dust and clay fractions of the soil (< 0.25 mm) help to fix an average of up to ~ 20% of the radionuclides in the crystalline structures of the soil minerals and organic component. It was fully established that as the portion of this fraction increases, the maximum concentration of radionuclides shifts upwards (Table 3, Fig. 1).

It is interesting to note that the caesium-134/caesium-137 ratio in soils with the quasi-fuel type of contamination varies from 0.23 to 0.55, apparently reflecting the various radionuclide occurrence forms and topographies in the fallout matrix, including isotope separation.

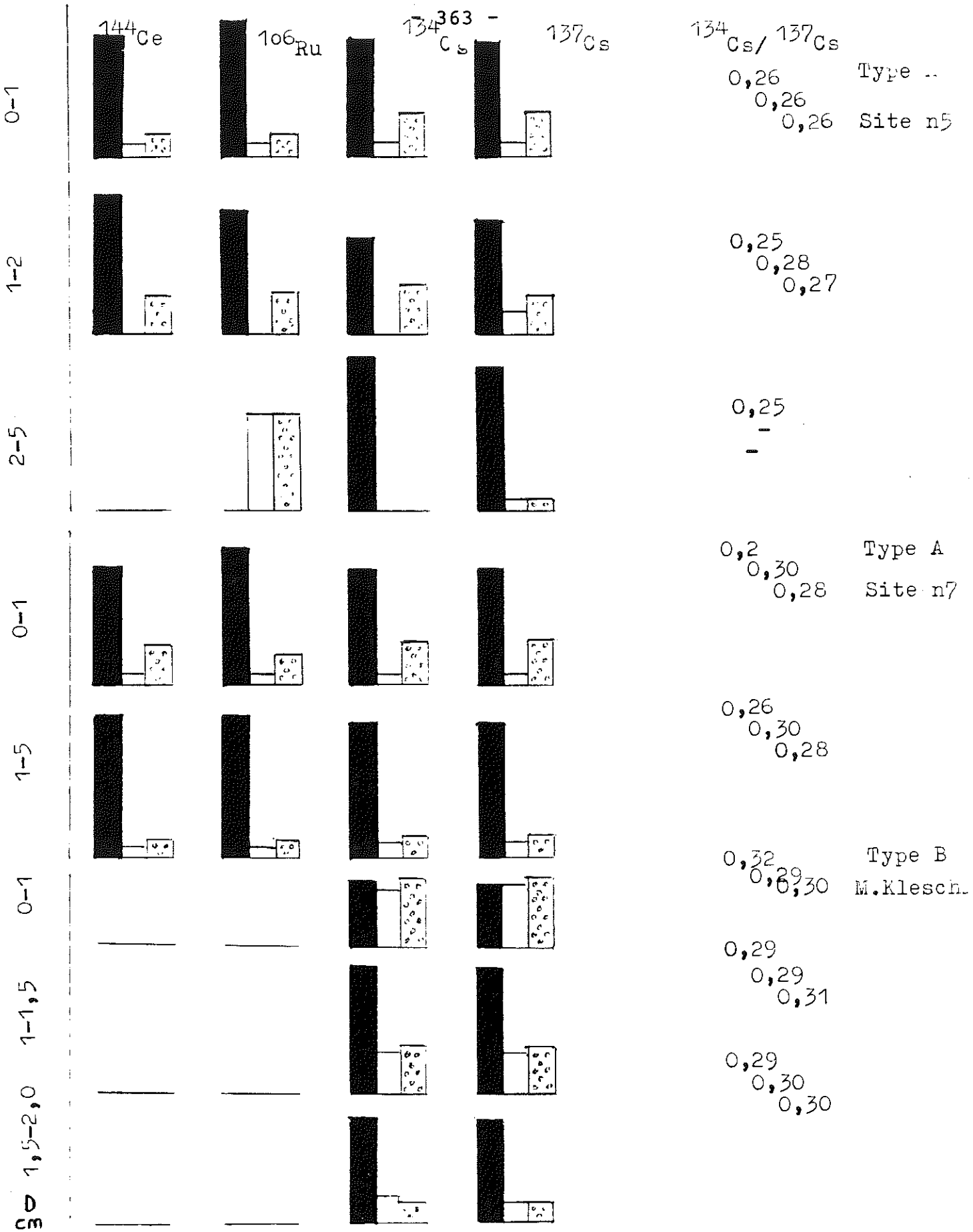

Fig.1 VERTICAL DISTRIBUTION OF LONG-LIVED RADIONUCLIDES IN THE FRACTIONS BY DEPTH (CONCENTRATION IN THE LAYER TAKEN AS 1)

MM: >1.0; 0.25÷1.0; <0.25

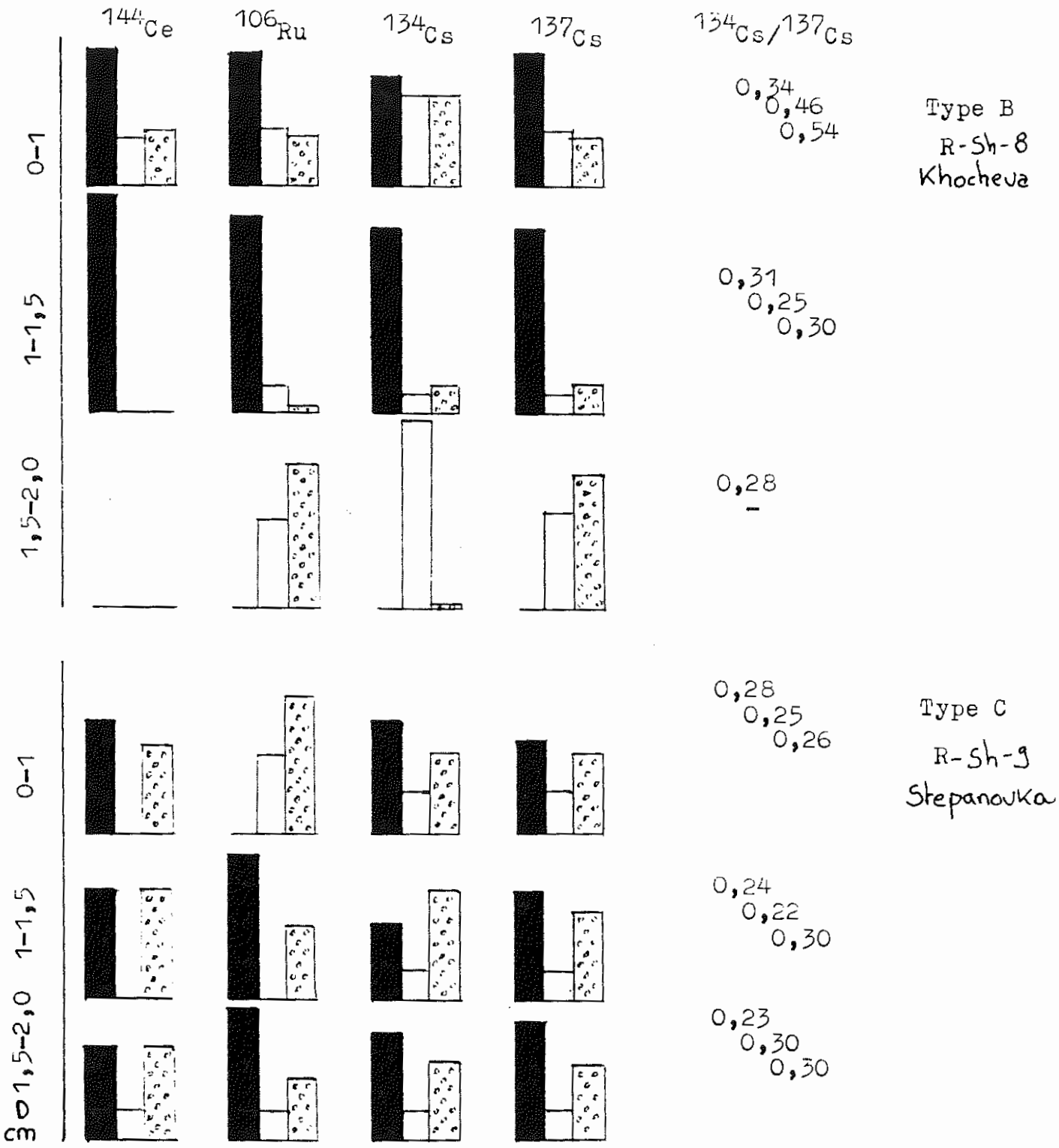
144Ce
106Ru
134Cs
137Cs
134Cs/137Cs
0-1
1-1,5
1,5-2,0
0,34
0,46
0,54
Type B
R-Sh-8
Khocheva
0,31
0,25
0,30
0,28
–
0-1
1-1,5
1,5-2,0
0,28
0,25
0,26
Type C
R-Sh-9
Stepanovka
0,24
0,22
0,30
0,23
0,30
0,30

Fig.1 (cont.)

The very weak migration of the radionuclides which are at present the main cause of the radiation and radioecological situation in the contaminated areas, is explained primarily by the short space of time which had elapsed since the accident and the peculiarities of the radioactive contamination (emitted over 10 days) in the area, and, consequently, by the presence of a large number of hot particles (the highly distinctive feature of the Chernobyl disaster).

We examined two samples of soil of similar morphology affected by radioactive contamination types A and B - fuel and quasi-fuel contamination.

The result of modelling the processes of desorption of the radionuclides caesium-134,137, cerium-144 and ruthenium-106 are given in Table 4.

As noted earlier, the fact that the radionuclides concentrate in the roots and the soil around the roots is evidence of their availability for uptake by plants. For this reason we used only three eluents, since we believe that, in order to assess how available these elements are in the soil for the plants, it is sufficient to determine the basic forms of radionuclide occurrence (water-soluble, exchangeable and mobile) which have a bearing on the soil processes.

The investigations showed that over 70% of the radionuclides studied are fairly strongly bound to the soil complex, and that no more than 1-2% of each element is in the water-soluble form.

In the readily soluble form (after elutriation using ammonium carbonate, pH 7, and a 1M solution of hydrochloric acid) the various elements line up as follows: cerium-144, caesium-134 > caesium-137, ruthenium-106, and in the mobile form they behave unconventionally. One would think that, as the most mobile, ruthenium-106 ought to be first in the sequence, not last. Evidently a role is played here by the presence in these samples of (hot) fuel particles rich in ruthenium and low in of cerium.

Consecutive elutriation reveals that the concentrations of the relatively firmly bound (and thus not readily available) radionuclides in the soil samples studied for contamination types A and B are as follows on average: cerium-144 ~ 0.26, caesium-134 ~ 0.25-0.45, caesium-137 ~ 0.24-0.40, ruthenium-106 ~ 0.30-0.70.

Conclusions

1. Regardless of the type of soil and radioactive contamination the bulk of the long-lived radionuclides/gamma-emitters is concentrated in the upper 0-1 (2) cm soil horizon. As a corollary to this, up to 1989 the boundaries of the contaminated areas had not changed from what they were immediately after the accident.

2. The maximum concentration of radionuclides is found in the > 1 mm fraction - tillering nodes, roots, root matt, litter and soil around the roots - which is proof of the growing role of biogenic migration processes in the mass transfer of radionuclides in biogeocenoses. The dust and clay fractions of the soil fix the radionuclides in the crystalline structures of the minerals and organic component of the soil.

3. The caesium-134/caesium-137 ratio in soils with quasi-fuel contamination differs from the calculated value, which might be due to differences in the topographies and forms of occurrence of the radionuclides in the fallout matrices.

4. In the soils studied the radionuclides cerium-144, caesium-134,137 and ruthenium-106 are found in the relatively available form. In the low-solubility state, depending on the type of radioactive contamination, the concentrations were found to be as follows: cerium-144 ~ 0.26, caesium-134 ~ 0.25-0.45, caesium-137 ~ 0.24-0.60, ruthenium-106 ~ 0.30-0.70.

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Table 4: MODELLING OF FORMS OF OCCURENCE OF LONG-LIVED RADIONUCLIDES BY MEANS OF THEIR CONSECUTIVE DESORPTION

Sample, collection: Depth, cm	Eluent	Fraction, mm	Relative content of radionuclides (initial concentration prior to processing = 1)			
			Ce -I44	Cs -I34	Cs -I37	Ru -I06
Site 105		>I	0,99	0,99	0,99	0,99
type A	Distill.	0,25-I,0	0,99	0,99	0,99	0,99
0 - I cm	Water	<0,25	0,98	0,99	0,99	0,99
I - 2 cm		>2	-	0,98	0,99	0,99
		I-2	-	0,99	0,90	0,99
		0,25-I,0	-	-	0,99	0,99
		<0,25	0,99	0,99	0,99	0,99
0 - I cm	CH_3COONa	>I	0,84	0,73	0,85	0,83
		0,25-I,0	0,80	0,7I	0,84	0,70
	pH 7	<0,25	0,87	0,70	0,89	0,70
		>2	-	0,8I	0,86	0,72
		I-2	-	0,99	0,80	0,75
		0,25-I,0	-	-	0,99	0,99
		<0,25	0,78	0,87	0,90	0,72

Table 4 (cont.)

I	2	3	4	5	6	7
0 - I см	HCl 1 M	>I	0,26	0,33	0,26	0,34
		0,25-I,0	0,27		0,22	0,30
		<0,25	0,22		0,24	0,18
I - 2 см		>2	-	0,19	0,19	0,31
		I-2	-	-	-	-
		0,25-I,0	-	-	-	-
		<0,25	0,63	-	0,28	0,39
R. Khocheva type B	Distill. Water	>I	0,99	0,98	0,98	0,98
		0,25-I,0	0,99	0,98	0,98	0,98
0 - I см		<0,25	0,99	0,99	0,99	0,98
I - I,5 см		>I	-	0,99	0,98	0,99
		0,25-I,0	-	-	-	-
		<0,25	-	-	-	-
0 - I см	CH_3COONa pH 7	>I	0,86	0,74	0,85	0,76
		0,25-I,0	0,73	0,78	0,83	0,68
		<0,25	0,79	0,75	0,89	0,71

Table 4 (cont)

I	2	3	4	5	6	7
I - I,5 см	CH_3COONa pH 7	>I	-	0,76	0,83	0,74
		0,25-I,0	0,75	0,42	0,78	0,75
		<0,25	-		0,80	0,80
0 - I см		>I	0,34	0,45	0,44	0,67
		0,25-I,0	0,26	0,33	0,46	0,86
	HCl	<0,25	-	-	0,38	0,76
I - I,5 см	1 M	>I	-	0,47	0,36	0,52
		0,25-I,0	-	0,30	-	-
		<0,25	-	-	-	-

Comparative Analysis of Chemical Forms of Long-Lived Radionuclides and their Migration and Transformation in the Environment Following the Kyshtym and Chernobyl Accidents

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ABSTRACT

This paper presents data from systematic research into the chemical forms of occurrence of long-lived radionuclides in atmospheric fallout, soils, and bed sediments and suspended particles in water bodies in contaminated regions of the Soviet Union after the Chernobyl accident. The work concentrates on the ratios between the different forms of the most ecologically hazardous radionuclides ^{90}Sr and ^{137}Cs. The main specific feature of Chernobyl fallout was the presence of fuel particles, and this is what determined its subsequent behaviour, such fuel particles being found mostly in the zone close to the Chernobyl NPP. In the initial years following the accident the main feature of soil contamination within the 30-km zone around the Chernobyl NPP was the large proportion of non-exchangeable forms present as compared to Kyshtym contamination and global fallout. This was due to the nature of the atmospheric fallout, as more than 70% of the ^{137}Cs and more than 85% of the ^{90}Sr in the fallout in the town of Chernobyl was in non-exchangeable forms, whereas the radionuclide fallout in Kyshtym mainly comprised soluble compounds.

The difference in the forms of occurrence of the radionuclides in the environment caused differences in their migratory properties (coefficients of washout, dispersion and distribution) and prevailing migratory patterns.

We analyse the mechanisms behind changes in chemical forms in the environment, and determine rate coefficients for the main processes involved in the transformation of chemical forms from one to another. This work highlights the need to take account of the processes involved in the transformation of radionuclide forms when studying and modelling radionuclide propagation.

The distinctive feature of radioactive contamination of the environment following a nuclear accident is the multiplicity of chemical forms of radionuclides in atmospheric fallout, soils and water bodies. When a reactor building is destroyed, particles of nuclear fuel (UO_2) and the decay products in their composition are released into the environment. The rest of the radionuclides are deposited on the surface layer as aerosol particles formed in the atmosphere by condensation (condensation particles). These particles differ in their dimensions, chemical composition and behaviour in the environment.

Hot condensation particles generally have surface contamination and a lower specific activity than fuel particles. The properties of condensation particles are similar to those of particles formed in the last stage of a nuclear explosion and to the global fallout of radionuclides in the period of nuclear weapons testing. This means that it is possible to obtain a reasonably reliable forecast of the behaviour of radionuclides in the composition of condensation particles from data on the behaviour of global fallout.

The behaviour of fuel particles, which were the distinguishing feature of the Chernobyl trail and were concentrated in the zone near the Chernobyl NPP, posed a serious scientific problem following the accident.

In the case of the Kyshtym accident, radioactive substances in the form of liquid pulp were ejected up to a height of 1-2 km and formed a cloud composed of liquid and solid aerosols. The radioactive substances in these aerosols were present in readily soluble compounds (nitrates) [1].

In order to forecast the exchange of the many forms of radionuclides between various components of the environment and their dispersion in the "soil-water" system, they should first be divided up into water-soluble, exchangeable and non-exchangeable forms. These forms of radionuclides were determined in soils, bed sediments, suspended material and atmospheric fallout by means of successive extractions:

1) the water-soluble forms were separated out by means of distilled water, the phase ratio being 1:5;
2) the exchangeable forms were extracted with the aid of a 1N solution of CH_3COONH_4, in the ratio of 1:8;
3) the non-exchangeable forms were separated out by boiling in a 6N solution of hydrochloric acid;
4) the firmly bonded, irreversibly sorbed forms were extracted by means of a mixture of hydrofluoric acid and nitric acid.

The soluble complex compounds of radionuclides and soil components (in neutral or anionic form) and their cations (which are desorbed by the ion exchange mechanism) transfer to the aqueous extract. The concentration of radionuclide cations in the aqueous extract solution depends on the ion-exchangeable equilibrium, which is determined by the exchange capacity of the soil and the concentration of exchangeable ions in solution. The main exchangeable ions in the soil are Ca^{2+}, Mg^{2+}, Na^{+} and K^{+}. In the case of the soils in the zone around the Chernobyl NPP, the amount of exchangeable calcium is generally considerably higher than that of other ions.

Radionuclides sorbed into the soil by means of ion exchange transfer to the acetate extract.

Non-exchangeable forms of radionuclides, i.e. forms which do not transfer to water in the environment under normal conditions, transfer to acid extracts. The non-exchangeable form includes radionuclides found in the composition of fuel particles and those known as 'irreversibly sorbed' forms (those in the crystal lattice of minerals and radionuclide-organic compounds with an insoluble natural organic substance).

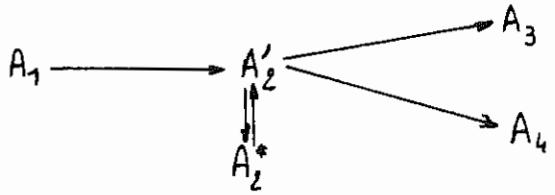
Table 1 shows data on the chemical forms of the most ecologically dangerous radionuclides - ^{137}Cs and ^{90}Sr - in atmospheric fallout during the period of radionuclide release from the damaged reactor between 26.04.86 and 10.05.86. It is evident that the composition of the fallout changed considerably during the accident period. These changes were caused by physico-chemical processes in the damaged reactor and in the atmosphere once the radioactivity had been released from the reactor.

After being deposited on the ground surface, the water-soluble component of the radionuclides interacts with the absorptive soil complex, is sorbed onto particles and is involved in a complexing reaction.

After ^{90}Sr is deposited on the soil, there is no significant redistribution of mobile and non-mobile forms. The water-soluble fraction of fallout switches primarily to the exchangeable state.

The situation is completely different for ^{137}Cs. After the water-soluble fraction of the ^{137}Cs fallout is absorbed by the soil complex, it switches to the exchangeable form, which is then rapidly fixed, however, as it switches to the irreversibly sorbed state due to its being incorporated into the crystal lattice of the minerals. The model experiments we carried out immediately after the Chernobyl accident in the summer of 1986 showed that it normally takes several dozen days for ^{137}Cs to become fixed in the soil. These values tally with those obtained for ^{137}Cs from global fallout and other sources.

The sum total of the radionuclide transformation processes in the "soil-water" system may be shown schematically as follows:



(1)

$$A_1 + A'_2 + A^*_2 + A_3 + A_4 = A_0$$

where A_0 is the overall content of the radionuclide
A_1 is the radionuclides in the composition of the fuel particles
A'_2 is the cation form of the radionuclide in solution
A^*_2 is the radionuclides absorbed by the soil in ion exchange
A_3 is the irreversibly sorbed form of the radionuclide
A_4 is the dissolved complex compounds of the radionuclide
K is the constant for the ion-exchange equilibrium
K_{ij} is the rate constant for the corresponding transformation process.

The main forms of occurrence of radionuclides shown in this diagram differ considerably in their migration mechanisms and rates. The A_2 and A_4 forms migrate with water in solution, but the rate of movement of A_2 is considerably lower than that of A_4, due to moderation of adsorption and desorption under ion-exchange interaction with the solid phase. The A_1, A_2 and A_3 forms migrate only with particles in whose composition they are present. The main movement processes for these forms are the transport of suspended load (in water courses and reservoirs), and seepage through pores, fissures etc. and what are known as 'biomixing processes' (in soil and bed sediments).

When describing the migration of radionuclides, it is essential to consider the movement of each form separately. Moreover, it is necessary to take into account the on-going processes of transformation between forms, i.e. the mutual transfer of one form to another.

The equations which describe radionuclide migration in soil and bed sediments in this case are as follows:

$$\frac{\partial A_i}{\partial t} = \frac{\partial}{\partial x}\left(D_i \frac{\partial A_i}{\partial x}\right) - v_i \frac{\partial A_i}{\partial x} + \sum_j k_{ij} A_j - \sum_j k_{ij} A_i \qquad (2)$$

with initial conditions of

$$A_i = A_i^o \delta(x-0)$$

and boundary conditions of

$$A_i\big|_{x=\infty} = 0$$

where D_i and v_i are the effective coefficients of migration of each form. Experimental studies in the Chernobyl accident area have shown that $v_i \approx 0$ for fuel particles. The rate of convective movement of the dissolved radionuclides is determined by the moisture infiltration rate.

The exchangeable forms migrate with the infiltration flow, their retention factor depending largely on the distribution coefficient. The magnitude of the distribution coefficient for cations in the radionuclides is determined by the exchange capacity of the sorbent (soil, bed sediments) and the concentration of exchangeable ions (Ca^{2+}, Mg^{2+}, Na^{+}, K^{+}) in solution.

In the case of the Kyshtym accident, the main long-lived radionuclide responsible for contamination was ^{90}Sr. The ^{137}Cs content in the mixture of fission products amounted to less than 1%.

In the Chernobyl NPP near zone the level of ^{90}Sr and ^{137}Cs contamination was about the same, whereas at greater distances from the Chernobyl NPP the contamination was caused primarily by ^{137}Cs.

Table 2 [1] shows the amounts of ^{90}Sr in exchangeable forms in various types of soil in the seventh year after the Kyshtym accident. The proportion of the exchangeable form in the upper soil layer is relatively high, ranging from 72 to 96%.

No significant change over time in the forms of occurrence of ^{90}Sr from the Kyshtym accident was observed. ^{90}Sr is found primarily in the exchangeable form and only an insignificant part of it transfers into mobile complex compounds with organic soil substances. Fixing and transfer of the exchangeable form into the non-exchangeable form do not play a major role for ^{90}Sr.

For the purposes of comparison, Tables 3 and 4 provide data on the amounts of the various forms of ^{90}Sr and ^{137}Cs in the main soils of the 30-km zone around the Chernobyl NPP in 1986-87 [2]. The high proportion of non-exchangeable forms of ^{90}Sr in the initial years after the accident in the soils of the Chernobyl NPP near zone stemmed from the fact that most of this radionuclide was deposited in the form of fuel particles.

As time passed after the accident, the fuel particles broke down, thus effectively increasing their surface area in contact with the aqueous medium, and an additional quantity of radionuclides transferred into solution (see Fig. 1). This led to an increase in the proportion of exchangeable forms of ^{90}Sr. Experiments concerning the content of the forms of occurrence of ^{90}Sr and ^{137}Cs in the main soils of the 30-km zone around the Chernobyl NPP made it possible to establish the constant for the rate at which these radionuclides leached from fuel particles (K_{12}) and the constant for the ^{137}Cs fixation rate (K_{23}). The figures obtained are shown in Table 5. Table 6 contains figures on the effective dispersion coefficients of the exchangeable and non-exchangeable forms of ^{90}Sr and ^{137}Cs in soils in the 30-km zone around the Chernobyl NPP.

Fig. 1 shows experimental profiles for various forms of ^{137}Cs in soddy-podzolic soil 700 days after the accident and the results of model calculations. Fig. 2 illustrates the migration of exchangeable forms of ^{90}Sr and ^{137}Cs in this soil, account being taken of their transformation processes.

The transfer of radionuclides into surface runoff is the most important process in their egress from contaminated territory into water bodies. The washout coefficient, which is equivalent to the proportion of the radionuclide in the soil entering the water body with surface runoff, defines radionuclide washout. It is best to examine separately the washout of radionuclides in dissolved form and in suspension-sorbed form.

$$K_l = \frac{\int_0^T C_w(t)Q(t)dt}{\int_S C_s dS}; \quad K_s = \frac{\int_0^T C_a(t)Q(t)dt}{\int_S C_s dS} \tag{3}$$

where K_l and K_s are the coefficients of "liquid" and "solid" washout,

C_w is the concentration of the radionuclide in the runoff solution,

C_a is the concentration of the radionuclide on the suspensions,

C_s is the soil contamination level at the surface,

S is the area of the catchment area,

T is the duration of runoff,

Q is the intensity of runoff,

M is the intensity of removal of suspensions.

In order to compare the results of the experiments with various hydrological scenarios and use them to forecast secondary contamination of water bodies, K_l and K_s can be related to the main hydrological features of runoff: K_l over the runoff layer and K_s over the mass of suspensions removed from one unit of area:

$$K_l^* = \frac{K_l}{h} = \frac{\overline{C}_w}{\overline{C}_s}; \quad K_s^* = \frac{K_s}{m} = \frac{\overline{C}_a}{\overline{C}_s} \tag{4}$$

where h is the runoff layer, m is the mass of suspensions removed from one unit of area, and C_w, C_a and C_s are the weighted mean concentrations in solution and on suspensions and the weighted mean amount of the radionuclide in the catchment area.

The first studies regarding washout of Chernobyl radionuclides were carried out as early as the summer of 1986, and were designed to yield washout coefficient values for rapid forecasting of the prevailing radiation situation in water bodies. These studies involved artificial sprinkling of specially equipped runoff areas. Table 7 shows standardised coefficients of "liquid" washout for the main long-lived radionuclides in pluvial and spring-time runoff. With the passage of time the radionuclides move deeper into the soil as a result of vertical migration processes. This should lead to a decrease in the washout coefficients.

However, there was no significant decrease in K_l for ^{90}Sr, which can be explained by the fact that the depletion of ^{90}Sr (which can transfer into solution) in the upper soil layer is balanced out by transformation of its non-exchangeable forms into exchangeable forms due to leaching of fuel particles.

For the purposes of comparison, Table 7 shows the normalized ^{90}Sr washout coefficients obtained in model experiments on the Urals contamination trail left by the Kyshtym accident in an area contaminated after nuclear explosions tests.

The experiments at Kyshtym were carried out over a period of two years following the introduction of ^{90}Sr into the soil. It can be seen that the washout coefficients obtained in these experiments generally exceed the values for the Chernobyl trail. This was due to the fact that, in contrast to the Chernobyl accident, up to 90% of the ^{90}Sr fallout was present in the exchangeable form in these cases.

The fast rate at which equilibrium concentrations of ^{90}Sr were attained in the runoff points to the decisive role of ion-exchange processes in the formation of radionuclide concentrations in runoff. Given that calcium is the dominant cation in the soils of the Chernobyl NPP 30-km zone, the following equation must be satisfied in order to obtain an equilibrium concentration of ^{90}Sr in these conditions:

$$[^{90}Sr]_s = \frac{[^{90}Sr]_e \cdot [Ca]_s}{K_{Ca}^{Sr}[Ca]_e} \qquad (5)$$

where K_{Ca}^{Sr} is the exchange selectivity coefficient for Ca and Sr ions in the soil;

$[Sr]_s$ is the concentration of ^{90}Sr in the solution;

$[Ca]_s$ is the concentration of calcium in the solution;

$[^{90}Sr]_e$ is the amount of exchangeable ^{90}Sr in the upper soil layer;

$[Ca]_e$ is the amount of exchangeable calcium in the soil.

In accordance with (5) the concentration of ^{90}Sr in runoff should, for the same soils, be directly proportional to the concentration of Ca. Experiments showed that the concentration of ^{90}Sr in runoff depends on the amount of Ca in runoff (Fig. 3).

During snowmelt runoff (in contrast to pluvial runoff), we observed that the concentration of ^{90}Sr is largely dependent on runoff intensity (see Fig. 4). This can be explained by the fact that when the soil is partly frozen some of the water flows off without touching the soil, which reduces the ^{90}Sr concentration in the runoff. In this case $[^{90}Sr]_s$ is not directly proportional to $[Ca]_s$ (see Fig. 5).

$$[^{90}Sr]_s = \frac{[^{90}Sr]_e}{K_{Ca}^{Sr}[Ca]_e} \left\{ \frac{[Ca]_{s1}}{[Ca]_{s1}-[Ca]_{s2}}[Ca]_s - \frac{[Ca]_{s1}[Ca]_{s2}}{[Ca]_{s1}-[Ca]_{s2}} \right\} \qquad (6)$$

Where $[Ca]_{s1}$ is the concentration of calcium in water flowing over thawed soil;
$[Ca]_{s2}$ is the background concentration of calcium in snow.

When the upper soil layer has fully thawed, runoff continues due to the previously frozen water being released in the soil and to precipitation. The ^{90}Sr concentration therefore remains constant and is not dependent on runoff intensity, as in the case of pluvial runoff.

The distribution coefficient (K_d), which is the ratio between the concentration of the radionuclide in the solid phase and its concentration in solution, is the most important value for describing and forecasting the behaviour of radionuclides in water bodies. The K_d value depends primarily on the radionuclide's chemical nature, the adsorption mechanism and the physico-chemical characteristics of the solid material determining its adsorption properties.

Each chemical form of the radionuclide has a specific K_d value. Thus, in the case of soluble complex compounds of radionuclides and fulvic acids (A_4), the K_d is close to zero because these compounds are not easily sorbed and exist primarily in solution. The distribution of the ionic form

of the radionuclide is governed by the processes of ion exchange and, as a rule, the K_d for ^{90}Sr ranges from several units to several tens of units. The K_d for non-exchangeable forms is very high (> 1 000). The actually observed distribution coefficient is the weighted mean of the contributions from the various chemical forms of the radionuclides.

In the case of Chernobyl radionuclides, the effective K_d value for ^{90}Sr in the "suspensions-water" system is between 50 and 1 000, and for ^{137}Cs it ranges from 1 000 to 100 000.

The significant difference in the values of the ^{90}Sr and ^{137}Cs distribution coefficients is caused by the difference in the predominant adsorption processes affecting these radionuclides. Reversible adsorption via ion exchange is characteristic of ^{90}Sr. Processes of fast fixation as a result of incorporation into the crystal lattice of minerals are characteristic of ^{137}Cs.

The presence of fuel particles in the Chernobyl release leads to a major increase in the K_d value for each radionuclide in its composition.

The ratio between the phases (solution:solid phase) has a considerable influence on the magnitude of the distribution coefficient. The K_d increases when the ratio between the phases increases. The results of model experiments to study this relationship for ^{90}Sr are shown in Fig. 6. The same relationship is observed for other radionuclides as well. The fact that the K_d is dependent on the ratio between the phases explains the considerable difference between the K_d for the "bed sediments-water" system and that for the "suspensions-water" system.

It should be pointed out that in the case of low phase ratios the adsorption-desorption equilibrium is reached far more slowly than in suspension, where desorption equilibrium is attained after no more than 15 minutes. A temperature change (within the range 4-45°C) has no significant influence on the K_d value.

Table 8 shows the average values for the distribution coefficients of ^{90}Sr and ^{137}Cs in the "suspensions-water" system of surface runoff in catchment areas of the 30-km zone around the Chernobyl NPP after the accident. The ratio between the phases in the surface runoff was more than 1 000.

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Table 1

Chemical forms of ^{137}Cs and ^{90}Sr fallout, Chernobyl, 1986

Date of exposure	^{137}Cs, %					^{90}Sr, %				
	Water-soluble	Acet. exch.	Total mobile	Acid exch.	Undissolved residue	Water-soluble	Acet. exch.	Total mobile	Acid exch.	Undissolved residue
25-26.04	4.3	25.8	30.1	24.0	21.3	0	0	0	37.5	62.5
26-27.04	14.3	4.9	19.2	30.9	24.2	1.0	1.5	2.5	80.2	17.7
27-28.04	0.3	4.3	4.6	–	–	0	1.8	1.8	85.2	13.0
28-29.04	5.9	26.2	32.1	27.1	5.9	6.3	21.0	27.3	65.1	7.6
29-30.04	3.4	14.8	18.2	42.2	3.2	1.0	5.0	6.0	91.0	3.0
1.2.05	13.0	22.2	35.2	55.4	5.0	4.0	9.0	13.0	82.3	5.0
2-3.05	7.8	19.7	27.5	55.4	0.9	1.3	3.6	4.9	92.0	3.0
3-4.05	1.3	11.0	12.3	34.1	–	0.72	7.9	8.6	88.6	2.8
21-23.06	1.2	1.76	2.96	85.4	10.7	0.2	1.5	1.7	93.4	4.9
25-26.06	3.4	3.4	6.8	80.1	5.9	–	–	–	–	–
26-27.06	3.1	3.5	6.6	86.4	4.3	–	–	–	–	–

Table 2

Percentage of exchangeable forms of ^{90}Sr in various types of soil in the 7th year after the formation of the Kyshtym trail [1]

Type of soil	Exchangeable strontium as a percentage of the total quantity of strontium
Soddy-podzolic	95
Grey forest	89
Leached chernozem	86
Meadow solod	79
Solonchak	72
Marshy	76

Table 3

^{90}Sr chemical forms in soils in the Chernobyl NPP 30-km zone, 1986-1987 (after Konoplev A.V. et al., 1988)

Site No	Soil type	Date sample collected	Water-soluble, %	Exchangeable, %	Non-exchangeable, %
1.	podzol	16.07.87	0.06±0.02	11.1±3.2	88.8±3.2
2.	moderately podzolic	09.07.87	0.67±0.12	35.4±7.5	64.0±7.7
3.	"	09.07.87	0.72±0.03	34.0±2.4	65.2±2.5
4.	"	15.07.87	0.18±0.06	16.1±2.6	83.8±2.7
5.	slightly podzolic	09.07.87	0.39±0.25	23.4±5.1	76.2±5.4
6.	sod podzolic	14.07.87	0.10±0.05	16.8±4.2	83.1±4.5
7.	cultivated land	15.07.87	0.04±0.01	5.20±2.1	94.8±2.6
8.	"	13.07.87	0.40±0.14	23.9±3.9	71.8±1.6
9.	"	12.07.87	0.05±0.02	7.5±2.5	94 ±5
10.	"	15.07.87	0.06±0.01	8.0±0.25	92 ±1
11.	"	11.07.87	0.4 ±0.1	15.3±1.8	84.3±1.8
12.	"	24.07.87	0.5 ±0.1	38.8±2.1	60.7±2.0
13.	primary sandy	14.07.87	0.8 ±0.1	4.7±1.0	94.5±1.0
14.	sod-alluvial	20.11.86	0.8 ±0.5	24 ±5	75.2±5.3
15.	"	13.07.86	1.0 ±0.8	13 ±11	86 ±12
16.	"	10.07.87	0.2 ±0.1	13.6±2.6	86.2±2.6
17.	"	16.07.87	0.1 ±0.02	5.5±2.8	94.4±2.8
18.	"	17.07.87	0.1 ±0.04	24.9±3.0	75 ±2
19.	"	08.07.87	0.3 ±0.1	31.8±5.4	67.9±5.5
20.	"	12.07.87	0.3 ±0.1	30.5±0.6	69.2±0.7

Table 3 (contin.)

Site No	Soil type	Date Sample collected	Water-soluble, %	Exchangeable, %	Non-exchangeable, %
21.	meadow alluvial	08.07.87	0.1±0.02	23.2±2.7	72.7±2.9
22.	sod	16.07.87	0.8±0.1	7.2±0.2	92 ±2
23.	swamp alluvial	08.07.87	3.2±0.3	31.1±1.6	65.7±1.8
24.	"	08.07.87	4.5±0.5	23.5±3.2	72.0±3.3

Table 4

^{137}Cs chemical forms in soils in the Chernobyl NPP 30-km zone, 1986 – 1987 (after Konoplev A.V. et al., 1988)

Site No	Soil type	Date Sample collected	Water-soluble, %	Exchangeable, %	Non-exchangeable, %
4	moderately podzolic	15.07.87	0.9±0.5	13.8±0.9	85.3±0.2
5.	slightly podzolic	09.07.87	0.4±0.1	5.2±0.2	94.4±1.0
7.	cultivated land	15.07.87	0.2±0.1	6.3±0.1	93.5±5.0
8.	"	13.07.87	1.0±0.2	10 ±1	89 ±1
10.	"	15.07.87	0.1±0.02	8.9±0.6	91 ±1
11.	"	11.07.87	0.6±0.2	6.9±0.3	92.5±2.5
14.	sod-alluvial	20.11.86	0.1±0.02	1.7±0.4	98.2±0.4
15.	"	13.07.86	0.2±0.02	4.7±0.6	
19.	"	13.07.86	0.2±0.05	0.6±0.1	99.2±0.2
23.	swamp alluvial	08.07.87	0.1	1.5	98.4
24.	"	08.07.87	0.1	0.5	99.4

Table 5

Constants of rate of ^{90}Sr and ^{137}Cs leaching from fuel particles and of ^{137}Cs fixation in various soils of the 30-km zone around the Chernobyl NPP

Site number	Place	Type of soil	K_{12}/day	K_{23}/day
1.	Chernobyl	Soddy, slightly podzolic	$1.5 \cdot 10^{-3}$	$1.5 \cdot 10^{-2}$
2.	Benevka	Alluvial, soddy, acidic	$1.7 \cdot 10^{-3}$	$1.2 \cdot 10^{-2}$
3.	Kopachi	Slightly podzolic	$4 \cdot 10^{-4}$	-
4.	Korogod	Soddy-podzolic	$3.6 \cdot 10^{-3}$	$1.5 \cdot 10^{-2}$

Table 6

Effective migrotion coefficients for exchangeable and non-exchangeable forms of ^{90}Sr and ^{137}Cs in certain soils of the 30-km zone around the Chernonbyl NPP

Place sample taken	Type of soil	(^{90}Sr), 10^{-8} cm^2/s		(^{137}Cs), 10^{-8} cm^2/s	
		Exchangeable form	Non-exchangeable form	Exchangeable form	Non-exchangeable form
1. Town of Chernobyl	Cultivated, soddy-podzolic, sandy	5.4 + 2.0	2.8 + 1.2	2.6 + 0.6	1.9 + 0.6
2. Settlement of Benevka	Alluvial, soddy, acidic, sandy-loam	3.2 + 1.0	2.1 + 0.6	1.6 + 1.1	1.4 + 0.5
3. Settlement of Korogod	Cultivated, soddy-podzolic, sandy-loam	2.1 + 0.5	1.5 + 0.8	2.1 + 0.6	1.5 Ω 0.5

Table 7

Coefficients of "liquid" washout of certain nuclear fuel fission products, standardized by runoff layer

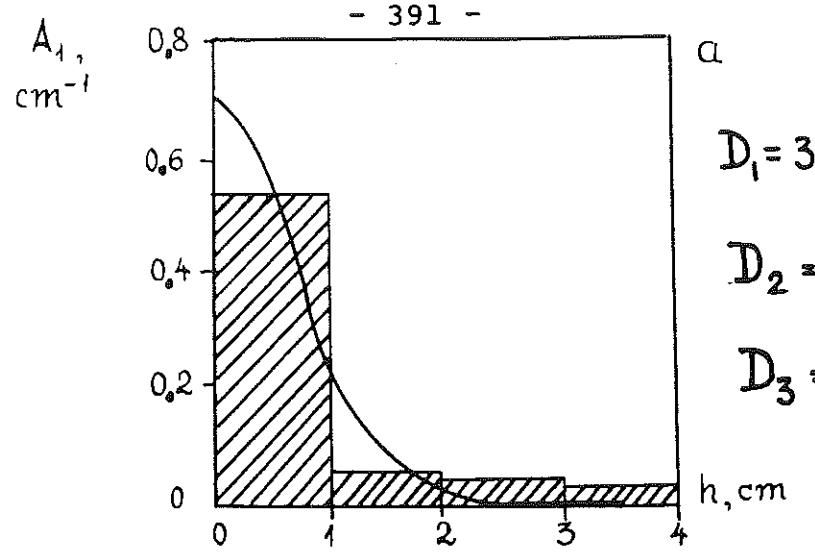
Location	Site number	Date	10^{-5} mm $^{-1}$					
			^{137}Cs	^{134}Cs	^{103}Ru	^{106}Ru	^{144}Ce	^{90}Sr
			Pluvial runoff					
Settlement of Dovlyady	1	16.07.86	3.4 + 1.2	3.5 + 1.3	–	0.7 + 0.3	–	10 + 4
	"	12.10.86	1.6 + 0.7	1.3 + 0.6	1.6 + 0.6	–	0.6 + 0.3	8.4 + 2.8
	"	"	0.5 + 0.2	0.4 + 0.2	0.7 + 0.4	–	–	8.4 + 3.0
	"	"	1.4 + 0.5	1.3 + 0.6	–	–	0.5 + 0.3	6.5 + 2.0
	"	"	0.6 + 0.4	0.4 + 0.3	–	–	–	5.2 + 1.7
Settlement of Benevka	4	14.10.86	0.6 + 0.2	0.6 + 0.3	–	–	–	5.8 + 2.1
	"	10.87	0.3 + 0.1	0.3 + 0.1	–	–	0.2 + 0.1	18 + 0.4
			Snowmelt runoff					
Settlement of Benevka	4	March–April 1987	0.2 + 0.1	0.3 + 0.2	–	0.3 + 0.2	0.2 + 0.1	0.6 + 0.3
Settlement of Kopachi	5	"	0.4 + 0.1	0.3 + 0.1	–	–	0.5 + 0.4	1.1 + 0.3
	"	"	5.7 + 2.5	5.0 + 2.4	–	0.7 + 0.3	0.4 + 0.2	6.6 + 1.6
Settlement of Korogod	6	"	0.7 + 0.4	–	–		0.6 + 0.2	0.8 + 0.3
Kyshtym			–	–	–	–	–	13
Nuclear explosions			–	–	–	–	–	83

Table 8

Coefficients of distribution (K_d) of ^{90}Sr and ^{137}Cs in surface runoff, 1986 [2]

Site Number	Date	$K_d(^{90}Sr)$	$K_d(^{137}Cs)$
1.	16.07	293	2 380
	"	1 240	9 870
	12.10	317	2 380
2.	"	534	3 800
3.	18.07	-	370
4.	14.10	493	4 760
	"	325	3 100

^{137}Cs

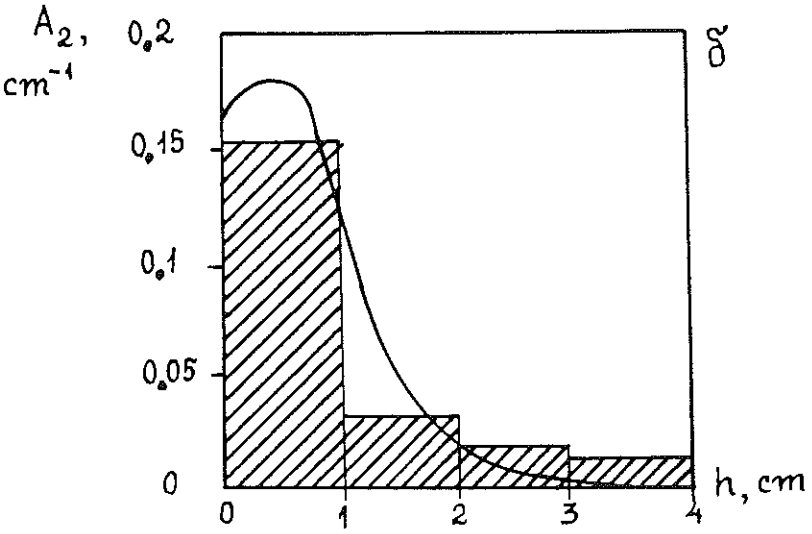


$D_1 = 3.5 \cdot 10^{-4}$ cm²/day

$D_2 = 5.0 \cdot 10^{-4}$ cm²/day

$D_3 = 1.5 \cdot 10^{-4}$ cm²/day

$V_1 = V_3 = 0$

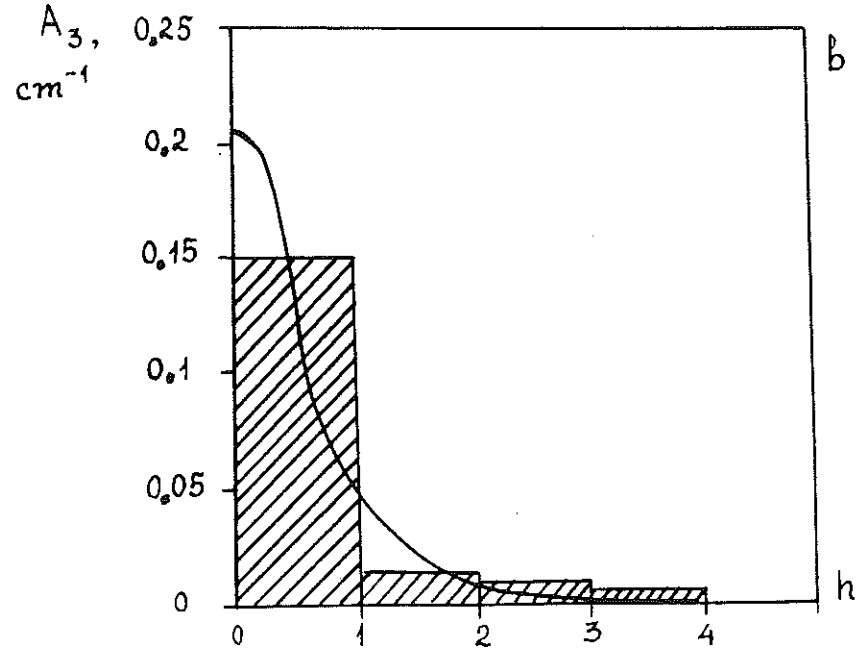


$V_2 = 3 \cdot 10^{-4}$ cm/day

$k_{12} = 1.5 \cdot 10^{-4}$ day^{-1}

$k_{23} = 6 \cdot 10^{-4}$ day^{-1}

$A_1^o = 0.68$



$A_2^o = 0.27$

$A_3^o = 0.05$

Fig. 1: Comparison of model and experimental vertical profiles for different chemical form of ^{137}Cs on 700th day after the accident.

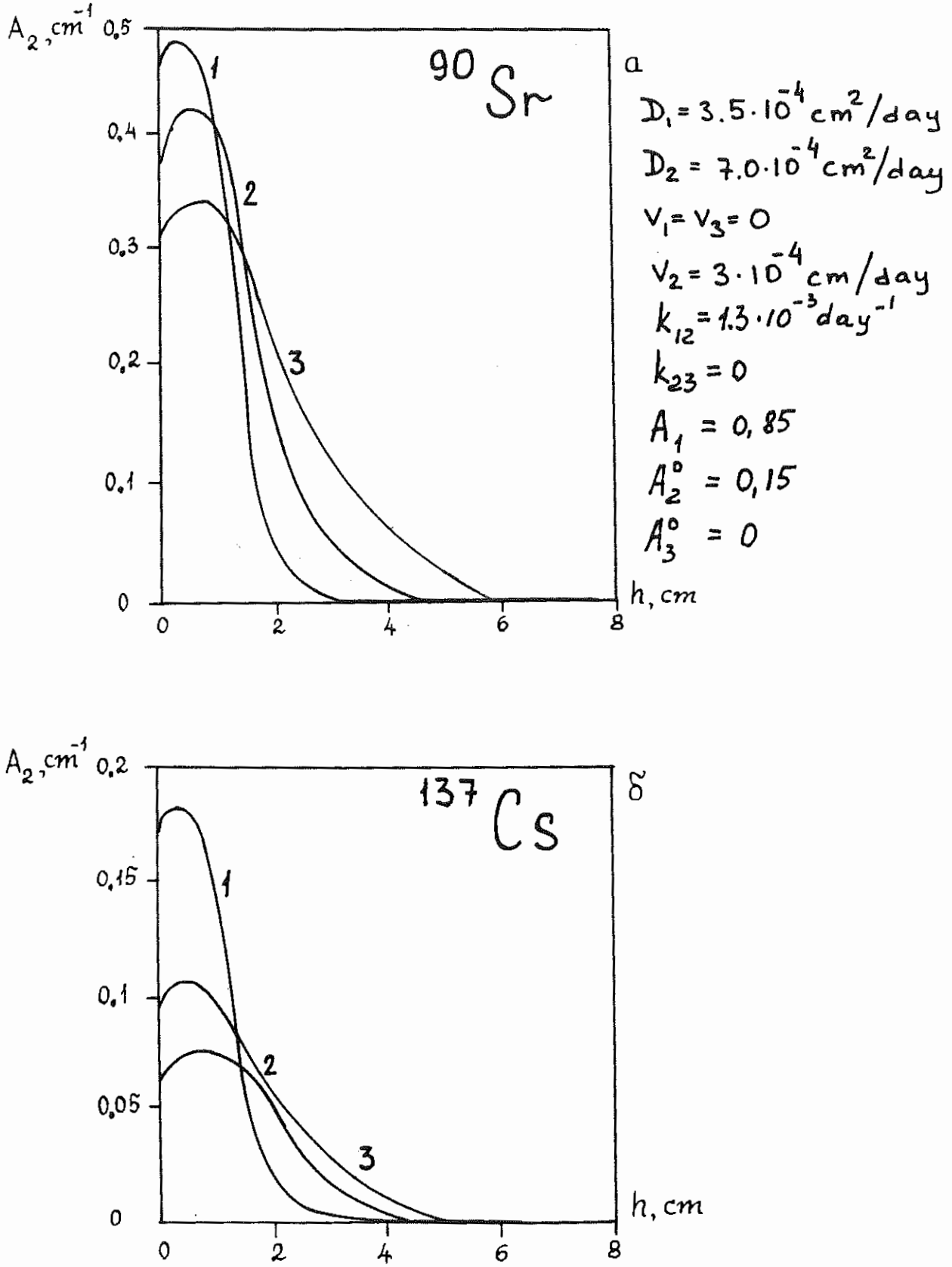


Fig. 2: Model vertical profiles of exchangeable forms of ^{90}Sr (a) and ^{137}Cs (б) on 700th (1), 1400th (2 and 2100th (3) day after the accident.

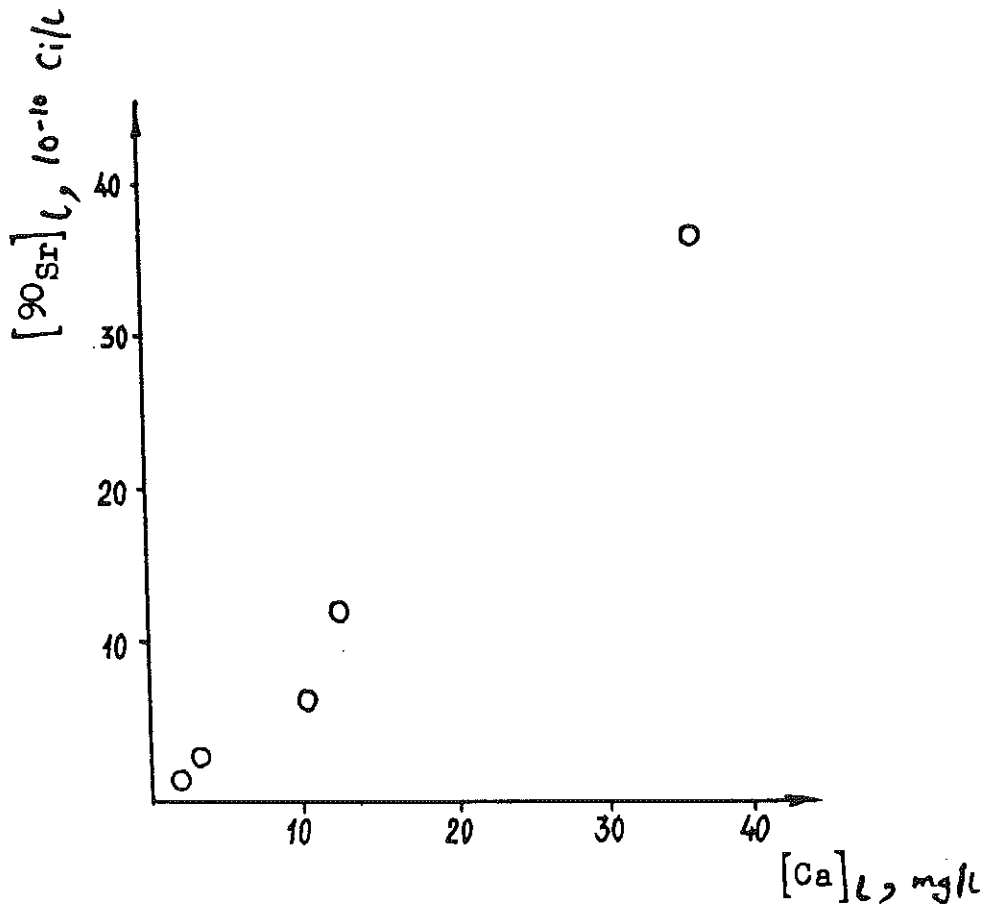


Fig. 3: Experimental dependence of ^{90}Sr runoff concentration on Ca concentration in solution

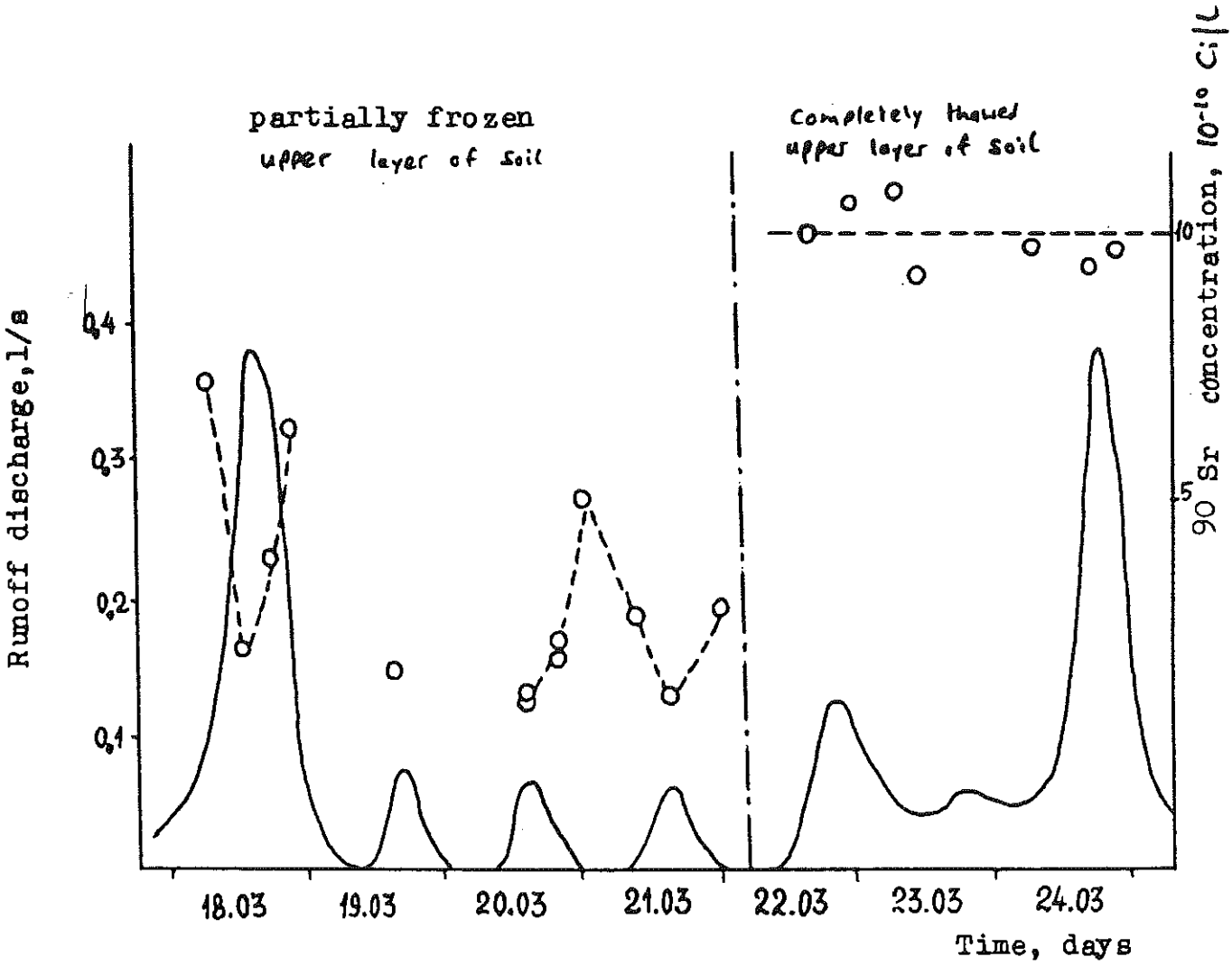


Fig. 4: Dependence of ^{90}Sr concentration on runoff intensity during snowmelt

— runoff discharge

o ^{90}Sr concentration in solution of runoff

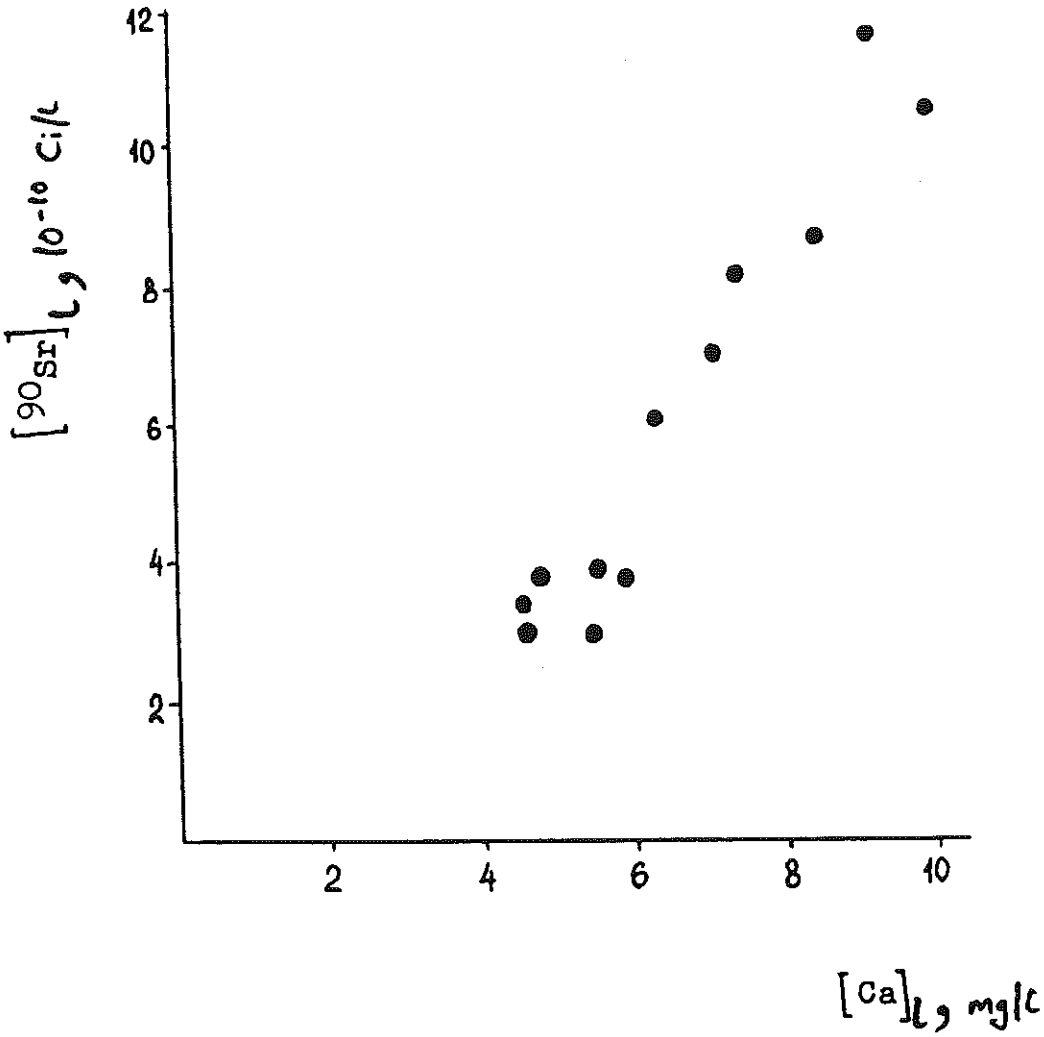


Fig. 5: Dependence of ^{90}Sr concentration on Ca concentration during snowmelt runoff when soil is partially frozen

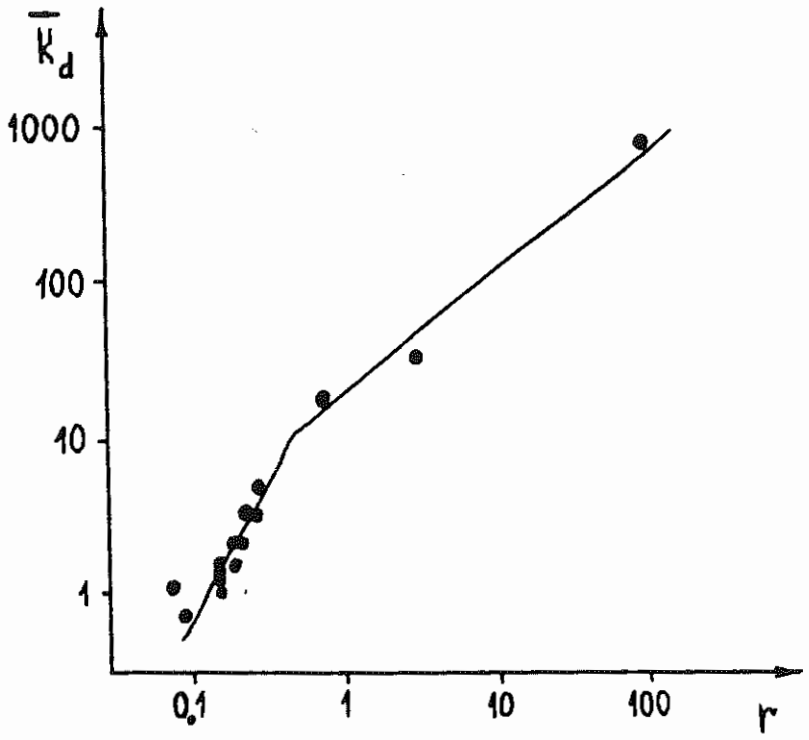


Fig. 6: Dependence of distribution coefficient (K_d) on water-soil ratio (r) [9]

Behaviour of Pu-239 and Pu-240 in Soils Following the Southern Urals and Chernobyl Accidents

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ABSTRACT

In addition to determining the plutonium content of soils we focused our attention on studying its forms of occurrence and migration in topsoil, comparing the data obtained with other sources relating to plutonium deposition on the earth's surface.

The study revealed that plutonium is quickly and lastingly absorbed by soils, and that its distribution between water-soluble, exchangeable, mobile, amorphous and silicate forms (as well as between the various components of organic and inorganic soil matter) is determined by soil type and depositio source.

It was shown that plutonium distribution and migration in the landscape and soils is determined by their location, type, genctic structure, species of vegetation and other natural factors, as well as forms of uptake and occurrence in soils. We established zones of plutonium removal and plutonium presence in geochemical barriers.

We established two types of plutonium transfer in soils: the predominant, slow mode (which accounts for 65-100% of plutonium transfer in the various soil types), and the rapid type (with migration coefficients one or two orders of magnitude greater).

The depth, coefficients and intensity of plutonium migration in soils increase with increasing amounts of water-soluble organic matter and light mechanical fractions, as well as in more hydromorphic soils containing higher concentrations of carbonates.

We evaluated plutonium removal from the surface horizons of various types of soil, and established that such layers are cleansed naturally, mainly via vertical migration through the soil profile.

The data obtained on the forms of plutonium occurrence and distribution in soils helped explain the observed processes and mechanisms governing its migration in the soil profile, leading us to conclude that "Southern Urals" and "Chernobyl" plutonium does not behave in a specific way: such behaviour depends both on the forms of occurrence and landscape-geochemical conditions. The only difference could be in the rate of migration of plutonium when it is deposited on topsoil together with finely dispersed fuel particles.

This article contains a summary of the results of investigations into the behaviour of plutonium in soils following two accidents which differed with regard to the radionuclide and chemical composition of the material released. The study of the processes governing the distribution and migration of plutonium, apart from its radioecological significance, is important for our understanding of the radiogeochemistry of this element, whose isotopes are unstable under natural conditions.

Since the Southern Urals accident was the result of a fault in a container for storing highly radioactive waste from the production of plutonium, it was important to evaluate the amount of this element in the surrounding area and determine its distribution characteristics in soils.

The investigation showed that, compared to ^{90}Sr and other fission products, there were only traces of plutonium fallout, most of which was concentrated close to the site. At a distance of a few kilometres, the plutonium content in soils was close to normal background levels. The $^{238}Pu/^{239}$, ^{240}Pu ratio in soils was 0.03-0.05.

Over the course of 24-26 years there was noticeable plutonium migration to deeper soil horizons (Fig. 1); between 47-82% and 82-97% of plutonium fallout remained in the upper 0-5 (8) and 0-18 (20) cm layers respectively, depending on soil and plant cover type. For example, the depth of migration in meadow-chernozem soil on carbonate loam (final profile) where plutonium was observed at a depth of 45 cm, is due not only to the hydromorphic nature of the soil but also to the formation of soluble carbonate compounds. The influence of the genetic structure of the soil profile was seen most clearly in grey forest soil under park-type birch forest (first profile). The plutonium removed from litter and then from the eluvial horizon A_2 accumulated in humic and illuvial horizons. In other soils there was also a tendency towards secondary migration of plutonium at a depth of 13 (14) - 18 (20) cm.

The ratio between the plutonium content in grasses and soil in meadows and under forest cover is close to that for other contamination sources (Table 1).

The plutonium content in soils in the European part of the Soviet Union following the Chernobyl accident varied, according to distance and direction from the power station, from a few dozen to four thousand Bq/m^2 outside the 30 km zone and from tens to hundreds of thousands within it. The considerable patchiness and diversity of plutonium content in the soils of a single region and even of a single sample resulted not only from the laws governing atmospheric transfer and fallout onto the earth's surface, but also from the presence of particles of different sizes and different radionuclide and chemical composition.

The average $^{238}Pu/^{239}$, ^{240}Pu ratio in the majority of the soil, aerosol and fallout samples examined was 0.3 (variations between 0.25 and 0.35), although in some samples it was lower than 0.20 and even values of 0.01-0.05 were noted. These soils may have been contaminated either by fresh fuel or were not contaminated by material from the accident. Such sites were found both close to and far from the Chernobyl NPP.

Data on the ratio between volatile and non-volatile radionuclides in environmental features and the amount calculated to be in the irradiated fuel at the time of the accident made it possible to define three types of contamination;

Type A: mechanically released finely dispersed fuel with unchanged composition, mostly found in the central zone (mainly in the lower section) and up to 60-70 km to the west.

Type B: finely dispersed fuel particles and other emissions, enriched with volatile radionuclides of ruthenium, caesium and barium by 4, 8 and 2 times respectively. Plutonium enrichment is up to twice as much as calculated and four times as much as in type A. This type of contamination is observed up to a distance of 100 km.

Type C: Emissions heavily enriched with radionuclides of caesium and ruthenium (up to 900 and 80 times respectively) and also with plutonium (up to approximately 14 times).

It is typical for distances between 100 and 150 km and is observed at distances up to 500 km. Even higher soil enrichment with plutonium (up to 200 times the calculated value) observed at considerable distances from Chernobyl may be due to relatively volatile compounds formed during the accident.

At sites contaminated with finely dispersed fuel particles a correlation was noted between the content of plutonium and ^{144}Ce and between the content of plutonium and volatile ^{103}Ru in soil (Table 3). The paired correlation coefficients of Pu-Ce decline with increasing distance from the accident site and with the change to contamination by more volatile emission products, while those of Pu-Ru increase.

Vertical migration of plutonium in soils becomes apparent quite soon after it settles on the surface (Fig. 2); as in the case of the Kyshtym accident, it is more pronounced in soils with a high carbonate content. Retention of plutonium in the upper 1-1.5 cm layer is made possible by the steppe root matting and by the tillering nodes of cereals, with a higher concentration in soils with a humic or clayey mineral content.

The influence of natural factors and the depth of plutonium penetration both increase with time (Fig. 3). Where the maximum plutonium content in non-hydromorphic soils remains in the 0-2 cm layer (73-92%), most intensive migration is observed in formerly arable soddy podzolic soil. Where a layer of moss (1 cm thick) covers the soil surface in pine forest, it was found to contain 92% of the plutonium.

A different pattern was observed in other geochemically connected landscapes (Fig.4). While a high proportion of plutonium has remained in the upper profile of soils in drained marshes on high flood plains with a higher plutonium content, the content in hydromorphic soils in terraces above flood plains and particularly in the terrace-edge hollows has been lower and vertical migration has been more intensive. The reason for the latter, in addition to inundation, may be the high proportion of water-soluble organic ligands and the anaerobic conditions causing the reduction of tetravalent plutonium to the more mobile trivalent form. The secondary concentration of plutonium in illuvial and silty horizons is very pronounced for soils of this landscape.

The data presented, and the discoveries we had made beforehand, which have been published in the literature, about the processes governing the behaviour of global and technogenic plutonium lead us to the conclusion that it quickly becomes incorporated into the biogeochemical migration cycles accompanying its redistribution in landscapes and soils. In this case, plutonium behaves according to the general mechanisms governing the migration of chemical elements and artificial radionuclides of strontium, caesium and others, which also accumulate in geochemical barriers - depressions in the relief, illuvial and muddy horizons - in the course of horizontal and vertical transfer.

Table 4 shows two types of plutonium movement in soils: slow (which predominates) and fast, with migration coefficients varying by 1 - 2 orders of magnitude. The plutonium migration coefficients found are of the same order as those for other artificial radionuclides, and they also increase with depth as the amount of humus and heavy fractions in the mechanical composition of the soil declines.

Assessments of plutonium removal from surface soil horizons, averaged over a long period of time, showed no substantial dependence on the source of contamination (Table 5). Maximum removal was observed from litter on forest soils and from turf on peaty soils. More intensive removal of Chernobyl plutonium was most probably linked to the formation of soluble compounds with natural organic ligands through the decomposition of contaminated litter and turf. The depths at which thousandths of 1% of plutonium may be observed in different types of soil are estimated to be 40-80, 50-160 and 100-440 cm after 25, 50 and 100 years respectively.

In addition to studying the influence of landscape and geochemical conditions in order to understand the processes taking place in the soil and vegetation cover which are responsible for the processes governing migration, a great deal of attention was paid to the forms of occurrence of plutonium in soils and to ascertaining its possible links with components of organic and inorganic soil components. Where it enters the soil in the form of soluble compounds, for example nitrates, it is mainly in acid-soluble and amorphous forms, irrespective of soil type (Fig. 5).

Plutonium content in more mobile forms, though the quantities involved are smaller, depends on the soil type, decreasing from soddy podzolic soil to leached chernozem. As expected, the proportion of mobile forms of Chernobyl plutonium increases, even in carbonate soil, from finely dispersed fuel to volatile emission products. In the case of the latter, the content of plutonium in amorphous form, i.e. potentially mobile, is virtually the same as that from other contamination sources.

The absorption of plutonium by amorphous compounds, covering the mineral fraction with a film, is a very rapid process regardless of soil type and local conditions; more than 90% is absorbed in this way (Table 6). Most of the plutonium (Fig. 6) was found in the two uppermost films in all soils, the films being compounds of natural organic substances with chemical elements and hydroxides of iron and aluminium. The presence of a small amount of plutonium in silicate form is evidence of its slow penetration into the crystal lattice of soil minerals.

In organic soil matter, plutonium is linked both with humic acids and with non-specific organic substances. Plutonium distribution between separate groups of organic matter depends on the physico-chemical properties of soils and, above all, on the qualitative and quantitative composition (translator's note: text becomes unreadable here)

Plutonium is attached to all groups of organic soil matter (Fig. 7), its maximum content being in the humic acids, where it correlates with the amount of humus in the soil.

Within the groups of humic acids (Table 7) most of the plutonium is found in the humates of calcium, and within the group of fulvic and low-molecular acids most of the plutonium is found in free acids and their compounds with mobile hydroxides of iron and aluminium on the one hand and fulvates and low-mobility hydroxides on the other.

Data obtained on the forms of occurence of plutonium have made it possible to explain the mechanisms governing its behaviour in soils.

For example, increased vertical migration of plutonium in soils and in the soil and plant system in the following order:
leached chernozem < grey forest < soddy podzolic
correlates with the increased quantity of the most mobile fulvic and low-molecular acids and their complex compounds with chemical elements; it also increases with the formation of soluble carbonates. Secondary accumulation of plutonium in humic, illuvial and silty horizons is caused by its presence in the structure of low-solubility humic acids and humates, primarily of typomorphic elements of the forest and forest-steppe zones investigated (of calcium or iron) and hydroxides of iron and aluminium.

The basic form of vertical plutonium migration in soils is movement with finely dispersed particles washed out of soils by atmospheric precipitation; in these particles it is found mainly as a part of the low-solubility compounds mentioned. In the area around the Chernobyl NPP the part of the plutonium which was deposited with the finely dispersed fuel particles may migrate in the form of oxides and other compounds with low solubility.

In conclusion, the behaviour of "Urals" and "Chernobyl" plutonium in soils in the form of volatile fallout products with which a large area of the European part of the country is contaminated, is non-specific. Forms of uptake into the surface of the earth and into soils are the factors determining its fate, similar to other sources of uptake and to other fission products[2, 4, 6, 7, 9-14, 18, 21], including the state of dispersion and the actual landscape and geochemical conditions it meets. Forecasts of plutonium distribution in soils made in the first days following the Chernobyl accident on the basis of already-revealed mechanisms governing its migration have proved largely correct. The specific behaviour of plutonium fallout in the form of finely dispersed fuel and "hot" particles may in the final analysis only show up in the intensity of its migration in surface ecosystems.

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Table 1. $^{239,240}Pu$ content in soils

Accident zone		Background areas	
Soil	Bq/m^2	Soil	Bq/m^2
~ 8 km from the source near the trail axis		Chelyabinsk district in various directions from the accident site	
Leached chernozem, meadow	10566	Leached chernozem, meadow	411
Grey forest, birch forest of park type	12724	Ditto	138
		Ditto	114
~ 4 km from the source near the trail boundary		Dark grey forest, meadow	105
		- , meadow	74
Leached chernozem, birch forest	665	Sverdlovsk district	
Ditto, skrit	523	Brown forest, glade	118-142
Between the axis and the trail boundary		Ditto, forest	117-128
		Other countries [1]	
Meadow chernozem, dry meadow	1292	Italy	159-537
		Austria	44-566
		Japan	55-131
		USA	63-192

1 - quotation from [4]

Table 2. $^{239,240}Pu$ concentration ratios

Southern Urals'	Byeloyarsk NPP	Other regions [6-8]
10^{-4} - 10^{-3}	10^{-5} - 10^{-4}	10^{-6} - 10^{-4}

Table 3. Correlation coefficients

Nuclides	Soils (n > 100)		Aerosols (n = 9)	Fallout (n = 10)
	pollution type		Various pollution types	Various pollution types
	A	B, C		
$^{239,240}Pu$ - ^{144}Ce	0,71	0,36	0.75	0,71
$^{239,240}Pu$ - ^{103}Ru	0,71	0.88	0.65	0.81

Table 4. $^{239,240}Pu$ migration coefficients in soils, slow (M_1) and rapid (M_2) transfer, 10^{-8} cm^2/s

Type of pollution	M_1	Share ,%	M_2	Share ,%
Southern Urals' accident	1,2 0,4-2	88 76-96	21 2-40	12 4-24
Technogenic, USA[1]	8 7-9	86 65-94	15 11-19	14 6-35
Chernobyl accident (type B)	4,7 0,6-3,4	-	68 26-103	-
The same (type C)	0,8	-	57	-
Global fallout[2]	1,3	77	14	23

Comments: 1 - Calculated from [14]

2 - Calculated from [15]; M_1+M_2 values for the Soviet Union's soils were estimated to be $(0,6-21).10^{-8}$ cm^2/s

Table 5. $^{239,240}Pu$ removal from the upper horizons of various soil types, %/ year

Horizon, depth, cm	Global fallout (Δ t > 20 years)	Accident in the Southern Urals (Δ t=24-26 years)	Byeloyarsk NPP (Δ t ≥ 20 year)	Accident at Chernobyl NPP (Δ t=15 months
Turf, litter, 0-2 (5)	3,3-4,2	2,2-3,8	2,9-3,8	1,4-9
Ditto + humus horizon 0-5 (8)	2,3-2,9	0,9-2,2	2,3-3,4	6

Table 6. Plutonium distribution between amorphous and silicate compounds in soils, %

Soil	Time and conditions of occurence in soil	Amorphous compounds, soluble in:		Silicate compounds
		0,1 N NaOH+ 0,05 N $Na_2C_2O_4$; Tamm's solution	Tamm's solution	
Sod-pod-zolic	5 days-3 years; laboratory experiments	97.6±0,7	85.7±4.7	0.8±0.3
Grey forest	5 days-4 years; ditto	96.0±0.6	85.0±4.9	1.0±0.02
Leached chernozem	17 hours-3 years; ditto	98.2±0.8	88.3±4,5	1.0±0.3
Ditto	Experimental area; 10 years	97.0±1.7	82.3±5.2	0.6±0.4
Silty loam [19]	global fallout	91.0±8.9	82.0±7.9	9.0±0.8

Table 7. Plutonium distribution in humic acids, %

Group of compounds	Humic acids	Fulvic acids
Free acids and their compounds with mobile iron and aluminum hydroxides	-	$\frac{0.3\text{-}11}{3.4\text{-}51}$ [1) 2)]
Humates and fulvates of calcium	$\frac{14\text{-}55}{72\text{-}87}$	Not found
Humates, fulvates and mobile hydroxides of iron and aluminium	$\frac{2\text{-}11}{4\text{-}16}$	$\frac{2\text{-}5}{16\text{-}24}$
Humates, fulvates and slightly mobile hydroxides of iron and aluminium	$\frac{0.6\text{-}5}{4\text{-}18}$	$\frac{3\text{-}6}{27\text{-}60}$

Comments: 1 - % of the content in soil;

2 - % of the content in humic acids.

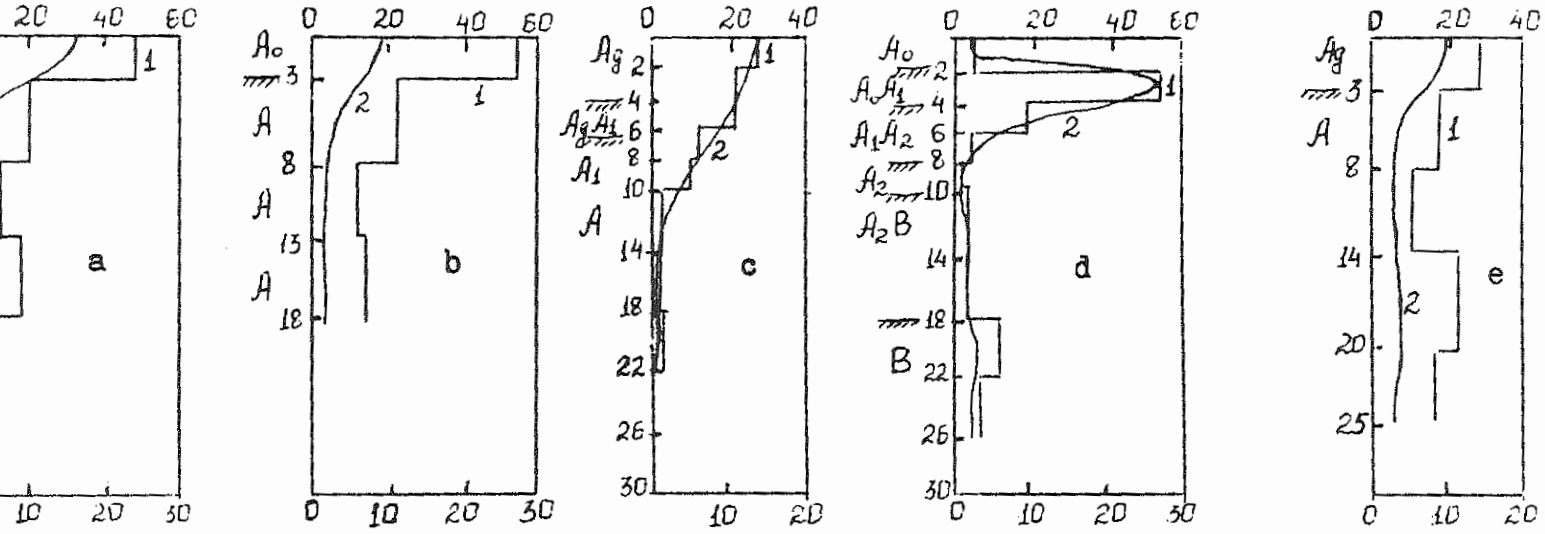
THE LIST OF FIGURES

and leached chernozem (laboratory experiments); 4 - leached chernozem (experiment plot on the Southern Urals' trail); 5 - different soil types (Chernobyl accident, finely dispersed fuel of unaffected composition); 6 - ditto (finely dispersed fuel enriched with volatile products); 7 - ditto (volatile products); 8 - carbonate soil (volatile products); ■ - water-soluble; ▨ - exchangeable and easily soluble; ▤ - mobile; ▦ - acid-soluble; □ - amorphous

Fig. 6. Plutonium distribution between organic and inorganic soil components, % of content in soil [19]; A, B and C - sod-podzolic, grey forest and leached chernozem (laboratory experiments); D - leached chernozem (experimental plot); ▥ - free humic acids and their compounds with Ca, Fe, Al etc.; ▩ - Fe and Al hydroxides; ▧ - organic substances released from bonds with hydroxides; ▤ - organic substances connected with clay minerals; □ - mineral fraction; in [1] [2] [3] - content in Fe and Al hydroxides (1), amorphous form (2) and organic substances and their compounds with chemical elements (3) considerable.

Fig. 7. Plutonium distribution between components of organic matter, % of content in soil: 1 and 2 - leached chernozem (laboratory experiments and experimental plot); 3 and 4 - grey forest and sod-podzolic; ■, ▨ and ▤ - soluble in 0,05, 0,25 and 0,5 M H_2SO_4 respectively; ▩ - fulvic acids; ▧ - humic acids; ▥ - unhydrolizable residue (humines).

^{239,240}Pu content in each layer, % (1)



^{239,240}Pu content, %/cm (2)

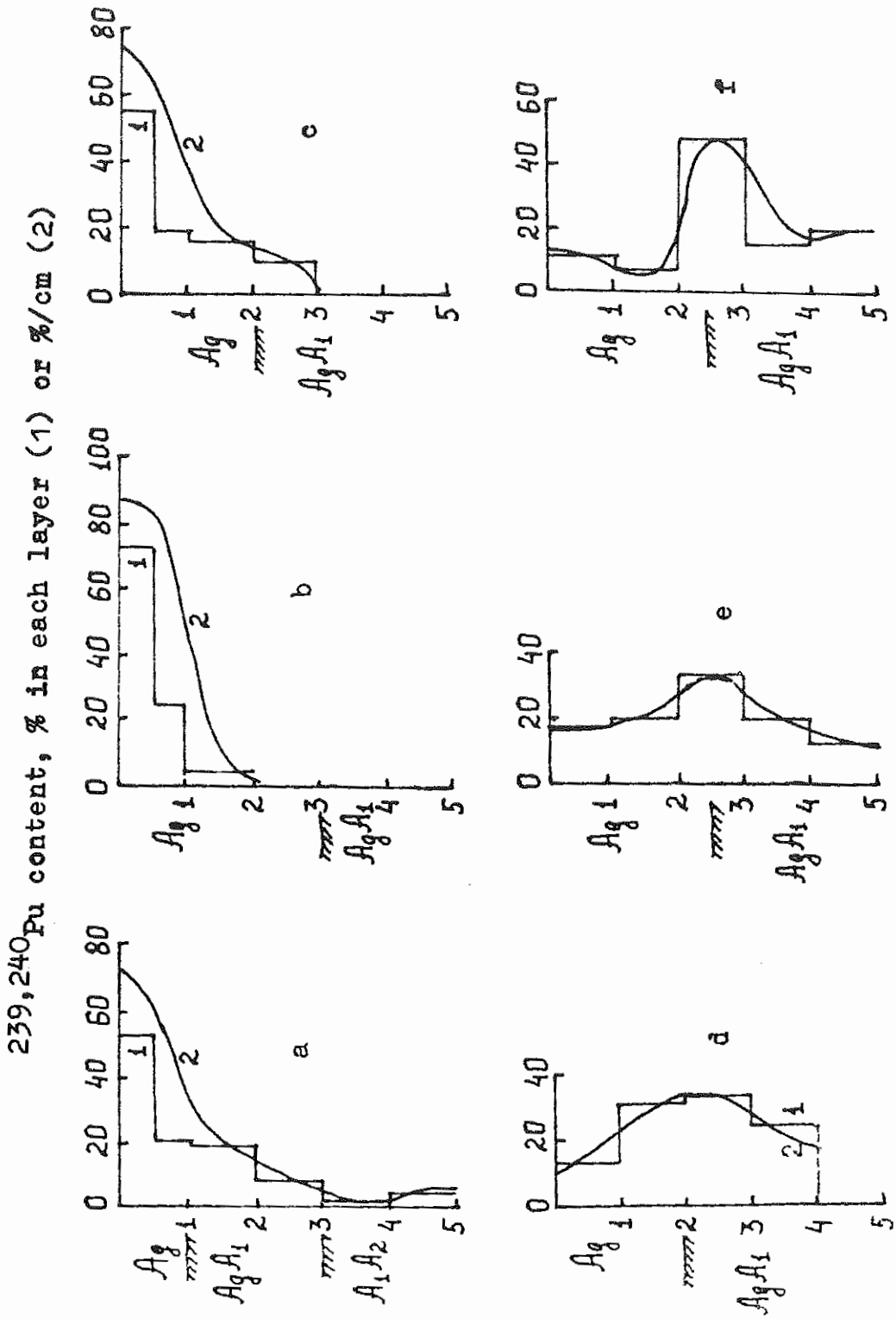

239,240Pu content, % in each layer (1) or %/cm (2)
Horizon, depth, cm
a
b
c
d
e
f
Ag
AgA1
A1A2
A2

Fig. 2

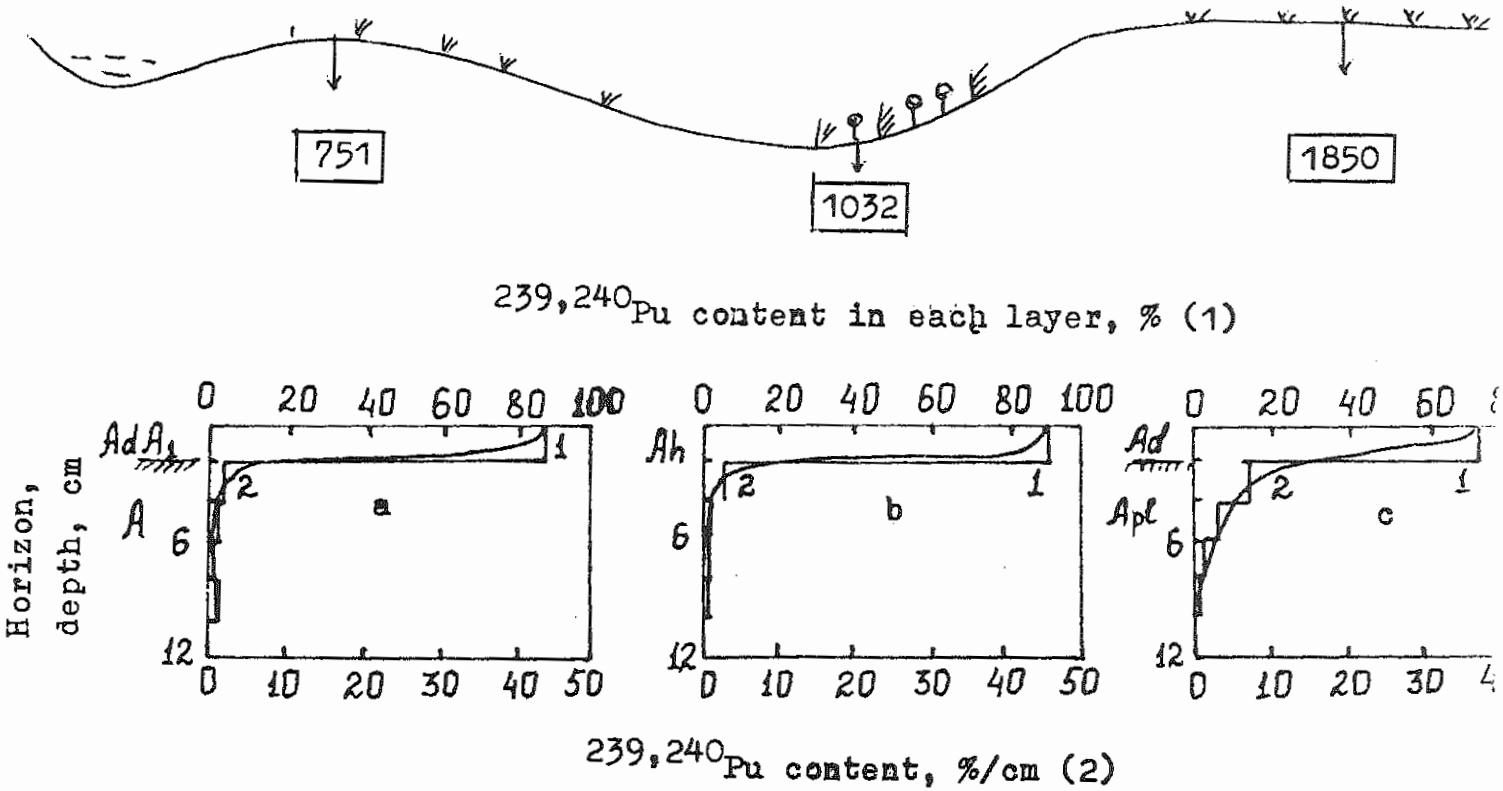

751
1032
1850
239,240Pu content in each layer, % (1)
Horizon, depth, cm
AdA
A
Ah
Ad
Apl
a
b
c
0 20 40 60 80 100
0 10 20 30 40 50
6
12
239,240Pu content, %/cm (2)

Fig. 3

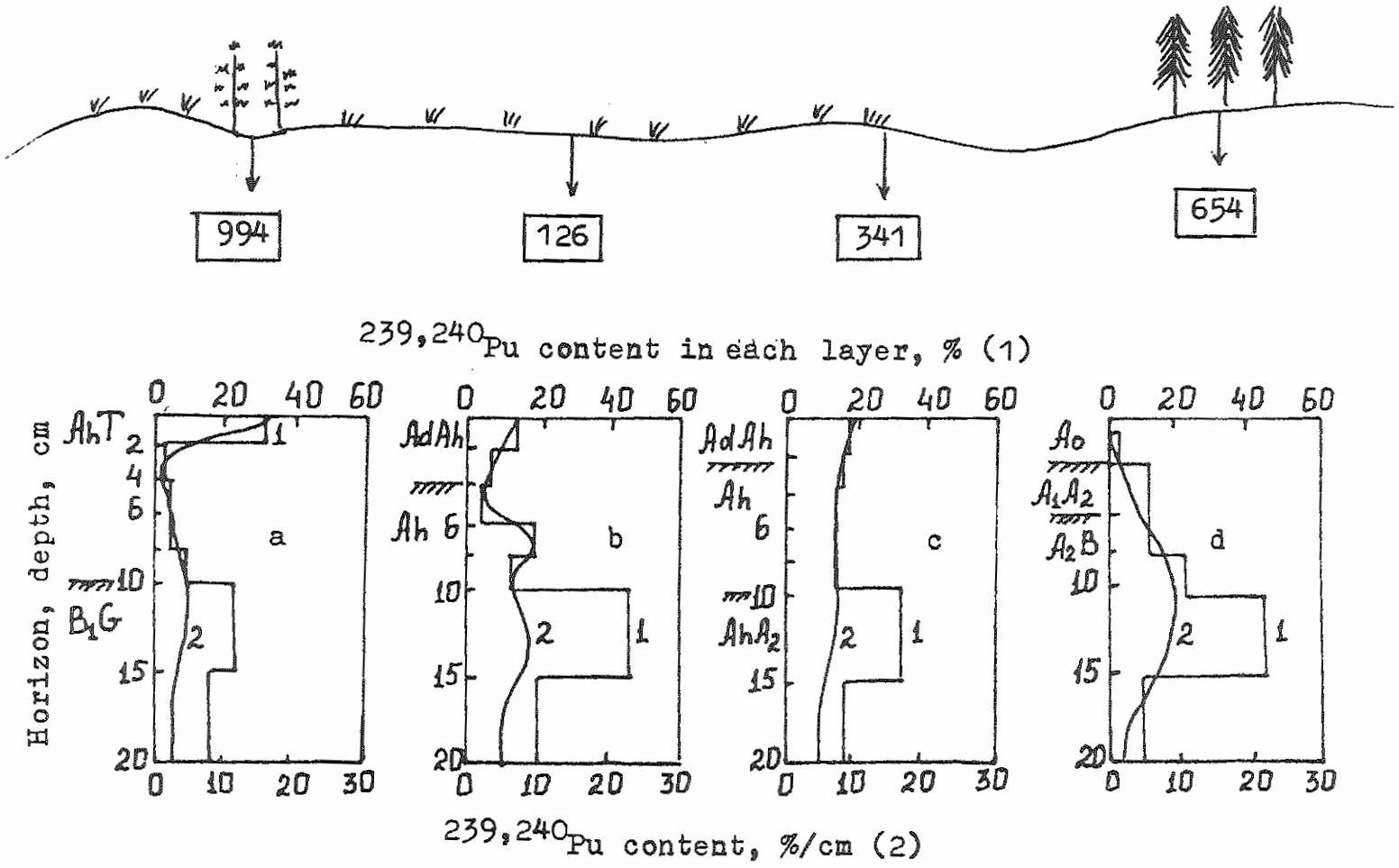

994
126
341
654
239,240Pu content in each layer, % (1)
Horizon, depth, cm
AhT
B1G
AdAh
Ah
AhA2
Ao
A1A2
A2B
a
b
c
d
0 20 40 60
0 10 20 30
2
4
6
10
15
20
1
2
239,240Pu content, %/cm (2)

Fig. 4

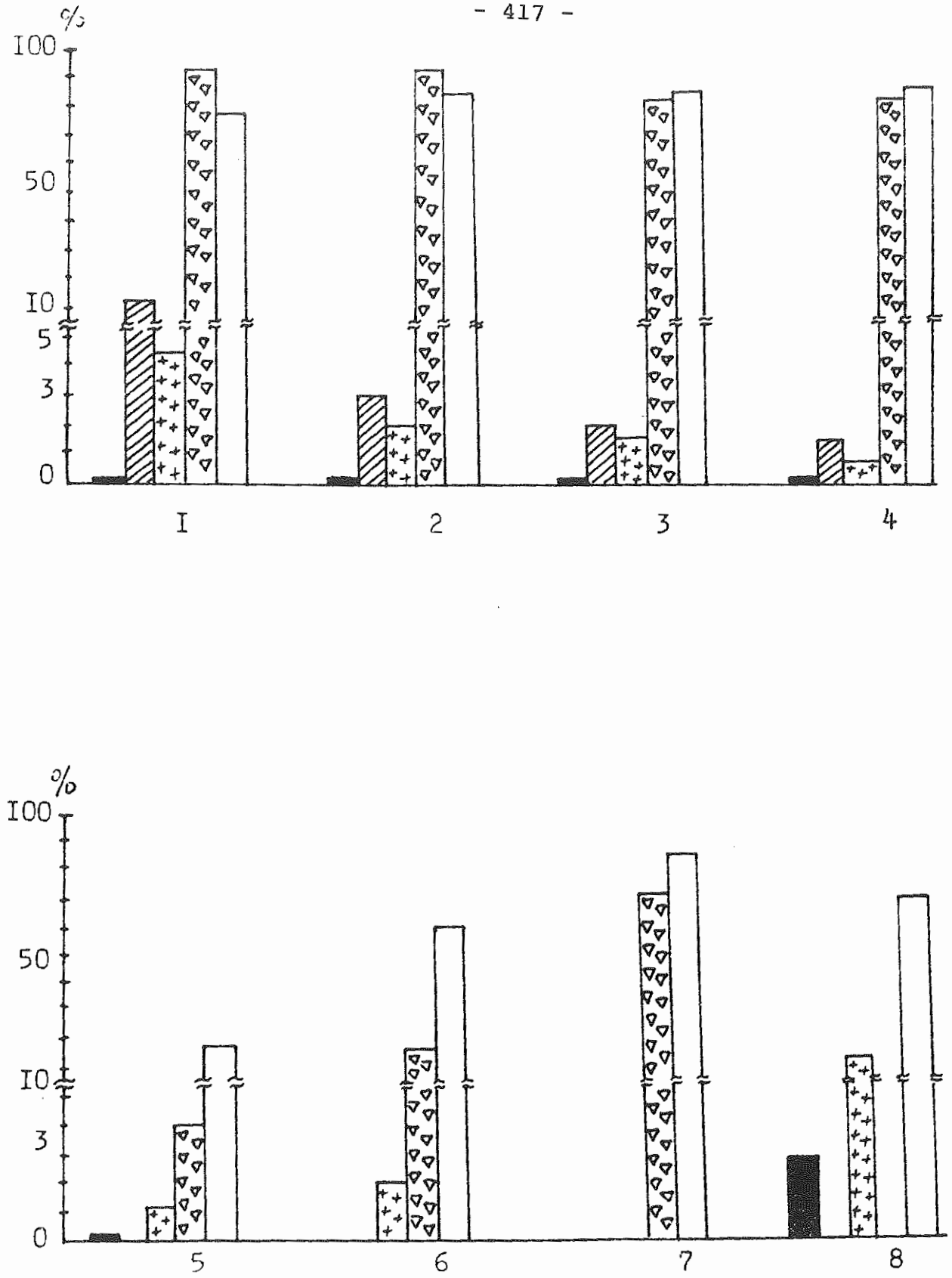


Fig. 5

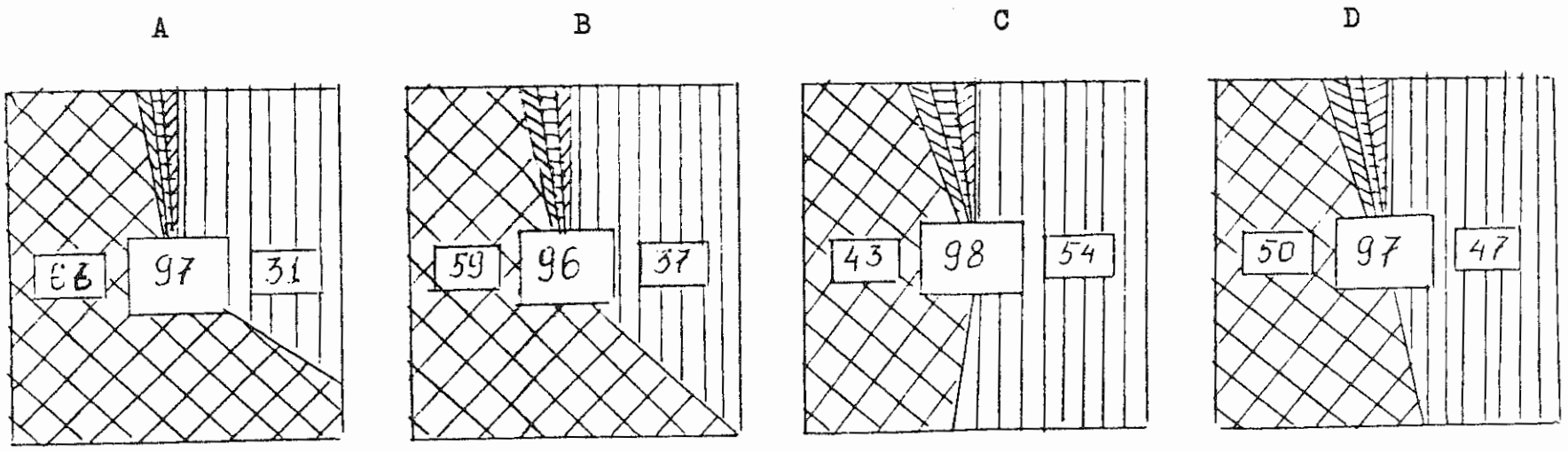


Fig. 6

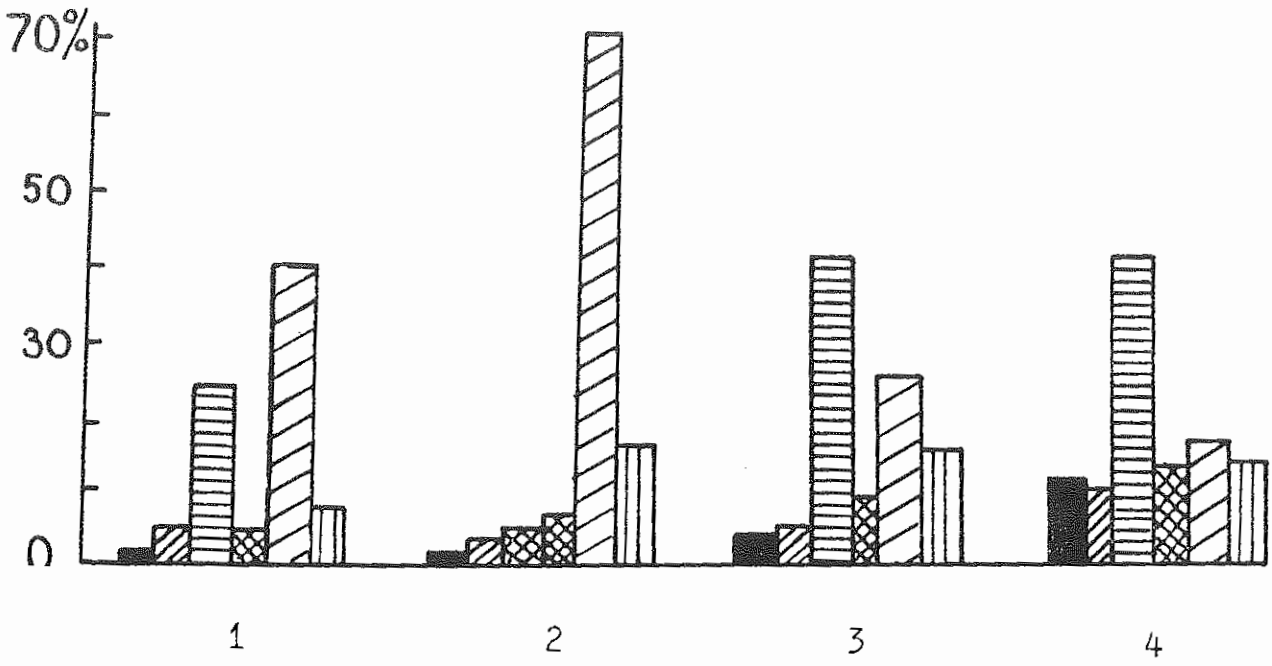


Fig. 7

Peculiarities of Sr-90 Migration in the Environment

G.N. ROMANOV, D.A. STUKIN, R.M. ALEKSAKHIN

ABSTRACT

The Eastern Urals radioactive track which formed as a result of the Kyshtym accident constituted a natural experimental base for studying the dynamics of Sr-90 behaviour and migration in the environment. Sr-90 behaviour in soil depends on the processes involved in its physical migration horizontally (water runoff, wind transport) and vertically (effective diffusion), on the intensity with which the physico-chemical forms of strontium change, and primarily on Sr-90 incorporation into the soil-absorptive complex. Over 30 years the effective diffusion processes led to downward displacement of Sr-90 in undisturbed soil to a depth of 30 to 50 cm. In 1988 84-94% of the Sr-90 was located in the 10 cm layer as against 90% in the 2 cm layer in the initial 1 or 2 years. The amount of Sr-90 in mobile forms in leached chernozem and grey forest soil hardly changed over time, amounting to between 76 and 90%. In the first 5-10 years Sr-90 surface water runoff was about 0.2% (and Sr-90 wind resuspension 0.1-1.0%) per year of the Sr-90 inventory per unit area. Due to Sr-90 loss from the top soil layer the water runoff and wind resuspension processes decreased exponentially, halving every 4 to 5 years.

In most cases, Sr-90 transfer from soil to plants via the roots does not involve discrimination of Sr-90 with regard to calcium, and therefore the Sr-90 accumulation level in various species of natural and agricultural plants depends on their calcium requirements and the amount of exchangeable calcium in the soil. This made it possible to work out quantitative indices for forecasting Sr-90 accumulation in various species of plants for given types of soil.

As Sr-90 moves through the trophic chains (including the agricultural one) we find - depending on whether two adjoining links discriminate against it or not as regards calcium - that some organisms reject it while others concentrate it. When restoring farming on the contaminated territory the fact that cattle discriminated against Sr-90 vis-a-vis calcium was the main reason for opting for meat production instead of grain or fruit production.

The Eastern Urals radioactive track formed after the Kyshtym accident constituted a natural experimental base for studying the dynamics of the behaviour and migration of Sr-90 in the environment.

The contaminated territory is part of the Zauralskaya wooded steppes with the relatively flat relief characteristic of this zone. Two types of plant growth predominate - grassland and woodland and there are a large number of water bodies, mainly in the form of shallow, precipitation-fed lakes.

The soils of the radioactive track are heterogeneous. Grey forest soil, leached chernozem and soddy-podzolic soils, including excessively wet and saline soils, predominate.

In examining the behaviour and migration of radionuclides in the environment and the food chain in the aftermath of the accident, it is essential to distinguish between two phases: the phase of initial dispersal of the radioactive substances in the area or systems under consideration, and the subsequent redistribution of the radionuclides under the influence of natural (including biogenic) and anthropogenic factors.

Immediately after being deposited the radioactive fallout started to redistribute under the influence of natural forces, whose vertical component, where mass transfer predominates, conditioned the subsequent displacement of radioactive material from the tree crowns and herbaceous cover to the soil surface and (in lakes) from water to bed sediment. In the course of time the soil and sediment became the basic accumulators of radioactivity, and the long-term behaviour of the radionuclides in different natural systems reflected the role of these depots in the overall mobility of radionuclides in the environment.

1. Soil cover

Radionuclide behaviour and speed of migration in the soil depend on the soil's physico-chemical properties, which themselves are a function of the type of soil, (absorption capacity, including the quantity of exchangeable calcium, acidity, humus content, silt fractions, sands, soil class, permeability to water, mineral composition), and the way it is wetted.

Each radionuclide's mobility in soil is determined by the water-solubility of the nuclide's chemical compounds and the soil's sorptive capacity in respect of the nuclide. Both these factors determine the intensity of horizontal migration (runoff) and vertical migration of radionuclides in the soil cover.

The fallout radionuclides line up as follows when ranked by increasing mobility in soil: Cs-137< Sr-90< Ce-144< Ru-106. Cs-137 has lowest mobility because of its high irreversible sorption in soil particles, with Cs-137 atoms being incorporated into the crystalline lattice of clay minerals. The content of mobile Sr-90 - i.e. forms that are water-soluble and displaced by ammonium acetate - remains virtually unchanged over time and accounts for 76% to 90% of total Sr-90 in the surface layer. The mobility of Sr-90 and Cs-137 in the soils of the track increases from soddy-podzolic soils through grey forest soils to leached chernozem, in line with differences in the intensity of soil formation processes in these types of soil.

Vertical downward migration of Sr-90 and Cs-137 in undisturbed soils is determined by a combination of diffusion (displacement together with moisture in the soil) and mass transfer (mechanical displacement together with soil particles). The linear speed of total downward migration of Sr-90 in the undisturbed 15-cm soil layer lies between 0.2 and 0.4 cm/year. For Cs-137 the range is 0.15 to 0.3 cm/year.

The past 30 years have seen a redistribution of radionuclides in the undisturbed soil profile, the main feature being Sr-90 and Cs-137 enrichment of the 2-10 cm soil layer (Figure 1). Table 1 shows the distribution of radionuclides in the profile of characteristic soil types 30 years after the accident. While in the first one to two years after the accident up to 90% was concentrated in the 0-2 cm layer, at the present time the surface layers are to all intents and purposes free of radioactivity. In leached chernozem half the Sr-90 is removed in 5 years from the 0-2 cm layer and in 19 years for the 0-5 cm layer; in the case of soddy-podzolic soils, the corresponding periods are 2 and 7 years.

Table 1

Sr-90 and Cs-137 in soil layers (1988) - % of total

Type of soil	STRONTIUM-90			CAESIUM-137		
	0-2 cm	0-10 cm	10-50 cm	0-2 cm	0-10 cm	10-50 cm
Chernozem	31	94	6	41	93	7
Grey forest soil	11	84	16	51	89	11
Soddy-podzolic	8	54	46	44	84	16

Migration processes differ considerably for ploughed fields, swamps and soils of lakeside meadows waterlogged by groundwater. In ploughed soils annual ploughing mingles the radionuclides deposited on the soil surface, so that after 30 years there is uniform distribution of radionuclides throughout the ploughed soil layer. After 30 years only a few hundredths of a percent of the radionuclides in the soil had migrated below the ploughed layer.

In swampy (peaty) soils Sr-90 and Cs-137 migrate through the soil profile very rapidly. After just six years, 40% of the total radionuclide inventory had migrated more than 10 cm below the surface, while after 30 years the radionuclides are uniformly distributed throughout the entire peat layer (up to 1-1.5m deep). In waterlogged soils evaporation inhibits virtually all vertical migration and all the radionuclides remain in the surface layer of the soil (0-2cm).*

2. Migration of radioactive material through water and wind erosion.

Water runoff and wind deflation are the predominant natural processes affecting migration of radioactive material over the earth's crust.

Water runoff - which is low because of the flatness of the terrain and the moderate amount of precipitation - accumulates in undrained lowlands, lake basins and a small number of minor rivers. We distinguish between surface runoff, subsoil runoff and channel runoff.

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* predominance of evaporation over precipitation

Surface runoff, which is most common in spring, when up to 70-80% of annual runoff occurs, leads to the runoff of radioactive material in its dissolved (ionic) and solid states. Among the factors influencing water runoff, the surface runoff of radioactive material depends on the quantity of radionuclides in the soil's surface layer and their water solubility. To express surface runoff of radioactive material in quantitative terms, use is made of a runoff coefficient indicating the proportion of activity in the catchment area which is removed by runoff annually.

In the five to ten years immediately following the accident the mean value of the runoff coefficient for Sr-90 was approximately 0.2%/year, while subsequently, due to binding of the radioactive material in the soil and downward migration, there was an exponential decrease in runoff with a halving approximately every 4 to 5 years. Currently, the Sr-90 runoff coefficient is estimated to be approximately 0.05%/year. Runoff of Sr-90 in its solid state makes up approximately 10-60% of aggregate Sr-90 surface runoff. Runoff varies with soil surface, being lowest in wooded areas (.001%/year) and highest in meadows and ploughed land (0.3%/year).

Subsoil runoff, which is characteristic of raised, drained and non-wooded watersheds and is mainly influenced by the infiltration of precipitation, plays a secondary role in the affected territory. The subsoil runoff coefficient for Sr-90 is reckoned to be one order of magnitude less than that of surface runoff. The low specific activity of the investigated groundwater and underground waters in the contaminated area substantiate this conclusion: in the early 1960s the concentration of Sr-90 in groundwater lay between 4 x 10-12 and 2 x 10^{-10} Ci/litre.

Channel runoff of radioactive material via permanent watercourses as an integral characteristic of surface and subsoil runoff depends on the location of the water catchment basin. Channel runoff in the affected territory has much the same dynamics as surface runoff, decreasing from 0.3%/year in the first decade to 0.05-0.1%/year at present. Runoff from marshes into rivers is more intensive than surface runoff.

In the 1960s the radionuclide runoff coefficient was approximately 4% per annum, because of the increased content of ionic forms of radionuclides in the marsh waters.

The total quantity of activity removed through runoff is estimated at 500 Ci, of which 60% was removed during the first five years.

Wind resuspension of radioactive material located on the surface (soil, vegetation, other elements of the landscape) and its subsequent transport and secondary deposition downwind are the key features of wind migration, whose role is particularly pronounced in the initial fallout phase, when the radioactive material is concentrated on the surface in weakly bound form.

The intensity of wind resuspension A, which is determined by the ratio between the intensity of the vertical flow of radioactive material (or intensity of dust creation), Ci/(m2.sec) and surface radioactive contamination at the site, Ci/km2, in general characterizes the proportion of the inventory of activity per unit area raised into the air during a unit of time.

Just after the accident the intensity of wind resuspension A was estimated at $10^{-9}.sec^{-1}$, while subsequently it decreased exponentially, with a halving every four years. The maximum value established for A for the spring, summer and autumn periods is $10^{-11}.sec^{-1}$.

Overall annual wind resuspension of radioactive material is estimated at 10^{-3} to 10^{-2} of total activity per unit area for the first year and 10^{-5} to 10^{-4} thereafter. In all, during the first three years some 2% of the radioactive material per unit area was removed by the wind.

Water and wind migration of the radioactive material did not decontaminate the territory, alter the macro-structure of contamination in the area, or change the axis and boundaries of the radioactive track. The combined contribution of these factors to the redistribution of activity is put at 1-2% of the inventory of material in the area in the initial period, and at fractions of one percent during the entire subsequent period. Changes in the contamination microstructure under the influence of water and wind migration, in particular the accumulation of radioactive material at certain localities, were in large measure offset by radioactive decay of the fallout, which occurred at the rate of 40-65% per annum from 1957 to 1960 and 2% per annum thereafter.

3. Plant cover

The behaviour of radionuclides in plant cover, being subordinated to the processes involved in the biogeochemical cycle of mineral elements, depends on the ratio between the intensity with which such substances migrate from plants to soil and that with which they migrate in the reverse direction. Initially (during the first one to three years) residual surface contamination and exorhizic contamination caused the downward flow of radioactive material to prevail under the impact of leaf fall, the death of plants and their organs, and dispersion through wind and rain. However, in the following period root uptake of Sr-90 from the soil created a fairly stable ratio between the radionuclide inventories in plants and in the soil.

In pastures, hayfields and fallow land almost all fallout was initially concentrated on the plant surfaces, the resulting concentrations of radionuclide mixtures depending on the amount of plant biomass and the degree of projecting cover. Rainfall during the initial weeks transferred approximately 90% of the activity to the sod, litter and soil under the plant cover, with seasonal plant death also contributing to this phenomenon. In the following growing season ·20-65% of radioactive contamination of herbaceous plants was caused by exorhizic contamination redeposited by the wind, 25-70% by root uptake from the sod and approximately 10% by root uptake from soil. The sod was the main source of root uptake for the first three years, after which - due to its mineralization in conjunction with the reduced role of wind resuspension - root uptake from the soil began to predominate (currently 95%). The maximum concentration of Sr-90 in herbaceous plants was observed five years after fallout - this being the time it took for mobile Sr-90 to reach the nutrition zones of the roots of most plant species with surface root systems. In plant species with deeper roots, Sr-90 concentrations peaked after 8 to 15 years (Figure 2).

The root uptake of Sr-90 by various herbaceous plants depends largely on the calcium requirements of different plant species - calcium being strontium's non-isotopic chemical analogue - and this is why there are substantial differences in Sr-90 accumulation in plants of different species growing on soils of different kinds. Generally, the quantitative index of Sr-90 accumulation in herbaceous plants ranges from 7-30 nCi/kg of dry mass in grey forest soil to 4-15 nCi/kg of dry mass in leached chernozem, assuming an Sr-90 activity of 1 Ci/km^2.

In forest plantations, after initial retention of approximately 80-90% of the radioactive fallout in the tree crowns, downward migration also began immediately after contamination. The process was conditioned by the action of the wind, precipitation, falling leaves (birch) and needles (pine). During the first few months activity in the crowns dropped by 20-40% due to washout and blowoff of retained radioactive substances. Approximately 80% of the activity in the crowns was transferred to the litter with falling leaves in autumn. Subsequently, the basic factors in the redistribution of the radionuclides in the forest biogeocenoses were biogenic migration (root uptake of radionuclides from litter and soil, falling needles, leaves and twigs, herbaceous vegetation), the downward migration of radionuclides to the litter and mineralized part of the soil, and exorhizic contamination (which fell rapidly in the first years) due to wind resuspension.

In the first three years Sr-90 content in forest was determined by residual contamination and exorhizic contamination, and thus concentration values were maximum. During the subsequent 20-25 years Sr-90 uptake in trees was almost exclusively via the roots, gradually decreasing from year to year in leaves and branches, while increasing in wood and bark.

Twenty to thirty years after the accident the concentration of Sr-90 in the forest cenosis, assuming Sr-90 activity of 1 Ci/km2, was as follows (in nCi/kg of dry material): pinewood - 0.3; birchwood - 1.8; pine needles - 2.7; birch leaves - 12.

The distribution of Sr-90 in herbaceous and forest biogeocenoses is shown in Table 2. It can be seen that natural herbaceous and arboreal vegetation do not play much of a role in the redistribution of radioactive strontium in ecosystems. The percentage of Sr-90 in arboreal vegetation is much the same as in herbaceous vegetation, bearing in mind that there is relatively more arboreal biomass.

4. Water bodies

A number of endorheic, steppe-type lakes and three smallish rivers are located within the radioactive track. The basic source of radioactive contamination of surface water bodies was the initial deposition of radioactive material on the water surface, and the subsequent surface runoff of radioactivity from the basin over a period of many years.

Natural cleansing of the water took place reasonably quickly until a dynamic equilibrium in the distribution of radionuclides in the lake hydrocenoses was achieved. The semi-elimination period in water was 120-190 days for Sr-90, 18-110 days for Ru-106 and 1-24 days for Ce-144. Because of the small quantity of biomass in the lakes the further distribution of radionuclides depended almost entirely on the interaction between the water and the bed sediment. The concentration

of radioactive material in the lake waters dropped, with a semi-decrease period of approximately 5-6 years; 30 years on, the concentrations are less than 1/1500 of the initial values. During this period Sr-90 was transferred to silts to a depth of more than 30 cm, most of it being concentrated in the 0-15 cm layer.

In the initial period, radioactive contamination of waterways was determined by the degree of dilution of the total quantity of fallout on the water surface by the mass of flowing water. Subsequently, surface runoff of radioactive material from the basin was the predominant factor. Thus, the maximum seasonal concentration of Sr-90 in the river waters occurred during snowmelt. In the river, whose water source is a swampy depression in the affected area, the peak spring concentration of Sr-90 in the water was considerably less. For over 30 years the dynamics of Sr-90 concentration in river water have been similar to those of lake water.

5. Fauna

The behaviour and migration of radionuclides in different ecosystems and biological systems, including the characteristic quantitative and qualitative diversity of their biological species and trophic levels, are in general subordinate to the processes involved in the biogeochemical cycling of substances in nature.

In flora, the behaviour of Sr-90 and Cs-137 is largely identical to that of their nm-isotopic chemical analogues - calcium and potassium - in the soil-plant system. Fauna, however, discriminate between strontium and calcium on the one hand and caesium and potassium on the other, and thus the levels of retention of Sr-90 and Cs-137 in living organisms, and consequently the intensity of transfer of these radionuclides from one trophic level to another, are determined by more complex mechanisms. As a result, the food chains through which Sr-90 and Cs-137 migrate include both organisms that concentrate radionuclides and organisms that discriminate against them.

The general quantitative measure of the concentration of radionuclides in living organisms is known as the 'accumulation coefficient' (AC), this expresses the ratio of the concentration of a radionuclide in the organism to its concentration in the preceding trophic link (or soil, this being the original depot). The ACs for terrestrial and aquatic organisms in the fallout track are shown in Tables 3 and 4.

The total uptake of radionuclides into the biological cycle of living organisms is quite small because of their low biomass, and is two to three orders of magnitude lower than uptake in plant communities.

The fact that certain links in the biological chains can distinguish Sr-90 from calcium was the reason for concentrating on livestock-keeping when resuming agricultural activity in the contaminated territory. As distinct from plants, which do not distinguish between strontium and calcium in root uptake, the organisms of farm animals distinguish between the two, and farm animals constitute a discriminatory barrier in the migration chain of Sr-90 (soil-plant-animal-livestock produce). The discrimination of Sr-90 in cattle (as a ratio of Sr-90 concentration to that of calcium, or in strontium units) is, for a given quantity of feedstuffs consumption, 0.5 in blood, 0.25 in the bones, 0.4 in muscle and 0.1 in milk. This is why, when reorganizing and establishing agriculture in the affected territory, it was decided to opt for meat production (pork and beef).

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Table 2

Distribution of Sr-90 in herbaceous and forest biogeocenoses, % of inventory by unit area)

Components	Herbaceous		Forest		
	Contribution	Annual release into cycle	Pine contribution	Birch contribution	Annual release into cycle
Grass cover	1.6 – 4.0	0.08 – 0.3			
Dead vegetation	0.06 – 0.3				–0.5
Sod, litter	0.04 – 5.7		3.5 (+ grasses)	6.7 (+ grasses)	
Arboreal storey			0.2	6.7	+0.8
. needles, leaves			0.04	0.4	
. branches and twigs			0.05	1.6	
. bark			0.06	2.7	
. wood			0.06	2.0	
Mineralized part of the soil	38.7 – 98.3		96.3	89.8	

Table 3:

Coefficients of transfer of Sr-90 and Cs-137 in living organisms via the land ecosystem

	Sr-90			Cs-137		
	Coefficient of proportionality with contamination level Ci/kg/Ci/km^2	Accumulation coefficient		Coefficient of proportionality with contamination level Ci/kg/Ci/km^2	Accumulation coefficient	
		in relation to preceding link	in relation to soil		in relation to preceding link	in relation to soil
	2	3	4	5	6	7
INVERTEBRATES					2.4	1
Soil microfauna	3.10^{-9}↓	0.5	0.2			
Worms	3.10^{-9}	0.5	0.2	1.10^{-8}		
Insects - Phytophaga	3.10^{-9}	0.5	0.2	1.10^{-8}		
AMPHIBIA	6.10^{-9}	3	0.45	1.10^{-8}	0.75	0.75
REPTILES	2.10^{-7}	10	150	1.10^{-8}	3.0	0.75
BIRDS						
Plant-eating	3.10^{-7}	45	20	3.10^{-9}	0.5	0.25
Insect-eating	7.10^{-8}	30	5	5.10^{-9}	0.5	0.4
Predatory	7.10-7	3.5	50	1.10^{-10}	0.4	0.1

TABLE 3 (contd.)

	2	3	4	5	6	7
MAMMALS						
Small rodents	2.10^{-7}	40	15	3.10^{-9}	0.5	0.25
Ungulate animals	5.10^{-8}	10	4	3.10^{-9}	0.5	0.25
Insect-eating	4.10^{-8}	20	3	8.10^{-9}	0.85	0.6
small predators	$8/10^{-8}$	0.4	6	4.10^{-9}	1	0.3

NOTES

1. The coefficients are calculated in terms of air-dry matter.
2. In the case of vertebrates, only Sr-90 in the skeleton was taken into account.

Table 4

Coefficients of accumulation of Sr-90 and Cs-137
in living organisms in aquatic and related environments (1)

LIVING ORGANISMS	Sr-90		Cs-137	
	in relation to preceding link	in relation to water	in relation to preceding link	in relation to water
FISH				
Plant-eating	3	450	0.4	130
Mixed diet	3	450	0.4	130
Level I predators	0.6	350	4	540
Level II predators	0.2	70	2	1 200
BIRDS				
Plant-eating	4	3 700	-	-
Fish-eating	0.2	150	0.8	100

1) in terms of air-dry matter; only Sr-90 in the skeleton is taken into account.

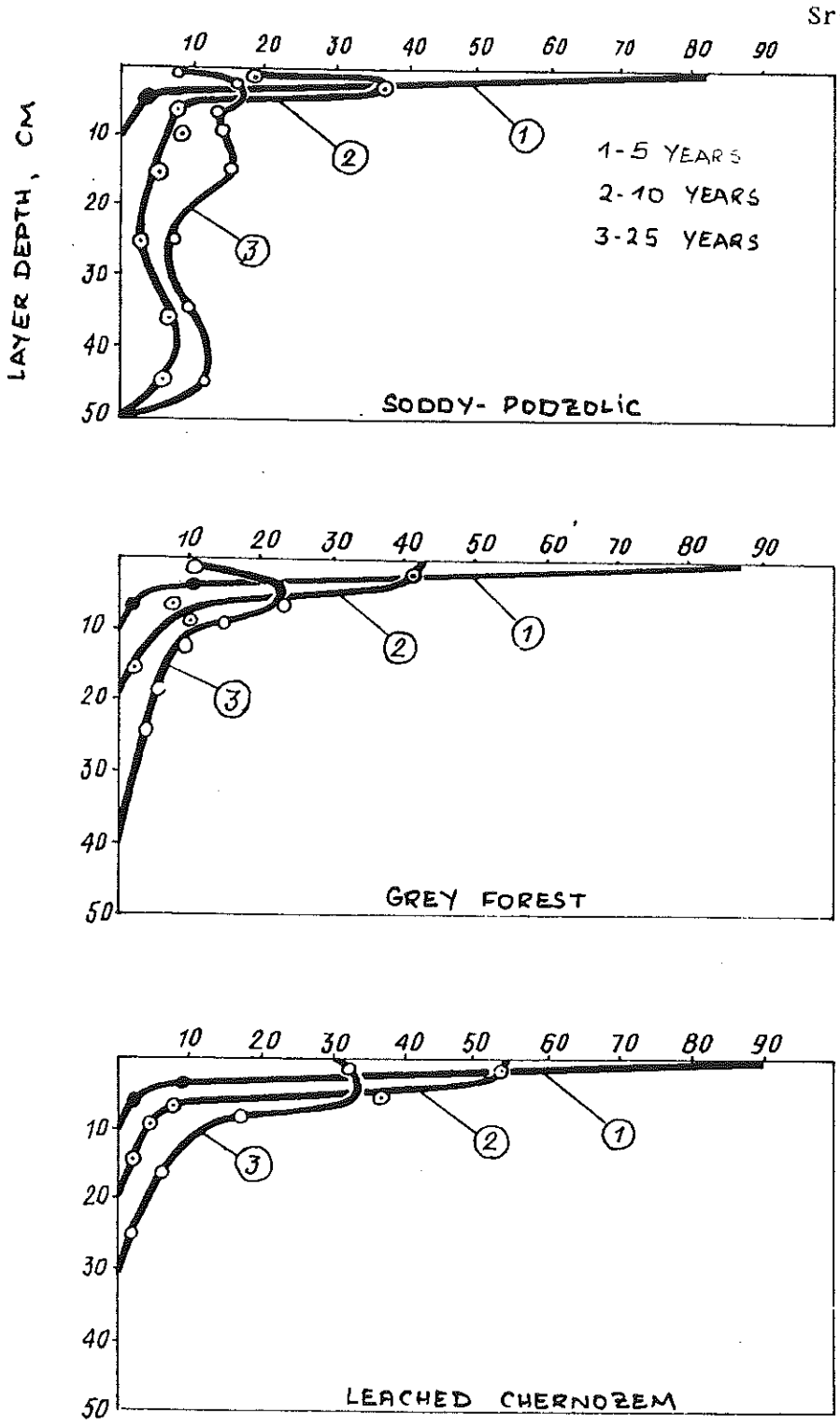

Fig.1: DYNAMICS OF DISTRIBUTION OF RADIOACTIVE SUBSTANCES THROUGH PROFILES OF DIFFERENT SOIL TYPES

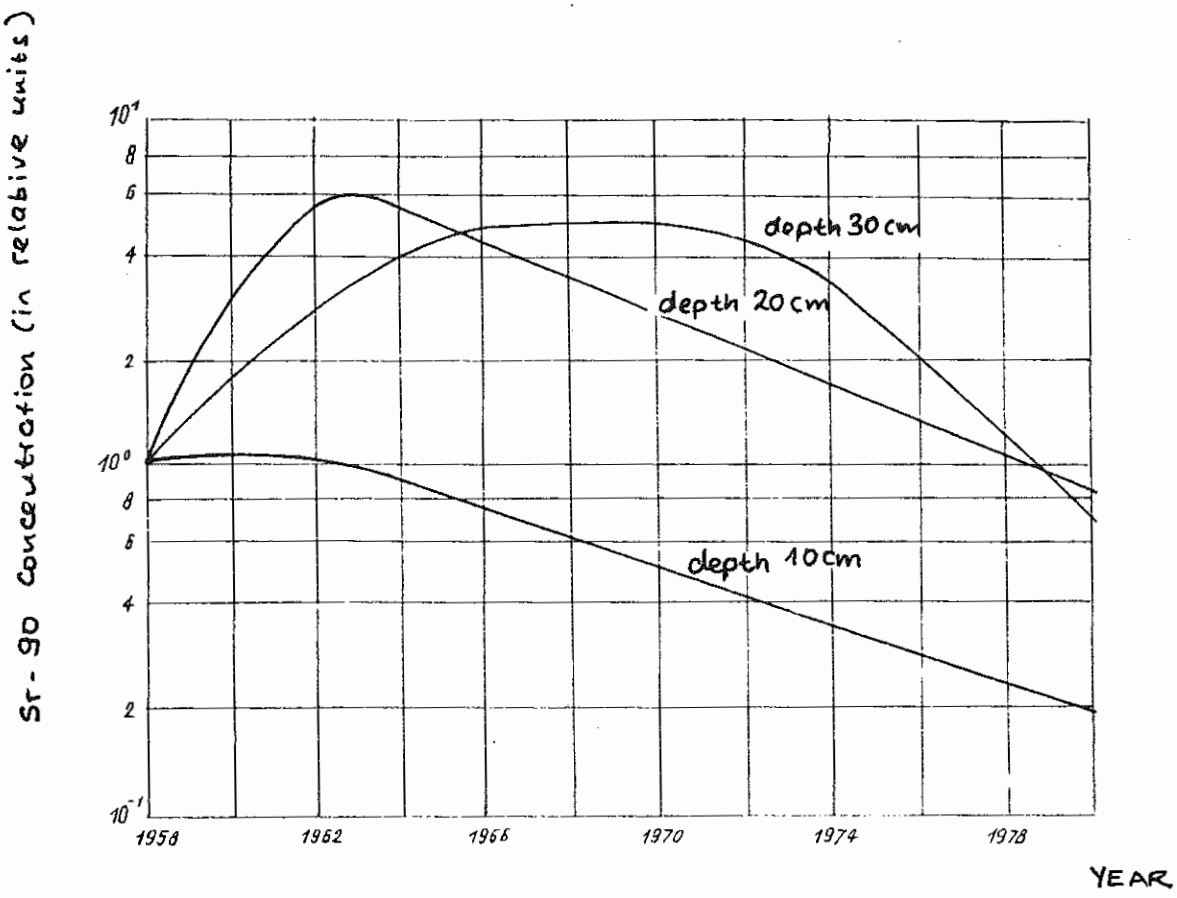

Fig.2: CHANGE IN Sr-90 CONCENTRATION IN HERBACEOUS VEGETATION AS A FUNCTION OF THE DEPTH OF THE ROOT SYSTEMS

Plutonium Release to the Environment During Chernobyl Accident

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INTRODUCTION

About 4,5 years passed since the Chernobyl accident. During this time the true scale of the catastrophe has become more and more clear, the data on the quantity of the radionuclides released, the structure of their environmental distribution and other aspects have been corrected and summarized. At present the following long-lived radionuclides can be considered responsible for the radiation situation in the contaminated areas : Sr-90 ($T_{1/2}$ = 29 years), Cs-137 ($T_{1/2}$ = 30 years), Pu-238 ($T_{1/2}$ = 88 years), Pu-239 ($T_{1/2}$ = $2.4.10^4$ years), Pu-240 ($T_{1/2}$ = $6.6.10^3$ years), Pu-241 ($T_{1/2}$ = 14 years), Am-241 ($T_{1/2}$ = 430 years), Cm-244 ($T_{1/2}$ = 18.5 years).

As has been found, the main feature of the Chernobyl accident is that apart from the release of the volatile and gaseous fission products, the fuel itself, small "hot" particles of dispersed fuel, structural materials and products of their interaction at high temperatures have been released to the environment. The largest longterm impact of the accident will be due to the release of the long-lived highly toxic radifonuclides of transuranium elements (and mainly of plutonium) to the environment. Evaluation of the quantity of plutonium release and the study of plutonium distribution in the environment at the first stage were especially important not only for the evaluation of the radioecological situation, but for the determination of the true scale of the accidental release as well.

1. Plutonium accumulation in the fuel and quantity of the plutonium released to the environment

The 4-th block of Chernobyl NPP was commissioned in December 1983. By the time of the accident it had 715 effective operating days. The burnup of most pat of 190.2 T nuclear fuel (about 75% reached the value of 11-17 MW days/kg (of U). The average burnup value for the core was 10.9 MWD/kg. As a result 660 kg plutonium had accumulated in the core of the reactor by 26.04.1986. (See Table 1).

Table 1

The quantities of some radionuclides accumulated in the fuel of ChNPP-4 by 26.04.1986 (Ref.1)

Radionuclides		Mass, kg	Radioactivity, Bq
Pu	238	1.479	$9.38\ 10^{14}$
	239	412.7	$9.48\ 10^{14}$
	240	176	$1.48\ 10^{15}$
	241	49.11	$1.84\ 10^{17}$
	242	14.16	$2.06\ 10^{12}$
Am	241	1.08	$1.37\ 10^{14}$
	243	0.73	$5.38\ 10^{12}$
Cm	242	0.256	$3.08\ 10^{16}$
	244	0.06	$1.78\ 10^{14}$

Preliminary evaluations of the released fuel fraction conducted in June 1986 at Kurchatov IAE have given the figure of about 3% (Ref.2). Later this figure was corrected and taking into account all the experimental data it was accepted as 3.5±0.5%. It means that the total plutonium isotopes quantity released to the environment reaches about 23 kg.

II. Techniques for the determination of plutonium isotopes in environmental samples

2.1 Radiochemical techniques for plutonium isolation and determination

The representative samples of contaminated soils were taken according to the data of preliminary γ-spectral measurements. After the mineralization for 5 hours at 500-550 C° the samples were homogenized by quartering and 20g fractions were weighed out and directed to the

analyses. Complete dissolution of samples by treatment with HNO_3 -HF- mixture as well as radionuclide leaching by treatment with 0.3 M solution of $KBrO_3$ in 7M HNO_3 at heating were applied. An average leaching efficiency of the last procedure reached the value of 86.5±1.8% at P=95. The routine ionexchange technique used at the very beginning for the plutonium isolation was replaced by express techniques based on the extraction of Pu(IV) by Trioctyl amine (TOA) (Ref.3) or by Trioctylphosphine oxide (TOPO) (Ref.4) depending on the task and instrumental availability. The solvent extraction with 10% TOA solution in oxylene was most widely applied. The flow sheet of the technique is shown in Fig.1. The plutonium isotope contents were determined by the isotope dilution technique with Pu-236 as a tracer. The activity of plutonium isotopes were calculated by using the following equation :

$$A = \frac{A_1}{N3} V (N_1 - N_2) \quad (Bq) \qquad (1)$$

where A is tht total α-activity of the plutonium isotopes (Pu-238+Pu-239+Pu-240) in the sample analysed (Bq); A1 -is the specific α-activity of the tracer solution (Bq/ml);; V - is the volume of the introduced tracer solution (ml); N1 - is the fraction of the total α-activity contributed by Pu-238 + Pu-239 + Pu-240 to the α-spectrum of plutonium obtained from the sample (%); N2 - is the fraction of the α-activity of the tracer, contributed by the heavier plutonium isotopes (Pu-238 and Pu-239) to the α-spectrum of the tracer (%); N3 - is the fraction of the total α-activity contributed by Pu-236 to the α-spectrum of the sample analysed (%).

The total plutonium α-activity in these express techniques can be determined by measuring an aliquot of the extract evaporated on specially prepared spectrometric targets (Ref.3), or the whole extract volume when using a liquid scintillation counter (Ref.4). In the latter case the sensitivity of plutonium determination can be essentially increased by measuring the ß-activity of Pu-241.

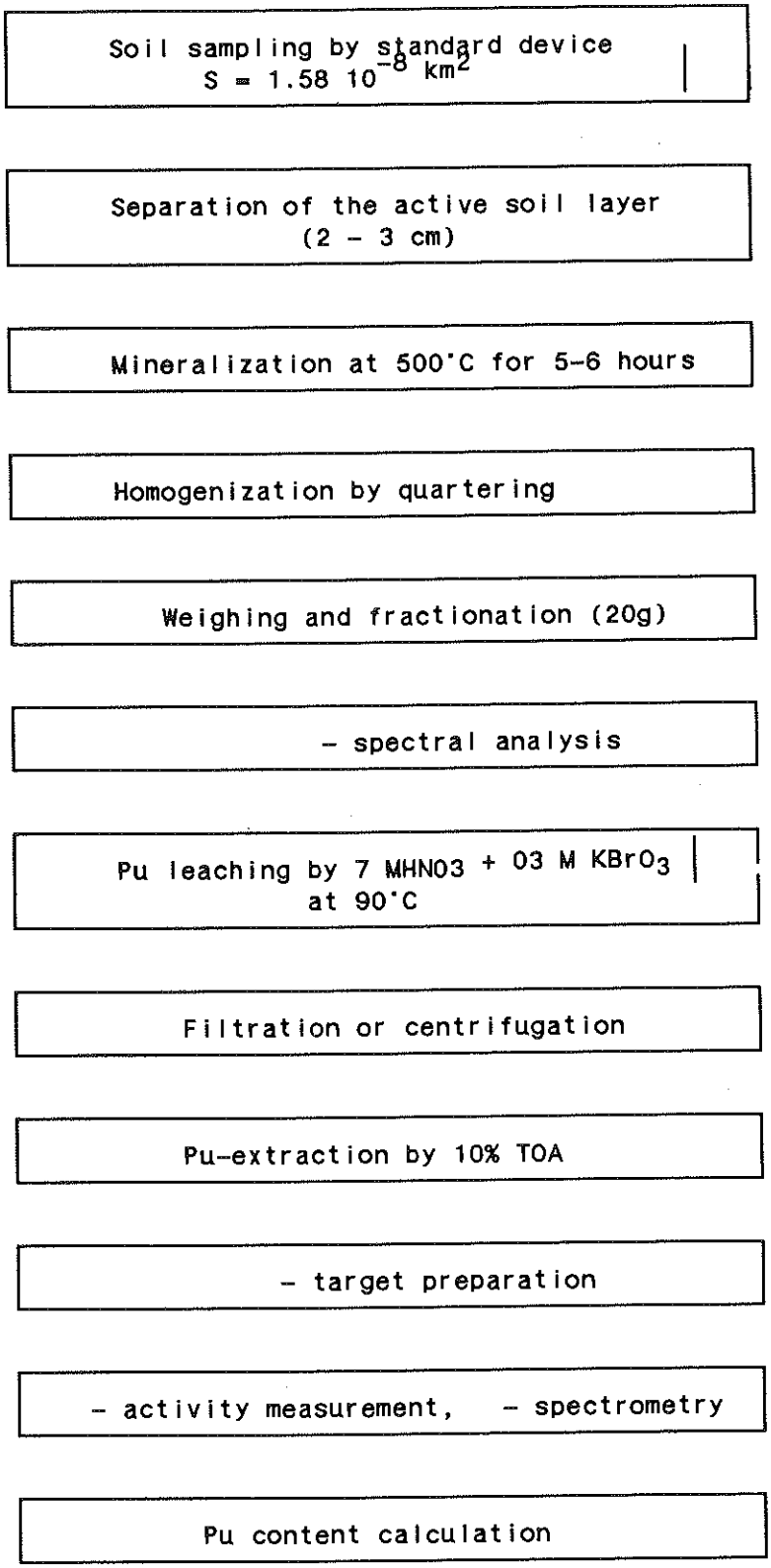

Fig. 1. The flow sheet of an express technique for plutonium determination in soil samples

Table 2

The values of Pu/Ce correlation factor calculated for the time of the accident

NN	Determination time	Sample type	Number of samples	Chemical treatment	$Xcor^{x10^{-4}}$
1.	May 1986	aerosols	2	solution	9.8 ± 1.5
2.	May-Sept. 1986	soil	12	solution	9.3 ± 1.0
3.	April 1987	soil	7	solution	10.1 ± 1.0
4.	April 1988	soil	25	leaching	10.2 ± 0.7
5.	December 1988	"Entombment"	14	solution	9.4 ± 0.8
6.	December 1988	soil	27	leaching	9.7 ± 1.5
7.	April 1989	soil	13	leaching	10.2 ± 1.0
8.	October 1989	soil	22	leaching	9.6 ± 1.0
9.	April-Oct. 1989	bottom sediments	6	leaching	10.1 ± 1.0
10.	October 1989	plants	2	leaching	9.6 ± 1.0
11.	October 1989	organic comp. of soil	1	leaching	7.8 ± 1.5
12.	October 1989	mineral comp. of soil	2	leaching	9.4 ± 0.9
13.	October 1989	heavy fraction	2	leaching	9.4 ± 0.8
14.	April 1990	soil "hot particles"	2	solution	9.4 ± 1.0
15.	April 1990	"Entombment"	6	solution	9.1 ± 0.4
16.	April 1990	peat soil	8	leaching	10.5 ± 1.8
17.	April 1990	sand soil	7	leaching	9.1 ± 1.1.
	Average				9.6 ± 0.6

2.2 Plutonium determination by using γ-spectrometric data

Analyses of the first samples of aerosols and soils taken at contaminated areas immediately after the accident have shown the presence of dispersed fuel particles with a radionuclides composition typicial for RBMC fuel in all samples. Significant deviations from this composition appeared only for gaseous and volatile radionuclides. The bulk of raidonuclides were strongly associated with the UO2 fuel matrix and relatively stable to atmospheric influences. These results, together with calculations of the accumulation of plutonium and fission products, taking into account their diffusion during reactor operation, demonstrated the possiblity of applying the γ-spectrometric data obtained during radiation assessment of contaminated areas for the determination of plutonium. Plutonium isotope activity may be determined by using certain relations between plutonium isotopes ans some γ-emitting radionuclides, which can be used as tracers for RBMC fuel of similar burnup. Zr-95, Nd-145 and Ce-144 can be applied as such tracers. Most of the analyses were accomplished using the following correlation factor between the contents of plutonium isotopes and Ce-144:

$$K_{cor.} = A_{\alpha} (\sum Pu)/A\beta\ (Ce\text{-}144) \qquad (2)$$

The value of this correlation factor averaged for the entire core at the time of the accident was $9.0\ 10^{-4}$. Wide application of this correlation factor the γ-spectrometric measurements of the contaminated areas allowed the data of various institutions to be corrected and compared and the maps of surface plutonium contamiantion of areas to be produced and the entire plutonium release to be quantitatively evaluated.

III. Experimental verification of the Pu/Ce correlation factor stability

Real plutonium content determinations using radiochemical techniques were carried out periodically in parallel with γ-spectrometric measurements of the samples. Such periodical verifications were

important both for elucidation of the possibility of separation of plutonium and cerium during the accident and confirmation of applicability of the correlation factor. It is obvious that the radionuclides released to the environment are gradually involved into the existing natural migration process and their behaviour will be mostly detemrined by physico-chemical properties of the elements which are sufficiently different.

A large number of samples of various kinds of soil taken at contamianted areas, individual ground components, aerosols, bottom sediments, various fuel containing debris from the "Entombment" (Containment of destroyed reactor building), "hot particles" taken in the "Envtombment" and in soil samples have been analysed since April 1986. Some data on the correlation factor determination using the Pu/Ce-144 relation are summarized in Table 2. It is seem from the table that the correlation factor values obtained for the samples taken from the reactor core and from the environment are in a rather good agreement.

IV. An automated system for data treatment and contamination maps drawing

An automated Information System (AIS) "Proba" was developped in 1986 at Kurchatov IAE for the expert evaluation of the information received from Chernobyl and other areas (Ref. 5). It gave us the possibility to accumulate, evaluate and store various actual and digital information on the radiation contamination of the areas associated with certain geographical coordinates by electronic maps. At the present time AIS converted to radioecological monitoring system with the purposes, tasks and possibilities designated to developpe techniques for verification and evaluation of the data reveiced from the areas with various contamination levels (from practically clean to heavy contaminated).

Not only the Chernobyl area, but also some areas in Belorussia, the Ukraine, RSFSR are involved in the assessment sphere of AIS "Proba".

The information-analysis system includes developed calculation and analytical programs utilizing the techniques and concepts of expert evaluation of incoming data and their statistical and model treatment. The system allows to evaluate the quality (representativeness) of samples and measurements, reliability level of the data, to carry out statistical polyparametric analysis and to give the information in form of video and hard copies of coloured maps, where area distribution of plutonium, strontium and caesium is shown with different colours in accordance with the temporary standards for soil radioactive contamination density (established in May 1986). The scales of measurements and the volume of information to be treated to obtain the contamination distribution maps are seen from the following figures. The natural background in the European part of the USSR is 0.01-0.02 mR/h and the total area contaminated up to a level of 0.2 mR/h was about 200.000 km^2 in the first days after the accident.

The first evaluations of the levels of radioactive contamination delineating the most critical spots had been accomplished by the summer 1986. The application of the correlation factor allowed to obtain the maps of surface contamination by plutonium for the large areas around Chernobyl by 1987. These maps were directed to the Government. One of these maps are presented in figure 2.

At present AIS is used for continuous inspection of radionuclides behaviour in the environment (migration mechanism, penetration levels, correlation factors stability). Some prognoses for dose values are prepared on the basis of AIS memory and mathematical models (Monte Carlo calculations) for the areas inspected.

At present radioisotopes of Cs and Sr are the main components of the radioactive contamination outside the 30-km Chernobyl zone and plutonium isotopes - inside the zone. Aero- and ground inspection accomplished in 1988 covering about 350.000 km^2 area did not reveal significant changes in Pu isoline positions in comparison with the past

years. The depth of plutonium contaminated layers averaged over various soils is not more than 5-7 cm (for 4 years). This layers contains about 90% of plutonium.

The results of regressive analysis and the maps of space-time distribution (from Chernobyl around Europe) were also obtained with the help of AIS. We suppose that these data would allow us to describe more precisely the accident process and release characteristics and to test the model of radioactivity transport in the atmosphere.

The AIS programm makes it possible to prepare passports for large areas and to replace the inspection by monitoring.

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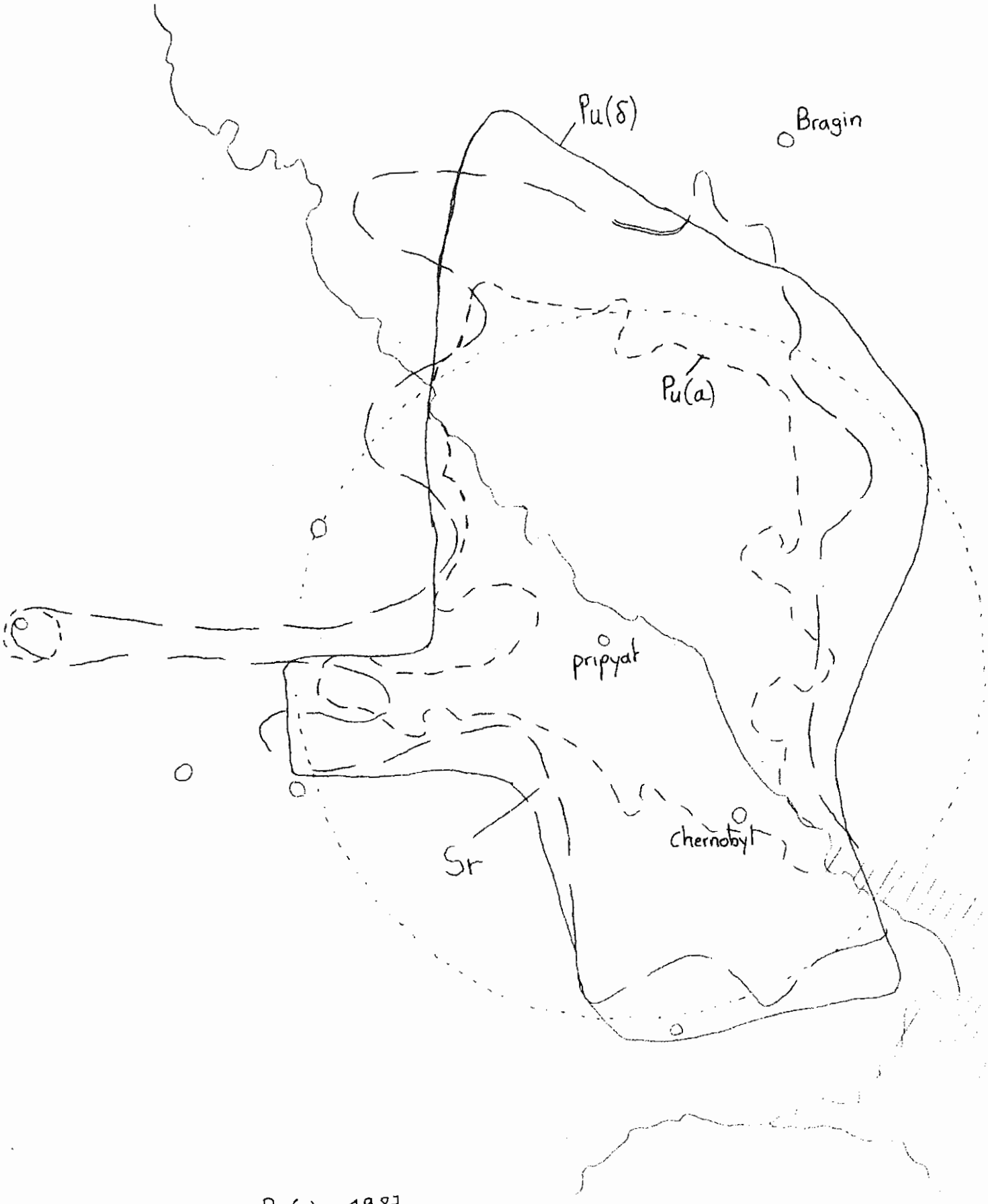

Fig. 2. Pu(a) - 1987
Pu(δ) - 1989

Agricultural Aspects of the Radiation Situation in the Areas Contaminated by the Southern Urals and Chernobyl Accidents

Boris S. PRISTER

ABSTRACT

Being different in nature, the accidents in the Southern Urals and at Chernobyl gave rise to radiation situations with their own specific features, affecting, in particular, agricultural activities in the contaminated area. The main specific features of the Chernobyl accident were the vast scale of contamination, the large contamination gradients even at considerable distances from the accident site, the heterogeneity of radioactive fallout distribution at micro-level, the inconsistent nature of changes in soil contamination levels, and separation of the radionuclides from the fallout.

In spite of the fundamental differences in the chemical character of the types of radioactive fallout, the radionuclides of ^{90}Sr and ^{137}Cs were in both cases readily available for assimilation by plant root systems. In both the Southern Urals and the Ukraine the coefficients of radionuclide build-up in soils with identical agrochemical properties fall within the observation accuracy limits. As a result of the Chernobyl accident, light soils of soddy-podzolic composition were subjected to the greatest contamination, their radionuclide build-up coefficients being 8-15 times higher than those of the chernozem soils in the Southern Urals. An abnormally high level of radiocaesium accumulation was observed in meadow grasses, which explains the leading role of milk contamination in the radiation situation on private holdings.

The greatest danger during the initial period after the Chernobyl NPP accident was the "iodine hazard", which was mainly reflected in ^{131}I contamination of milk, whose consumption by the population accounts for up to 90% of the total intake of this nuclide into the human body. The only really effective way of avoiding high doses to the thyroid gland of humans on a wide scale was to take the precaution (as in the Chernobyl case) of banning milk consumption in the various areas, where the radiation situation was monitored on the basis of the gamma-radiation dose rate.

Prohibitive measures are difficult to maintain for any major length of time, but the iodine hazard period did not last more than 45 days. After this time no ^{131}I was detected in milk, this being due to both radioactive decay and natural cleansing of pasture vegetation. The process of removal of ^{131}I and of other radionuclides in fields is expressed well by an exponential model with two components.

Both in simulation experiments involving plants contaminated with a soluble form of the nuclide in I^- ionic form, and under the actual conditions resulting from the Chernobyl accident, some 70-90% of the nuclide is removed from the vegetative plant mass with a half-loss period of $T_{\delta}^{I} \approx 0.8\text{-}3.0$ days, while some 10-30% is removed with T_{δ}^{2} = 7-14 days, depending on the plants' biological characteristics and age, the type of fallout and the weather conditions. [1] The fraction which fixes more strongly in plants is basically the sole component playing a role in ^{131}I intake by cows, and then by man, and this feature is also observed with respect to other nuclides.

Caesium intake into the human body via food products in the first post-accident year is mainly via the "non-root" contamination pathway. Calculations are based on the assumption that the coefficient of radionuclide retention on pasture vegetation with a biomass of 1 kg/m_2 is 0.2; the caesium content in grass is described by a two-component exponential model with half-periods of natural cleansing of 3 days (70%) and 50 days (30%):

$$q_T(I) = 0{,}2\left[0{,}7\exp(-0{,}231t) + 0{,}3\exp(-0{,}114t)\right]\cdot\delta$$

where δ is a fallout density of 10^{-6} Ci/m_2.

The grazing season lasts 6 months, with haymaking taking place in the last 3 months of this. A cow consumes 50 kg of grass per day, the biological half-life of ^{137}Cs in a cow is 30 days, and the amount of radionuclides in the daily milk yield is 0.13 or 0.013 per litre of milk (given an average daily milk yield of 10 litres). The caesium content in milk before the cows are stabled is calculated by the formula:

$$q_M(t) = 0{,}013\, q_M \cdot \lambda_K \cdot \left[\frac{0{,}7}{\lambda_1 - \lambda_K} \left(e^{-\lambda_K t} - e^{-\lambda_1 t}\right) + \frac{0{,}3}{\lambda_2 - \lambda_K} \left(e^{-\lambda_K t} - e^{-\lambda_2 t}\right) \right]$$

where q_n is the daily intake of ^{137}Cs by a cow, equal to $q_T^0 \cdot 50$ (q_T^0 is the initial contamination of the grass); λ_K is the constant of radionuclide biological half-life in a cow - 0.023 day^{-1}, λ_2 = 0.014, λ_1 = 0.231.

During the time the cows are stabled the caesium concentration in the hay is taken as being a constant equal to the average concentration in the grass during the haymaking period, determined by the formula:

$$\bar{q}_P = \frac{0{,}2}{t_e} \left[\frac{0{,}7}{\lambda_1} \left(1 - e^{-\lambda_1 t_e}\right) + \frac{0{,}3}{\lambda_2} \left(1 - e^{-\lambda_2 t_e}\right) \right] \cdot \sigma$$

where t_e is the period of averaging (haymaking).

The ^{134}Cs content in grass and milk in the first year is calculated on the assumption that the ratio of its concentration in fallout to the concentration of ^{137}Cs is maintained at 1:2 - 1:1.5.

The models described were used to predict the radiation situation in the zone of influence of the Chernobyl accident. [2]

Experimental research into the process involved in non-root contamination of vegetation, carried out in 1986 in the 30-km zone and the environs of Kiev [3], showed that the quantitative parameters of this process (retention coefficients and half-loss periods) came close to the values we had observed in experiments involving contamination of

grocenoses with radionuclides in soluble forms. This demonstrates the high degree of relative biological mobility (RBM) of radionuclides in the composition of the matter contained in Chernobyl fallout. The RBM of ^{137}Cs outside the 30-km zone is 80-100%.

Over-irradiation of the thyroid gland in man was avoided through restrictive measures, primarily stopping the consumption of whole milk with a total volumetric β-activity of over $1 \cdot 10^{-7}$ Ci/l (3 700 Bq/l). In places where such measures were introduced late due to a lack of information on the radiation situation, or were not strictly implemented, the thyroid gland irradiation doses were extremely high in some children.

Banning milk consumption during the iodine hazard period also limited intake of the long-lived nuclides $^{134,137}Cs$ and $^{89,90}Sr$ into man. This is particularly important as regards ^{90}Sr, which - when deposited in the skeleton even following just brief intake - will cause irradiation of the bone marrow till the end of a person's life.

Estimates of the radionuclide composition of fallout on the territory of the Ukrainian SSR - based on data obtained in May/early June 1986 - showed that the radiation hazard outside the 30-km zone stemmed mainly from external gamma-irradiation (gamma-radiation from short- and medium-lived radionuclides) and from internal gamma-irradiation by $^{134,137}Cs$. The $^{137}Cs/^{90}Sr$ concentration ratio in soil varied widely, from 10 for the "western" fallout trail to 1.2 (from 2.0 to 0.5) for the "southern" trail on average. A $^{137}Cs/^{90}Sr$ ratio near 1.0 was usually observed at ^{90}Sr soil contamination levels below 3.0 Ci/km$_2$ (3.0 TBq/km$_2$), which was taken as the limit value. Therefore, the chief factor in the radiation situation after the "iodine period" is ^{137}Cs contamination, which causes both external and internal irradiation of humans.

The main specific feature of the radiation situation in the Polesye (Pripyat Marshes) is the absence of soil contamination gradients displaying the consistent changes in certain directions characteristic of the radioactive trails formed in the Windscale and Southern Urals accidents. Similar studies of soil contamination levels in the Ukrainian part of the Polesye revealed a large degree of heterogeneity in the radiation field pattern; this makes it virtually impossible to

rely on maps plotting contamination levels by means of isolines to predict the radioactive contamination levels of agricultural products from a given piece of land - field, pasture or haymeadow. Fields with a mean contamination level of from 40 to 100 Ci/km$_2$ occur in areas bounded by ^{137}Cs soil contamination isolines of from 5 to 15 Ci/km$_2$, while fields some 50-150 ha large with a mean contamination level of under 15 Ci/km$_2$ have been recorded in areas intersected by 40 Ci/km$_2$ isolines.

The contamination levels of the Republic's soils were rapidly estimated in 1986 by taking representative samples from each administrative area, including 20-25 collective farms. For this purpose, 25 samples of soil were taken from the arable horizon at each farm and carefully averaged. An average sample for each area was prepared from the average samples taken at each farm, and the ^{134,137}Cs concentration in these determined by a gamma-spectrometric method and the ^{90}Sr concentration by a radiochemical method. In this way we were able to carry out a preliminary analysis of the situation in the Republic's agricultural areas as a whole within 10 days, using the facilities of a number of government departments. In 1987 a thorough gamma-survey was carried out of all agricultural lands in some 20 areas where ^{137}Cs contamination exceeded 5 Ci/km$_2$. At the same time, samples were taken to study the radionuclide composition of the contamination. In subsequent years areas with a contamination level of up to 1 Ci/km$_2$ were also gamma-surveyed.

A detailed study of the soils led to those with contamination exceeding 40 Ci/km$_2$ being taken out of use, and to a reduction in produce, primarily milk, with a radionuclide concentration above the set limits.

A forecast of contamination levels in agricultural produce in 1987 was made using previously obtained values for coefficients of soil-to-plant radionuclide transfer (mostly obtained from studies of the Eastern Urals fallout trail). [5] From 1987 on, a study was made of the ^{137}Cs concentration in all the links of the human food-chains and in animal feedstuffs in 31 areas of the Ukrainian Polesye, i.e. in Rovno, Zhitomir, Kiev and Chernigov oblasts (regions). The ^{137}Cs concentration in the soil was measured in averaged samples taken from each field from which plant samples were taken.

Table 1 gives the transfer coefficient (TC) values, averaged for zones with similar agrochemical soil properties, these soils being divided into 3 groups according to their saline extract pH: I - from 4.5 to 5.5, acidic; II - from 5.6 to 6.5, slightly acidic; III - from 6.6 to 7.2, neutral. This parameter best reflects ^{137}Cs uptake by plants, as confirmed by other studies. [6]

As Table 1 shows, for a given contamination level the ^{137}Cs concentration in products from Group I soils will be 2-8 times higher than in those from Group III soils.

Group I includes soddy-podzolic sandy and sandy-loam soils, Group II - light-grey and grey podzolised soils, and Group III - dark-grey and chernozem soils with signs of podzolisation.

Table 2 gives the TC for ^{90}Sr transfer into the main agricultural plants from the soils classified above. The differences in the ^{90}Sr TC values for the soil categories are 2-5 times lower than those for ^{137}Cs.

Overall, radionuclide transfer into plants from the soils of the Polesye reflects the particular properties of the soils: light mechanical composition, low in micaceous and argillaceous minerals, low in humus (0.4-2.1%), and with low absorptive capacity (1-10 mg equiv. per 100 g of soil).

The averaged values for the coefficients of radionuclide transfer from different types of soil into the main types of agricultural produce, observed in the Kyshtym and Chernobyl accident zones, tally with one another quite well despite considerable differences in soil and climatic conditions (Table 3). This confirms the high biological availability of the ^{90}Sr in the Chernobyl fallout and, in our opinion, is of key significance, since it means we can use the large amount of previously obtained experimental material on radionuclide behaviour in food chains to predict the consequences of different types of radiation accidents to a degree of accuracy sufficient for planning protective measures, at least in the period immediately after an accident.

One general conclusion to emerge from the voluminous factual material, was that crop-farming produce with a radionuclide concentration below the established limits can be obtained from land in the Ukrainian Polesye with a ^{137}Cs soil contamination level below 40 Ci/km_2. Farming operations have been discontinued on land with higher contamination levels, while forest has been planted on land where contamination exceeds 80 Ci/km_2.

From 1987 to 1989 there was no significant decrease in the rate of ^{137}Cs transfer from soil to plants (Fig. 1). Evidently, contamination of agricultural produce will decrease slowly in future years as a result of radioactive decay and migration processes.

Introduction of special agrotechnical and agrochemical measures reduces ^{137}Cs accumulation in the main agricultural crops harvested by a factor of 1.5-5.0. Most effective in this respect is the application of lime, potassic and phosphoric/potassic fertilisers, while application of organic fertilisers gives the greatest effect.

One radioecological peculiarity of the zone contaminated by the Chernobyl accident is the substantial increase (three-fold in some cases) in ^{137}Cs accumulation in plants upon application of nitrogen fertilisers, usually in the form of acid salts. In the Southern Urals, nitrogen fertiliser caused a much smaller increase in radionuclide concentrations in plants - by 20-30%. In low-humus sandy soils an increase in crop yields is impossible unless nitrogen fertilisers are used, but these should be in the ammonium form as far as possible with an N:P:K ratio of 1:1,5-1,8:2,0.

Intake of ^{137}Cs into man in the Ukrainian Polesye zone is mainly due (70-90%) to consumption of milk and beef, and attention should therefore focus on the conditions under which these foods are produced. An analysis of the structure of livestock rations has shown that contaminated hay accounts for contamination of milk and meat produced in the public sector to the following degrees: 35% in the winter season (cows stabled) and up to 97% in summer (grazing on cultivated pastures). Differences in ration structure lead to substantially

greater (up to 5 times) intake of ^{137}Cs by cattle on private holdings, something also observed in the Southern Urals accident zone.

The complex radiation situation in the Polesye area was further complicated by the fact that in most populated areas the people grow hay and graze cattle on various types of unsuitable land, at forest edges and even deep inside forests. Such places have contamination levels between 1.5 and 8.0 times higher than the adjacent arable land, this being due to air turbulence as the radioactive cloud passed over them, leading to the capture and deposition on the soil of radioactive substances from higher atmospheric layers.

Similar mechanisms played a role in formation of the Eastern Urals fallout trail.

This was one of the reasons why cows produced milk with a ^{137}Cs concentration substantially exceeding the established limit of 370 Bq/kg, even on contamination isolines very much lower than the 15 Ci/km_2 adopted as the limit level for soil contamination.

The main problem encountered in ensuring radiation safety stems from production of milk on private holdings, which obtain coarse feeds from natural lands. Research in the Southern Urals [7] has shown that on peaty soils ^{137}Cs accumulation by plants can be 30 times higher than on soddy-podzolic sandy-loam soil.

Peat-bog soils are widely found throughout the Ukrainian Polesye zone. For example, in the northern areas of Rovno oblast which suffered the worst contamination, peaty soils make up some 30% of the agricultural land. Many of the terraces above the floodplain and undrained depressions in the Chernigov, Kiev and Zhitomir oblasts also have peaty and peat-bog soils with a varying degree of gleying, where the coefficients of ^{137}Cs transfer into meadow vegetation are dozens of times higher than those in mineralised soddy-podzolic soils.

In field experiments, improvement of soils by liming, ploughing or rotavating and applying mineral fertilisers reduces ^{137}Cs transfer into plants by up to 10 times, but in actual production conditions this

approach is not as effective, the concentration of the nuclide in grass being reduced by 1.3-3.5 times.

The distribution of ^{137}Cs in biomass by herbage height is non-uniform: in the basal parts of plants (up to a height of 5 cm) it is found in concentrations which are 3-8 times greater than in the top part. Nuclide concentrations substantially greater than those in the basal parts are observed in the mat of dead, but not yet decomposed, plants. Thus, when grazing animals on low-yielding overgrazed pastures the ^{137}Cs concentration in cows' milk may be substantially higher than when grazing them or making hay on high-yielding or cultivated pastures with the same contamination level.

All this, together with the heterogeneous nature of the contamination in the area, explains why milk exceeding the permissible limit was produced on some private holdings on land with a ^{137}Cs contamination level of 2 Ci/km_2 or even lower.

The Chernobyl accident contaminated much vaster expanses of land than did the Windscale and Southern Urals accidents (in which only narrow fallout trails were formed); as a result, private holdings are hindered, and in some populated areas even prevented, from providing coarse feed with a permissible radionuclide concentration through changing the structure of sown areas and by grazing their cattle and growing hay in "clean" areas only.

An important radioecological feature of the zone of intense Chernobyl contamination is that it belongs to a geochemical province in which the amount of most trace elements in the soil is from several to dozens of times lower than the minimum amount required by plants and animals.

Liming soils for the purpose of reducing radionuclide transfer from soil to plants reduces the concentration of trace elements in feedstuffs and food products, which aggravates trace element deficiency in the diet of man and animals and facilitates the spread of plant, animal and human diseases. It is therefore necessary to devise and take measures to increase the amount of trace elements in soils, feedstuffs and food products.

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Table 1: Cs-137 PROPORTIONALITY COEFFICIENTS
(average for 1987-1989)
(Bq/kg natural product) / (kBq/m² soil)

Oblast	KIEV			ZHITOMIR	CHERNIGOV	ROVNO
Soil group Saline pH	I 4.5-5.5	II 5.6-6.5	III 6.5-7.5	I 4.5-5.5	I 4.5-5.5	II 6.5-7.5
Milk	1.9-2.1	0.3	0.15	0.5-1.6	0.1-1.5	0.3-0.6
Meat	1.1-1.8	0.6	0.3-0.4	1.3-2.2	0.6-4.8	1.0-2.2
Hay from cultivated pastures	3.9	2.8	1.7	4.1	4.7	2.2
Hay from natural hayfields	10.7	4.0	1.9	10-15	17	10-40
Lucerne (fresh mass)	0.8	0.4	0.2	0.8	0.6	0.9
Clover (fresh mass)	0.8-2.3	0.3	0.3	0.9	1.1	0.4
Vetch (fresh mass)	0.9-3.6	0.5	0.2	1.1	1.6	0.5
Lupin (fresh mass)	0.9-1.9	0.4	0.2	-	0.7	-
Fodder beet	0.5	0.4	0.2	0.7	0.5	0.4
Silage maize	0.5	0.2	0.1	0.5	0.5	0.2
Winter wheat (grain)	0.5	0.2	0.05	0.4	0.4	0.2
Winter rye (grain)	0.4	0.1	0.07	0.3	0.3	0.1
Winter barley (grain)	0.3	0.1	0.0	0.3	0.2	0.2
Potato	0.3	0.1	0.04	0.4	0.4	0.4
Edible beet	0.6	0.3	0.06	0.7	0.4	0.2
Cabbage	0.3	0.1	0.04	0.4	0.2	0.1
Tomato	0.2	0.09	0.03	0.2	0.19	0.09
Cucumber	0.1	0.06	0.03	0.15	0.1	0.06
Onion	0.6	0.2	0.11	0.7	0.5	0.16
Greens	0.3	0.05	0.02	0.2	0.2	0.05
Carrot (root)	0.3	0.12	0.05	0.4	0.3	0.2

Table 2: Sr-90 PROPORTIONALITY COEFFICIENTS
(AVERAGE FOR 1987-1989)
(Bq/kg natural product) / (kBq/m² soil)

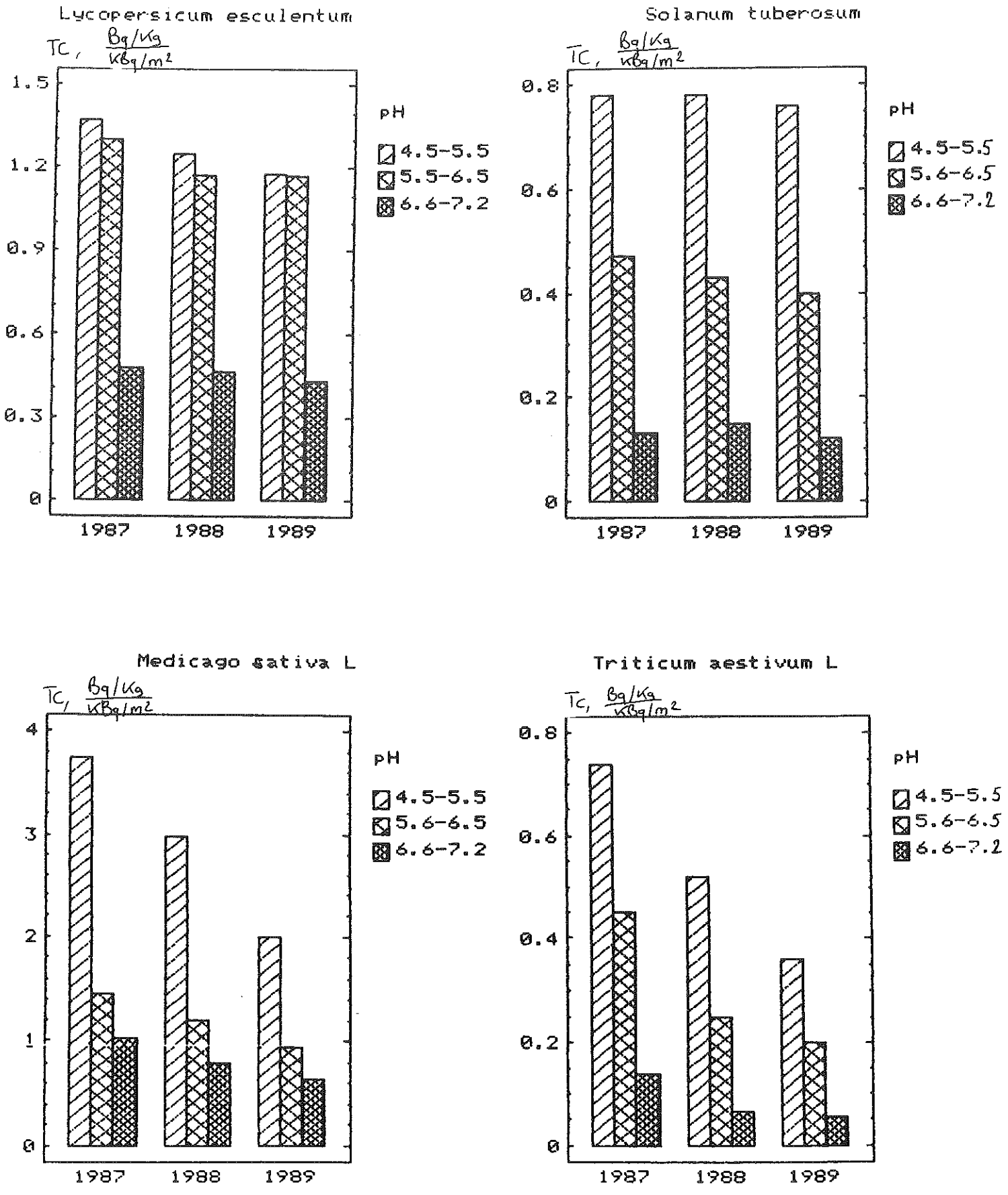
Oblast	KIEV			ZHITOMIR	ROVNO
Soil group Saline pH	I 4.5-5.5	II 5.5-6.5	III 6.5-7.2	I 4.5-5.5	II 5.6-6.5
Milk	3.1	1.4	0.47	2.5	2.0
Hay from grasses (meadow)	102	40	18	124	65
Lucerne (fresh mass)	73	58	16	61	37
Silage Maize	49	12	6	35	15
Fodder beet	15	12	5	17	12

Table 3: COMPARISON OF MEAN MULTI-YEAR VALUES OF COEFFICIENTS OF RADIONUCLIDE TRANSFER FROM SOIL INTO AGRICULTURAL PRODUCE IN THE SOUTHERN URALS AND UKRAINIAN POLESYE
(mCi/kg)/(Ci/km²) of product, $\sigma \pm 30$ %

Radio-nuclide	Product	Contamination area	Exchangeable Ca^2 in soil mg equiv./100 g (saline pH)		
			0.9 pH=4.5-5.5	4.0 pH=5.6-6.5	25 pH=6.6-7.2
Cs-137	Hay from sown grasses	Southern Urals	13	4.4	1.1-1.9
		Polesye	10	2.8	1.9
	Cows's milk	Southern Urals	0.50	0.20	0.04
		Polesye	0.35	0.19	0.13
	Beef	Southern Urals	2.0	0.80	0.20
		Polesye	1.4	0.76	0.55
Sr-90	Hay from sown grasses	Southern Urals	5.7-14	1.3-3.2	0.3-0.7
		Polesye	10	4.8	2.4
	Milk	Southern Urals	2.6	0.56	0.07-0.13
		Polesye	2.1	1.4	0.07

Fig. 1:

CAESIUM 463137



Processes Governing the Transfer of Radionuclides into Crops Following the Chernobyl Accident

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ABSTRACT

Given that the Chernobyl accident contaminated large areas of Byelorussia, we studied the processes governing - and the quantitative features of - radionuclide accumulation in meadow herbage and staple agricultural crops grown by farms in the Gomel Region.

The fields are situated on soddy-podzolic sandy and sandy-loam soils where radiocaesium contamination varied between 15 and 80 Ci/km^2.

The quantitative features detected in radionuclide accumulation levels in crops (depending on soil composition and plant biological properties) confirmed the previously established basic mechanisms governing the transfer of radionuclides from soil to plants.

Proportionality coefficients were determined for prediction purposes, and in 1987 these were (expressed in units of 10^{-9} Ci/kg per Ci/km^2) 0.24 for grains of winter rye, 0.19 for barley and 0.65 for oats, 0.16 for potato tubers, 0.10 for beetroot and 0.23 for silage maize.

These values were a factor of 4-5 lower in 1989.

Depending on meadow type and soil properties, contamination levels in meadow herbage differed by up to two orders of magnitude. When the contaminated turf was reploughed or disc-harrowed and lime and potassium phosphate fertilizers added, contamination levels in the cultivated herbage fell by a factor of 4-7.

As a result of the accident at the Chernobyl NPP, the Byelorussian and Ukrainian Polesye (Pripyat Marshes) - the soils of which are critical as regards the accumulation of radioactive caesium in plants - was the region most contaminated with radionuclides, among which the isotopes of caesium account for most of the dose.

The study of the processes governing the transfer of radioactive substances into crops following the radioactive contamination of Byelorussia is therefore not merely of scientific interest, but is essential for making long-term forecasts about radionuclide accumulation in plants and for devising measures to limit the uptake of isotopes into harvest crops.

The migration of radionuclides in the soil and plant system was studied and the quantitative aspects of radionuclide accumulation in meadow herbage and the main agricultural crops evaluated on farmland in the Gomel oblast (region) in 1987.

The fields concerned are situated primarily on soddy-podzolic sandy and sandy-loam soils, whose radiocaesium contamination density varied from 500 to 2 800 kBq/m^2.

The analysis of radiocaesium accumulation levels in crops showed that, generally speaking, the adoption of a series of soil improvement measures (liming, the application of higher doses of mineral fertilizers, deep ploughing) produces crops on a farm scale which do not exceed the maximum permissible radionuclide concentration (provisionally permissible level), even given contamination densities of up to 1 500 kBq/m^2.

The research and experiments conducted in the Gomel region following the Chernobyl accident confirmed the main, well-known mechanisms governing radionuclide accumulation in agricultural plants, i.e. the transfer of radioactive substances into crops depends on the quantity and the physico-chemical properties of fallout, the characteristics of the soil, the particular biological features of plants and the way the land is farmed.

Cereals can be classified as follows in terms of decreasing radiocaesium accumulation levels : oats -> winter rye -> barley, with the order for crops requiring tilling between rows being maize -> potatoes -> fodder beet.

As a rule, high radiocaesium accumulation in plants was observed where the soil was supplied with little mobile potassium and the soil solution produced an acid reaction.

For the purposes of forecasting, multiplier coefficients were determined, the levels in 1987 (expressed in Bq/kg /kBq/m^2) being 0.24 for the grain of winter rye, 0.19 for barley, 0.65 for oats, 0.16 for potato tubers, 0.10 for beetroot and 0.23 for silage maize.

These values had fallen 4 to 5 times by 1989 (Table 1).

The contamination levels of the meadow herbage differed by up to two orders of magnitude, depending on the type of meadow and the soil properties. With the passing of time since the accident, radiocaesium accumulation in grass fell by tens and hundreds of times, but the content of the radionuclide in natural vegetation is still fairly high (Table 2).

The study of the effect of different soil cultivation methods on pollution levels in pasture proves the effectiveness of approaches such as disc-harrowing with the application of complete mineral fertilizer and lime, and ploughing with prior application of potassium fertilizers at the rate of 250 kg active substance per hectare in comparison with traditional ploughing (Table 3).

The study of the action of soil improvers added to contaminated pasture prior to sowing to grass showed that double liming (on the surface and along the furrow) and the application of zeolite were more effective than traditional ploughing and the application of phosphogypsum (Table 4).

Our research has clearly shown that agricultural crops can be cultivated in areas polluted by the Chernobyl accident, provided that a number of measures are taken not only to reduce the extent of radionuclide transfer from soils to plants but also to preserve and increase soil fertility.

Table 1

COEFFICIENTS OF RADIOCAESIUM TRANSFER FROM SOILS TO THE MAIN AGRICULTURAL CROPS

Bq/kg / kBq/m^2

Crop	Part of crop	1987	1988	1989
Winter rye	Grain	0.24 ± 0.06	0.12 ± 0.04	0.05 ± 0.01
	Straw	0.44 ± 0.20	0.20 ± 0.05	0.11 ± 0.20
Barley	Grain	0.19 ± 0.09	0.06 ± 0.01	0.04 ± 0.01
	Straw	0.50 ± 0.30	0.20 ± 0.05	0.10 ± 0.01
Oats	Grain	0.65 ± 0.19	0.30 ± 0.10	0.22 ± 0.11
	Straw	0.90 ± 0.40	0.40 ± 0.10	0.48 ± 0.20
Potatoes	Tubers	0.16 ± 0.03	0.11 ± 0.01	0.04 ± 0.01
	Stem & Leaf	0.32 ± 0.04		0.11 ± 0.01
Beet	Roots	0.10 ± 0.02	0.06 ± 0.01	0.04 ± 0.01
	Stem & Leaf	0.60 ± 0.10		0.13 ± 0.02
Maize	For silage	0.23 ± 0.07	0.09 ± 0.02	0.06 ± 0.01

Table 2

DYNAMICS OF RADIOCAESIUM CONTENT IN THE HERBAGE OF NATURAL MEADOWS IN THE GOMEL REGION

Location of sites	^{134,137}Cs in soil kBq/m^2	Radiocaesium content in grass (kBq/kg)				
		Autumn 1987	Autumn 1988	Spring 1989	Autumn 1989	Spring 1990
District of Bragin, village of Krasnaya Gora	1 573	62.90	0.48	0.14	0.16	0.08
District of Khoyniki, village of Lomachi	5 120	not determined	17.39	6.66	5.18	4.44
District of Khoyniki, village of Lomachi	3 330	155.40	1.63	0.56	0.37	0.48
District of Narovlya, village of Verbovichi	2 827	111.0	48.10	5.92	5.92	3.18
District of Narovlya, village of Konotop	918	44.40	7.77	1.44	1.66	0.37
District of Vetka, village of Bartolomeevka	4 122	not determined	28.86	14.80	15.17	6.10
District of Vetka village of Bartolomeevka	2 490	not determined	6.66	1.89	1.37	0.59
District of Chechersk						

TABLE 3

THE EFFECT OF DIFFERENT METHODS OF SOIL TREATMENT
ON POLLUTION LEVELS IN PASTURE
(flood plain meadow, 1 110 kBq/m^2)

Experiment variant	Radiocaesium content in fresh mass (kBq/kg)		
	1989		1990
	1st crop	2nd crop	1st crop
1. Control (natural state)	2.36 ± 0.26	1.30 ± 0.07	1.07 ± 0.41
2. 6t/ha lime on the ground, disc-harrowing	0.96 ± 0.33	1.59 ± 0.33	0.96 ± 0.26
3. N30-P90-K200 + 6t/ha lime on the ground, disc-harrowing	0.08 ± 0.04	0.21 ± 0.02	0.11 ± 0.02
4. Ploughing, along the furrow N30-P90-K120 + 6t/ha lime	0.78 ± 0.33	0.81 ± 0.11	1.11 ± 0.22
5. K250 on the ground, ploughing, along the furrow N30-P90-K120 + 6t/ha lime	0.10 ± 0.02	0.28 ± 0.10	0.30 ± 0.07

TABLE 4

EFFECT OF LAND IMPROVEMENT MEASURES ON THE TRANSFER OF RADIONUCLIDES INTO PASTURE
(dry valley meadow, 1 590 kBq/m^2)

Experiment variant	Radiocaesium content in fresh mass, kBq/kg			Sr-90 in hay, kBq/kg
	1st crop, 1988	2nd crop, 1989	1st crop, 1990	1989
1. Control (natural herbage)	9.25 ± 2.96	3.33 ± 0.48	2.33 ± 0.37	1.66
2. Tilling	2.41 ± 0.52	1.66 ± 0.48	0.33 ± 0.04	0.29
3. 10t/ha zeolite on the ground, tilling	1.37 ± 0.26	0.70 ± 0.04	0.33 ± 0.22	0.30
4. 5t/ha phosphogypsum on the ground, tilling	2.55 ± 0.52	1.48 ± 0.74	1.04 ± 0.59	0.17
5. 5t/ha lime on the ground, tilling	1.30 ± 0.18	0.67 ± 0.15	0.56 ± 0.11	0.72

Environmental Distribution and Transport of Radionuclides in West Cumbria Following the Windscale and Chernobyl Accidents

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ABSTRACT

Data are summarised for the pattern of deposition of Cs-137 and I-131 in West Cumbria, UK, following the Windscale and Chernobyl accidents and on the apparent environmental behaviour in terms of retention on plants, transfer from soil to plant, and transfer to milk and meat. For the 1957 accident, peak deposits of Cs-137 several kilometres south of Windscale are estimated to be in the range ~16000-40000 Bq/m^2. Peak recorded concentration of Cs-137 in milk in October 1957 was 592 Bq/l, two weeks after the accident. The ratio of concentration in milk (Bq/l) to content in herbage (Bq/m^2) was ~0.4 to 0.6 m^2/l for Cs-137 and ~0.06 to 0.1 m^2/l for I-131. Peak recorded concentration of Cs-137 in milk in May 1986 was 356 Bq/l and the corresponding ratios of concentration in milk to content in herbage were 0.37 m^2/l for Cs-137 and 0.31 m^2/l for I-131. The reason for the higher value for I-131 recorded after Chernobyl compared to that for Windscale is not clear. Whereas the initial decline in Cs-137 and I-131 content in herbage after the Windscale accident occurred with a biological half-life of ~16 d, initial loss after the Chernobyl accident was much more rapid, reflecting both the effects of rainfall and the different times of year of the two inputs. No data appear to exist for radiocaesium concentrations in meat after the Windscale accident. After Chernobyl, radiocaesium concentration ratios for soil-to-plant transfer in upland ecosystems ranged from 1 to 10. Radiocaesium concentrations in sheep at some locations did not show any substantial decline from 1986 to 1989. Experience on soil-plant-animal transfer after the Chernobyl accident, coupled with measurements on contents of radiocaesium in soil from the 1950s and 1970s, indicates that Cs-137 concentrations in individual lambs from a few locations in 1958 and the middle 1960s may have been similar to concentrations measured in individual animals in summer 1986.

INTRODUCTION

West Cumbria (Fig. 1) has received deposits of radioactivity since the early 1950s from several sources. These include uranium oxide particles from the Windscale piles over the period 1954 to 1957 due to damaged cartridges lodged in the outlet air ducts [1], releases of volatile fission products during the Windscale accident of 1957 [2], weapons-testing fallout [3], routine discharges to atmosphere from fuel reprocessing by British Nuclear Fuels plc. at Sellafield [4], and deposits during May 1986 following atmospheric dispersion of material from Chernobyl [5]. This paper evaluates data that exist for Cs-137 and I-131 from these sources in relation to retention by vegetation and transfer to animal products.

PRE-1957 RELEASES

The environmental impact of particles released prior to 1957 has been evaluated by Chamberlain [1] who estimated that ~12 kg of uranium was discharged containing (amongst other nuclides) ~7.4×10^{11} Bq Cs-137. Chamberlain concluded that the impact of these releases on the food chain would have been relatively less significant than for other forms of deposit because of their large particle size (i.e. 85% between 10 and 100 μm). No published information exists for Cs-137 concentrations in soils, vegetation or animal products for the 1954-1957 period. However, data for Sr-90 concentrations in soils in November 1957 were used by Booker [6] to provide estimates of Cs-137 deposits prior to 1957. These ranged from 12200 Bq/m^2 at 0.5 km to <185 Bq/m^2 at >13.1 km to the north east of Windscale.

OCTOBER 1957 RELEASES

Chamberlain [7] estimated a release of 8×10^{13} Bq Cs-137 from fuel during the October 1957 accident. Of this, ~10% was retained on the stack filter, allowing ~7×10^{13} Bq to be released to atmosphere. Assuming an overall ratio of I-131/Cs-137 of 20 (i.e. similar to that in the fuel), Chamberlain estimated that ~1.4×10^{15} Bq I-131 would have been released to atmosphere. Environmental measurements following the Windscale accident have been reported and evaluated by several authors [1,2,6,8-11]. Booker [6] used data on I-131 levels in herbage during the period 13-28 October

1957 to construct contours for the relative distribution of Cs-137 assuming an I-131/Cs-137 ratio of 50:1 (Fig. 2). His analysis showed total Cs-137 deposits reaching 7000-12000 Bq/m² at some locations several km to the south and south east of Windscale. Booker neglected to include the original I-131 contour to the south which equated to 16300 Bq/m² Cs-137 - this has been included in Fig. 2. Though Booker assumed an I-131/Cs-137 ratio of 50:1, Chamberlain (above) suggested that a ratio of 20:1 can be supported by the available data. This ratio would increase the estimated Cs-137 deposits given in Fig. 2 by a factor of 2.5, i.e. for Black Coombe from ~16000 to ~40000 Bq/m².

Contents of Cs-137 reported for vegetation after the Windscale accident ranged from 500 Bq/m² at Millom to 1440 Bq/m² at Seascale on 28 October 1957, and represented 16 to 37% of the total observed deposit to 4 inches depth in soil [8]. Concentrations in spot samples of herbage ranged from <100 to 36000 Bq/kg dw. For herbage at Seascale, the peak concentration (66300 Bq/kg dw) was recorded on 13 October 1957 and the effective half-life for decline in I-131 concentration over the period October to November 1957 based on deposit per unit area was ~4.9 d [8], implying a biological half-life of ~16 d. Cs-137 showed a similar pattern to that for I-131 during October and November 1957 but a relatively slow decline from January to March 1958.

Booker [8] reported a peak I-131 concentration in milk at Sellafield of 48800 Bq/l on the afternoon of 12 October 1957; subsequently concentrations declined with an effective half-life of ~4.7 d (i.e. similar to that for herbage at Seascale). At Seascale, the ratio of I-131 concentration in milk (Bq/l) to content in herbage (Bq/m²) on 13 October 1957 was 0.06 m²/l. Chamberlain and Dunster [9] reported a corresponding ratio of 0.1 m²/l.

No data are available for Cs-137 concentrations in milk prior to 28 October 1957, reflecting difficulties in detecting Cs-137 in the presence of I-131 with the equipment then in use [2,12]. Samples from Seascale for 28 October 1957 showed a Cs-137 concentration of 592 Bq/l. The ratio of concentration in milk to content in herbage was 0.41 m²/l. In a more detailed survey of milk concentrations over the period 8-11 November 1957, the maximum Cs-137 concentration recorded was 555 Bq/l at Corney (on rising ground ~15 km south of Windscale). The herbage content at this site on 28 October 1957 was ~1300 Bq/m². Allowing for a decline due to

weathering from 28 October to 8-11 November, this implies a ratio of milk concentration to herbage content of ~0.6 m^2/l. Milk concentrations were lower than might have been anticipated because the accident occurred at a time of year when dairy cattle were receiving an increasing intake of conserved fodder harvested prior to the accident.

No published data appear to exist for I-131 or Cs-137 concentrations in meat of animals. The impact to the consumer would have been reduced because beef cattle and sheep can be expected to have been slaughtered at relatively long times after the original deposit.

WEAPONS-TESTING FALLOUT

Several publications provide information on the cumulative input of Cs-137 to soils of the UK as a result of weapons-testing fallout [3,13-15]. Fallout contributions of Cs-137 can be estimated from relationships between measured deposits and average annual rainfall. Maximum cumulative inputs to soils of West Cumbria from weapons-testing to the end of 1980 calculated on this basis are unlikely to have exceeded 6000 Bq/m^2 [11]. Cs-137 contents in soils in Cumbria in 1978 are given in Fig. 3 and show a complex pattern which combines the local influence of Sellafield, altitude, and pre- and post-1957 releases (Fig. 2).

Surveys in the UK in 1963, the period of maximum weapons-testing fallout, showed highest concentrations of Cs-137 in vegetation and animal products from unimproved upland pastures [6]. At one site in Cumbria, the reported Cs-137 concentration in vegetation in 1963 was 629 Bq/kg dw. Cs-137 concentrations in mutton from upland areas of the UK other than from Cumbria ranged from 50-200 Bq/kg during 1963. However, no published data appear to exist for Cs-137 concentrations in animals grazing unimproved pastures in Cumbria during the period of maximum fallout from weapons-testing. Studies in other parts of the UK and elsewhere generally suggested that Cs-137 concentrations in food products were dominated by recent deposits rather than by the cumulative inventory, but these studies were for soils with an appreciable mineral content.

ROUTINE RELEASES FROM SELLAFIELD

Maximum annual Cs-137 discharges to atmosphere from Sellafield (4.5×10^{8} Bq) occurred in 1980 [4] and corresponded with a maximum recorded concentration in milk of 13 Bq/l for farms within 3 km of the site. Routine discharges are small when compared with the 7.4×10^{11} and 7×10^{13} Bq released prior to 1957 and during the 1957 accident (above). By April 1986, concentrations in milk had declined to only a few Bq/l at most.

DEPOSITION FROM CHERNOBYL

Results of monitoring in the vicinity of Sellafield during May 1986 have been discussed by Jackson et al [17]. There is evidence to suggest that a large fraction of I-131 and Cs-137 initially intercepted by vegetation was removed rapidly, possibly as a result of rainfall. Data for a farm at Beckermet (within 2 km of Sellafield) collected over the period 3-12 May 1986 showed a total Cs-137 inventory in soil of 15000 Bq/m^2 [18]. For four farms within 1.5 km of the works sampled over the period 1979-1982, Bradford et al [19] reported Cs-137 contents in root matt plus soil in the range 9900 to 53000 Bq/m^2. Clearly, a significant fraction of the 15000 Bq/m^2 measured in 1986 must have been attributable to sources other than Chernobyl. A similar conclusion emerges for sites at greater distance to the south of Sellafield. For example, a location near Bootle showed ~12000 Bq/m^2 on 13 May 1986. Nearby locations showed measured contents of 6600 Bq/m^2 in 1957, 7800 Bq/m^2 in 1961 and 9700 Bq/m^2 in 1977.

Though it was recognised during 1986 that Cs-137 contents in soils of upland areas of West Cumbria were in the range 20000 to 30000 Bq/m^2, it was not until summer 1988 that the full extent of the contamination became clear as a result of an aerial survey [20]. This showed a central spine of Cs-137 contamination at >30000 Bq/m^2 (Fig. 4a). There were, however, some inconsistencies between the patterns observed for Cs-137 and Cs-134, particularly in upland areas to the south of Sellafield, where the aerial survey indicated Cs-137 inventories in the range 30000-50000 Bq/m^2 in some locations but corresponding Cs-134 inventories in the range 9000-12000 Bq/m^2 (Fig. 4b). Such inconsistencies were not entirely unexpected, especially if much of the Cs-137 present in the 1950s (Fig. 2) and in the 1970s (Fig. 3) remained in situ and was not removed by leaching or

erosion. Naturally, the aerial survey results do not provide any indication of the availability of radiocaesium.

The maximum I-131 concentration in milk recorded at farms near Sellafield during May 1986 was ~370 Bq/l [21]. This corresponded to an initial content on pasture of 1200 Bq/m^2 giving a milk concentration to pasture content ratio of 0.31 m^2/l. The maximum Cs-137 concentration in milk of 356 Bq/l occurred on 5 May 1986 and corresponded to a pasture content of 970 Bq/m^2 on 3 May 1986, giving a milk concentration to pasture content ratio of 0.37 m^2/l. Since the Chernobyl deposit occurred at a time of year at which animals were just beginning to graze fresh pastures, the patterns of concentration in milk at different farms were highly variable according to agricultural practice [21].

DISCUSSION

Concentrations of Cs-137 and I-131 recorded in milk in October/November 1957 after the Windscale accident were higher than for May 1986 after the Chernobyl accident. For I-131, this reflects a relatively low I-131/Cs-137 ratio for the Chernobyl deposit in Cumbria. For Cs-137, the explanation appears to be the relatively small fraction retained by herbage after the Chernobyl deposit [17,18]. The ratio of concentration of Cs-137 in milk to content on herbage observed after the Chernobyl accident (0.37 m^2/l) is close to that observed after the Windscale accident (0.41 m^2/l, but note that measurements of Cs-137 were not available until some time after the 1957 accident). Similar ratios after Chernobyl and Windscale contrast with the markedly lower equilibrium transfer factors (f_m) derived post-Chernobyl relative to weapons-testing fallout and tracer experiments [5]. For I-131, the ratio observed after Chernobyl (0.31 m^2/l) was considerably higher than that observed after Windscale (0.06 to 0.1 m^2/l). The reasons for this are not clear but could involve either differences in the relative contributions of organic and inorganic forms in the two source terms, or differences in dietary intakes between spring and autumn.

By far the major impact of the Chernobyl deposit in West Cumbria has been the effect on sheep which graze upland areas. Cs-137 concentrations in sheep in some areas did not decline from 1986 to 1989. This observation is particularly important since it implies that the cumulative inventory

of Cs-137 in soil is an important aspect of the environmental consequences of radiocaesium contamination in UK uplands. Given the fact that some upland areas of West Cumbria contained significant inventories of Cs-137 prior to the Chernobyl accident, it is interesting to speculate on the relative significance of the various components of these inventories.

The direct method for assessing the significance of the various components requires detailed information on the past history of Cs-137 deposits and assumptions concerning the extent to which radiocaesium remains available in soil or is removed from the rooting zone via erosion or leaching. There is emerging evidence from studies in the UK and Scandinavia that, in highly organic soils, radiocaesium remains available for plant uptake but not for physical or chemical removal processes [5]. Fells to the south of Windscale received maximum deposits of Cs-137 in 1957 in the range 16000 to 40000 Bq/m^2. Some of these fells are dominated by *Calluna vulgaris* (heather) which is known to be particularly effective at recycling radiocaesium and retaining it in surface layers [22,23]. Making the optimistic assumption that losses from soil represented 50% of the original input and taking into account radioactive decay, the maximum 1957 deposit would have reduced to 4000-10000 Bq/m^2 by 1986. Maximum weapons-testing fallout accumulated by 1986 and corrected for radioactive decay is unlikely to have exceeded 6000 Bq/m^2 (equivalent to 2 m rainfall), giving a maximum total inventory by 1986 of 10000-16000 Bq/m^2.

The second method makes use of the input of Cs-134 with the Chernobyl deposit and requires assumptions concerning the Cs-137/Cs-134 ratio in the initial deposit for the area considered. On a UK basis, this ratio appears to have been rather variable (a reasonable range being 1.5 to 2.0 [5]). However, when data for Cumbria alone are examined, the reported values include [11]: 1.59 (range 1.51 to 1.65) for air in the vicinity of Sellafield; 1.53 for rainwater at Sellafield; and 1.5 for water from two lakes. To these data can be added the value of 1.55 (range 1.35 to 1.79) for rainwater from South Cumbria [ITE, unpublished]. Assuming a ratio of 1.6 for the initial deposit in Southwest Cumbria, no more than 50% of the Cs-137 inventories measured in upland soils during 1986 at the most contaminated sites can be attributed to Chernobyl [23]. A maximum Cs-137 content in soil of 34500 Bq/m^2 was observed under *Calluna vulgaris* on fells to the south of Sellafield during November 1987 [23]. Correcting for radioactive decay, and assuming an initial Cs-137/Cs-134 ratio of 1.6 (range 1.8 to 1.4), indicates a pre-Chernobyl Cs-137 content at this site

of 16800 (range 14600 to 19100) Bq/m^2. This corresponds with the maximum value estimated above by the direct method.

Unfortunately, if the second method is applied to vegetation samples collected from 1986 to 1989, it becomes clear that some plant species must have had an appreciable concentration of Cs-137 prior to 1986 [24]. These species include native herbs and shrubs typical of acid grasslands in the UK (i.e. *Eriophorum vaginatum*, *Trichophorum caespitosum*, *Calluna vulgaris*, *Narthecium ossifragum* and species of *Carex*) [23-25]. For these species, soil-to-plant transfer factors observed in 1986 and subsequently were typically in the range 1-10 [26,27]. As a result, Cs-137/Cs-134 ratios derived for individual plant species varied quite widely according to the relative contributions of Chernobyl and other sources to the radiocaesium inventory in soil at different depths in the rooting zone [11,23].

Both methods imply that maximum Cs-137 contents in soil at selected upland locations in West Cumbria prior to the Chernobyl accident may have been in the order of 15000 Bq/m^2. Using the conservative assumption that this inventory was dispersed in the top 15 cm of soil, assuming a soil bulk density of 0.5 g/cm^3 and a soil-to-plant transfer factor in the range 1-10, the estimated Cs-137 concentration in vegetation would have been 200-2000 Bq/kg dw. Assuming a transfer coefficient for lamb of 0.6 d/kg and a daily dry matter intake of 0.6 kg/d, the estimated range of maximum concentrations in individual lambs would have been 70-700 Bq/kg fw. On the basis of the observed continuing availability of the Chernobyl deposit in upland areas, the pattern of Cs-137 concentration in sheep prior to the Chernobyl accident can be expected to have followed the cumulative Cs-137 content in soil.

Taking into account the higher concentrations of Cs-137 reported for milk in 1957 compared with those reported for 1986 and the known pattern of input of weapons-testing fallout, then maximum concentrations of Cs-137 in sheep after the 1957 accident and in the middle 1960s will have been similar to or higher than maxima recorded after the Chernobyl deposit. To our knowledge, this factor has not been considered when assessing the relative radiological impacts of the Windscale and Chernobyl accidents to the population of West Cumbria.

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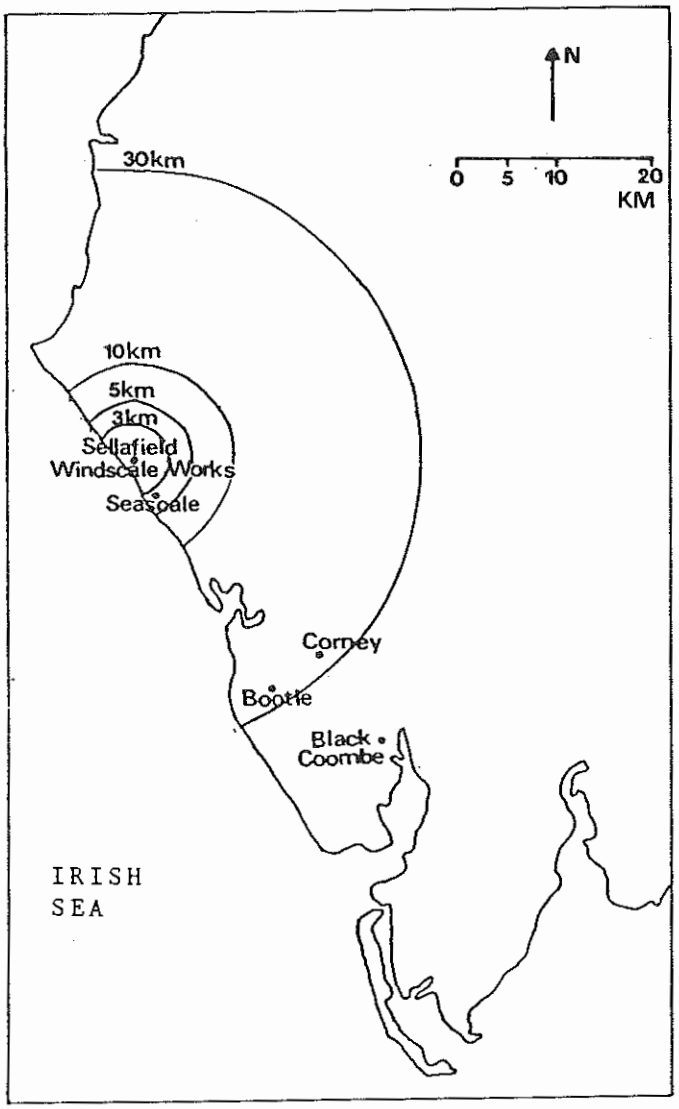
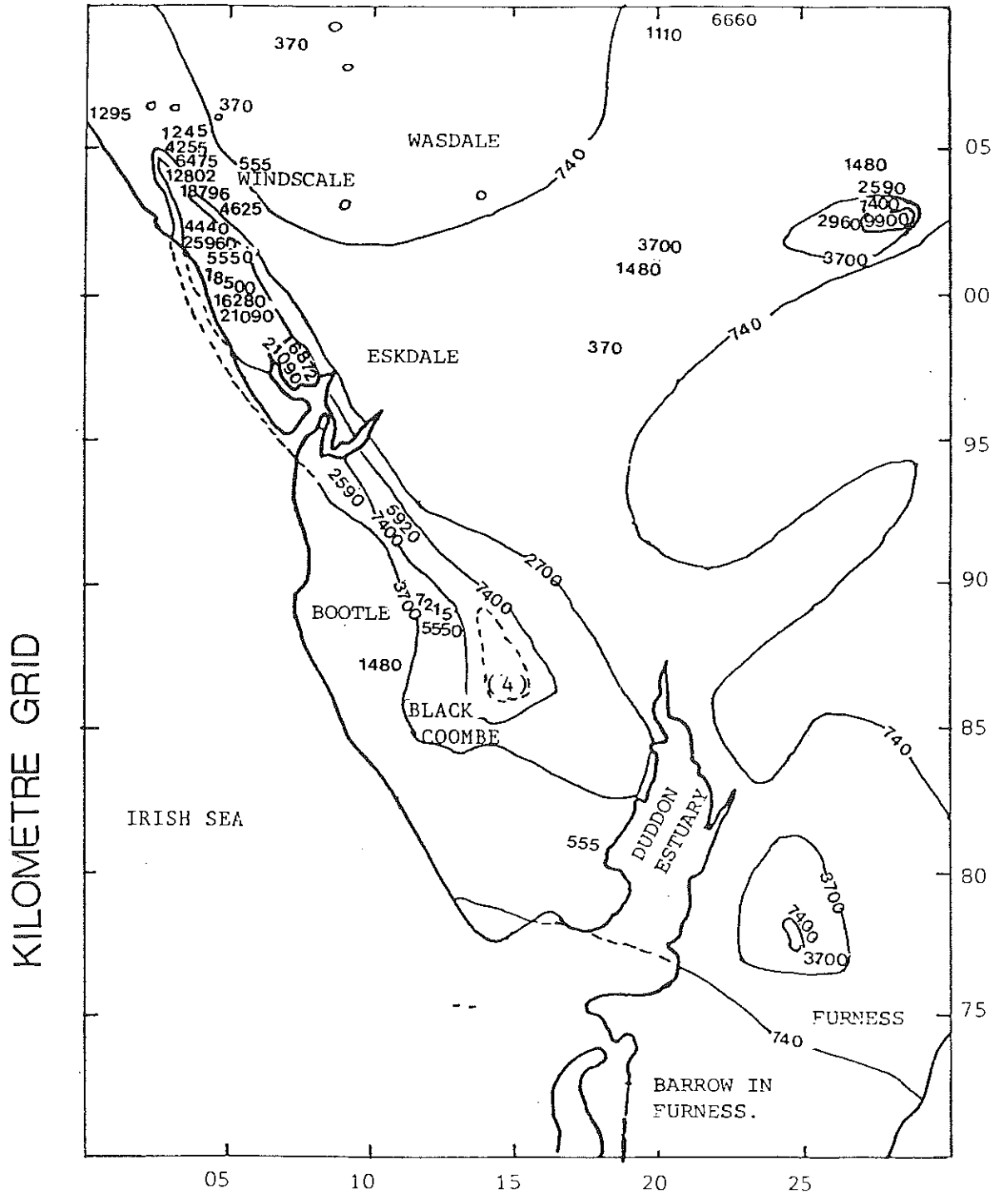


FIGURE 1. LOCATION OF SITES REFERRED TO IN TEXT.



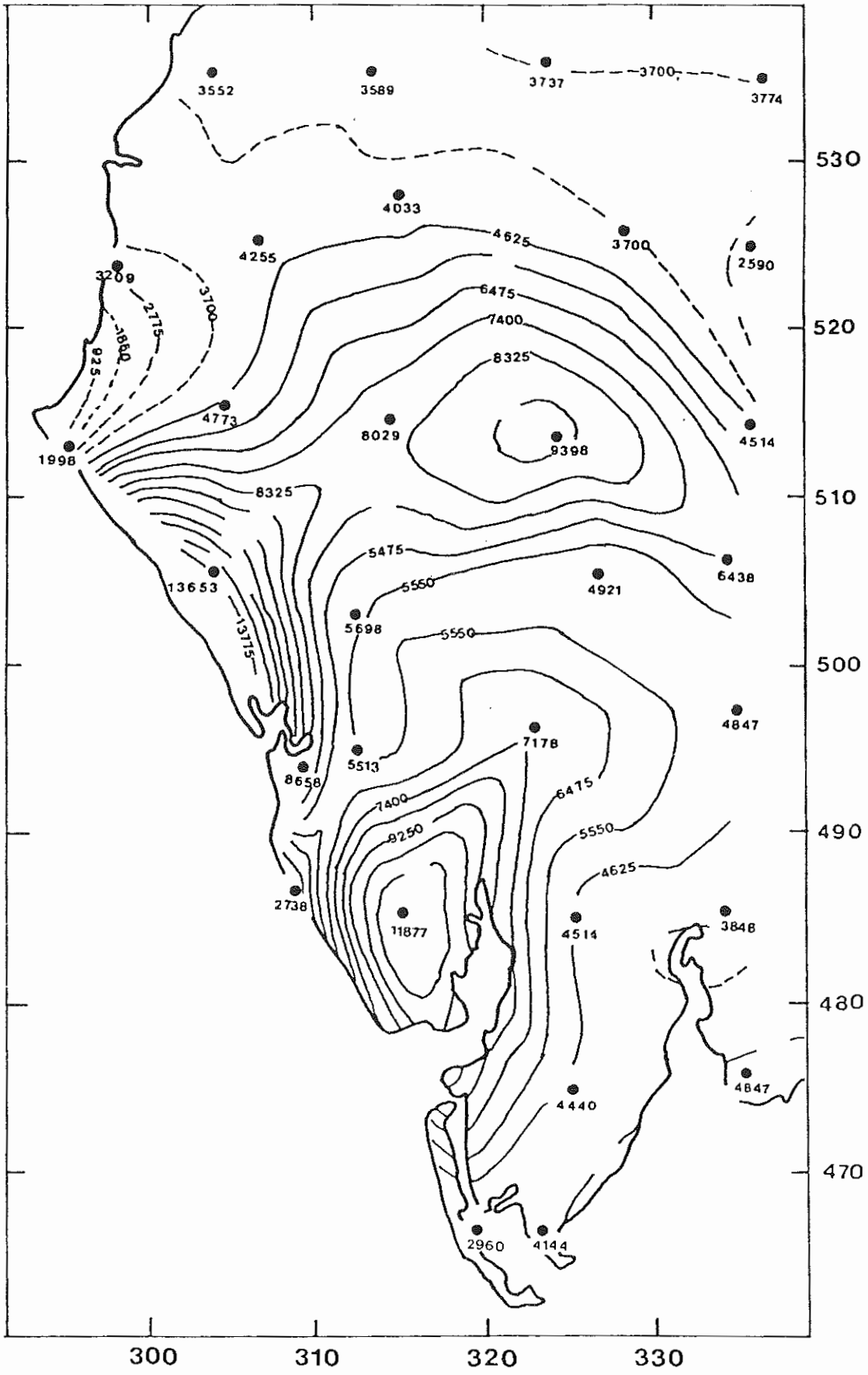
Notes 1. After Booker [6]

2. Figures are Bq m^{-2} of Cs-137 in soil

3. Contours are Bq m^{-2} reproduced from I-131 contours x 0.02

4. Additional contour based on observed I-131 deposits equivalent to 11,100 Bq m^{-2} Cs-137 assuming I-131/Cs-137 ratio = 50/1.

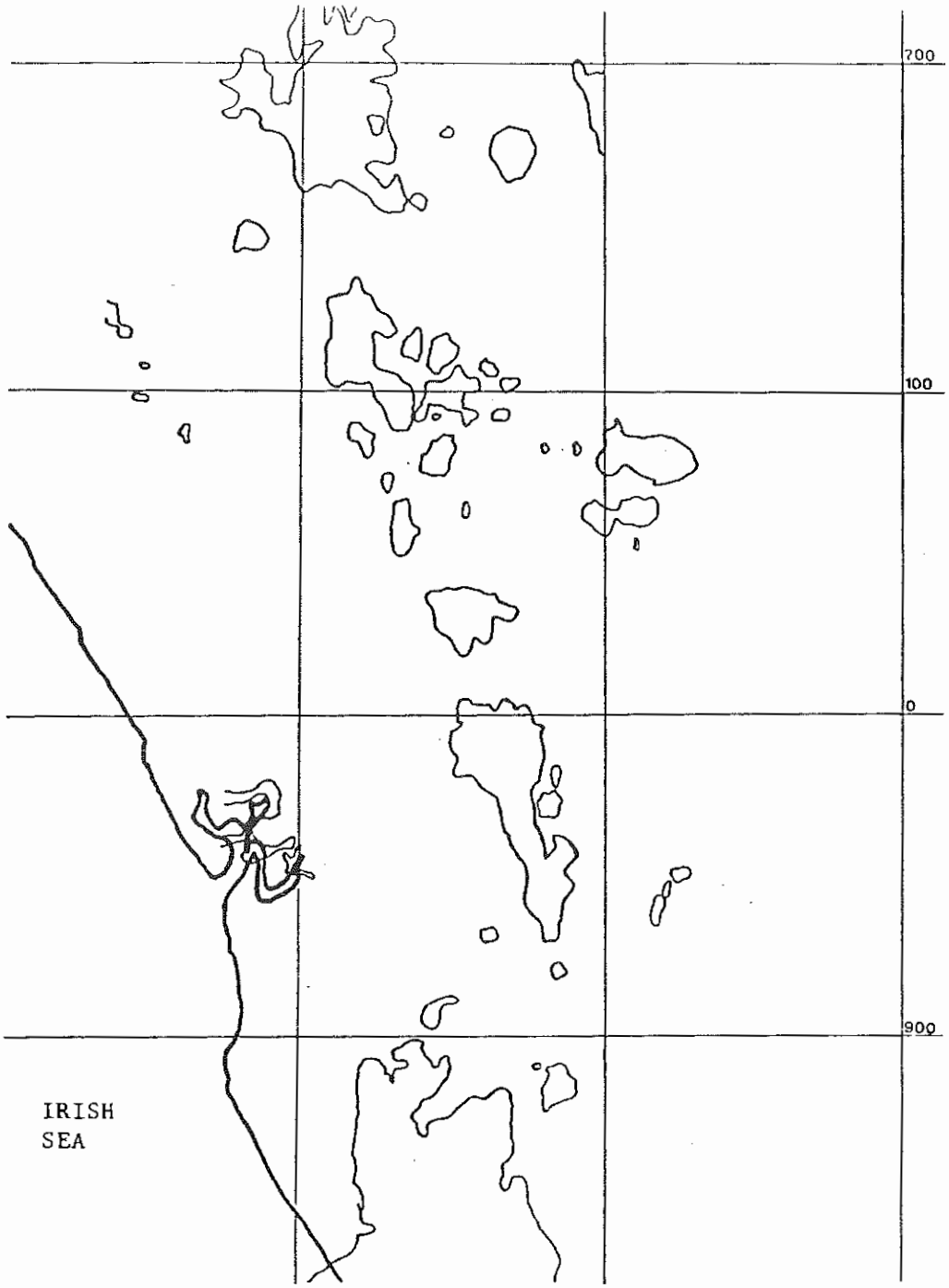
FIGURE 2. PATTERN OF DEPOSITION OF Cs-137 DUE TO THE WINDSCALE ACCIDENT.



Notes 1. From Cawse [3]

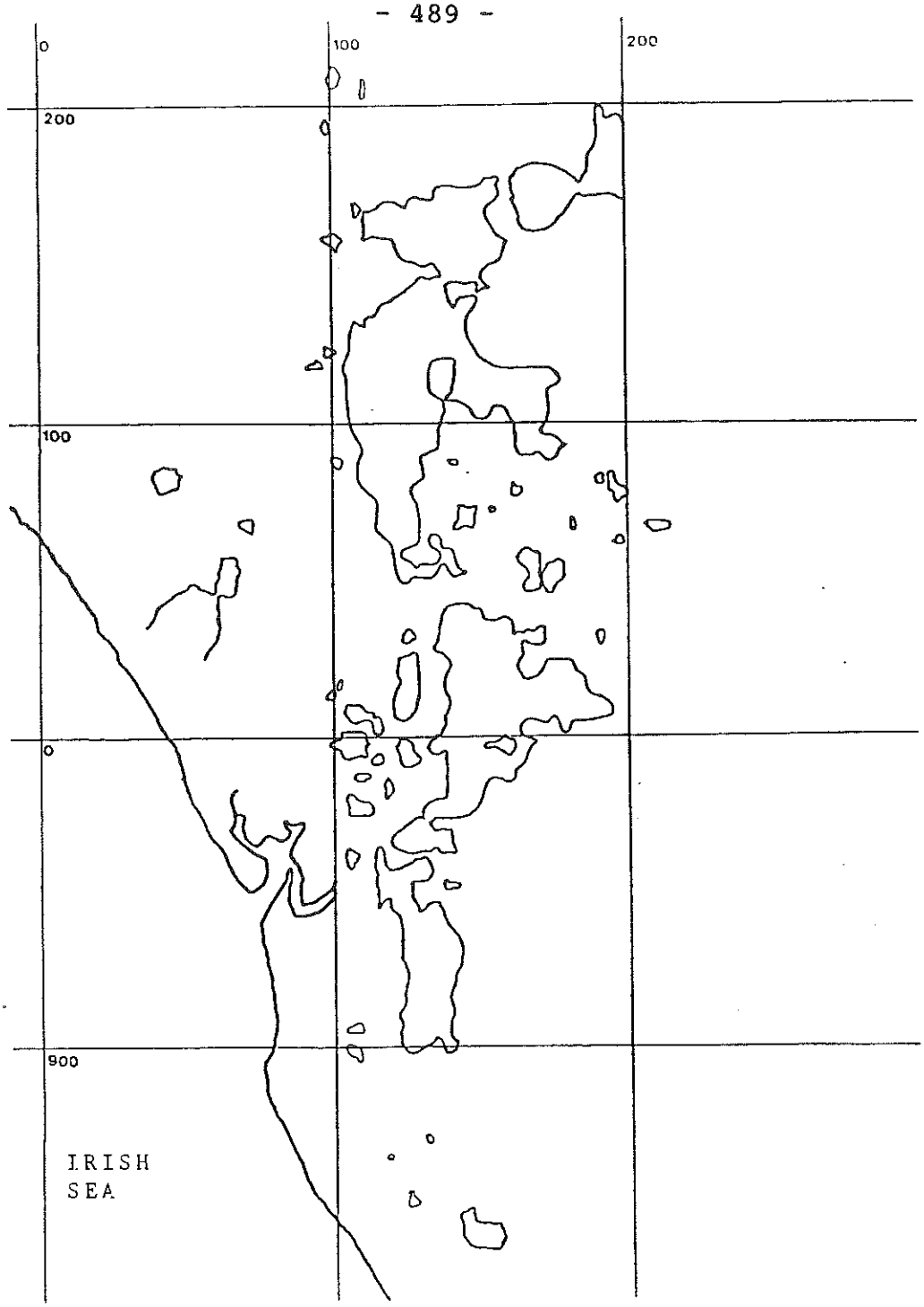
2. Deposits given as Bq m^{-2}

FIGURE 3. DEPOSITS OF Cs-137 TO 15 CM DEPTH IN THE 1978 GRASSLAND SURVEY.



Notes: 1. From Sanderson and Scott [20]
2. Contour shows Cs-137 > 30kBq/m²

FIGURE 4a Cs-137 GROUND DEPOSIT IN CUMBRIA IN NOVEMBER 1988



Notes 1. From Sanderson and Scott [20]

2. Contour shows Cs-134 > 12kBq/m^2

FIGURE 4b. Cs-134 GROUND DEPOSITS IN CUMBRIA IN NOVEMBER 1988.

Session V

ENVIRONMENTAL CONTAMINATION AND TRANSFER

Part II

Analysis of Chernobyl Fuel Particles and their Migration Characteristics in Water and Soil

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INTRODUCTION

Contamination of the environment in the wake of the accident at the fourth unit of the Chernobyl NPP differs quite considerably from global fallout after nuclear explosions. Apart from various volatile radioactive materials, dispersed nuclear fuel was also released into the atmosphere.

This study reflects the results of experimental investigations into the behaviour of fuel particles in the water and soil of the floodlands in the Chernobyl NPP near zone. In the initial fallout these products took the form of low-solubility matrices (UO_2–UO_3). In the case of the Chernobyl accident there was a most unfortunate constellation in that the territory most severely affected by the fallout was a floodplain with a high probability of flooding by river waters (Figure 1). Accordingly, the prime goal of our research was to determine the kinetics of radionuclide transfer from the fuel particles to the water as a basis for predicting the radioecological impact on the environment. The results are important for predicting the processes of mass exchange of exchangeable forms of radionuclides in soil and bed sediment, their washout from the soil surface and the filtration element in migration.

The floodlands area investigated is the plain adjacent to the Chernobyl NPP zone. Its characteristic features are well-developed floodplain vegetation, soddy-, podzolic, gleyey soils and low to medium podzolic sandy soils with underlying sands.

The hydrological conditions for inundation of this area are determined by the regimen of the Pripyat, with flooding of the patch of contamination studied occurring once every four to five years approximately. The water from such floods may remain on the floodplain surface for several weeks.

Once every 10–15 years on average this surface is fully flooded by a developed system of floodplain streams. Less often (once every 25 to 50 years, and most recently in 1979), the high water may cause up to 50% of the river flow to be transported across the floodplain (flood levels of up to 3 m), while flooding of the intermediate and low-lying parts may last for 6 to 8 weeks. During flooding the speed of the current may range from a few centimetres to one metre per second. However, the usual speed of the water flowing over the floodplain is 0.1 to 0.2 m per second, and thus it normally takes the water masses two days to pass over the worst contaminated part of the floodplain within the northern fallout track, although the actual exchange of floodplain waters with the river may take considerably longer (10 days or more) because of the large number of standing and circulating water zones on the floodplain.

Given the direct hydraulic link between the floodplain groundwaters and the River Pripyat, groundwaters formed over the year flow from the river towards the floodplain water bodies. The water table is usually no higher than 1.5 m below the surface, but in a number of places it reaches the surface, forming swampy lowlands and floodplain lakes. The chemical composition of the groundwater is similar to that of waters of the hydrocarbonate-calcium type, but with a different degree of mineralization and richer in organic acids and iron. The extreme wetness of the soil causes it to be washed through intensively and promotes migration of the dispersed nuclear fuel both in solution and in non-exchangeable and low-solubility physico-chemical forms.

Moreover, observations have shown that during high water substances are mainly removed from the floodplain in both floodplain channels and surface runoff. Since the accident (1986-1990) there has been no major flooding of the floodlands and the water flow of the Pripyat has been characteristic of low-water years. This allowed us to study in greater detail the potential source of contamination, including changes in the fuel particles and the mechanisms by which the resultant products are transferred to the river waters.

The object of the investigations was as follows: to study the processes of radionuclide floodplain migration and the kinetic parameters of the changes in the radionuclides' chemical forms, and then propose a number of protective measures with a view to reducing the potential washout of radioactive products into the river system of the Dnieper basin.

FORMATION OF FUEL AND CONDENSED FORMS OF FALLOUT

The destruction of the reactor was accompanied by the ejection not only of volatile fission products but also by a large quantity (2-5%) of dispersed nuclear fuel.[1, 2] The fuel was dispersed under the impact of a sudden energy release in the fuel elements due to a surge in neutron flow, the shock wave of the thermal explosion and the temperature gradients. Thus, in the case of high energy release values [3] (over 285 calories/g UO_2) the fuel element casing is destroyed, while at 320 calories/g UO_2 the fuel itself breaks up into fine particles. The high temperature in the active reactor zone resulting from the continued heat release and burning graphite led to further break-up of fuel and its ejection beyond the reactor zone. Subsequently - from 26 April to 6 May - fuel particles were ejected into the atmosphere together with volatile radionuclides . During and after the thermal explosion, the high-temperature oxidizing processes promoted the partial transformation of sintered UO_2 into U_3O_8, whose structure is more susceptible to mechanical break-up.

In all probability the further oxidization of UO_2 at a range of high temperatures took place in the following stages. At temperatures of 260 to 300° C, UO_2 with its cube-shaped lattice oxidizes at the rate of 0.3 mg/cm^2 per hour to U_3O_8 with its orthorhombic lattice.[4,5] The process occurred in two stages with the formation of an intermediate tetragonal phase U_3O_7. Then according to [6], in the 300 to 600°C temperature range oxidization to U_3O_8 should have occurred rapidly, resulting in the break-up of the fuel pellets into fine particles. From 600 to 850°C the oxidization process slowed down. At 800-900°C oxidization takes place in two stages, a slow one followed by a fast one. Depending on the properties of the specimen, the second stage begins 80-180 minutes after the commencement of oxidization.

The first stage is characterized by diffuse penetration of oxygen into the fuel pellet and takes the form $UO_2 - UO_{2+x} - U_4O_9$. The rapidity of the process during the second stage of oxidization is linked to the phase transformation of U_4O_9 to U_3O_8, and is accompanied by break-up of the specimen. UO_2 particles are brown, U_3O_8 are black and UO_3 are orange.

Greenish-yellow particles of a glass-like material may be created when UO_2 enters into contact with water and the partially hydrolised oxide $UO_3O.8H_2O$ forms.

The macro- and microstructural changes in UO_2 (during reactor operations) under the effect of neutron irradiation and radially directed thermal flow (neutron fluence, burn-up, temperature gradient across the fuel radius, irradiation duration, swelling, etc.) as well as temperature shocks had a major impact on the process of nuclear fuel dispersion. Thus, "hot" fuel particles formed as a result of the explosion shock waves, temperature gradients and UO_2 oxidization.

During ejection, over 90% of all particles formed in this manner were UO_2-UO_3 particles with a radionuclide composition similar to that of the irradiated fuel (Table 1) [7] and containing a variety of highly mobile volatile fission products.

The fractionation of the fission products in the nuclear fuel took place both during the accident (subsequent reactor heat effects, or "annealing") and during normal reactor operations. The relative migration rate of the fission products from UO_2 in the presence of high temperatures is reduced as their binding energy in respect of oxygen grows, in the following sequence:

Kr<0.09 eV; Xe-0.38 eV; I-0.91 eV; Ag-2.22 eV; Cs-3.09 eV; Te-4.06 eV; Sr>4.72 eV; Ru-5.0 eV; Ba>5.85 eV; Zr-7.9 eV; Ce-8.25 eV.[8]

Gaseous and volatile fission products such as Kr, Xe, I, Te, Cs, which have fairly high migration properties, are more readily fractionated in the nuclear fuel. During normal reactor operations, high temperature gradients ($3x10^5$ degrees/minute) are observed across the radial profile of the fuel pellet.[4] This led to a redistribution of fission products from regions with high temperatures (pellet centre) to regions with lower ones.

Table 1

Mean specific activity of radionuclides in fuel of the Chernobyl NPP fourth unit at the time of the accident (Bq/g UO_2)

Nuclide	Activity	Nuclide	Activity	Nuclide	Activity
Sr-90	1.2E+09	Cs-134	8.6E+08	Sb-125	1.8E+08
Zr-95	3.3E+10	Cs-137	1.5E+09	Eu-154	7.1E+07
Nb-95	3.4E+10	Ce-141	3.2E+10	Eu-155	1.0E+08
Ru-103	2.6E+10	Ce-144	2.4E+10	Pu-238	5.4E+06
Ru-106	7.0E+09	Pm-147	3.8E+09	Pu-239	4.5E+06
				Pu-240	6.3E+06

As a result of high-temperature "annealing" of the fuel particles, these lost their highly mobile fission products. On the other hand, at relatively low temperatures these fission products condensed and were adsorbed on the surfaces on all kinds of carriers. This explains the different quantities of caesium present in the hot fuel particles (mean coefficient of fractionation: 0.4-0.6) and, to a lesser degree, strontium (0.9-1.0).

Insoluble fission products in microsections of irradiated fuel take the form of second-phase inclusions with a characteristic metallic sheen. Most of the inclusions have a roundish form, and are generally sited on the margins of the granules and bound to the pores. The presence of inclusions may explain the abnormal quantities of refractory radionuclides in certain hot particles.

As a result of the destruction of the fuel elements and the "annealing" of the nuclear fuel, a substantial quantity of volatile fission products (I, Cs, Sr etc., the so-called steam-gas or jet components of the release) was ejected into the atmosphere. Some of these condensed on inert carriers - particles of soot, dust, construction materials, etc. Hot particles formed in this manner are characterised by surface contamination and low specific activity by comparison with the fuel particles.

These finely dispersed hot particles and the steam-gas component were drawn into the jet of ejected materials, and subsequently into the atmosphere. Their further dispersion was determined by the movements of the air masses - deposition of aerosols on land and washout of radioactive products through rain.

Of the known tracks of aerosol deposition in the area, the northern track is the worst contaminated, with much of the countryside in this fallout track 10-15 km from the power plant forming part of the Pripyat floodplain. It was here that the basic cycle of complex investigations was carried out, including analysis of the radionuclide inventories, contamination structure, landscape and geochemical analysis of the environment and hydrochemical flooding regimen (Figure 1).

In order to determine the inventories and distribution densities of the radioactive substances deposited, soil samples were taken using special 50 mm-long sampling tubes with a diameter of 140 mm. To investigate vertical migration, cores of bed sediment and soil were removed using two types of sampling device, of 50 mm and 25 mm diameters respectively, capable of removing cores up to 250 and 500 mm long. The samples were taken in May/June 1989 and June/July 1990.

Examination of the samples' nuclide composition was conducted with the aid of an ORTEC gamma spectrometer linked to an IBM PC/XT, using a HPGe coaxial detector.

The contamination levels in soils in the Krasnensky and Benevsky bayous are shown in Table 2.

The vertical distribution of fission products in the soil and bed sediment cores taken from the most representative parts of the floodplain was analysed. The samples had been removed using special tubular sampling devices, so as not to destroy their structure. Samples were taken from:

1. a non-flooded dry sector on the floodplain;

2. a regularly flooded sector at the perimeter of a floodplain lake;

3. bed sediments in a closed water body.

Samples for analysis were also taken from patches of abnormally high contamination. The cores were sliced into 1-2 cm thick layers, dried and homogenized. After gamma spectrometry they were subjected to radiochemical analysis of total strontium content.

The distribution characteristics are given in Figure 2. They show that three years after the accident the bulk of the radionuclides (95-98%) is distributed through the top 3 to 5 cm layers. This is confirmation that the fallout's main component - i.e. fuel (90-98%) - participates only in the convective migration mechanism, i.e. movement through pore spaces together with infiltrating waters. Autoradiographic photographs of the top layers suggest an exponential law of distribution of hot particles, both as regards their dimensions and the soil profile. Hot particles were not discovered at depths of more than 6 cm.

In bed sediments from closed and standing water bodies there was a more uniform distribution of radioactive substances through the profile. Specific activity in the top layers of the bed sediment (less than 5 cm) constitutes 80-90% of total activity. Hot particles have been discovered at depths of up to 8 cm and this, most probably, is linked with turbidity and load deposition/accumulation on the bed.

Table 2

Soil contamination in the abandoned Krasnensky and Benevsky channels, Pripyat left-bank floodplain in the Chernobyl NPP near zone (1989)

SAMPLING POINT	RANGE OF ACTIVITY A (Ci/km^2)			
	Ce-144	Cs-137	Sr-90	total Pu (38,39,40)
Krasnensky bayou	200-900 <425>	100-500 <230>	130-600 <280>	3.46-15.60 <7.90>
Benevsky bayou	20-150 <55>	10-100 <32>	13-100 <36>	0.3-2.6 <0.9>

Table 3

Weighted mean estimates of major gamma-emitting nuclides in total activity of the nuclear fuel (26 April 1986) - percentage figures

Ru-106	Cs-134	Cs-137	Ce-144	Sb-125	Eu-155
20.70	2.55	4.15	71.20	0.53	0.21

An analysis of the radionuclide activity ratios in terms of Ce-144 units shows that the sampling procedure was methodologically sound. The growth in the activity ratios vis-a-vis Ce-144 with increasing depth indicates increasing radionuclide migratory capacity in the following order: Ce-144 < Sb-125 < Ru-106 < Cs-137 < Sr-90.

The figures for the distribution of Sr-90 both in bed sediment and in soil show a relative shift towards Sr-90 as compared with Ce-144 and other gamma-emitting radionuclides through layer depth. From this we can conclude that Sr-90 has a greater migratory capacity than the other radionuclides.

With a view to learning more about the structure of fuel fallout, we investigated the size and radionuclide composition of the hot particles. Autoradiographic and micro-photographic analyses of the hot particles, which account for 95-98% of contamination in the area under investigation, showed that they consist of particles of micron or sub-micron dimensions, while their radionuclide composition resembles that of irradiated fuel. In the years immediately following the accident, the ratio between fuel and condensed forms in the soil samples was 90-98%:2-10% on average. Currently, because the physico-chemical forms of the fuel radionuclides change, the proportion of more mobile forms is increasing.

In determining the dimensions of the hot particles in various soils and bed sediments, use was made of the contact autoradiography method, and coefficients of correlation were established between the diameters of the black patches on the autoradiographs and the activity and dimensions of the fuel particles. Some 10,000 particles in the soil samples were covered.

Figure 3a shows the size distribution of soil sample particles. Figure 3b shows the cumulative frequencies for this distribution. On a probability-logarithmic scale the particle distribution adequately fits a straight line, so one can assume that the distribution is log-normal and hence estimate its parameters. The mean geometric diameter of the particles is D = 8 microns, while the median diameter of the hot particles was approximately 7 microns.

The mean geometric diameter of particles taken from bed sediment was found to be D = 14 microns, most probably because the finer particles had dissolved.

The process of hot particle separation by size could be observed at as early a stage as that of aerosol deposition and settlement in water bodies. Here, the larger particles and their carriers sank more rapidly, while the finest particles - under the influence of hydrodynamic factors - were carried closer to the shores. Then, as the high water receded and lake water levels dropped, they attached to the soils around the shore. This is confirmed by the existence of zones of increased contamination at the edges of water bodies.

Analysis of the size distribution of hot particles in the soil samples (Figure 4), taken from a site 10 km north of the power plant in 1987 and again in 1989, shows a reduction in the proportion of fine particles (< 2 microns) over time. Clearly, this is because the fine particles dissolve under natural physico-chemical conditions. The proportion of 2-4 micron particles increases, presumably because of the decrease in the number of fine particles and because the larger particles undergo mechanical break-up. Clearly, this deserves closer study.

Microscopic examination shows that the bulk of particles from soil and bed sediment is to be found in fragile aggregates containing fine particles of soil and a variety of organic and mineral inclusions. The hot particles themselves have irregular structures with well-developed surfaces. They easily break up under mechanical impact. Most are black, while some have an orange or matt sheen and a granular structure. Some particles were found to have a fused, uniform surface, and unlike the others were very hard. Spectral analysis showed that Ru-106 was the main source of activity in these particles.

Under the assumption that the particles consist of UO_2 with a density of 10.4 g/cm^3 and a burn-up of 11.6-12.5 MWd/kg UO_2, particle diameter was estimated by means of Ce-144 activity. Ce-144 was selected because it has a relatively small coefficient of diffusion in fuel (3 to 4 orders of magnitude less than other nuclides) and because of its capacity to replace uranium in the UO_2 lattice.

For the estimates it was assumed that the hot particles were spherical. Ce-144 activity at the time of the accident on 26 April 1986 was assumed to be 2.2 x 10^{10} Bq/g UO_2.[9] Hence the calculation coefficient for estimating the diameter of the fuel particles by Ce-144 activity is:

$$A(Ce\text{-}144/Dp = 11.43 \times 10^{-2} \exp(-2.44 \times 10^{-9} \times T)$$

where:

Dp: estimated particle diameter (microns)
A(Ce-144): Ce-144 activity at the time of measurement (Bq);
T: number of days since the accident.

In addition, De is the diameter of the particles as determined visually on a microscope scale (microns).

The mean ratio of measured to estimated values (De/Dp) is 1.2. This difference would suggest that besides errors in the visual measurement of the size of particles in conglomerate with inactive carriers, there was some change in the stoichiometric composition of the particles during oxidization to U_3O_8 with a density of 7-8 g/cm^3.

RADIONUCLIDE COMPOSITION OF THE HOT PARTICLES

Table 3 shows the weighted mean (%) estimated contributions of the main gamma-emitting isotopes to total gamma activity of "average" fuel on 26 April 1986.

From [7] and Table 3 it is clear that the mean weighted composition of the main gamma-emitting nuclides in "average" fuel at the time of the accident corresponds to a burn-up rate of 11.6-12.5 MWd/kg UO_2, i.e. to the 4th or 5th group of fuel element assemblies.

The radionuclide composition of the soil and bed sediment samples tallies well with the estimated figures for "average" fuel as shown in Table 3 (within margins of 10-15%).

To classify the hot particles by isotopic composition, we established the distribution of the ratios of experimental and estimated contributions ("average" fuel) to total gamma activity of the isotopes Ce-144, Ru-106, Cs-134 and Cs-137.

Figures 5 and 6 show the distribution of these ratios for hot particles from soil and bed sediment. For Ce-144 the contribution is higher than expected, with a mean of 1.1 for particles taken from soil, while for bed sediment it is 1.18. This can be interpreted as indicating a decrease since the accident in the amount of the other radionuclides contained in the fuel particles in soil and aquatic environments, particularly Sr-90 and Cs-137. Below we show the estimated values (%) of radionuclide impoverishment in fuel particles from soil and bed sediment:

	Cs-137,134	Ru-106	Sb-125	Sr-90
Soil	(20-25%)	(5-9%)	(15-20%)	(5-10%)
Sediment	(35-40%)	(5-9%)	(20-25%)	(10-15%)

Ruthenium impoverishment was estimated without taking into account pure ruthenium particles, which make up 5-10% of the total number of particles.

Similar estimates, previously carried out by the authors for particles in aggregate with soil and bed sediment material [11], showed that for caesium, radionuclide impoverishment did not exceed 5%, while in the case of Sr-90 impoverishment in the soil was close to that for individual particles, and in some of the 1989 samples exceeded 30%. The increased impoverishment of caesium and strontium in particles taken from bed sediment is evidence of their water-soluble and exchangeable forms leaching into the aquatic environment by comparison with dry parts of the floodplain. The difference amounts to 10-15%.

PHYSICAL AND CHEMICAL CHARACTERISTICS OF FUEL FALLOUT AND ANALYSIS OF ITS BEHAVIOUR IN THE AQUATIC ENVIRONMENT

The estimates of the distribution of radionuclide forms of occurrence in the aquatic and soil environment were obtained via classical consecutive lixiviation of soil and bed sediment samples in (1) a water solution (using distilled water and water from the Pripyat River), (2) a 0.1N solution of CH_3COONH_4, and (3) 6N HCL. The first extracts were used to determine the proportion of radionuclides in water-soluble form for different degrees of mineralization of the solvent (distilled water and river water); the second set of extracts made it possible to determine the quantity of radionuclides capable of participating in migration via the ion exchange mechanism; the third set of extracts, together with the quantity of radionuclides in the insoluble residue, made it possible to determine the proportion in non-exchangeable sorbed form and in the composition of the fuel particle matrix.

The mobile and non-exchangeable forms were determined for areas of soil and bed sediment with different degrees of surface contamination. Table 4 shows the results for the measurements conducted in 1989.

The data for 1990 show that the proportion of mobile radionuclides, in particular of Sr-90, continued to grow. Towards summer 1990 the total proportion of exchangeable and water-soluble Sr-90 in the floodplain soils in many areas amounted to as much as 60-75%, indicating that the fuel matrix was subjected to very intensive transformation processes.

Similar estimates were conducted for different down-core sections of soil and bed sediment samples. The data show that for Cs-137 the percentage both of water-soluble and exchangeable forms of the radionuclide increases with the depth of the soil layer in which migration takes place. At the same time the proportion of non-exchangeable sorbed and fuel forms of Cs-137 decreases with depth. The percentage of water-soluble and exchangeable caesium grows from 1-2% at the surface to 20-30% at a depth of 10-15 cm. At the same time, the water-soluble proportion of caesium for these horizons grows from 0.1% to 1.5% in dry non-flooded areas and to 5-10% in moist soils. For Sr-90 there is no statistically significant increase in its mobile form in the top 20-25 cm layer. The total proportion of exchangeable and water-soluble Sr-90 in the floodplain soil samples was 30-40% in summer 1989, but had grown to 50-70% in spring-summer 1990 in the same region. At deeper horizons exchangeable forms made up 90% and more. Less than 10% were fixed to soil particles as a result of adsorption. In absolute terms the radioactivity (Sr-90) of the soil sections was 10-25% at depths below 1 cm, 1-5% at depths below 5 cm and 0.1-0.5% at depths below 10 cm, while at depths below 20 cm the specific radioactivity of the sections did not exceed 0.1% of the total content in the floodplain soils.

Table 4

Mean data on radionuclide forms of occurrence (as a % of total content) in soil and bed sediment in the floodplain of the Chernobyl NPP near zone (1989)

	Percentage in extract soil/bed sediment			
Form of occurrence	Ce-144	Cs-137	Ru-106	Sr-90
Water-soluble	0.30/0.10	0.17/0.10	0.20/0.10	15.0/25.0
Exchangeable	2.8/1.0	1.1/0.5	0.05/0.5	30.0/25.0
Non-exchang.	97.9/99.5	98.7/99.4	99.8/99.4	55.0/50.0

For Cs-137 the corresponding figures are approximately 1% below 5 cm and 0.01-0.1% below 20 cm. The lakeside samples constitute an exception, the lower layers being contaminated due to a rise in contaminated groundwater and lake waters through capillary action. These results allow us to draw an indirect but important conclusion concerning the impact of water on the migration properties of fuel fallout. Bearing in mind that initially the floodplain was contaminated almost exclusively by fuel fallout, and then by convection transfer through pore spaces, four years after the accident the bulk of the fuel materials had not migrated to layers lower than 2-3 cm, total radioactive contamination in the lower levels possibly being mainly attributable to mobile strontium from the fuel matrix. At the same time, layers below 20 cm contained less than 0.1% of total radionuclides on average. Thus, the quantity of radionuclides in the lower levels may be ignored (Fig. 2).

The total content of radioactivity in the intermediate 3-20 cm layer is mainly attributable to the effective quantity of mobile Sr-90 in the years following the accident. This constitutes approximately 10% of the total inventory of Sr-90 within the limits of the potentially floodable Pripyat lowlands in the Chernobyl NPP near zone. Likewise, mobile strontium entered the soil at an annual rate of 2-3%, depending on water levels during the year.

As is well known, since the accident (1986-1990) most of the territory under investigation has not experienced direct flooding. Accordingly, washout of radionuclides from the fuel particles was exclusively due to the influence of rain and melting snow. Total precipitation from spring 1986 to spring 1990 was approximately 600 mm (ignoring evaporation). The indirect integral estimate of Sr-90 washout in the investigated area from an inventory of approximately 12 000 Ci is in the order of 1.0×10^{-4} mm^{-1}. Within a margin of 20%, this estimate coincides with the value of the integral washout coefficient obtained at water-sprinkling sites.[10]

The ratio of infiltration flow to surface runoff from total precipitation on the floodplain was approximately 30:70. Accordingly, the waters flowing off the surface of the floodplain during the flood periods of the preceding years must have carried a considerably smaller quantity of radionuclides to the river than was localized in the floodland soils during the same period. This is confirmed by the results of experimental observations concerning the radionuclide migration balance on the floodplain.[11]

Since the exchangeable forms of Sr-90 considerably exceed the actual amount of initial fallout washed out - and accounted for 40-70% in 1990 - it seems that a large part of the Sr-90 is already potentially ready for water migration. It is likely that very soon the key factor in the water migration of Sr-90 will be the size of the layer of water runoff occurring during inundation of the floodplain.

In conditions of washout by infiltrating waters after rain, the period of contact between the water masses and the radioactive material in the surface layer is limited; this prevents equilibrium concentrations of exchangeable forms of radionuclides from forming between the liquid and solid phases (infiltrate-soil). During protracted flooding of the floodplain surface the picture is completely different. In these conditions the fuel particles may be in direct contact with water for several months, and the quantity of exchangeable strontium in the soil-water solution capable of transferring to the water is determined by the quantity of exchangeable ions in the river water able to substitute Sr-90 in first-order chemical reactions.

In this connection experiments on the kinetics of radionuclide removal from soils provide useful information.

These were conducted in natural conditions. Three soil monoliths with mean dimensions 70 x 40 x 20 cm were removed from the floodplain. They were placed in special basins, and after their sides had been made impermeable, Pripyat River water was poured onto them to give overlying water levels ranging from 5 to 20 cm in depth. Subsequently, water samples were periodically taken from the containers over a 45-day period (the water being stirred each time) and measured for Sr-90 and Cs-137 content. The findings were used to calculate washout coefficients, these being the ratio of the concentration of radionuclides in the water to the surface contamination of the monoliths at the time they were dug out.

On completion of the experiment the water was poured off, and the total quantity of Sr-90 removed into the water during submersion was calculated. A curve showing the kinetics of Sr-90 removal during the experiment is reproduced in Figure 7. The results show that during the experiments anything from 2.0% to 2.6% of the strontium inventory in the soil fallout transferred into aqueous solution above the flooded soil. These results are 25% to 30% higher than those pertaining to another floodplain area for 1986 (reproduced in [10]). While in the 1986 experiments the washout coefficient for Sr-90 from floodplain soils (standardized for flooding depth) ranged in some cases from 3 to 13 x 10^{-5} mm^{-1} after 35 days, in our experiments the values for this coefficient after flooding the monoliths for 35 days ranged from 16 to 30 x 10^{-5} mm^{-1}. This tallies with data showing a 2.5- to 3.5-fold increase in the quantity of exchangeable Sr-90 in the period between these experiments. The results showed that during the experiment period no equilibrium concentration of the radionuclide formed in the soil solution. In contrast to experiments with sprinkling, where such an equilibrium concentration was observed very soon, in this case the process by which Sr-90 entered the water was largely determined by the diffusion flow of the radionuclide out of the flooded soil. Additional flooding of the soil monoliths over different periods of time showed that the average rate of mass exchange of exchangeable Sr-90 radionuclides amounts to 0.15-0.20% per day of the Sr-90 inventory in the near zone floodplain. Consequently, in conditions of extreme flooding lasting over 40-50 days, not more than 10% of the Sr-90 inventory in the investigated territory can enter the river waters. Nevertheless, this quantity can substantially increase the radiostrontium content of the waters of the River Pripyat and Kiev reservoir and cause highly undesirable radioecological problems. In this connection the conclusion drawn was that it is essential to isolate this particular area of the floodplain from direct contact with the river waters and take steps to dry the area, which would make it possible to substantially reduce the speed of migration of radionuclides into the aquatic system of the Dnieper basin.

CONCLUSIONS

1. 90%-95% of the contamination of the Pripyat floodplain in the northern part of the Chernobyl NPP near zone (radius of 10-15 km) was caused by fuel material with a nuclide composition corresponding to irradiated fuel from the 4th/5th group of fuel element assemblies. The mean geometric diameter of the particles was 8.0 microns for soil samples and 14.0 microns for bed sediment. The median diameter of the particles was on average 20%-25% greater than the estimated values, suggesting that there was an oxidization-induced change in the stoichiometric composition of the particles and a reduction in their density to approximately 7-8 g/cm^3.

2. The bulk of the particles (70-80%) are black and friable. Regularly shaped particles are also encountered, and these are firm and have a characteristic metallic sheen. Ru-106 is responsible for most of the activity of these particles. Pure ruthenium particles account for 5-10% and pure caesium for 2-3%.

3. The vertical distribition of the particles in the soil fits an exponential curve, both for quantity and size. Particles larger than 1 micron were not discovered below 5-6 cm in soil and below 7-8 cm in bed sediment.

4. In natural conditions in soil there is a change over time in the fine fraction of the fuel particles (under 2 microns), their share being down by 5-10% in 1989 compared with 1987. Apart from the processes of physical and chemical degradation of the fuel material, processes of mechanical break-up also occur with the formation of colloid fractions (under 0. 1 micron).

5. Since the accident, as a result of mechanical and chemical transformation of the fuel matrix, there has been a gradual increase in exchangeable forms of the radionuclides in the soils. Thus, while water-soluble and exchangeable forms taken together in the floodplain territory accounted for 30-40% in 1989, in 1990 they accounted for 70%.

6. An analysis of the kinetics of Sr-90 removal into water during inundation of the floodplain fuel fallout zone showed that in the event of full flooding during extreme high water, only 10% of the inventory of Sr-90 exchangeable forms (making up 50-70% of the total) can enter the river water.

7. Bearing in mind the critical conditions obtaining for a possible increase in Sr-90 migration from the fuel fallout zone within the floodplain, there is a need for urgent measures to prevent the washout of radioactive substances.

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Fig.1: Sr-90 CONTAMINATION IN CHERNOBYL NPP NEAR ZONE
(Ci/km²)

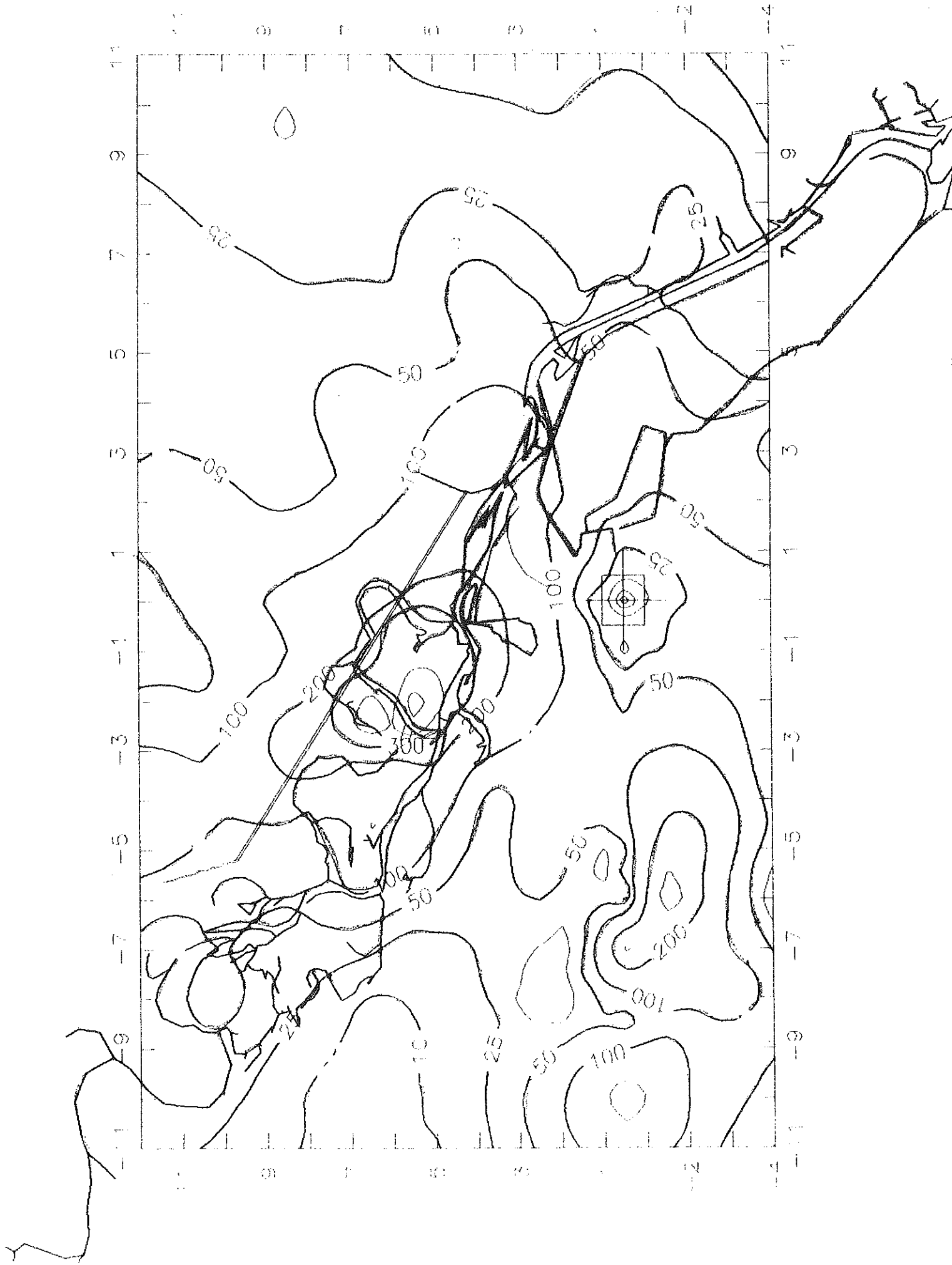
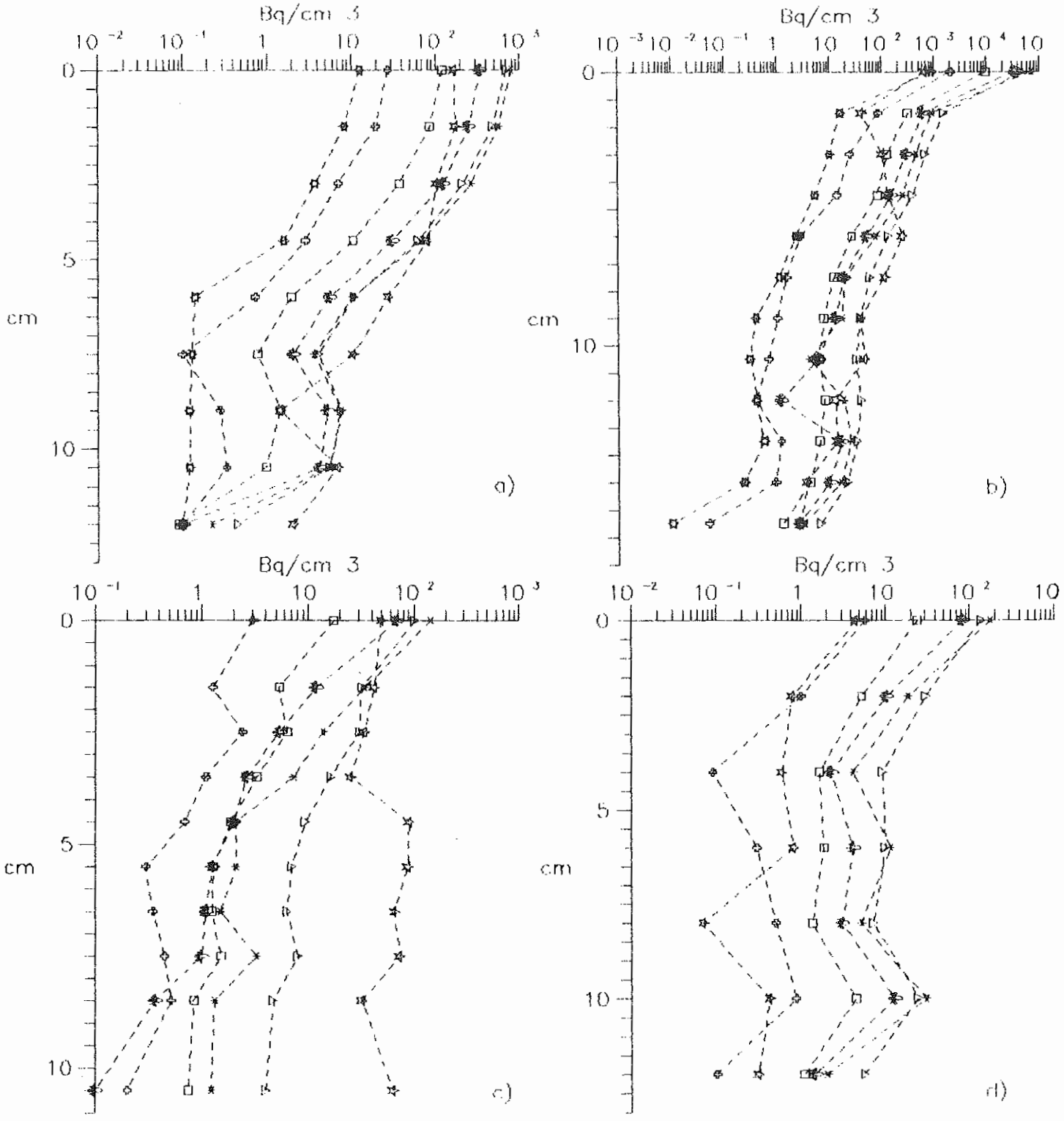


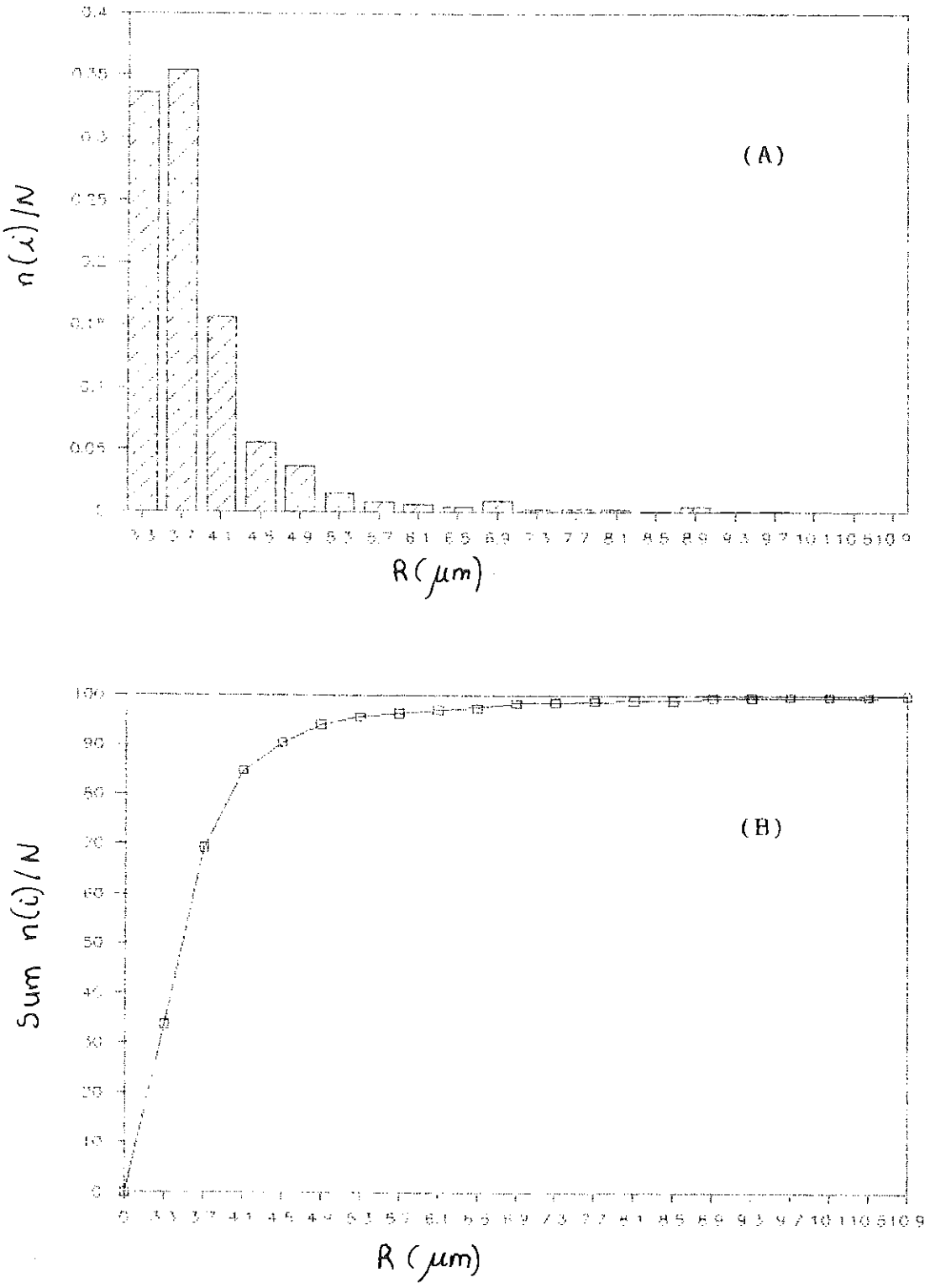
Fig.2: VERTICAL DISTRIBUTION OF RADIONUCLIDES IN SOIL AND BED SEDIMENT IN THE FLOODPLAIN (ZONE CLOSE TO CHERNOBYL NPP) 1989



a - dry soil
b - abnormal fallout
c - moist soil
d - bed sediment of lake

Ce-144 Cs-134 Cs-137 Ru-106 Sb-125 Eu-154 Sr-90

Fig.3: TYPICAL PARTICLE SIZE DISTRIBUTIONS FOR "HOT" PARTICLES OF SOIL TAKEN FROM THE PRIPYAT RIVER FLOODPLAIN IN THE CHERNOBYL NPP NEAR ZONE



A - Size distribution
B - Cumulative frequency (in %)

Fig.4: VARIATION OF "HOT" PARTICLES DISTRIBUTION AT DIFFERENT TIMES AFTER THE CHERNOBYL ACCIDENT
SAMPLES TAKEN FROM SOIL 10 km NORTH OF THE NPP IN 1987-1989

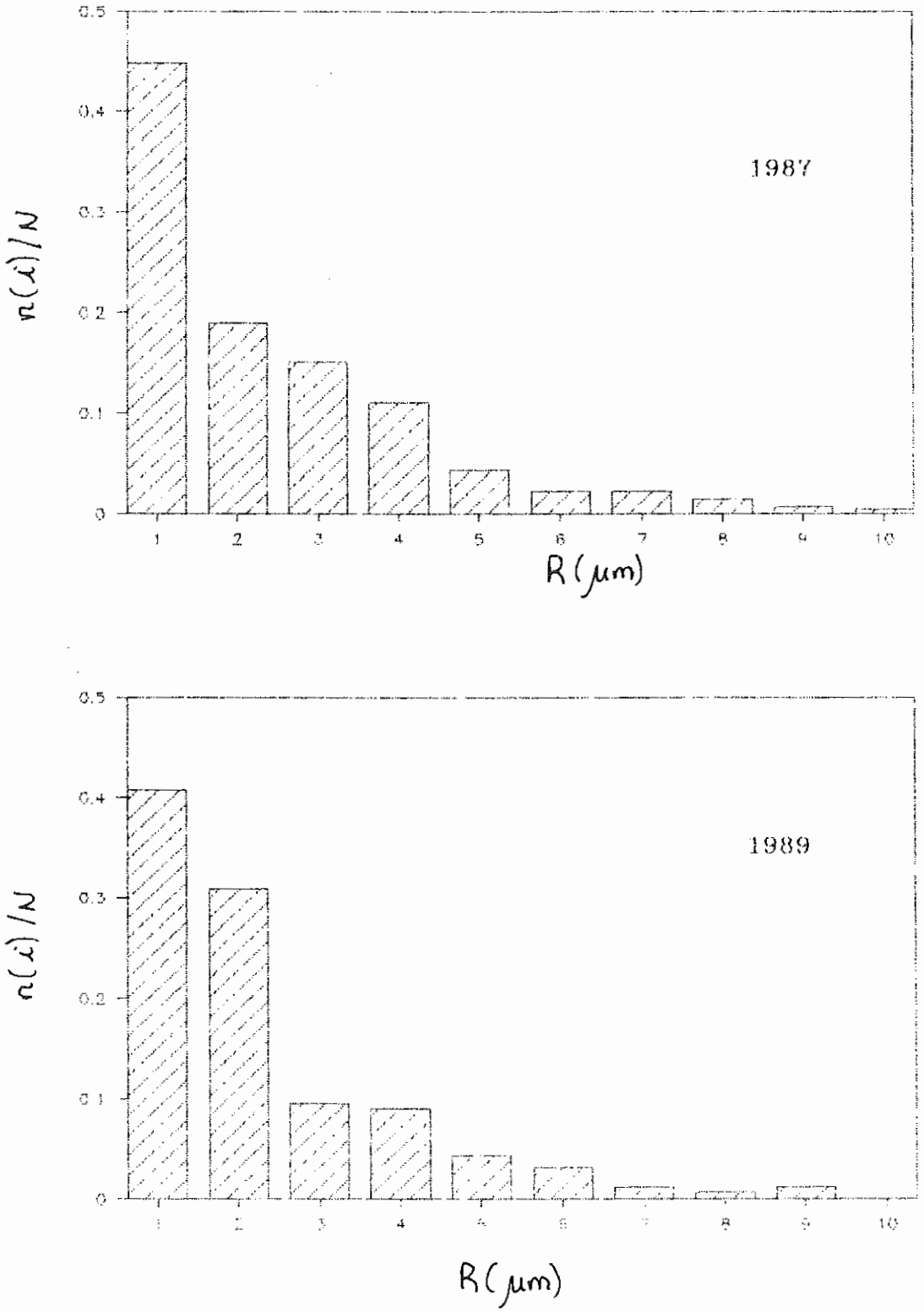
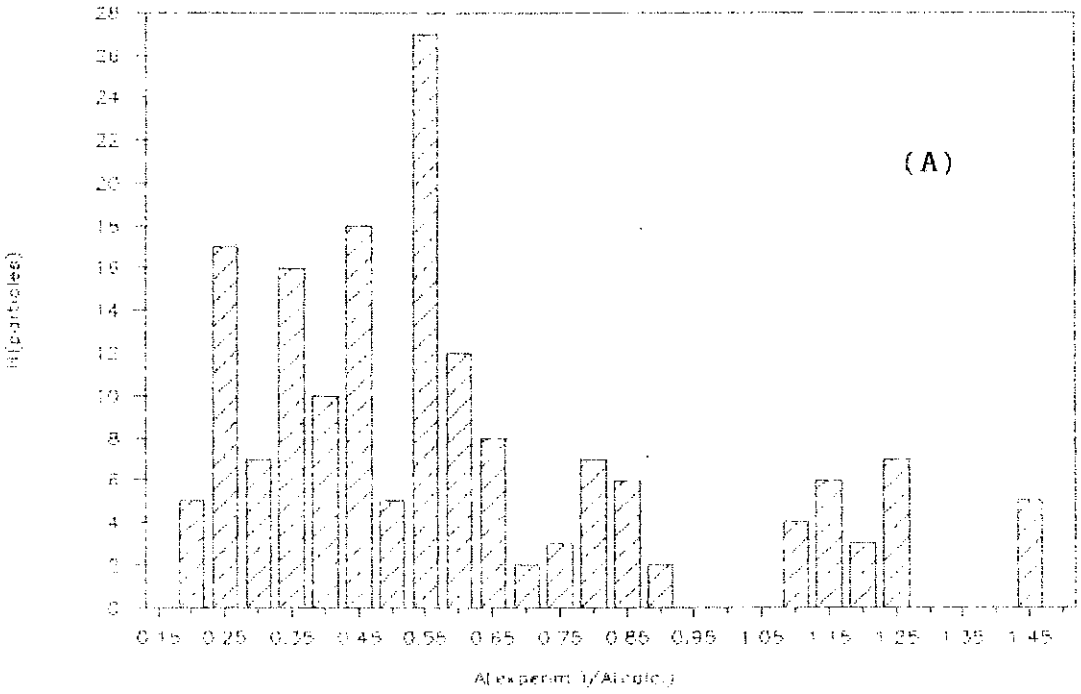


Fig.5: DISTRIBUTION OF ACTIVITY RATIOS FOR RADIONUCLIDES IN "HOT" PARTICLES. A (experimental) / A (calculated)
ZONE 10-15 km NOTH OF CHERNOBYL NPP. 1989

A: Cs-134 B: Cs-137

Distribution of activity ratios (Ae/Ac) for Cs-134



Distribution of activity ratios (Ae/Ac) for Cs-137

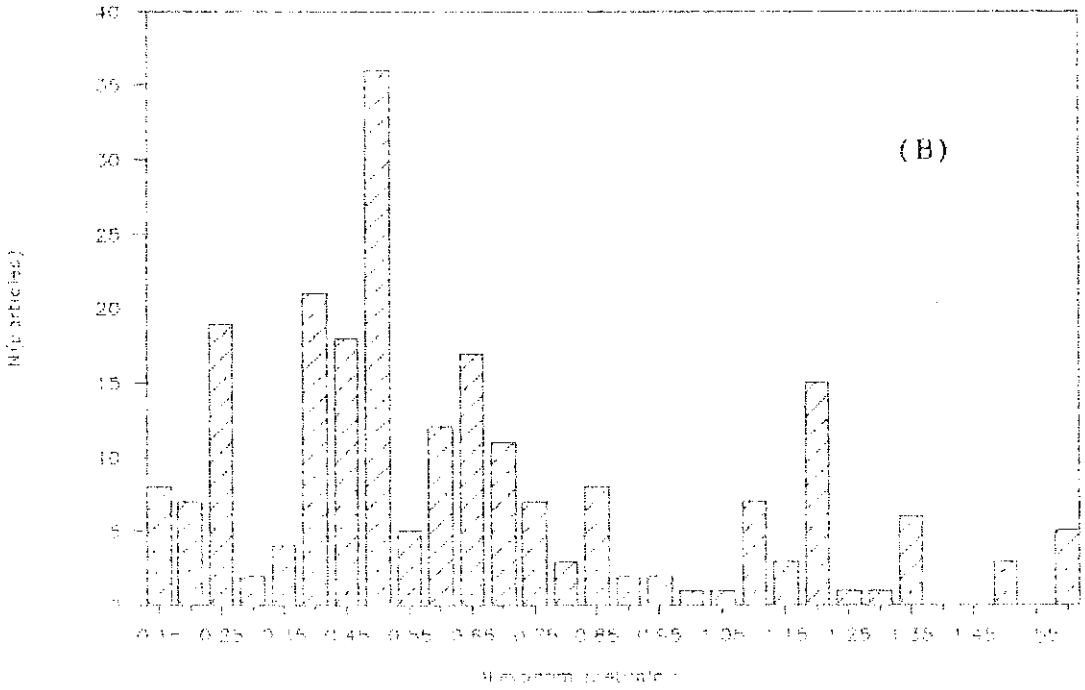
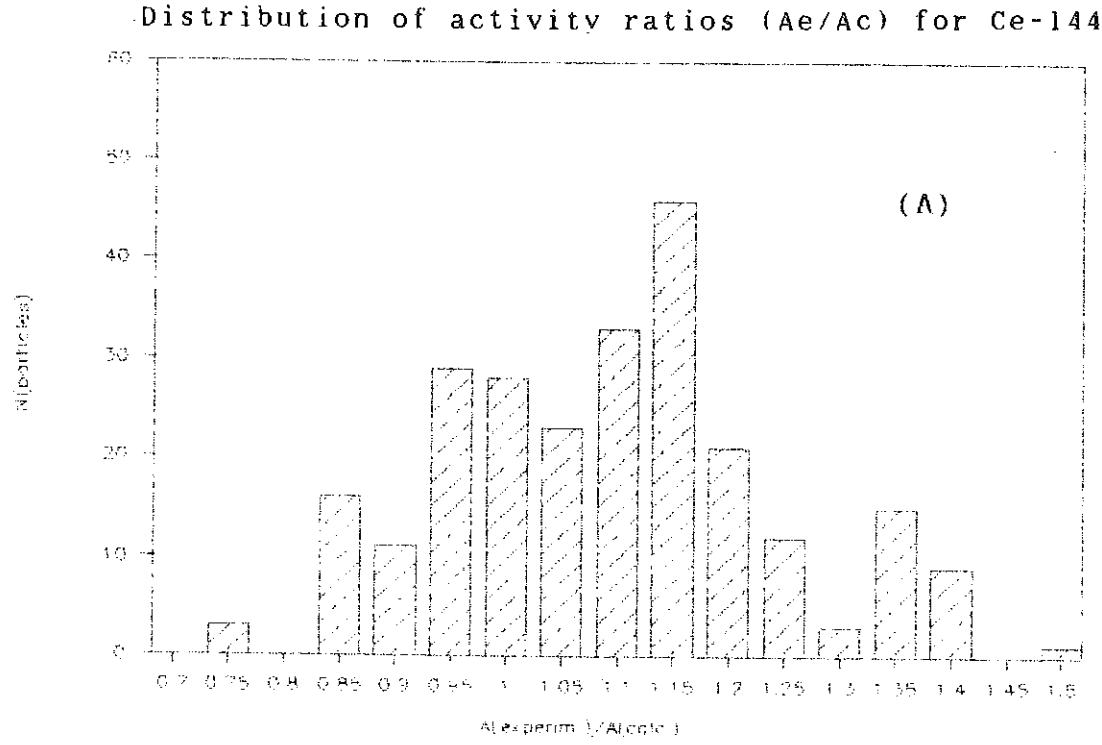


Fig.6: DISTRIBUTION OF ACTIVITY RATIOS FOR RADIONUCLIDES IN "HOT" PARTICLES, A (experimental) / A (calculated)
ZONE 10-15 km NOTH OF CHERNOBYL NPP, 1989

A: Ce-144 B: Ru-106



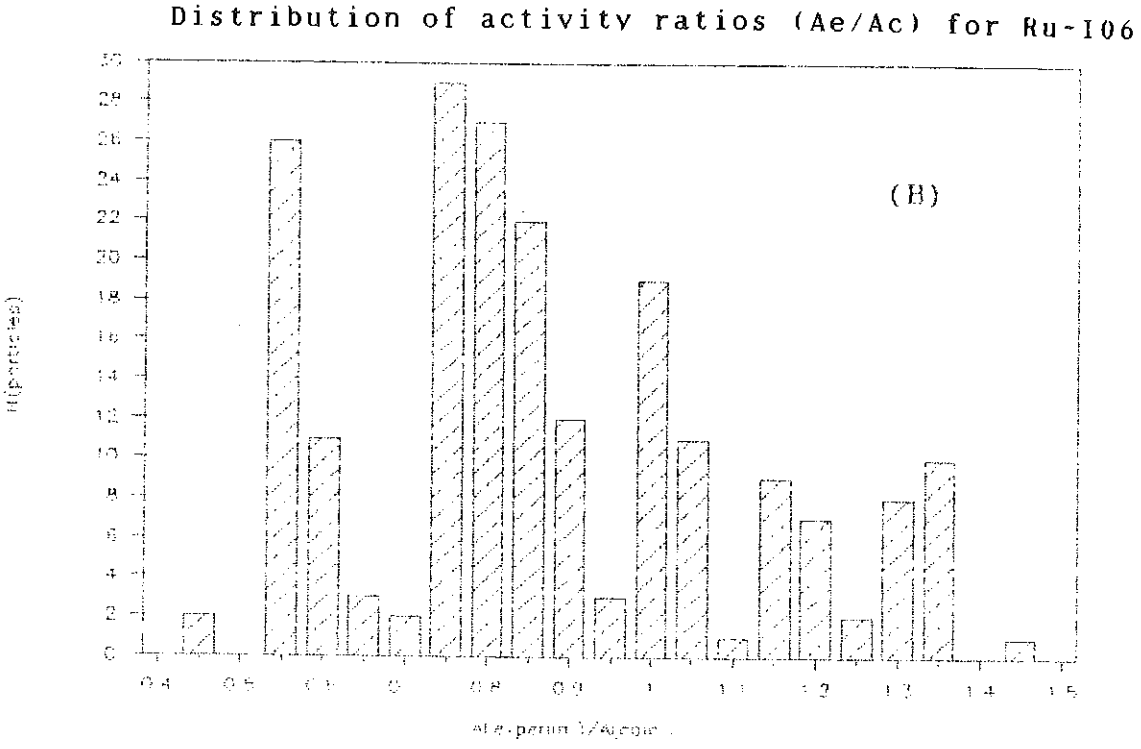
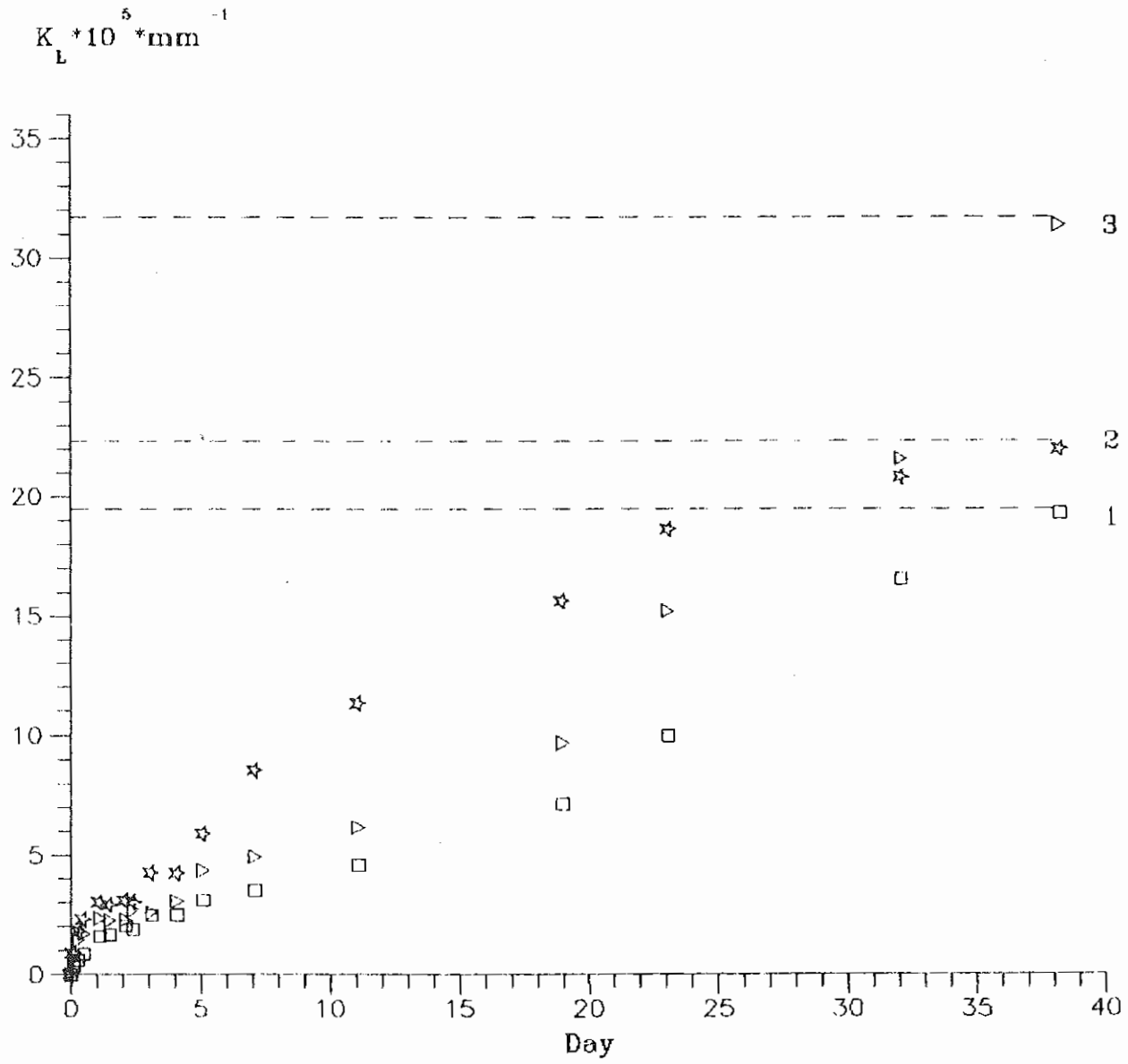


Fig.7: KINETIC CURVES OF RELEASE OF Sr-90 FROM SOIL AFTER WATERING



K : Liquid Washout coefficient standardized to run-off layer
1,2 : Sandy /Silty soils; 3 clay soils

Forms of Radioactive Fallout and Mobility of Radionuclides in Ukrainian Aqueous Landscapes

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ABSTRACT

The mechanisms underlying the kinetics of caesium-137 and strontium-90 washout from bed sediments were studied. Migratory radiostrontium was shown to be present mainly in the form of cations, and radiocaesium was present in both mineral and organic forms bound to both humic acids and oil products. Radiocaesium migration in surface waters was mainly technogenic in character.

Three illustrations; one table; bibliography: nine references.

As a result of the accidental release of radioactivity at the Chernobyl NPP in 1986, and the processes of surface runoff from contaminated catchment areas since then, bed sediments in water courses and reservoirs in the Dnieper Cascade are now a repository for large amounts of long-lived radioisotopes of caesium and strontium. In 1989 it was estimated that the bed sediments in the Dnieper Cascade reservoirs contained 1.5×10^{14} Bq of caesium-137, and that the Kiev reservoir contained 1×10^{14} Bq of caesium-137 and 1.5×10^{13}Bq of strontium-90. The subsequent behaviour of these radionuclides is governed by a range of natural processes; for instance, the overlaying of radioactive sediments with new, less contaminated layers reduces the availability of radionuclides for transfer into the water. The reverse process occurs simultaneously; silty waters, containing water-soluble forms of the elements found in the sediments, form in the initial stages of diagenesis as a result of compaction of the upper layers. The presence of an "upwards" transfer process determines the need for a detailed investigation of the forms of occurrence of radionuclides in bed sediments, followed by an assessment of the scale of secondary contamination of surface waters.

The attachment of radionuclides to the bed sediments of water bodies is mentioned in many papers: in connection with global fallout [1], with liquid radioactive wastes from enterprises involved in the nuclear energy cycle [2, 3] or with the operation of nuclear power plants using cooling ponds [4].

However, the fallout from the Chernobyl accident has specific characteristics, since the catchment areas and surfaces of rivers in the near zone (Pripyat, Uzh, Dnieper and others) and the northern section of the Kiev reservoir were contaminated chiefly by fission-produced radionuclides contained in the uranium dioxide matrix. As a result of this particular characteristic, each radioisotope can be liberated during the course of the following processes:

a) egress from the matrix (through diffusion or dissolution of the matrix substance);
b) interaction with the sediment matter (sorption or formation of chemical compounds);
c) migration of dissolved forms through mixing of silty and surface waters.

Naturally, the speed at which the successive processes occur is determined by the speed of the slowest stage, which, initially, constituted the processes taking place at the phase boundary between fuel matrix and sediment. At the present time, the accumulation of migratory forms of radionuclides in bed sediments indicates retarded mixing of interstitial solutions with the main mass of water. Removing these kinetic limitations under laboratory conditions through constantly mixing sediments with fresh water allows us both to establish the speed of desorption for migratory forms of radionuclides, and to estimate the parameters of the process governing the egress of radioisotopes from dispersed nuclear fuel.

Extensive information has now been accumulated relating to the composition and particle dimensions of radioactive fallout in the near zone. There is a significant amount of carbon (graphite) - up to 15% - in the uranium dioxide particles, and the distribution of particles by size in relation to distance from Reactor No 4 is log-normal. Data from radiochemical and instrument studies indicate significant oxidation of the uranium dioxide as a result of the high temperatures produced during the accident.

These factors are what cause the significant instability of the dispersed nuclear fuel in water. According to the authors' original calculations, alpha-radiolysis of the water and subsequent oxidation of uranium (+4) to uranium (+6) engender the disintegration and dissolution of approximately 3% per year of the uranium matrix.[5] Clearly, the diffusion mechanism contributes significantly to radionuclide egress from uranium dioxide, particularly for particles with dimensions measured in microns. Starting from a diffusion coefficient value of $D = 1 \times 10^{-19}$ cm_2/sec, as determined for the diffusion of inert gases from irradiated uranium dioxide at temperatures below 600°C [6], it is not difficult to estimate the likely scale of radionuclide egress. Thus, in the case of 2 μm-diameter particles of UO_2 the losses over three years of fission-produced radionuclides through diffusion amount to over 1%.

Naturally, using the diffusion parameters for inert gases will give very high values for the diffusion of radionuclides of caesium and strontium since, in contrast to inert gas atoms, these radionuclides are capable of dissolving within the molecular lattice of uranium dioxide. However, given the possible decrease in the density of the crystal lattice of UO_2 due to high-temperature carbon penetration and partial oxidation to U_3O_8, use of the parameters mentioned above is sufficiently justified.

Because both processes occurred simultaneously over four years, there was a significant redistribution of radioisotopes from the initial fallout particles into bed sediments. Thus, the processes of radionuclide interaction with the mineral and organic components of the sediments now play an increasingly decisive role in the formation of fixed and migratory forms of radiocaesium and radiostrontium. The dominant role played by natural layered minerals in fixing fission-produced radionuclides is widely known, and no explanation is necessary here. Quartz, with its characteristic cryptocrystalline and amorphous diversity, plays a significant role. As for the problems of interaction between technogenic radionuclides and natural organic substances, insufficient research has been done in this area. The paper listed at [7] shows that the fulvic and humic acids washed out of soils are capable of forming stable complexes with many metals present in macroconcentrations.

The authors of the report listed at [8] investigated the molecular and mass composition of organic compounds separated from soil using the radio gel chromatography method. Until now, however, the literature has not contained much information on the processes determining the kinetics of technogenic radionuclide washout from soils and bed sediments.

The aim of this paper is to investigate these processes as they relate to caesium-137 and strontium-90 washout from bed sediments.

Our attention focused on bed sediments from the floodplain of the Pripyat river (sample P5), silts from the northern sector of the artificial cooling pond at the Chernobyl NPP (sample R5), and silted sands taken from the southern sector of the same body of water (sample R1). Radioactive contamination in silts from areas adjoining the power plant site was mainly in the form of dispersed reactor fuel; this was clearly indicated by the results of mass spectrum analysis, as the uranium separated from the samples has an isotopic composition which differs from that of natural uranium. By means of the relationship

between the fission reactions of ^{235}U and the capture reaction of ^{235}U (n,γ) and ^{236}U, fuel particles showing a 50% ^{235}U burn-up (the initial amount being 1.9%) were identified in the separated fraction. The "fuel" origin of the fallout on the floodplain is also confirmed by the results of isotopic analysis of the uranium, the atomic ratio of ^{238}U and ^{235}U in the samples analysed being 87-92 [9].

The samples were processed over the course of 28 hours using water of natural hydrochemical composition, a check on the amount of organic substances and radionuclides washed out being made every 3 hours. The radionuclide composition of the samples was determined by a germanium-lithium semiconductor detector after passing the solution through a membrane filter with a pore diameter of 0.2 μm, evaporating it and then using radiochemical means to determine the strontium-90 content. Infrared spectra of dried aqueous extracts were obtained using the UR-20 infrared spectrometer on the fourth slit programme. 0.1 N solutions of ammonium acetate and sodium hydroxide were used to extract the ion-exchangeable forms of radionuclides and organic components.

The results obtained for radiostrontium leaching in kinetic conditions are the most suitable reflection of processes on the matrix-sediment phase boundary, since strontium ions are the ones least subject to sorption by silt components. The calculated speeds of strontium-90 egress are given in Fig. 1. In interpreting the results, the concept of two processes occurring simultaneously was used - a) the transfer of radiostrontium into solution from water-accessible positions through the ion-exchangeable desorption mechanism, and b) egress from the matrix through diffusion and simultaneous dissolution of the "hot" particle substance. Superimposing these processes yields the differential curves shown in Fig. 1, the downward portions of which are caused mainly by desorption, while the horizontal portions reflect the egress of strontium atoms from the matrix. These experimental curves are a long way from providing an ideal picture of the superposition of the two independent processes, reflecting the diversity of actual strontium desorption processes. The lowest V value should be used to evaluate the speed of egress of radionuclides from dispersed fuel particles. The large variation in the V_{min} values for samples R5 and P5 may be explained by the difference in the fallout particle sizes; typical particle sizes for the area where sample R5 was taken went up to 200 μm, while the average diameter in the area where sample P5 was taken was 3-5 μm. Bearing in mind that the amount of oxidised forms of uranium contained is in proportion to the surface area of the particles, it is not surprising that the matrix egress speeds for radiostrontium should be so high, or that the degree of leaching should be so significant (up to 50%) (Fig. 2a, curve 1).

The amount of mobile forms of radiostrontium found in all the samples examined does not correlate with the samples' organic substance content (see Table), which leads us to suggest that strontium is present mainly in the form of cations. Radiocaesium egress is governed by other processes.

The highest degree of caesium-137 egress is noted in soil with the lowest organic substance content (losses through calcination 1.8%) (Fig. 2b, curve 3), the smallest in silts with the highest organic substance content (losses through calcination 23.3%) (Fig. 2b, curve 2). Bed sediments taken from the Pripyat river floodplain (P5) lie between these two extremes in terms of caesium-137 and organic compound

content (Fig. 2b, curve 1). There is a relationship between losses through calcination and the distribution of this radioisotope by forms of occurrence. Thus, in sample R1 (smallest losses through calcination, i.e. 1.8%) the largest quantity of caesium-137 is found in the water-soluble (4.6%) and ion-exchangeable forms (9.6%), while the smallest amount is bound to humic and fulvic acids (see Table). For the sample R5 (with the greatest losses through calcination, i.e. 23.3%) the content of water-soluble forms of radiocaesium is lowest (0.03%), caesium-137 is not observed at all in ion-exchangeable form, and practically all the radioisotope is bound up with humic and fulvic acids.

It may be assumed that caesium-137 is found in two forms, mineral and organic, in contrast to migratory strontium-90, which is mainly found in ion-exchangeable form.

On the basis of the infra-red spectra obtained, which show the presence of functional groups of organic compounds with which radionuclides bind, a correlation was found between the intensity of the C-O and C-C bonds in organic compounds and the egress of caesium-137.

Fig. 3 (a, b) shows the relative egress of caesium-137 and the relative intensity of the C-O and C-C bond regions after processing a sample with water. Infra-red spectra from consecutive sample processing show that the C-C and C-O regions can belong to humic and aromatic acids. It was shown that the curve shapes in Fig. 3a fully coincide with the curve shapes in Fig. 3b; however, in the case of sample P5 (curve 1) the influence of the mineral fraction is apparent, since Si-O groups also absorb in the region 1 000 - 1 100 cm^{-1}.

It has been established that radiocaesium dissolved in water also binds to oil products, and in conditions of intensive technogenic loading its migration in surface waters is essentially technogenic in character. Study of the kinetic processes involved established the parameters of radionuclide egress from nuclear fuel, and also allowed an estimate to be made of the desorption speed and composition of migratory forms of radionuclides. It has been shown that migratory radiostrontium is mainly found in the form of cations, while radiocaesium is found both in mineral and organic forms and is combined with humic and aromatic acids.

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Table 1: DISTRIBUTION OF RADIONUCLIDES BY FORMS OF OCCURENCE IN BED SEDIMENTS

Sample reference	P-5				R-1				R-5			
Losses through calcination %	12.8				1.8				23.3			
Forms of occurence	RADIOISOTOPES DISTRIBUTION BY FORMS, %											
	Ru-106	Cs-134	Cs-137	Sr-90	Ru-106	Cs-134	Cs-137	Sr-90	Ru-106	Cs-134	Cs-137	Sr-90
Water-soluble	1.1	1.1	0.8	6.9	-	6.8	4.6	1.7	1.0	0.08	0.06	0.62
Non-exchangeable	-	0.72	0.54	25.4	-	9.4	9.6	8.8	-	-	-	1.25
Humic acid	4.4	0.64	0.27	0.09	-	-	0.5	3.0	0.07	0.1	0.6	0.05
Fulvic acid	1.3	0.86	0.07	5.8	-	0.97	0.65	0.8	0.05	0.03	0.15	0.14

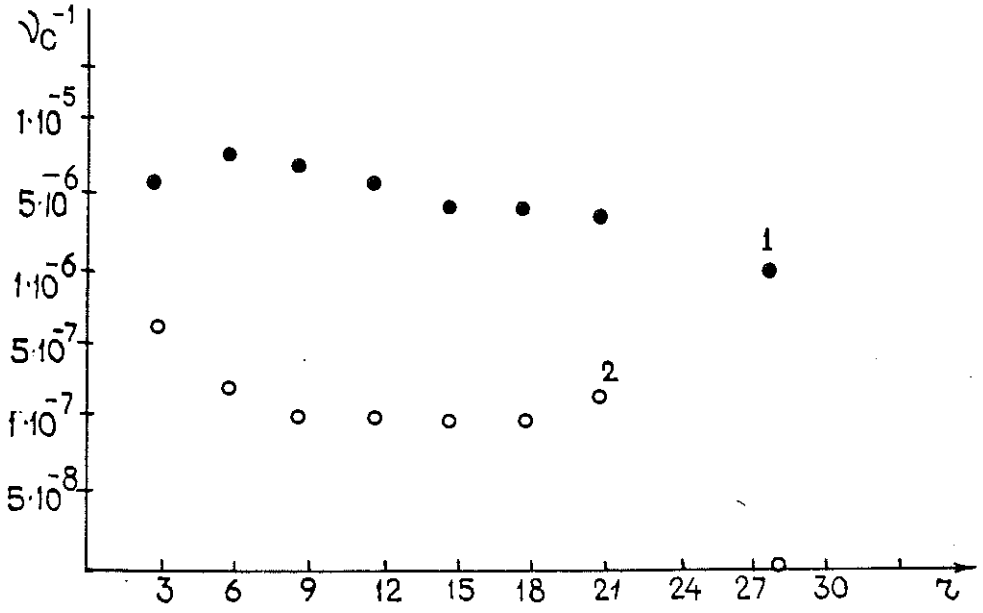


Fig. 1 Speed of strontium-90 egress from bed sediments:
(1) Floodplain (2) Cooling pond

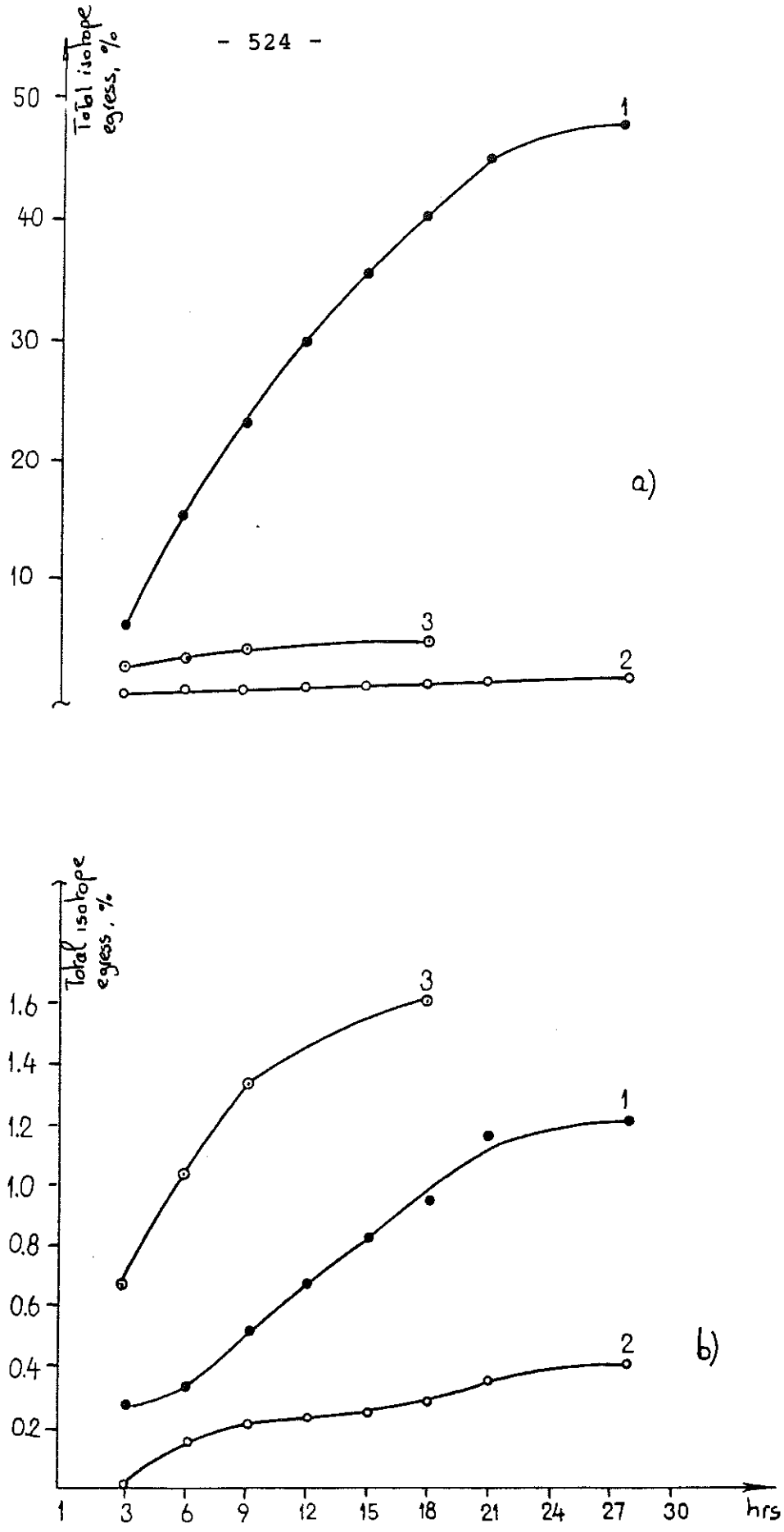


Fig. 2 Dependence on sample-processing time of overall egress of water-soluble strontium-90 (a) and caesium-137 (b): 1 = P5, 2 = R5, 3 = R1.

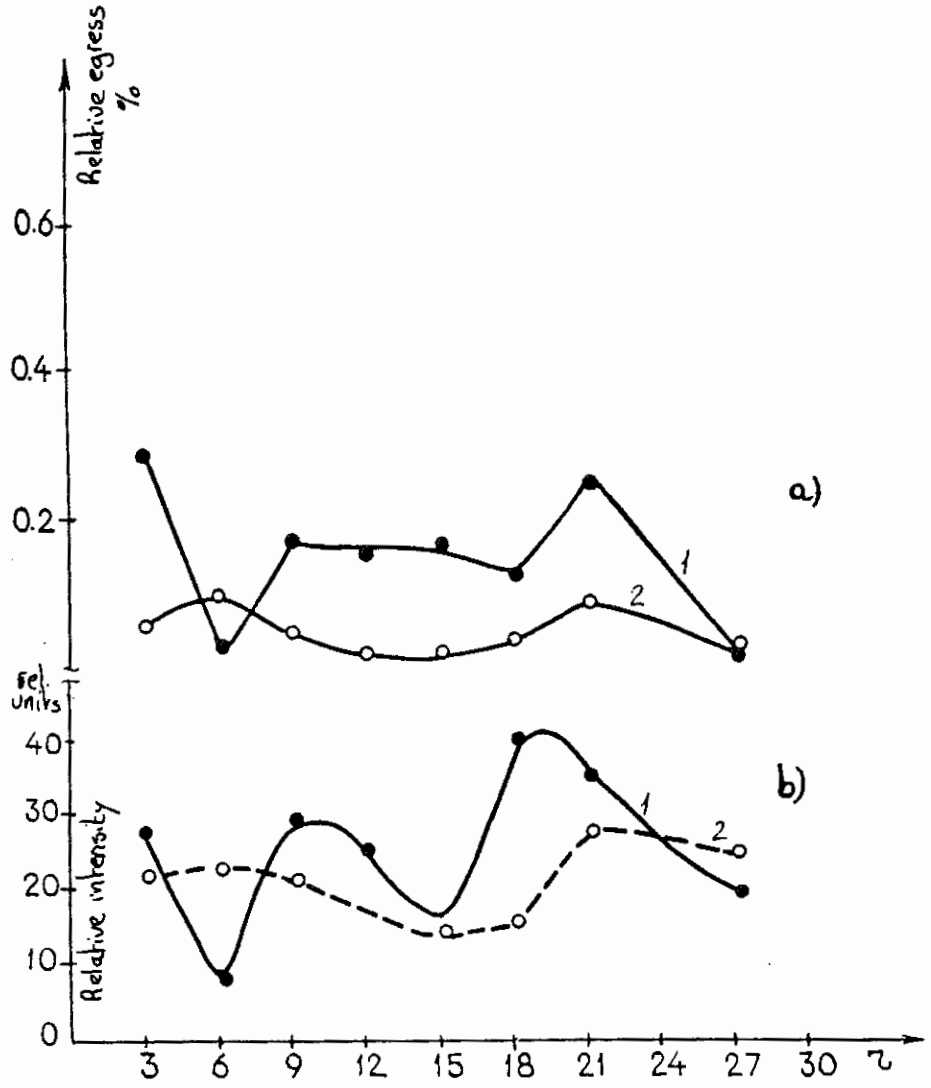


Fig. 3 Interrelationship between the relative content of water-soluble caesium-137 and the structure of organic compounds:
(a) Relative content of caesium-137
(b) Relative intensity of C-C and C-O bond regions. For key see Fig. 1.

Hydrological Aspects of Radionuclide Migration in Water Bodies Following the Chernobyl Accident

G.V. VOYTSEKHOVICH, V.A. BORZILOV, V.A. KONOPLEV

ABSTRACT

The variety of secondary effects determining migration processes stands out as the main specific feature of the radioactive contamination of water bodies following the Chernobyl accident. Of these the paper highlights the transformation processes of the various physico-chemical forms of radioactive fallout products as observed in catchment areas and water bodies, such processes taking place in different geochemical and hydrological conditions depending on the landscape. The following are considered: dynamics of physico-chemical forms of radioactive fallout, changes in the contamination pattern of soils and water-body bed sediments following the accident and - as a result of runoff formation in these areas - the radioactive contamination regime of the Pripyat and Dnieper rivers between 1986 and 1990.

The paper presents experimental data for evaluating retention factors for waterborne migration of Chernobyl radionuclides in surface washout, via seepage waters in soils, in silt solutions of bed sediments and via transport of suspensions in rivers and reservoirs. It also considers the different approaches to evaluating these parameters and the field research methods.

We consider the methods used in field studies of washout mechanisms and mass transfer parameters for water-soluble, exchangeable forms of radionuclides and contaminated particles of soil and bed sediments in runoff and floodplain flows, and provide data on the kinetics of these processes. We also analyse the observed processes of flow purification through sedimentation, and the role of catchment-area and river-channel load in the transport and deposition of radionuclides such as ^{137}Cs, ^{90}Sr and total Pu. The paper provides transport and accumulation balances for Chernobyl radionuclides in the large reservoirs of the Dnieper Cascade. It also takes a close look at the specifics of ^{90}Sr migration in Dnieper river water systems following the Chernobyl accident, comparing them with the specifics of ^{90}Sr dispersion in the river network and lakes following the Kyshtym accident.

INTRODUCTION

On 26 April 1986 a serious nuclear accident took place in the fourth unit of the Chernobyl NPP, destroying the reactor and its housing, and releasing into the atmosphere approximately 3.5% of the fission and activation products accumulated in the reactor.[1, 2]

A cloud formed and the fallout of aerosols from this left a radioactive trail in areas to the west and north depending on the meteorological conditions governing air mass transfer. From 26 April to 8 May 1986 the continuing discharge of a jet of gaseous, volatile and aerosol products led to the formation of what are known as the near and distant zones of radioactive fallout.

Data on the daily releases into the atmosphere [1] make it possible to determine the composition of the release and the radionuclides' physico-chemical properties, which changed during the time the various branches of the trail were being formed. For this reason, the composition of the radionuclides in the fallout on the territory studied was heterogeneous and depended on direction and distance from the NPP.

The shock wave of the Chernobyl explosion, the temperature gradients and oxidation of the nuclear fuel (uranium dioxide - UO_2) led to the formation of "hot" fuel particles, over 90% of which, with an activity of 10^{-9} Ci, had a radionuclide composition similar to that of spent fuel and whose content of highly mobile fission products varied. [3] The ingress of hot fuel particles into the environment is the main feature of contamination from the Chernobyl accident. The destruction of the fuel elements and the "annealing" of the nuclear fuel released into the atmosphere a considerable quantity of volatile fission products as well, some of which condensed on the inert carrier particles of the aerosols: mixtures of dust, construction materials, etc. The "hot" particles formed in the accident had properties similar to those formed in the last stage of a nuclear explosion, their surface contamination level and specific activity being lower than those of fuel particles.

The condensed particles formed as a result of the Chernobyl accident are similar to the global fallout of radionuclides after nuclear weapons tests, and it is therefore possible to predict their behaviour in water bodies to a reasonably accurate degree. The behaviour of the fuel particles, however, which are the unique feature of the Chernobyl "trail" and are concentrated primarily in the 60-km zone around the Chernobyl NPP, presents major scientific problems.

Most of the radioactive fallout was deposited on the catchment areas of the Pripyat, Dnieper and Desna Rivers, which are the main ones feeding the reservoirs in the Dnieper Cascade (Fig. 1). The radioactive trail has three distinct branches (northern, southern and western) which cover the southern rayons (districts) of Byelorussia, the western part of the Bryansk oblast of the RSFSR and the northern and central oblasts (regions) of the Ukraine. Estimates of the content of ^{137}Cs and ^{90}Sr in the catchment areas of the main tributaries of the River Pripyat and the Upper Dnieper are shown in Table 1. The accident contaminated the water bodies in two stages: via direct fallout and as a result of various secondary radionuclide migration processes via aquatic pathways.

Table 1

^{137}Cs and ^{90}Sr content

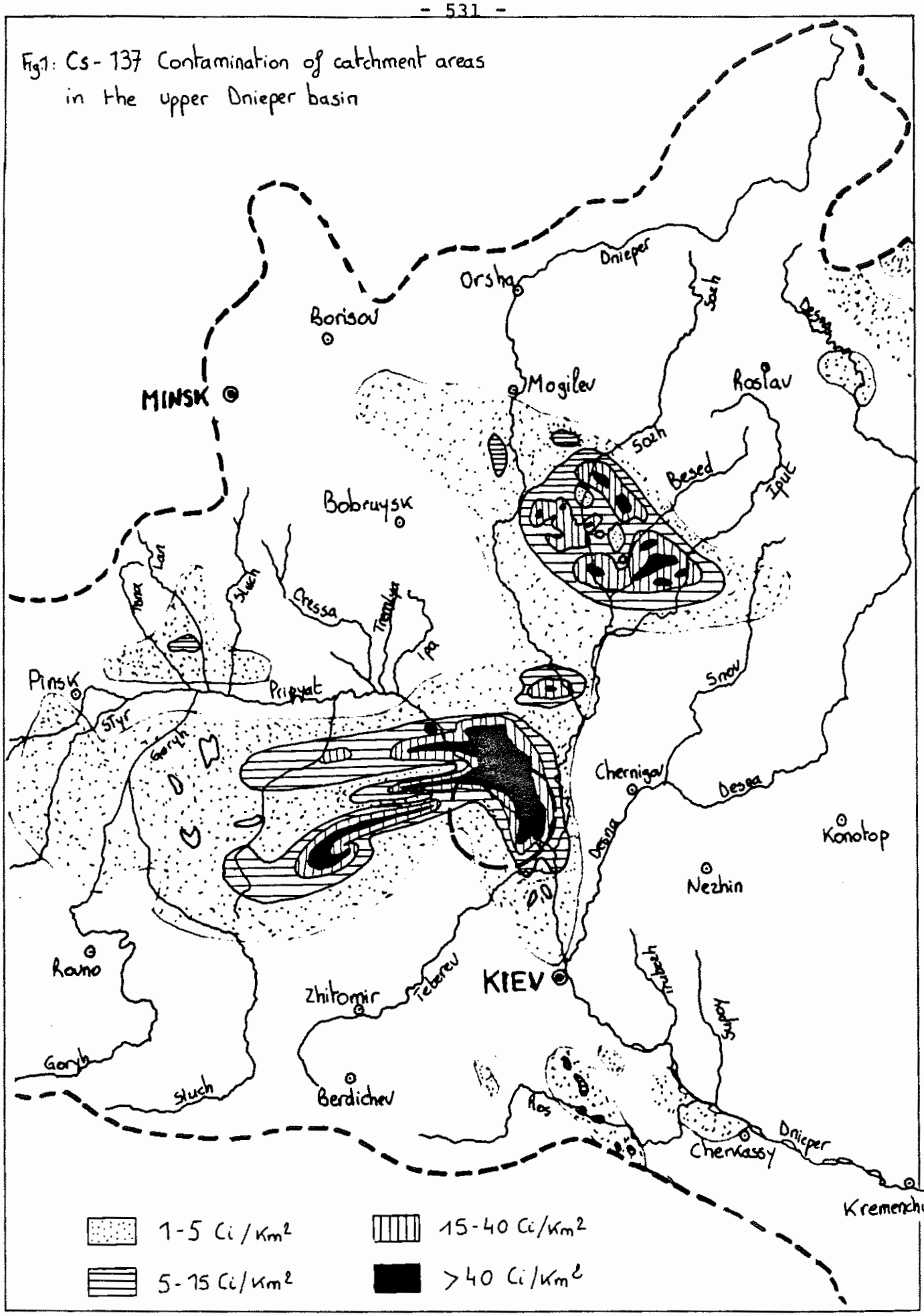
in the catchment areas of the main rivers/tributaries

feeding the Kiev Reservoir

River	Catchment area in thousands of km^2		Content of radionuclides in Ci	
	basin	A>1 Ci/km^2	^{137}Cs	^{90}Sr
Dnieper mouth	105	29	275	6
Pripyat mouth	115	27	180	42
Braginka and interfluve	2	2	55	12
Desna mouth	89	61	8	<1

* The radioactive contamination of the Chernobyl NPP site was not taken into account when evaluating the radionuclide content.

Fig.1: Cs-137 Contamination of catchment areas in the upper Dnieper basin



1. PRIMARY CONTAMINATION OF WATER BODIES

Immediately after the accident the water bodies were radioactively contaminated by direct fallout of accident aerosols onto the surface of rivers, reservoirs and seas. Table 2 shows the maximum concentrations of certain radionuclides in the water during the observation period (which started on 01.05) in various rivers, where water contamination levels changed considerably up to the end of the fallout period.[4] The total beta-activity of the Pripyat River fell from 10^{-7} Ci/l in the initial days after the accident to 4-6 x 10^{-9} Ci/l by the end of May. The maximum concentrations of ^{239}Pu observed in the Pripyat River in the initial days after the accident (10^{-11} Ci/l) fell to 2 x 10^{-13} Ci/l by August, which is four orders of magnitude lower than the maximum permissible level. Up to 98% of this was found insuspensions and bed load. For this reason, attention was then focused on studying the water migration processes of long-lived isotopes such as ^{90}Sr and ^{137}Cs.

Table 2

Radionuclide content in rivers in the initial period following the Chernobyl accident [4]

River	Date sample collected	Radionuclides content (10^{-9} Ci/l) ^{144}Ce	^{141}Ce	^{131}I	^{103}Ru	^{137}Cs	^{95}Zr	^{140}Ba
Pripyat	1.05.86	10	11	57	15	6.7	11	38
Chernobyl	2.05.86	--	--	120	22	15	42	60
	6.05.86	--	2.4	22	4.6	43	4.5	4.5
	3.06.86	--	--	0.9	0.7	0.6	0.3	--
	16.07.86	1.0	0.4	--	0.4	0.2	1.0	--
Teterev	3.05.86	--	18	54	19	--	39	34
Irpen	6.05.86	--	--	50	--	--	22	30
Dnieper, Kiev	3.05.86	--	9	35	--	--	19	19

The content of ^{90}Sr in various waters was determined from 1 May onwards. The highest levels were observed in the first few days of May in the Pripyat and reached 4 x 10^{-10} Ci/l. From the end of May until June its content in the Pripyat remained within the range 3-5 x 10^{-11} Ci/l.[4, 5]

The river creeks, lakes and floodplain water courses in the Chernobyl NPP zone were contaminated via direct deposition of radioactive substances onto the water surface and bed. In small rivers the initial period of contamination lasted the whole of May 1986. Their level of radioactive contamination was determined by the speed with which fallout was deposited onto - and dissolved in - the water, as well as by river flow itself, i.e. the throughput of water in the contaminated

area. Thus, this period lasted about 20 days in the case of the Lubyanka River, the ^{137}Cs content in its waters falling from 10^{-8} to 10^{-11} Ci/l.[6] The highest contamination levels in the waters of the reservoirs in the Dnieper Cascade were also registered in the initial period after the accident.[4] The observation data showed that the radioactivity of the water in this period came primarily from particles suspended in the water. The activity of the filtrate accounted for just some 10% of the total activity of the gamma-emitters in the sample. The total beta-activity of the water in the period of aerosol fallout reached 10^{-7} Ci/l.

From that time onwards, the sharp fall in the intensity of emissions from the source led to a reduction in the role played by the aerosol component in reservoir contamination. Suspended particles settled quite quickly onto the bed of the reservoir, and as early as 7 May the total beta-activity of the water in the area around the settlement of Lyutezh was of the order of 10^{-11} Ci/l.

Isotopes of ^{131}I played an important role in the contamination of water bodies in the initial period. The highest content of ^{131}I in the Dnieper, observed in the Dnieper water intake area, came from fallout of radioactive aerosols and, according to [4], amounted to 3×10^{-9} Ci/l on 3 May. Once atmospheric fallout of radioactive aerosols onto the water surface had ceased, secondary processes became the main source of water contamination, the most important being a) radionuclide washout from contaminated catchment areas and river floodplains and b) radionuclide exchange processes in the "bed-sediment/water" system. The intensity of secondary water-contamination processes was determined largely by the physico-chemical forms of the radionuclides in the fallout and by their rate of transformation in the "soil-water" system.

2. RADIONUCLIDE WASHOUT FROM CONTAMINATED CATCHMENT AREAS

Immediately after the Chernobyl accident there was a need for rapid forecasting of secondary radioactive contamination of water bodies. The question of radioactive contamination in the water courses of the Dnieper basin was particularly important, given that this is the Ukraine's main source of water and that migration of radionuclides via the river systems may contaminate areas a long way from the accident site. One of the main processes resulting in secondary contamination of surface waters is washout of radionuclides from the contaminated territory in surface runoff. The first studies into washout of Chernobyl radionuclides were carried out as early as the summer of 1986, the aim being to calculate washout coefficients in order to provide a rapid forecast of the radiation situation in water bodies.[7, 8] These studies were carried out using artificial sprinkling on specially equipped runoff sites. Table 3 shows the standardized washout coefficients for the main long-lived radionuclides as a result of pluvial and snowmelt runoff, these being obtained in experiments at runoff sites. In the spring and summer of 1987 studies were carried out in actual catchment areas in the Boguslav rayon of Kiev oblast into washout of ^{134}Cs, ^{137}Cs, ^{106}Ru and ^{144}Ce with surface runoff into rivers. The standardized coefficients of liquid washout (K_l) calculated using the data from these studies tally with the experimental findings from the runoff sites.

After the radionuclides fall out onto the surface of the soil, they

become fixed and migrate into the depths of the soil. These processes resulted in a reduction in the magnitude of the standardized coefficients of radionuclide washout in surface runoff. However, no significant fall in the K_l level for ^{90}Sr was observed between the summer of 1986 and the autumn of 1989. This can be explained by the fact that migration into the lower soil layers by this radionuclide's mobile soluble forms is balanced out by transformation of its non-exchangeable forms due to leaching of the fuel particles. As a result, the content of exchangeable forms of ^{90}Sr in the upper soil layer does not change much in the catchment areas in the zone, with the exception of the areas in the near zone of the fuel contamination trail where, in recent years, we have observed increased rates of radionuclide leaching from the matrix of the fuel particles and, at the same time, conditions have not been conducive to water-based migration of their exchangeable components into the depths of the soil.[9]

Table 3

Coefficients of liquid washout of ^{137}Cs and ^{90}Sr standardized by runoff layer, obtained at experimental sites in the 30-km zone around the Chernobyl NPP, k_l-10^{-5} l/mm

Experimental site	Date	^{137}Cs	^{90}Sr
Chernobyl	9.07.88	1.1	42
	10.07.89	1.0	45
Benevka	14.10.86	0.6	5.8
	.03.87	0.2	0.6
	17.09.87	0.3	21
	8.07.88	1.0	20
	11.07.88	0.7	13
	29.09.88	0.9	17
Kopachi	03.87	0.4	1.1
	04.87	5.7	6.6
	16.09.87	0.1	2.0
	10.07.88	0.7	12
	27.09.88	0.8	10
Korogod	03.87	0.7	0.8
	18.09.87	0.4	11
	7.07.88	1.4	13
	24.09.88	1.5	8.8
	7.07.89	0.7	15
Dovlyady	16.07.86	5.5	10
	16.07.86	3.4	10
	12.10.86	0.8	7.2
	16.10.86	0.6	12
	16.10.86	1.6	8.4

Experiments involving sprinkling runoff sites and flooding soil monoliths were carried out around the Benevka settlement during various seasons of 1986-1987 to study the washout of radionuclides under various conditions of water-runoff formation in the catchment areas. The monolith flooding experiment showed that a balance between the concentrations of dissolved ^{137}Cs and ^{90}Sr formed within some three weeks. The K_l values calculated from these tallied, within a given margin of error, with the standardized coefficients of "liquid" washout

obtained at the runoff sites. In spite of considerable differences in the sprinkling regimes (intensity and duration of experiments), the radionuclide concentrations in the runoff at these sites did not change much. The duration of sprinkling ranged from under one hour to 24 hours. All this shows that equilibrium concentrations are produced in the "soil-water" system during contact between rain water and the uppermost soil layer, in which surface runoff is formed.[10] In the turfy sections of the soils where the experiments were carried out, this layer comprised surface vegetation and the uppermost root-supporting layer of soil particles, i.e. where most of the radioactive fallout is concentrated. In washout experiments involving artificial sprinkling, the uptake of radionuclides into the waters of surface runoff resulted mainly from direct desorption and ion-exchange of radionuclides in the upper contact layer of the soils. At the same time, in the case of complete prolonged flooding, diffusive flows of contaminated interstitial waters in the upper soil layer were also involved in the mass exchange between water and contaminated soil. Speedy formation of equilibrium concentrations points to the decisive role of ion-exchange processes in the formation of radionuclide content in water runoff. Where considerable time is taken to establish equilibrium conditions, the major role in mass exchange between radionuclides and water is played by diffusive migration processes.

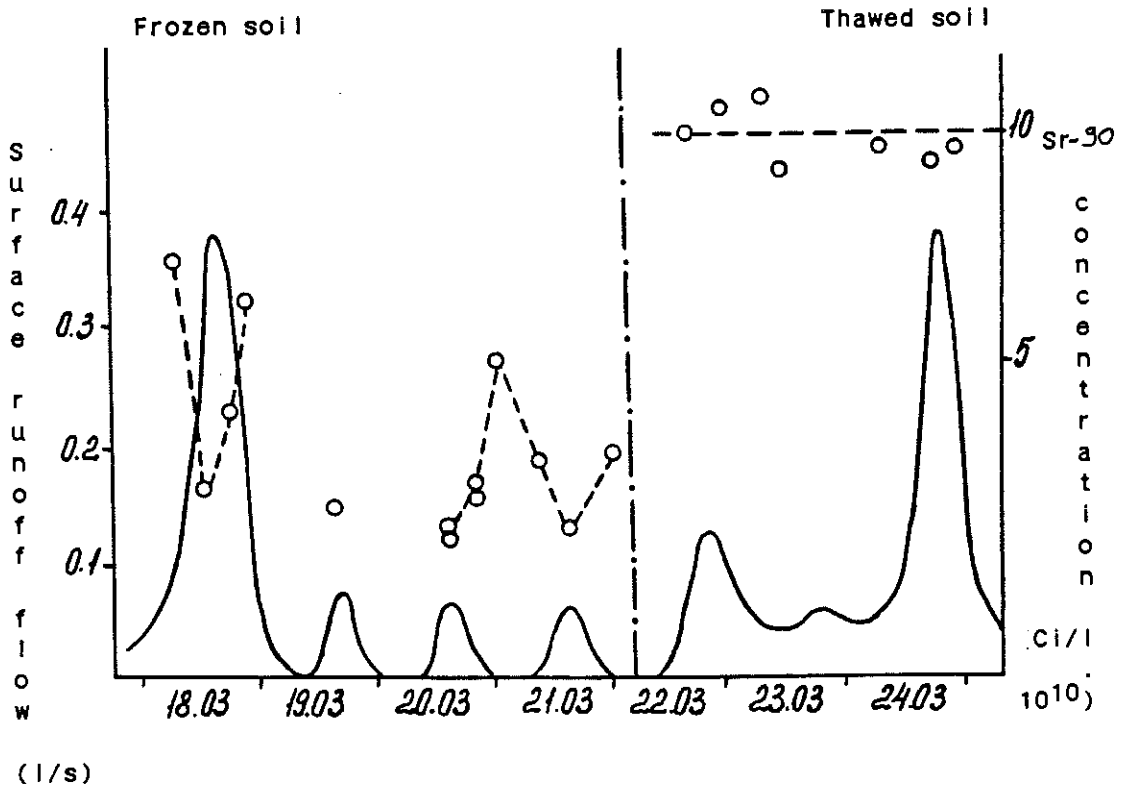
Calcium, which largely determines the equilibrium content of ^{90}Sr in the waters forming or accumulating in the 30-km zone around the Chernobyl NPP, is the main exchangeable ion in the soils of this zone and is extremely important for the migration of this radionuclide in the soils of the areas studied. The studies therefore also included data on the chemistry of the waters in the zone.

In the case of formation of snowmelt (in contrast to pluvial) runoff, the ^{90}Sr content in the water is clearly dependent on the intensity of runoff in radioactively contaminated catchment areas. This can be explained by the fact that when snow melts, part of the melted water drains along the surface of the lower layers of snow and frozen soil and has little contact with the latter, and therefore dilutes the contaminated runoff. When the upper, most contaminated soil layer has completely thawed, the incorporation of radionuclides into the snowmelt (via mass exchange with the soil) is balanced out since the less contaminated snowmelt water dilutes the radionuclide content (Fig. 2). The ^{90}Sr concentration therefore remained constant, and, as in the case of the experiments with pluvial washout, did not depend on the intensity of surface runoff formation. In contrast to the processes of formation of radionuclide washout in snowmelt and pluvial runoff, we identified - through in-situ observations - a number of features specific to radioactive contamination of river waters during flooding of soils in the floodplain landscape of the Chernobyl NPP near zone.[11] The main hydrological features of floodplain washout in this area are usually prolonged flooding, a wide range of ways in which the water masses move along the relief surface of the floodplain landscape, and the multi-directional nature of water movement in the aeration zone and in subsoil flows where there is a direct hydraulic link between the levels of river and floodplain waters. The particular physico-chemical features of radionuclide migration in this area were determined by the high content of fuel particles in the trail of fallout deposited, by the favourable conditions for their breakdown

given the presence of organic substances in floodplain soils, by the large amount of hydrocarbonaceous substrates, and by the relative scarcity of geochemical barriers to their migration in alluvial subsoils. As a result of the first of these factors, a major role in river contamination was played by the processes of water yield and post-high-water runoff of the contaminated floodplain flows via the surface and filtration pathways.

Dependence of ^{90}Sr level in surface runoff water

on snowmelt intensity



——— Surface runoff flow

o concentration of ^{90}Sr in solution

Fig. 2

It should be pointed out that in the years following the accident the low flows and levels of water during high water meant there was no significant flooding of the floodplain area in the near zone (Figure 3). As high water developed and reached its peak, floodplain runoff of insignificant volume was diluted by the high water in the river. When the high water fell, the intensity of floodplain water runoff increased because of the fall in river water level, which led to a significant rise in the ^{90}Sr content in the channel flow (Figure 3). The longer the water forming part of river channel capacity spent on the floodplain landscape, the higher the content of radionuclides in floodplain runoff. The kinetics of this process were determined by the speed of flow of the river waters across the surface of the floodplain, and by the quantity of radionuclides in exchangeable form in soils in contact with the floodplain waters. In the closed floodplain lakes in this zone equilibrium between the ^{90}Sr content in water and its content in bed sediments formed over a period of several months after the accident and has not changed significantly since, having stabilized at 8-10% of its content in the bed ground. Data on typical contamination levels of water bodies in the zone near the Chernobyl NPP are given in Table 4. The findings showing how strontium is distributed among the various components of the water bodies are close to those observed for lakes in the area contaminated by the Kyshtym accident. Thus, flushing of these numerous floodplain water bodies in the high water period may also have a considerable effect on the level of general removal of radioactivity into the river when the floodplain territory is flooded.

In the years after the accident when no more than 10% of the floodplain area within the near zone was submerged, annual ^{90}Sr washout from its surface amounted to 0.5-2% of its content in the given territory, while the contribution of runoff from its surface to contamination of the river (at the mouth) amounted to 30-40% of all the other sources of ^{90}Sr uptake into the Pripyat River. The integral coefficient of radionuclide runoff is the integral indicator of the radionuclide washout processes due to all water runoff processes - from catchment areas, via flooding of floodplains and via filtration. For the Pripyat this coefficient was used to work out how the annual river removal of radiocaesium and radiostrontium in the Chernobyl reach related to the amount of such radionuclides in the catchment area and to the strata of annual water runoff from the surface of the catchment area. The same was done for the mouth of the Dnieper River where it flows into the Kiev Reservoir. The figures obtained for the basins of the Pripyat and Dnieper Rivers ranged from 2 to 5 x 10^{-4} l/mm, which is not significantly different from the experimental sprinkling findings, and we therefore conclude that the standardized runoff coefficients did not undergo major changes in the years following the accident.

3. RADIONUCLIDE RUNOFF AND THE RADIATION SITUATION IN DNIEPER BASIN WATER BODIES BETWEEN 1986 AND 1990

The most important agents in the formation of radionuclide runoff from the contaminated regions were the climatic and landscape-geochemical factors involved in the formation of water runoff in the catchment areas of rivers flowing across their surface. Figure 3 chronologically charts the changes in the flow of radioactivity in the Pripyat waters on the basis of 10-day average values from 1986 to 1989. Within the limits of the typical hydrological seasons, Fig. 3 reveals a close connection between type of river flow formation in contaminated areas and the content of radioactive substances in the Pripyat and Dnieper

waters. Moreover, the removal of ^{137}Cs by the river considerably depended on the content of suspended sediment flowing in the water and on the intensity of erosional washout of fine soil particles from the surface of the catchment areas, whereas transport of river load had no significant bearing on the removal of ^{90}Sr. Figure 4 shows an example of the observed ratio between ^{137}Cs removal on suspended load and in solution for the period following the accident ($d<1\mu m$). It is evident that the proportion of ^{137}Cs contained in sediment flow may vary between 20 and 80% depending on the roiling conditions, catchment area erosion and the transportational ability of currents.

In 1987-1989 between 30 and 40% of the annual amount of caesium removed by the river was on suspended load. At the same time, whereas the ^{137}Cs activity of the suspensions in the Pripyat River was 10^3-10^2 Bq/year at the end of 1986, by 1990 it had fallen to 10^1-10^0 Bq/year. This effect is probably related primarily to the marked decrease in erosional removal of contaminated soils into rivers after discontinuation of ploughing and sodding of contaminated catchment areas in the Polesye (Pripyat Marshes), and to the gradual downward displacement of the most contaminated soil layer in the catchment areas as a result of vertical migration processes. In addition, the specific activity of river load was - depending on the point at which it entered the river current and on its material composition and size - still 10_x Bq/year or more in individual samples even four years after the accident.

The distribution of the specific activity of certain isotopes following their sorption from water to load and soil particles of various dimensions in the Chernobyl NPP zone is shown in Fig. 5. It follows from this that, given the river channel conditions obtaining, the bulk of radionuclides cannot settle on suspended material, and that channel load (usually sands in the case of flatland rivers) will remain relatively clean, as a result of the fine fraction being flushed by the current and it having lower sorption properties than muddy and clayey river load.

Channel changes gradually caused finely dispersed aerosols which had settled onto river beds to be removed into the reservoir, and part of the heavy fraction of the fuel component in the radioactive fallout also became buried under a layer of sand as a result of ridge-type load movement. Another trend observed was a decrease in the overall amount of radionuclides such as $^{134,137}Cs$, ^{144}Ce, $^{290,240}Pu$ removed by the rivers into Kiev Reservoir via load in river flow. A similar effect in decreasing the contamination level of the Pripyat and Dnieper waters (although less significant) was also observed in liquid-phase migration of these radionuclides, including radiostrontium. However, whereas in the years since the accident over 70% of the ^{137}Cs in the Pripyat has come from the region above the Chernobyl NPP 30-km zone, the contribution of the near zone to ^{90}Sr contamination of the river in recent years (1988-1989) has been 60% and more. This was mainly due to the removal of radionuclides from floodplain areas in the near zone and by contaminated waters filtrating from dammed river creeks near the town of Pripyat and from the cooling pond of the Chernobyl NPP. The relative increase in the contribution to contamination of the Pripyat from contaminated landscapes in the Chernobyl NPP zone is due primarily to continuing physico-chemical changes in the fuel component of the fallout and to the breakdown of the "hot" particles, both mechanically and by the lixiviation of radionuclides.

Chronological representation of water discharge in the Pripyat River and the content of certain radionuclides in water near the town of Chernobyl (from observations)

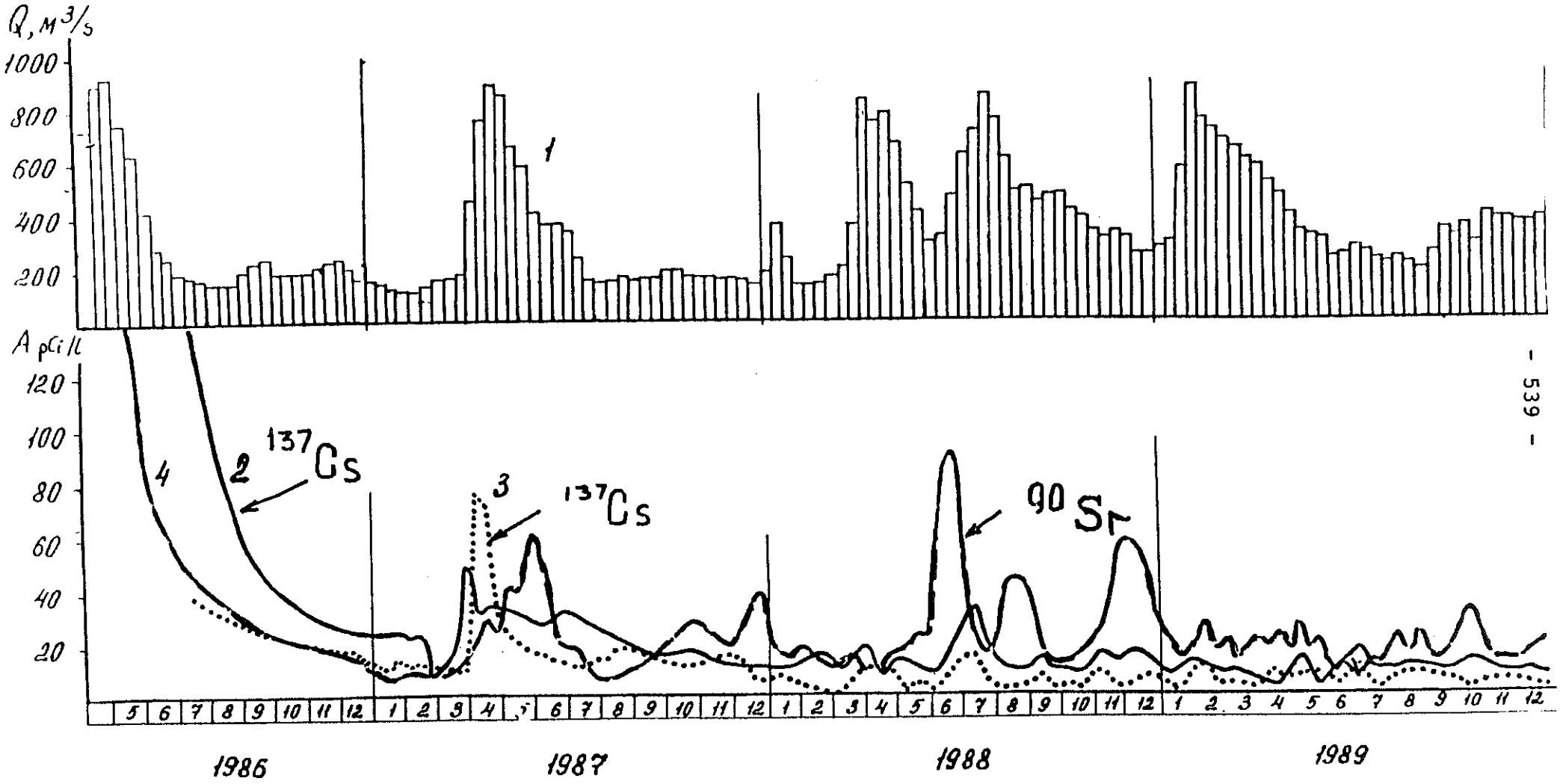


Figure 3

1 – water discharge
2 – ^{137}Cs (in solution)
3 – ^{137}Cs (in suspended load)
4 – ^{90}Sr

Table 4

Certain typical levels of radioactive contamination of surface waters in water bodies and rivers in the 30-km zone around the Chernobyl NPP in 1989-1990 (pCi/l).

Study site	^{137}Cs		^{90}Sr
	water	suspension	water
Pripyat River, Benevka	2-10	1-6	5-20
Chernobyl	5-20	2-15	15-36
Kiev Reservoir	5-15	2-5	10-15
Uzh River, Chernobyl	5-20	5-10	20-30
Sakhan River, Novoshepelichi	10-30	5-20	60-80
Chernobyl NPP cooling pond	100-250	40-60	250-600
Pripyat creek	80-400	25-100	$(3-4)10_3$
Standing floodplain water bodies in the near zone (left bank)	$(1-3)10_3$	100-200	$(1-10)10_3$
Ozero Glubokoye (lake)	800-1 200	100-140	$(6-10)10_3$
Ozero Vershina (lake)	200-300	10-30	$(6-8)10_3$
Goluboy ruchey	150-300	10-40	$(2-4)10_3$
Murovka channel	20-50	5-30	40-130
Polder waters	120-350	30-60	$(1-3)10_3$
Clay pits	10-30	2-8	200-350
Lake, Buda-Varovichi settlement	10-20	2-5	200-250
Lake, Lubyanka settlement	10-20	2-5	200-250

Ratio between ^{137}Cs removal on suspended load and overall flow in the Pripyat and Dnieper Rivers (1987–1989)

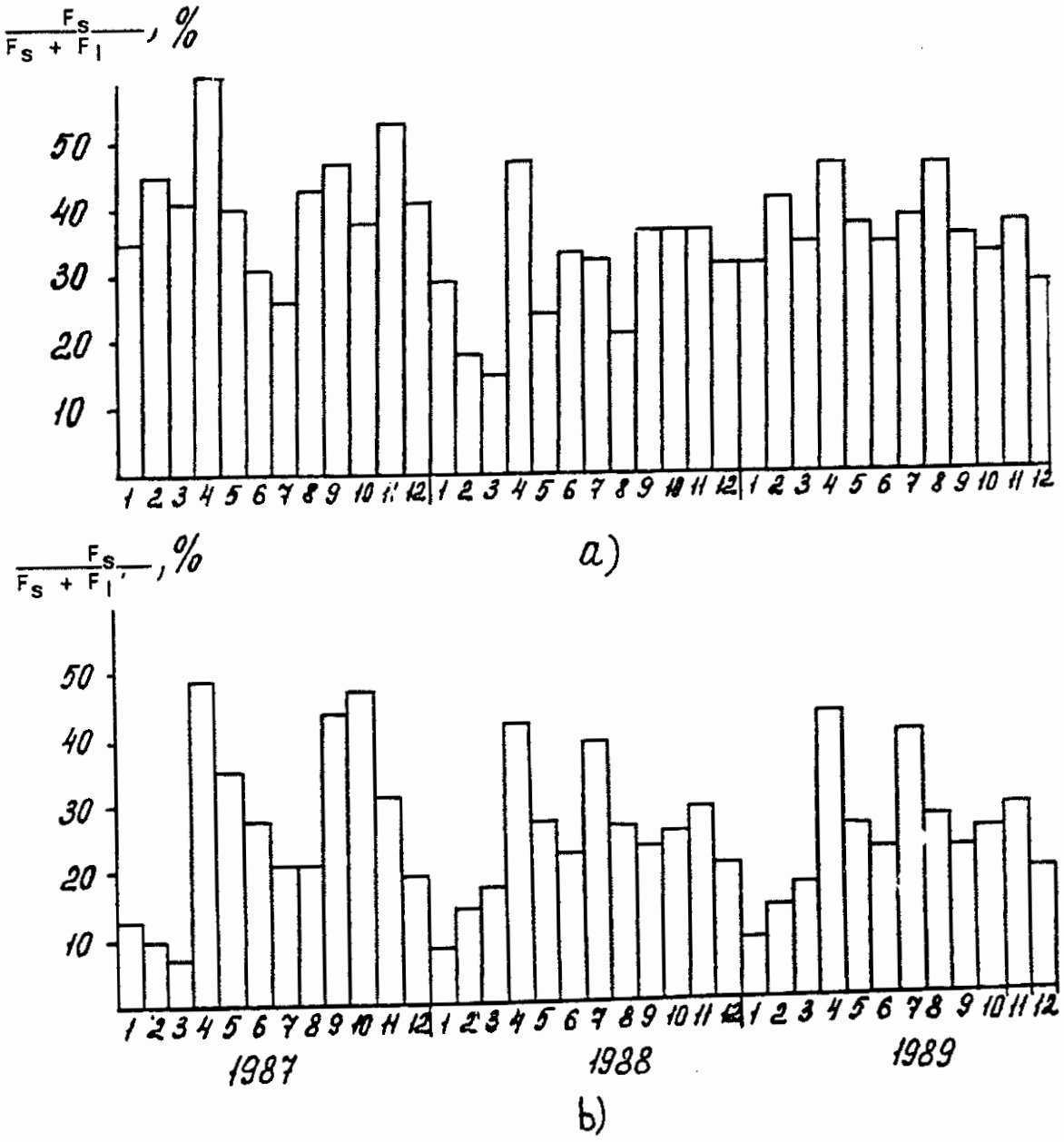


Fig. 4

F_s = concentration of ^{137}Cs in suspended matter flow

F_l = as above for the liquid phase;

a) Pripyat River at Chernobyl; b) Dnieper River, Nedanchichi

The reservoirs of the Dnieper Cascade are - and will remain - the main receptacles for the contaminated water present in the basin. Given significant dilution in these areas and in other running water courses, the radioactive contamination level of the water is tending to fall. The typical trends over time in different geographical areas are shown in Fig. 6 (on the basis of data from in-situ observations).

The main processes involved here in natural cleansing of the water are sedimentational removal of the radionuclides of caesium and cerium etc. into the bed sediments and sediment accumulation. Typical examples of this process are given in Figure 7, which shows various forms of vertical radionuclide distribution in reservoir bed sediments under different roiling conditions and sedimentation rates. The mean rate of sediment accumulation as a result of river load, bank erosion and internal water-body processes ranges from a fraction of a millimetre to ten millimetres per annum for the various regions. Therefore, in certain parts of the Kiev Reservoir the worst contaminated layer is already buried under the layer of sediment which has formed on the bed since the accident. It bonds less firmly with the bed ground and its behaviour in water depends largely on the presence of calcium macro-ions. The fact that high levels of ^{90}Sr accumulation were observed in the valves of molluscs, in particular of Dreissenacea, proves that strontium, like calcium, may transfer to carbonaceous insoluble forms. Thus, the exchange selectivity coefficient of the Sr-Ca pair - which indicates their relative ability to participate in substitution reactions - is close to (or even above) one.

Therefore, the reservoirs of the Dnieper Cascade are clearly undergoing natural water-cleansing processes as a result of adsorption and sedimentational burial of contaminated load and accident fallout by cleaner products of natural sediment accumulation. The main sources of sediment accumulation material settling on the bed of the reservoirs are allochthonous river load, the products of bank abrasion and those of bed erosion. Currently, more than 70% of the radioactive substances attached to the solid ground particles of the Dnieper Cascade reservoirs are located in the Kiev Reservoir's bed sediments. A substantial spillover of radionuclides into the lower reservoirs is observed only in storms and transitory river flow during high water; in the case of ^{137}Cs this can be as high as 50% of the flow into the reservoir with the finest load particles. When this happens, up to 80% of the mass of solid river load entering with the inflow precipitates out in the reservoir's headwaters.

Between 1986 and 1989 the overall discharge of soluble and suspended forms of ^{137}Cs in the waters flowing into Kiev Reservoir from the Pripyat and Dnieper Rivers and other small affluents amounted to some 4 000 Ci. The discharge of ^{90}Sr in the same period amounted to approximately 2 500 Ci.

The amount of caesium which moved across the Dnieper Cascade into the Black Sea between 1987 and 1989 accounted for no more than 20% of the caesium flowing into Kiev Reservoir, while the corresponding figure for ^{90}Sr was over 70%. This points to the considerable role played by the "sorptive capacity" of the bed sediments and ground of reservoirs in cleansing contaminated water masses. This process is continuing and in the coming years the tendency for radionuclides to accumulate in reservoirs will probably continue (Figure 6). In 1989-1990 the radiocaesium contamination level of the reservoir waters lay between 10^{-12} and 10^{-13} Ci/l, which is similar to the background levels during the global fallout period. The value for ^{90}Sr was slightly

higher at 5-10 pCi/l in 1990. During 1990, ^{90}Sr content - measured from beginning to end of the Cascade - varied on average from 20-30 pCi/l in the Pripyat River mouth to 1-5 pCi/l in the lower reservoirs.

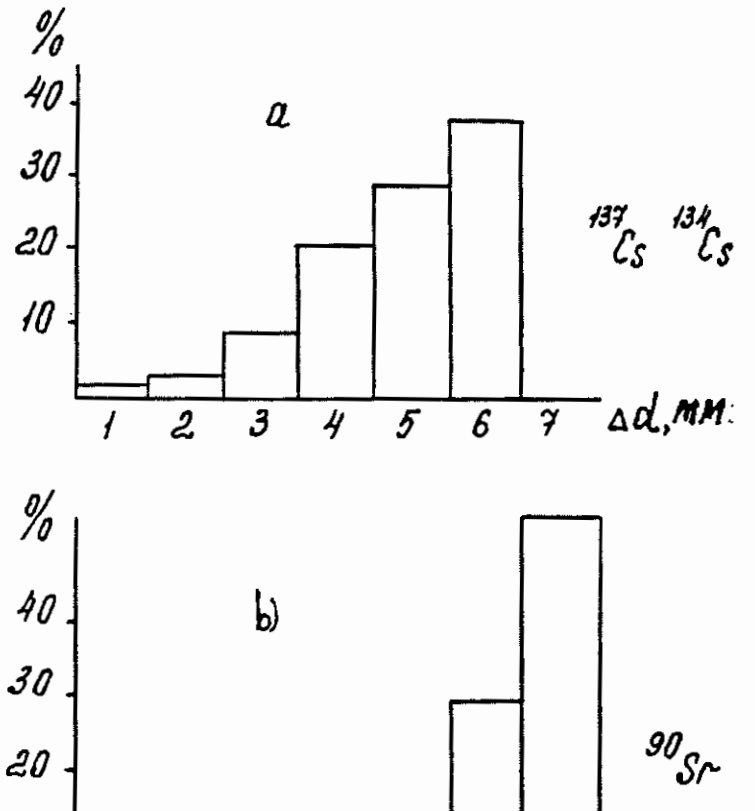
From now on the water contamination levels in the reservoirs will depend largely on the development of the natural processes governing the migration of radionuclides. They will be determined a) by the stability of the processes of internal water-body localization of radioactive substances in the various bed sediment components and by the rate of inter-phase transfer of non-exchangeable forms of radionuclides into water, and b) by the amount of additional radionuclides entering the water from contaminated catchment areas. The latter will depend, in turn, on the effectiveness of the water protection measures being carried out in the Chernobyl NPP's zone of influence, while the effectiveness of any planned water protection measures will greatly depend on the ability to forecast correctly the aftereffects of their large-scale introduction into natural systems.

Forecasting the prevailing radiation conditions in water bodies in the zones of influence of Chernobyl contamination was founded primarily on a system of basic data on the scale and forms of radioactive contamination in the affected areas: data from analysing the processes involved in formation of water runoff and washout of radioactive substances from catchment areas in the liquid and solid phases; analysis of the hydrological conditions governing flow in rivers and of the exchange of water in the Dnieper Cascade reservoirs; data on the most important elements involved in natural cleansing of the water bodies, on the lithology and geochemistry of the ground and bed sediments in the water bodies in the zone of migration of radioactive substances; and a set of physico-mathematical models for describing their migration processes.

The very first rough forecasts were prepared as early as May 1986. They were based on approximations of radionuclide solubility, the work done to determine washout parameters following the Kyshtym accident and hydrological forecasts of the spring high-water flow in the Pripyat and Dnieper Rivers. Even at that early juncture it was estimated that the radionuclide content levels in river waters at the Dnieper's main water intake points would not exceed the maximum permissible levels [12]. After that, approximate forecasts were needed covering pluvial washout of radionuclides from contaminated catchment areas, levels of secondary contamination of rivers, dispersion of radioactive substances in reservoirs, etc.

To this end, a series of physico-mathematical models relating to the transport of radionuclides in various water systems was developed for the catchment area, rivers and reservoirs.[8, 12, 13] Comparing the forecasts made of the radiation situation in rivers in the accident's zone of influence with the findings of in-situ monitoring of the state of the water bodies showed that the forecast data had quite a high degree of accuracy. This was achieved through a multi-facetted approach in organizing the work, involving closely coordinated studies to develop mathematical models of migration, planning of in-situ and laboratory experiments and devising a system for collecting data for all the major contaminated water bodies in the Dnieper River basin. The main forecasting principles, the basic mathematical equations used in the migration models, and the experience gained from their use and parameterization in practical work after the Chernobyl accident have already been described in sufficient detail in [8, 12, 13], to which the authors of this paper contributed.

Distribution of the specific content of radionuclides in load particles of various dimensions in the Pripyat River



1 -> 0.25 мм
2 - 0.25 - 0.1
3 - 0.1 - 0.05
4 - 0.05 - 0.01
5 - 0.01 - 0.005
6 - 0.005 - 0.001
7 - < 0.001

Fig. 5.

Radioactive contamination of Dnieper reservoirs following the Chernobyl accident

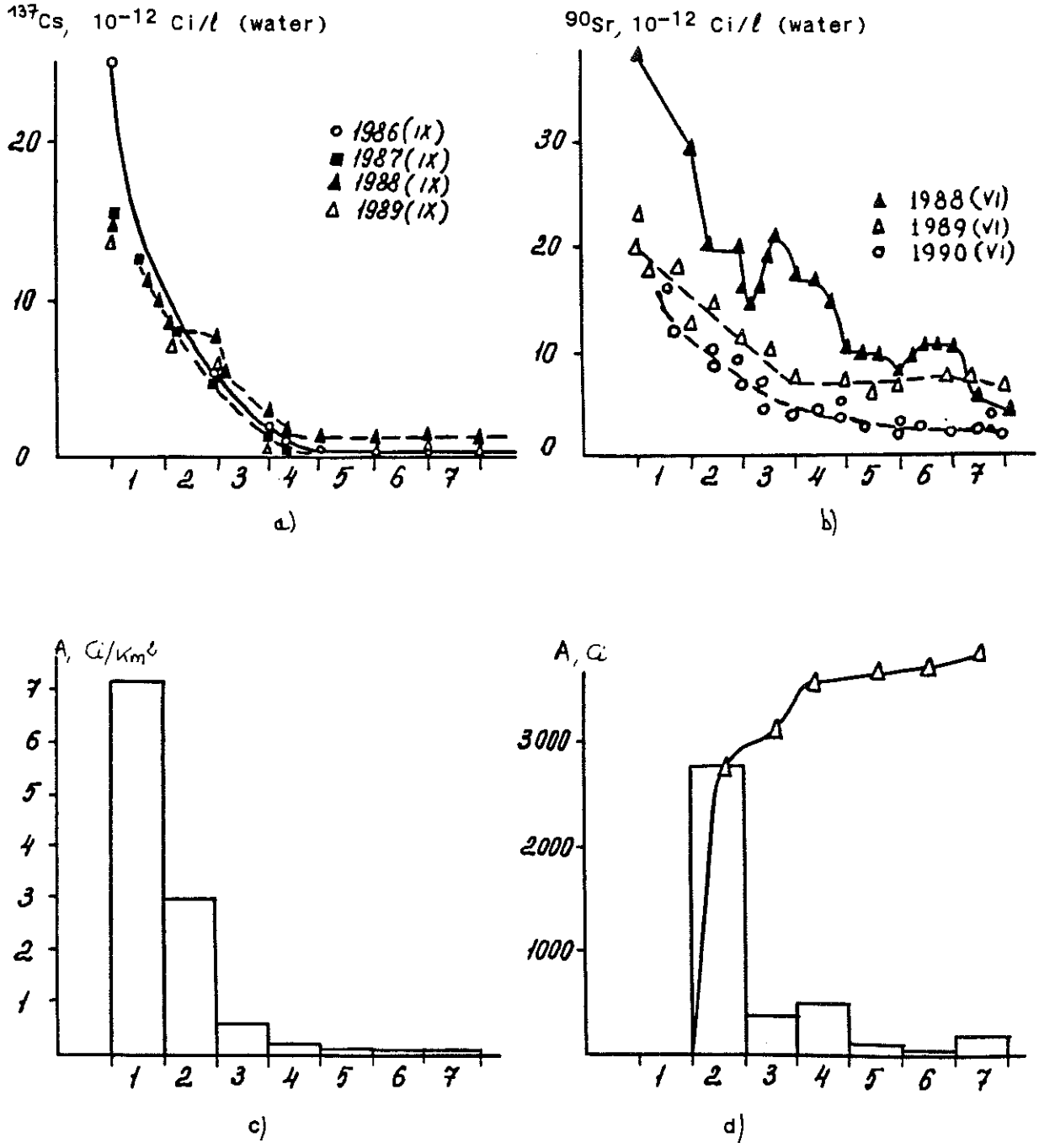


Figure 6

1. Mouth of the Pripyat and Dnieper rivers
2. Kiev reservoir
3. Kanev reservoir
4. Kremenchug reservoir
5. Dneprodzerzhinsk reservoir
6. Kakhovka reservoir

a, b) Distribution of ^{137}Cs and ^{90}Sr in the reservoirs of the Dnieper Cascade
c) Mean bed contamination
d) Overall content of ^{137}Cs in bed sediments

Typical vertical distribution of ^{137}Cs
in bed sediments of the Kiev Reservoir (1989)

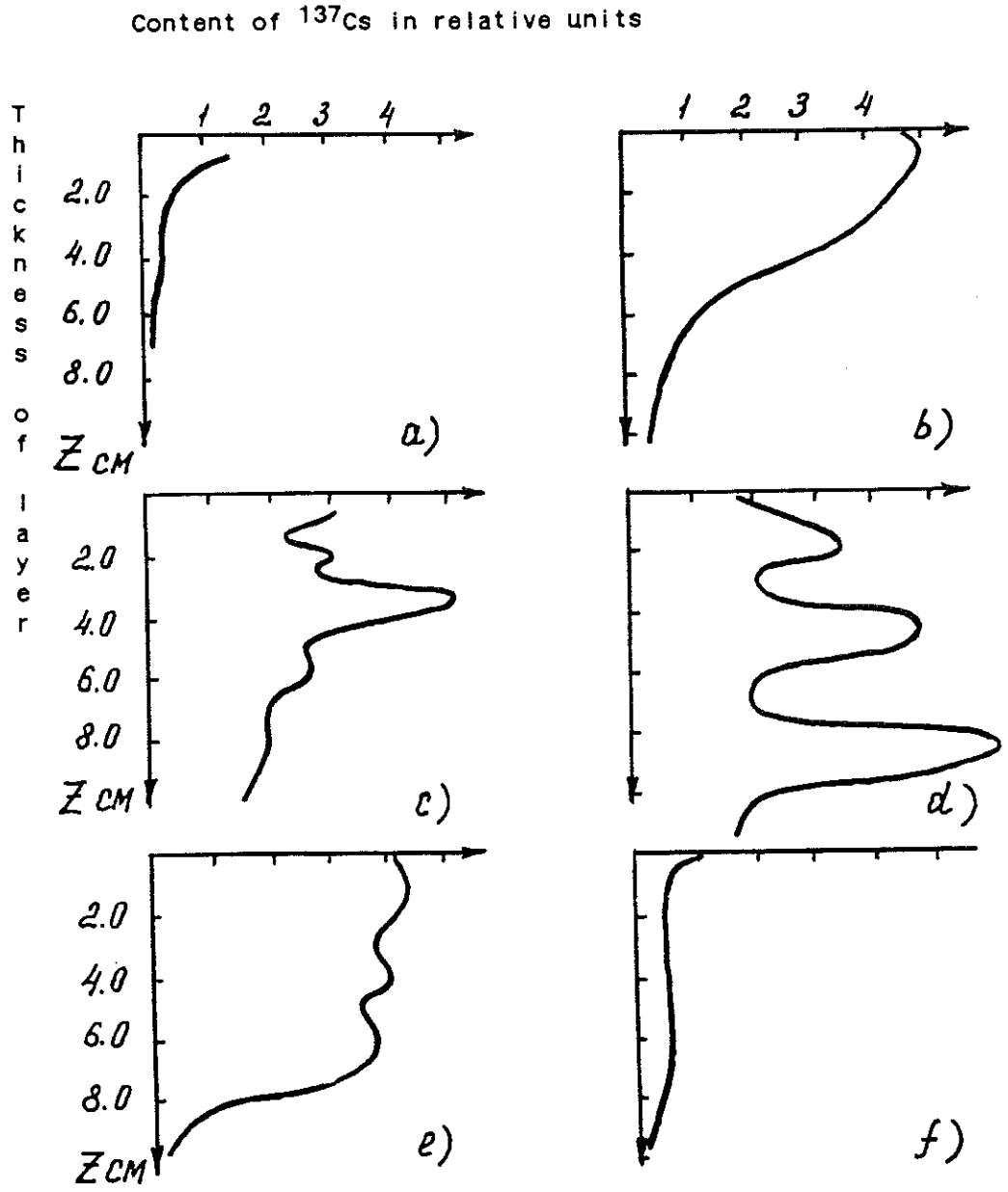


Figure 7

a,b – areas of the bed with no significant sediment accumulation

c – contaminated areas of the bed characterized by storm roiling processes

d – areas with typical seasonal sediment accumulation cycles

e,f – channel flow areas of the bed with ridge-type sand movement and good filtration properties

By way of conclusion, it should be pointed out that, when it came to dealing with the consequences of the accidental radioactive contamination of the flooded areas and water bodies, no-one in the world had had experience of working in such difficult natural water-soil landscapes in conjunction with such a scale and variety of physico-chemical forms of radioactive contamination. The years that have elapsed since the accident have been very benign in terms of the amount of water in the rivers in the contaminated zone. This has largely conditioned our understanding of the actual natural processes involved in the self-cleansing of water systems. However, it also allowed us to study in more detail the processes governing - and the factors involved in - the migration of Chernobyl radionuclides via the water pathway, and to gain experience in forecasting the long-term processes of secondary contamination of water bodies, and consequently to draw up active measures to counter radionuclide migration processes, to attempt to prevent radionuclides from dispersing beyond the initial fallout areas and thereby devise optimum water protection measures to limit the negative after-effects on the environment.

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Migration in the Pripyat and Dnieper River Basins of Caesium and Strontium Radionuclides Accidentally Released from the Chernobyl NPP

E.V. SENIN

The accident which occurred on 26 April 1986 at the IVth unit of the Chernobyl nuclear power plant following a surge in radioactivity, shattered the nuclear fuel into discrete particles varying considerably in size - from a minimum of some tenths of a micron to a maximum of 100 microns. Contact between this dispersed fuel and the (water) coolant caused a steam explosion and, in consequence, the process channels were destroyed, the reactor breached and the dispersed fuel ejected into the atmosphere, a significant part of it falling in the basins of the Pripyat and Dnieper rivers. The material composition of the matrix of most of these particles constituted oxides of uranium, while the radionuclide composition matched the composition of the irradiated fuel.

The radioactive fallout in the basins of the Pripyat and Dnieper rivers, which consisted of solidified aerosol products from the molten fuel, the coolant material, etc., and which radioactively contaminated large and fairly distant territories in the nine days that followed, plays a lesser role in the 30-km zone as such, although in the northwestern and northeastern parts of the evacuation zone its contribution to overall activity is highly significant.

In the soils of the 30-km zone more than 98% of the activity was concentrated in the uppermost layer in the first few months after the accident. In lower horizons (5-10 cm) activity decreased by several orders of magnitude, almost logarithmically. The isotopic composition was similar in each layer, testifying to vertical migration of radionuclides - in the form of very fine solid radioactive particles - through the soil pores along with infiltrating moisture, i.e. by leaching.

Contact between the discharged radioactive particles and the soil in the first few weeks after fallout deposition led to the formation of water-soluble forms of the radionuclides, mainly through leaching. A more active transfer of radionuclides into soil solutions was observed in soils contaminated with condensation-type precipitation in the distant zone. The least mobile radionuclides were in the fuel particles of comparatively small size (50-100 microns) deposited in the near zone.

Solid-phase radioactive fallout, commonly known as "hot particles", thus formed the first barrier to the further spread of radionuclides in the environment during the first three years following the Chernobyl accident.

The migration of radionuclides in the 30-km zone is currently proceeding both via "hot particles" and in water-soluble form.

The migration of radionuclides via "hot particles" is governed by the laws of gravitation, just like the transport of any kind of material on the earth's surface. The dynamics depend on a whole range of factors: the morphology, size and density of the "hot particles", the morphology of the surface of the land, the transport mechanism involved, the climate, etc.

The migration of water-soluble forms of radionuclides in the 30-km zone depends on the chemical composition of the "hot particles", the rates at which radionuclides are leached out of them, the mineralogical, petrographic and geochemical characteristics of the soils, the specific hydrological, hydrogeological and geological features of this area (the composition of the rocks in the sedimentary mantle, their water content, the structure of the crystalline substrate and the neotectonic development of the area).

It is now becoming clear that in the 30-km zone the main vectors of radionuclide migration in the Pripyat and Dnieper basins are washout and blowoff (i.e. water-borne and wind-borne transport mechanisms respectively) from the entire contaminated catchment area into the floodplain and channel of these rivers, followed by migration with the current at the bottom of the channel into the Black Sea.

It is also clear that studies of strontium-90 and caesium-137 migration in the basins of these rivers are most important from the medico-biological and geochemical point of view. The present report is devoted to some findings from such studies.

In the initial months after the accident ^{131}I was the main source of gamma activity in the surface waters, and we used it as a tracer to monitor how the water masses moved through the reservoirs of the Dnieper Cascade.

Apart from ^{131}I, the radioactive isotopes ^{106}Ru, ^{95}Nb, ^{95}Zr, ^{90}Sr and $^{134,137}Cs$ also played a substantial role in the early stages.

The short half-life of ^{131}I, plus the transfer of medium-lived isotopes into sediment and their subsequent decay, meant that once atmospheric radionuclide fallout directly onto the water surfaces had ceased (by the summer of 1986), the nature of the subsequent contamination of the water systems depends on the various mass transfer mechanisms.

To study the nature and dynamics of these processes a network of fixed observation points was set up in the Kiev Reservoir's main tributaries, near the water intakes and in the upper reaches of all the Dnieper Cascade reservoirs. Monitoring was also carried out from floating stations.

The overall beta activity of the water was measured daily, while the radionuclide composition of the waters and their ^{90}Sr content were determined periodically. The radioactive contamination of the river/reservoir bed sediments and catchment areas was also studied.

It was not until late 1986 that this monitoring system was developed in full.

The radionuclide transfer processes in the water differ considerably from one radionuclide to another. Removal of cerium-144 and plutonium was observed only in the solid phase, via suspended and bed load on which these radionuclides had settled. Particles suspended in water play the essential role in caesium-134 and caesium-137 transfer, whereas migration of strontium-90 mainly involves solutions (90-95%) and particles 0.001 mm in size (with suspensions and the transport of bed sediment and load playing no role here).

Fig. 1 shows the distribution of the specific radioactivity of Cs and Sr on their sorption from water to soil and load particles in the Chernobyl NPP area. It follows from this that, given the river channel conditions obtaining, most of the radionuclides attached to the suspensions are unable to settle, and the fine fraction of the sandy

channel sediment will remain relatively clean as a result of washing by the flow.

Fig. 2 shows the portions of ^{137}Cs removed on suspended particles and in solution. It can be seen that caesium-137 sediment discharge varies between 10% and 60% depending on turbidity conditions and the transporting ability of currents. It should also be remembered that the activity of these suspensions fell by a factor of 25 from late 1986 to 1989.

In Fig. 3 the changing pattern of radioactivity in the water flow of the River Pripyat at the town of Chernobyl can be seen from the mean ten-day values for the 1986-1989 period. This diagram clearly shows that the inter-relationship between the nature of river flow, the extent of flooding and the amount of radioactive substances in the water are quite complex. In 1986, when there was not much precipitation and the water flow rate was correspondingly low, the fall in radionuclide concentrations was due primarily to the effects of dilution and transfer of radionuclides into bed sediments. The insufficient number of observations in 1986 prevents us from determining the process in detail, and reflects its general character only.

From 1987 onwards observations were carried out systematically, and more detailed information was obtained. The lowest concentrations of radionuclides in the river were noted during periods of river freeze-over and freezing of soils in the catchment areas, when radionuclides no longer entered the river with surface runoff (this is obvious in January/February of 1987 and 1988).

This Fig. shows, firstly, that there is no clear-cut correlation between water flow rate peaks and radionuclide content peaks; and secondly, that the different temporal spread and nature of the Srand Cs behaviour curves reflect the different ways they enter the river and the different spatial spread of the radioactive contamination sources.

In 1987 radionuclide runoff was primarily due to washout processes in the Byelorussian Polesye (Byelorussian part of the Pripyat Marshes).

The specific way in which the river removed radioactivity in 1988 is linked to a decrease in the proportion of radionuclide washout from the

catchment areas in the distant zone during the high-water period, and a simultaneous increase in river contamination from the (worst contaminated) Chernobyl NPP near zone.

The greatest contamination of the waters at the town of Chernobyl in 1988 was observed as flooding abated and when high-water ended.

A major role in determining the river's radiation regime in 1988 was also played by the summer rain floods, which produced a flow volume greater than that of the spring high-water.

The simultaneous occurrence of peaks in the river's strontium-90 content and its water content was accompanied by a significant dilution of runoff waters, and was connected with the specific features of surface runoff from the catchment areas in the distant contamination zone, as well as with the flushing of the channels in the floodplain landscape, while the surface of the floodplain in the Chernobyl NPP near zone remained dry and unflooded. The autumn/winter peak in radioactivity was caused solely by pluvial washout and the shedding of contaminated waters accumulated during the spring and summer in the Zimovishensky polder system.

The relatively stable and low levels of radioactive contamination of the River Pripyat waters in 1989 may be explained by the very early arrival of extreme high-water (in February) when air temperatures were low, by surface water runoff (with relatively low radioactive contamination levels) formed in the River Pripyat's upper catchment areas, by the radionuclides continuing to be chemically fixed in the catchment area soils, and by the absence, more or less, of snow and of water in the catchment areas adjacent to the Chernobyl NPP.

The beginning of 1990 was also a time of low moisture levels, while the River Pripyat high-water peak occurred in the second half of January (the earliest high-water recorded during the whole monitoring period).

Similar radioactivity runoff processes are also found in the River Dnieper, but with the difference that here a large role in radionuclide migration is played by washout from the catchment area, rather than by removal from the floodplain. This leads to corresponding differences in the relationships between water runoff and removal of caesium and strontium radionuclides during the year.

Particular attention should be paid to the ^{90}Sr peaks in December 1987 and June, August and December 1988. An analysis of the mean ^{90}Sr content figures and of the water flow rate in reaches upstream of the Chernobyl NPP (Benevka) and downstream (Chernobyl) produces a figure of 140 curies per annum. This quantity of radioactive strontium was removed from the near zone in 1989, while in 1987-88 the quantity was substantially greater. The ^{137}Cs figures in 1989 for the same reaches are practically the same, i.e. the near zone does not have such a marked effect on the ingress of Cs as it does for Sr.

The highly active source of this ^{90}Sr turned out to be the waters of the many sluggish and stagnant bayous, lakes and streams of the floodplain on the left and right banks in the near zone.

The shift in the phase of occurrence of peak water content and peak ^{90}Sr activity is related to the rise of contaminated waters on the floodplain during high-water and their reflux from the side of the river, since maximum activity occurs later than maximum water content.

A study of the distribution of the ^{90}Sr isotope in the various bodies of water in this region has shown that the mean annual concentrations in them vary from 80 to 9 000 pCi/l.

These figures show that the category "b" permissible concentration for ^{90}Sr - 400 pCi/l - is substantially exceeded in some places in the water bodies of the near zone. Determining the concentration and mean annual flow rates for a number of water bodies communicating directly with the River Pripyat, and for filtration from the cooling pond, allowed us to ascertain the sources of some 70% of the ^{90}Sr entering the River Pripyat, with the remaining 30% coming from sluggish and stagnant bayous and lakes directly exchanging water with the River Pripyat during wind setup and permanently connected with it via filtration.

The near zone's overall contribution to ^{90}Sr contamination of the Kiev Reservoir amounted to approximately 60% in 1988 and to some 40% in 1989. This justifies the special attention devoted to this sector in the plan to protect the Dnieper Cascade reservoirs from radioactive contamination.

The ingress of ^{90}Sr and ^{137}Cs radioactivity into the Kiev Reservoir is shown in Fig. 4. A substantial contribution to the activity of the Dnieper river waters is made by the River Sozh, which carries ^{137}Cs removed from the Bryansk-Byelorussian patch of radioactive contamination. The River Sozh's share in River Dnieper removal falls with time - from 80% in 1987 to 35% in 1989 - reflecting the way Cs is fixed in its deposition areas and transport path.

The figures given reflect the dynamics of the process, since - according to other estimates - in 1986-1989 total removal by the Rivers Pripyat and Dnieper was 4 000 Ci of ^{137}Cs and 1 200 Ci of ^{90}Sr.

The Kiev Reservoir is the first in the Dnieper Cascade's chain of reservoirs and also the main accumulator of radioactive isotopes. As much as 80% of the mass of solid material entering from the Dnieper and Pripyat is deposited in its headwaters. The natural cleansing processes occurring in the water of the reservoirs mainly involve transfer to sediment of the radionuclides of caesium, cerium, etc. Sediment accumulation does not play much of a role in the case of strontium-90 (it binds less firmly to bottom soils), but calcium content considerably influences its behaviour. High levels of strontium-90 accumulation have been noted in the shells of river molluscs, especially in the species Dreissenacea.

The radionuclides deposited on the bed, especially in the early years, are now being covered over by cleaner sediment. At the present time more than 70% of the radioactive substances bound up with the soils of the Dnieper Cascade are located in the bed sediment of the Kiev Reservoir. Considerable shedding of radionuclides into the lower reservoirs is observed mainly during storms and high-water, when caesium-137, which is carried along with the most finely dispersed particles, may account for up to 50% of the radionuclide influx into the reservoir.

The slide shows diagrams relating to caesium and strontium content in the Dnieper Cascade water bodies.

As the data show, when substantial dilution occurs the level of radioactive contamination falls. By 1989 this level was 10^{-12} to 10^{-13} Ci/l for caesium-137, which is close to the background levels

obtaining prior to the accident. The level for strontium-90 is somewhat higher, i.e. 10^{-11} to 10^{-12} Ci/l.

Of the caesium which entered Kiev Reservoir from 1987 to 1989, 20% or less reached the Black Sea, while the corresponding figure for strontium-90 was over 70%.

In the first few months after the accident, surface water contamination was highly heterogeneous, with maximum contamination levels of 14 pCi/l of caesium-137 and 3.3 pCi/l of strontium-90. By 1989 these levels had come down (as a result of horizontal and vertical intermixing) - more or less to the pre-accident level for strontium-90, while for caesium-137 they stabilized at 1.5 pCi/l from 1987 onwards, i.e. some three times higher than before the accident.

The amounts of caesium-137 and strontium-90 in the Black Sea (determined by depth distribution) are approximately 85 000 Ci and 47 500 Ci respectively. Of these, 45 000 Ci of caesium and 7 500 Ci of strontium entered the sea after the Chernobyl accident.

Direct Continuous Multichannel γ -Spectrometric Measurements- One of the Main Methods for Control and Study of Radioactive Environmental Pollution

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In Chernobyl along with usual methods of environment radiation control there were used methods and equipment of direct continuous multichannel measurements. The necessary equipment was installed both on permanent observation stations (river Pripyat, Chernobyl, river Dnepr, Kiev) and on mobile units (helicopters, scientific river-boats, automobiles). Together with continuous control of radioactive situation and its estimation in time and space this equipment enabled to carry out the following:

- determination of time-spatial structure of radioactive pollution in stationary points and on space (mapping);
- selection of representative samples for subsequent radionuclide analysis;
- direct data input into the computer, data storage and data base creation.

The results and conclusions drawn are important not only for the situation on Chernobyl atomic station - they may and should be used for a continuous radioactive monitoring of the environment. Though the method and its realization remain to be modernized and unified.

Indeed, according to /1/ by the end of 1987 197 nuclear power plants were registered in the world with the total electric power of 120 GW. The number of nuclear reactors used for scientific research purposes is also increasing and in 1990 will reach 61, out of which 40 are situated in the developing countries /2/.

As the late academician V.V. Legasov /2,3/ noted, all safety measures (increasing the structure and cost of the equipment or the process) only decrease the risk of an accident, diminish the possibility of catastrophic consequences of the hard-ware failure or personnel mistakes to very small quantities but still this possibility is never nil. The zero risk is possible only in systems without stored energy. In a number of papers (for example in /4/) the probability of serious accidents on nuclear power plants is estimated with the account of their global increase (fig. 1). The estimation shows that for 500 active nuclear power plant accidents are due every 4 years. With all possible optimistic amendments to the schedule the picture is still quite tragic. Thus, the necessity of employment of continuous radioactive environment monitoring is stipulated by:

- firstly, the possibility of accidents on NPP (nuclear power plants) with all the consequences;
- secondly, there is always a possibility of damage of nuclear power plants and other nuclear industry enterprises owing to other circumstances (local non-nuclear wars, natural disasters) when the great amount of radionuclides is released in the biospere;
- thirdly, the development of "transport" nuclear power engineering (nuclear power ships), employment of autonomous nuclear power sources, the presence of a large amount of nuclear missiles also enlarges the probability of accidental situations, the pollution of the environment;
- Forthly. Even the non-failing (safe, normal) work of NPP causes the release of artificial radionuclides in the environment. The employment of thermal power stations, the amount of mining and construction works lead to increase and re-distribution of the natural radionuclides in the environment. Global radioactive fallout after nuclear weapons tests (though minimal) still exists;

- Fifthly. Wide utilization of radioactive isotopes (sources) in the different fields of economy, science and technology stipulates the possibility of situations (for example, in Brazil in 1987) even locally but, nevertheless, strongly affecting the man and biosphere.

Naturally, scale and character of the factors enumerated above are quite different - from the global transfer of radioactive contamination to local effect. The common feature is the impossibility to forecast such accidents and disasters. On the other hand, determination of their consequences, especially on first stages in case of "medium" and "small" contamination, demands distinct criteria of definition of "fresh-spread" radioactivity from eventual fluctuation of natural radioactive background.

So, in the sixth case, direct continuous methods of radioactive monitoring of the environment are necessary for its understanding, for creation of physical models of the possible radionuclide migration and distribution.

this, in the long run, enables to present an objective information for evaluation of the amount of inner and outer irradiation of the population, for the safety of which the International Commission on Radiological Protection (ICRP) recommends /5/ - "the level of the irradiation doses should be as low as can be obtained only with the account of economic and social factors".

Some Features of Direct Continuous -Spectrometry Measurements

In direct continuous measurements "in situ" one can use registration of radionuclides, gamma-irradiators because - and -irradiations have very small penetrating ability. This gives considerable mistakes caused by screening by inhomogeneous surroundings (soil) or hermetic capsule of the seasing element (data unit) pick-up in the water.

The quantum character of the gamma-irradiation enables to take gamma-spectrum of the surrounding, i.e. measure its radionuclide composition. Moreover, the major part of radionuclides important for their role in pollution and biological effect are gamma-irradiators. As to - irradiator strontium-90 and -irradiators of plutonium type their concentration may be determined in separate samples as well as in correlation with radionuclide gamma-irradiators if the initial ratio is known and the time passed from the moment of pollution is short. (The small period of time excludes radiogeochemical differentiation due to migration).

Registration of repeated scattering of gamma-quantums of radionuclides in stretched and volume surroundings /6/ seems also important in gamma-spectrometry "in situ".

In all the cases we investigate irradiating-absorbing surroundings where identification and quantitative evaluations of gamma-irradiators are made by registrating peaks of complete photoabsorption of initial gamma-quantums. Scattering increases Kompton's part of gamma-spectrum of each radionuclide and deteriorates the ratio "effect - background" for gamma-irradiators which energy is less than the boundary energy of Kompton's part gamma-spectrum of any radionuclide. This leads to deformation of gamma-spectra and a certain difficulty in their interpretation.

Such deformation of the spectrum in measurements "in situ" taken in homogeneous irradiating-absorbing surroundings (water) was shown

in /7/. These results for monoenergy source zinc-65 are given in Fig. 2. Fig. 3 gives deformation of gamma-spectrum when passing from point source Sodium-24 to bigger volumes with the same specific activity. One may see how increases the Kompton's part of the spectrum and decreases the relative area of photopeaks 1,38 and 2,76 Mev for the point, 3 litre-, 300 litre- and 10^4-litre volume sources.

For plane-extended sources (soil) in measurements of "fresh" fallouts when radionuclides do not yet migrate vertically the gamma-spectrum deformation will be caused by dispersion in the air. In both cases the necessary corrections are made either by theoretical calculation or when the graduated analysis of radionuclide composition of water (if it changes) is made or on testing areas where the radionuclide composition is determined by independent methods.

Dispercing properties of the medium considerably diminish the expediency of semi-conductor detectors. Moreover, they have lesser efficiency and their application (especially in water) is difficult.

When measuring low-level contamination (for example < 10^{-11} Ci/l for water) with the aim of constant and continuous control it is reasonable to carry out measurements in wide energy ranges (200-500 Kev) in order to raise threshold sensitivity. More selective data can be then received by samples selection or taking down the whole gamma-spectrum (this done if some anomalies are registered in the course of measurements). The deformation of spectrum in the dispersing medium should also be taken into account when the gamma-field intensity is evaluated in dose level.

Practical realization of the method of direct continuous measurements of gamma-activity in situ may vary according to aims set:

Single detectors	- "field" of detectors (> 10)
Cable transmission	- telemetry
Transmission of all the data	- level saturation signal
Changes in several ranges of energy or gamma-spectrometry	- changes in "integral"
Automated measurements (operator)	- automatic measurements.

Measurements in stable (stationary) points:
radioactive level control in rivers, testing areas, major waterworks, waste (reject) works of APS.

Measurements on mobile vehicles:
river-boats and ships - square (areal) gamma-shooting of water surface, control of reject and emergency situations, determination of "mixing zones" of water with different activity, determination of dependence on natural phenomena (i.e. high waters, etc.);
aviation carriers - active mapping;
surface-carriers - correction and detalization of aerogamma-shooting data, mapping of routs of moving, specification of radionuclide composition of revealed radioactive anomalies, measurements in "semi-stationary" points.

Practically all above operations have been and are being carried out in the course of restoration at Chernobyl APP.

Application of the Method and Facilities Used in 1986-88.
Measurements in Stationary Points.
Direct continuous measurements have begun in May 1986 in the river Pripyat (Chernobyl region) and in river Dnieper (Kiev region). Two separate points, spaced more than 100 km apart, could secure to control the radioactive "cloud" moving towards Kiev should such had taken place. Detection blocks from sodium iodine crystals measuring 80 x 80 mm and 80 x 400 mm were installed on buoys. The buoys were placed 50-100 m from the shore at the depth of 2 m from the water surface and not less than 4 m from the bottom of the river. the spectrometry information and the power supply to the detection blocks (-30 D) were passed by the same wire. The results were registered on auto-recording display and printer having regard to astronomic time count at expositions 60 sec (1986-1987), 600 sec - as from Summer 1987. The value of immeasurable radioactivity (beyond control) considering actual debit of r. Pripyat (400 m^3/sec) and concrete concentrations of radionuclides in water could not exceed 5.10^{-3} Ci/l (during exposure).

Regular gamma-spectrometric and radiochemical analysis of water samples enabled to determine the radionuclide composition (Fig. 4) and, if necessary, make corrections in detector readings.

The results of continuous measurements were presented in units Ci/l of the total gamma-activity.

All time of continuous (24 hour) run of the units made 13000 hours.

Generalized results of the measurements are presented in Fig. 5 where the fall of the total radioactivity level with time can be seen. For the whole observation period (1986-1988) radioactivity level never exceeded the weekly mean value more than 20-30%. From May 1986 (when total radioactivity level in Pripyat river was 2.10^{-9} Ci/l /8/) till August 1987 it fell 40-50 times. Gamma-spectrometry and radioachemical analysis showed considerable change in radionuclide composition of water: in 1987 Caesium radionuclides constituted the major part while in Summer 1986 a lot of fission fragment radionuclides were registered (Fig. 4).

The direct method permitted to carry out measurements under ice cover in Winter 1987, when considerable decrease of gamma-activity took place as well as to perform control during the flood. Consequently, continuous measurements in r. Pripyat and r. Dnieper made it possible to secure efficient control over the level of radioactivity in river waters in two points. The received results were taken into consideration also in the similar operations of Goskomhydromet of the USSR and other organizations on sampling water for analyses. Besides, the distribution of radioactivity between these two points was registered by direct continuous methods on a special boat (see below).

Measurements on Mobile Vehicles. Boat, research ship, helicopter and specially equipped car were used as mobile vehicles. On the river-boat "Academician Vernadsky" two detector blocks (Sodium crystals 80 x 80 mm or 80 x 400 mm large) were installed: one on the board of the vessel with the help of special suspernder-device placed at the depth of 1,5 m serving for water radioactivity control and another one was placed on the bow of the boat for radioactivity control of the fallout in the region. The equipment of direct measurements were a computer and a multichannel amplitude analyzer. The research equipment also included devices for radiochemical analysis of large amount of water (100-

150 l). The boat moved on the tack of 15 km/hr. The time of exposition was 20 or 60 sek, thus making possible integration of the results from regions 80 or 250 m long. Gamma-spectrography, water sampling, sediments and suspension were taken regularly.

During 1986-1988 eight area gamma-shootings of Kiev reservoir were performed as well as three wide-scale shootings of r. Dnieper cascades (from river mouth to the Black sea), shooting of the navigable part of the Pripyat river and up-river parts of r. Dnieper. The results are given in Fig. 6 (distribution of gamma-activity in the Kiev reservoir) and in Fig. 7 (gamma-shooting of Dnieper cascades). As it is seen, the radioactivity level fall was quite considerable. Thus, in the Kiev reservoir from June 1986 till May 1987 the radioactivity level of water in the estuary of r. Pripyat decreased 15-fold and 20-fold near the weir. continual measurements performed in the zone where waters of r. Pripyat mix with those of the Kiev reservoir showed a specific microstructure of flows with high radioactivity level of no more than 100-200 m in width (see Fig. 8). This effect is of considerable interest for hydrology. Moreover, it illustrates the possibility to select non-representative samples when selection is "blind".

On the ship "Academician Boris Petrov" /9/ there were used two detection blocks, one placed on sub-keel device (5 m-depth), second was towed 70 m under water surface. On drifting oceanographic stations deep water gamma-spectrometer was used. This enabled to registrate gamma-spectra on different levels. The investigation of radioactive situation in the region was carried out on the oceanographic stations and ships. Integrated scientific system of data collection and processing on the major part of hydrophysical fields made clear the dependence of vertical distribution of technogenic radionuclides on the hydrological conditions of the region.

In 1987 gamma-shooting of a number of regions in the Black Sea and the Baltic Sea was carried out. It showed the presence of Caesium-135 with concentrations of 1-5 x 10^{-13} Ci/l down to 100 m in the Black Sea and in the surface layers of the Baltic Sea.

Regular gamma-shooting of next and distant zones of accidental reject from Chernobyl APP was made from helicopter by means of Sodium Iodine detector of 3000 cm^3 volume. The results were used for mapping of the radioactive pollution of the region /10/. The investigation showed the necessity of correction of the results obtained by ground shooting in order to define more exactly the boundaries, pollution level and radionuclide composition. It is necessary for alteration of standard geophysical aerogamma-methods applied for greater inhomogeneousness of radioactive anomalies on the surface, for more complicated and variable radionuclide composition and per registration of possible radioactive fallout on the trees. It is important to note that for such aerogamma-shootings most essential is graduation of devices on special testing ground with defined and various radionuclide compositions and morphology.

Surface Mobile Gamma-Spectrometry Complex

On land we used a microbus where we installed complex measurement equipment. With it we were able to carry out continuous gamma-shooting in movement, radionuclide analysis of soil and water and continuous dose measurements. In order to decrease the dispersed irradiation of soil and plants we used collimating detection blocks and blocks similar to those on ships were used for measurements in water. The speed was

usually 20-40 km/hour. This complex enabled us to fulfill the following procedures:

- to estimate the dose values and variations in gamma-measuring radionuclide composition in regional and local analysis of contaminated territories in the Ukraine and Byelo-Russia;
- to identify a number of local "spots" of approximately 1 km which have not registered by aerogamma-shooting;
- to make corrections of the boundaries of radioactive contamination and isodoses and evaluate their dynamics;
- to identify the main radionuclide composition of the controlled regions in different "spots" (this simplified the procedure of sampling and their consequent gamma-spectrometry in laboratories);
- to provide collection of representative samples for their consequent radiochemical analysis on β- and γ-irradiating.

As it was shown, the correlation between the results of direct measurements on "spots" and lab. analysis of samples from the same "spots" was: for caesium-134, caesium-137 $\pm$ (15-20%), for ruthenium-106 $\pm$ (25-30%), caesium-144 $\pm$ 50%.

Fig. 9 shows results of gamma-ray measurements by means of the complex along one of the routes across the Chernobyl region.

Examples illustrate the efficiency of direct continuous gamma-ray measurements carried out by our Institute. this work is only a small part of wide-scale activity in the Chernobyl atomic power plant zone in 1986-88 carried out by more than 260000 people representing 40 Ministries and other organizations of the USSR /17/.

Perspectives of Method Application

Any serious accident causing radioactive contamination of the environment orders immediate actions for its liquidation. This calls for objective and prompt information on existing radioactive situation and its dynamics.

In 1984 in /11/ it was shown that in an accident when dehermetization of reactor core and destruction of circular pipeline take place in 1,5-2 hrs on the border of the safety zone around APP a zone of outer irradiation forms which reachs 10 zem. Another example of unsufficient promptness of radioactive control of the environment is the activities undertaken after the "Three Mile Island-2" accident /12/.

Evidently, all nuclear states have created wide systems of radiation control that function, usually, on the territories of APPs and provide (according to ICRP) "source monitoring". This system includes measurements around APP, detection of radionuclides in the air, fallouts, water of the surface reservoirs and ground waters, in soil, plants, botton deposits. The system gives objective information concerning the mean annual radioactive situation and, as a rule, satisfies the radioactivity level standards accepted in different countries. But these radioactive control media have one considerable drawback: the information refers to the past. It can be neglected when the exploitation regime of the APP is normal but thoroughly unacceptable in accident situations with uncontrolled emission of radionuclides in the environment. In such cases prompt information is necessary and is to be received within short time.

Nowadays, especially after the Chernobyl accident, many countries work out and introduce systems of automated radioactive ecological

monitoring. Thus, Great Britain announced national net (RIMNETO incorporating 90 surface stations all over the country for continuous gamma-ray measurement with Gaiger-Müller counters. The current information together with aerogamma shooting data and results of samples analysis is then passed over to the central data base /13/.

Denmark /14/ plans to install 10 stationary observation posts on the territory of the country which are to work in one-line regime. The equipment is to register 10-20% increase of natural background which makes up 0,02 $\mu c\ Sv.h^{-1}$. But such fluctuation may be due to increase in radon concentration after rains. Thus, observation posts, besides ionizing cameras with the broad measurement range from 0,01 $\mu c\ Sv/h^{-1}$ to 1 mSv/h^{-1} are provided with sodium iodine crystals and 256-channel analysers which have to extract technogenic nuclides out of natural background. The results will be accumulated on PC and will be transmitted every hour to the central station in Copenhagen and in case of an accident the data will be transmitted every 10 minutes.

In FRG /15/ a system of radioecological monitoring through continuous control of a radioactivity of the air is developed as well as technical requirements to its precision and climatic characteristics. Besides, FRG decided to create integrated information and measuring system to control radioactivity in the environment /16/. Powerful computer will collect, process and transmit this information to management. This information will be collected at normal and emergency situation as well. Similar systems of the highest level are developed in the USSR /18/.

The method of direct continuous gamma-spectrometric measurements thanks to its promptness, efficiency and security is the main in developed systems. As this method is used in different countries it is necessary to calibrate existing and developed equipment.

In conclusion it should be noted once more that besides problems of radioactivity control the method may be used for wide-scale research of the environment. It should be, evidently, recommended for the implementation of the international program "Biogeochemical pathways of artificial radionuclides" /19/.

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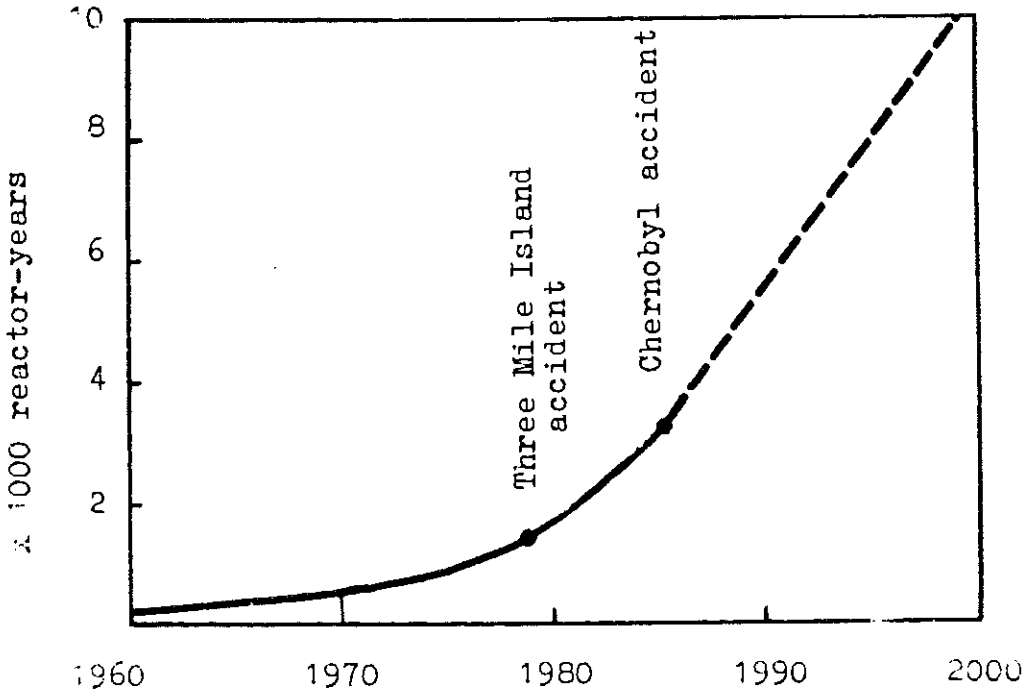

Fig.1. The global employment of nuclear reactors, 1960-2000

_ _ _ Dashed line indicates projection for 1985-2000

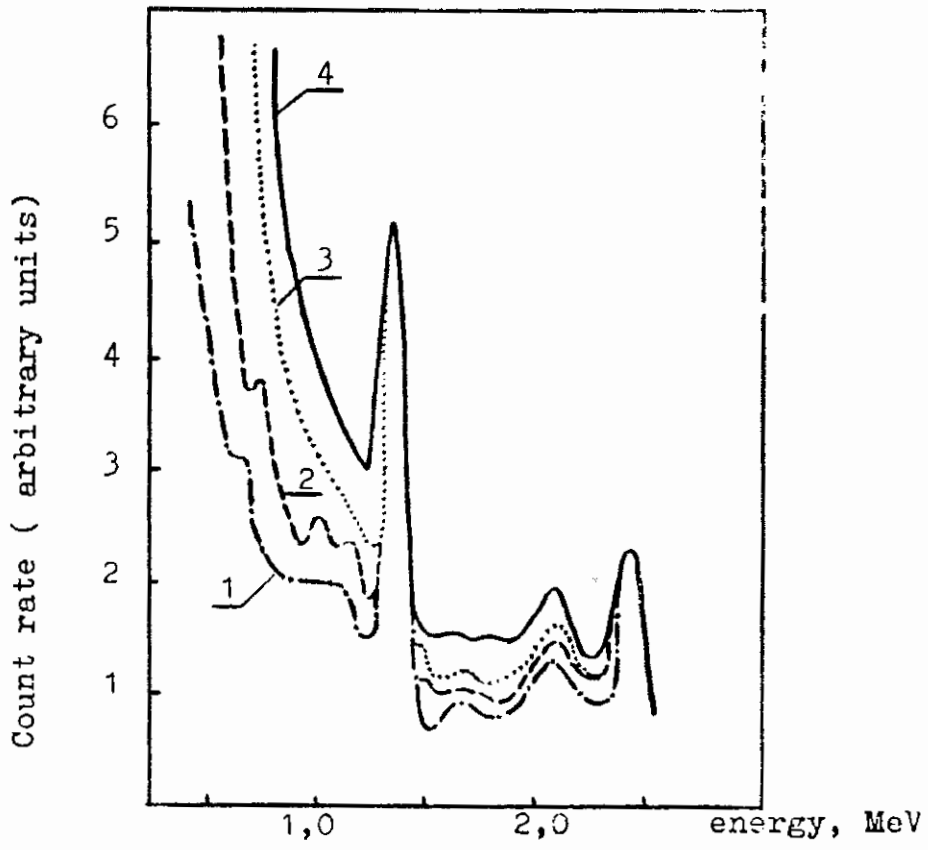

Fig.3. Sodium-24 gamma spectra transfered from the point source to the volume one.

The spectra are standardized by the photopeak 1.37 MeV.

1 - point source
2 - 3 l volume
3 - 300 l volume
4 - 10 000 l volume

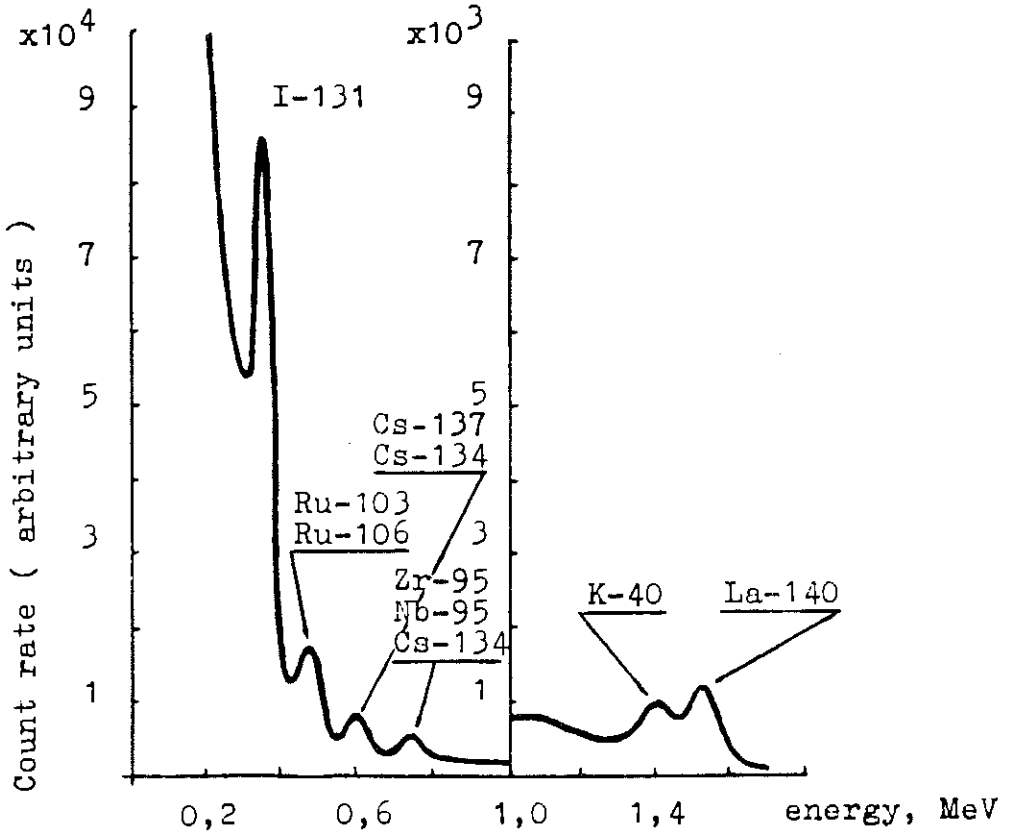

Fig.4. Gamma spectrum of water in Kiev reservoir (May 30, 1986)

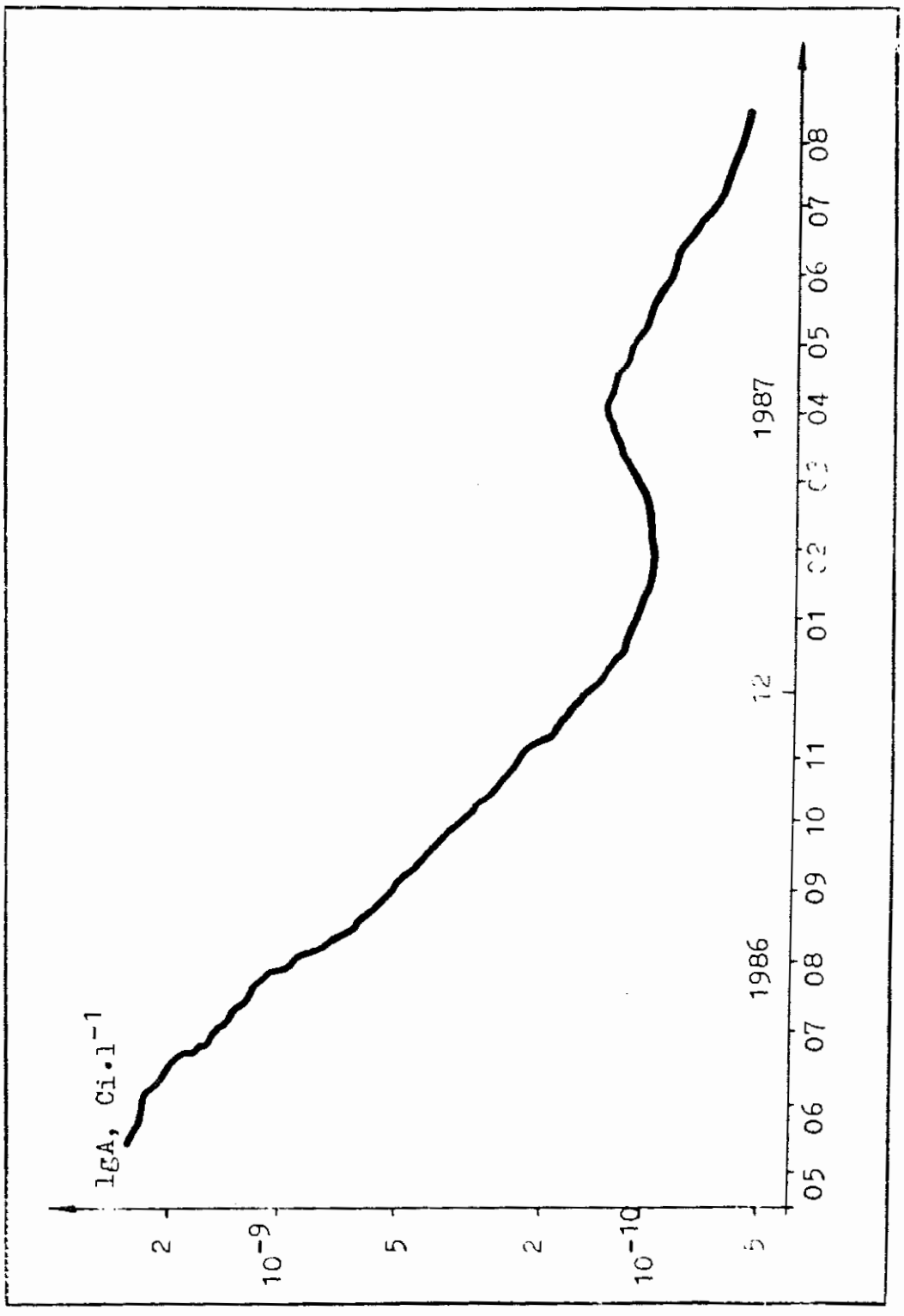

Fig.5. Summerized results of measurements in point №1 from Ju. 1986 to August 1987 (river Pripyat, Chernobyl)

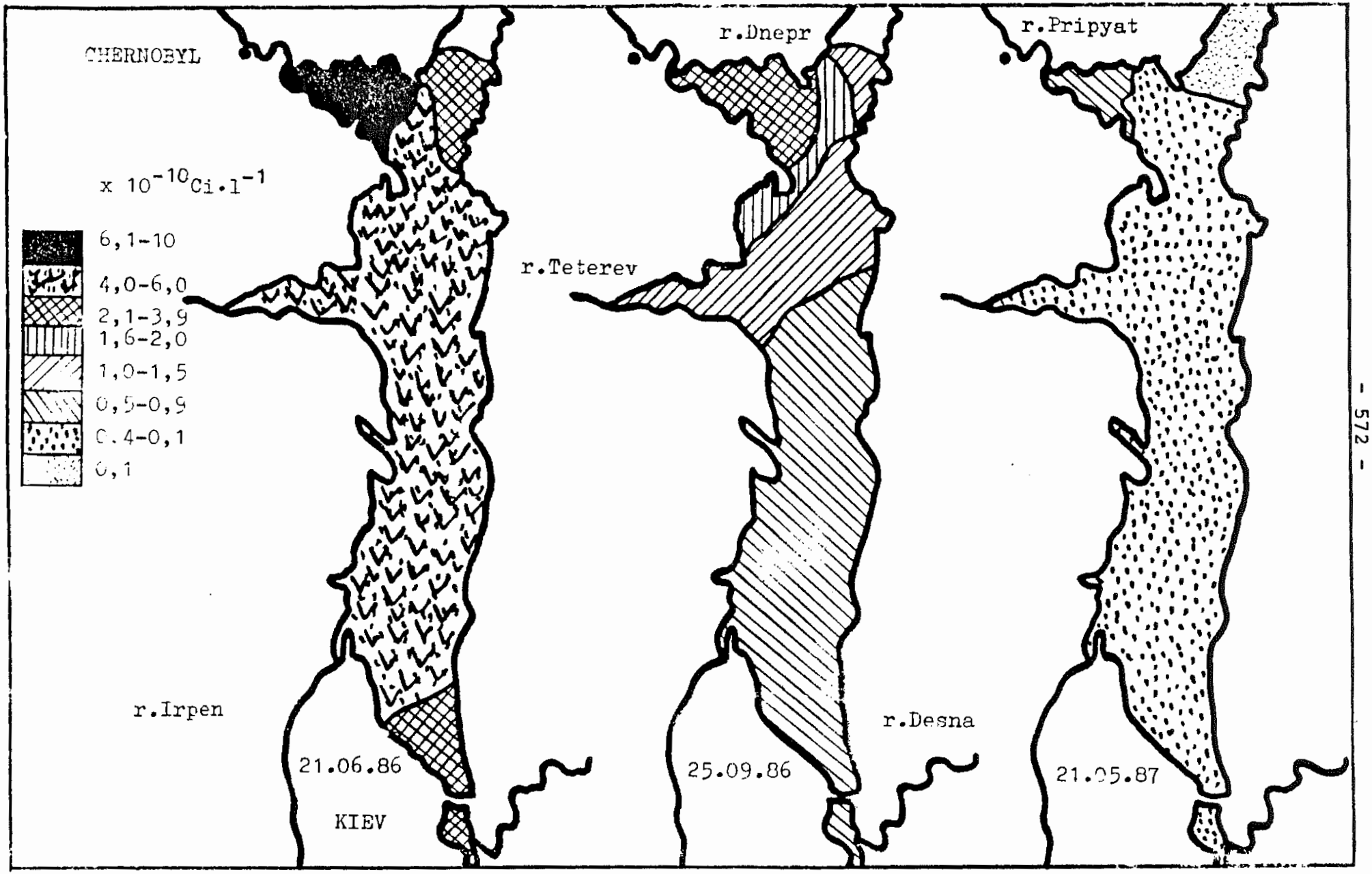

Fig.6. Gamma activity distribution in Kiev reservoir (according to 1986-1987 results)

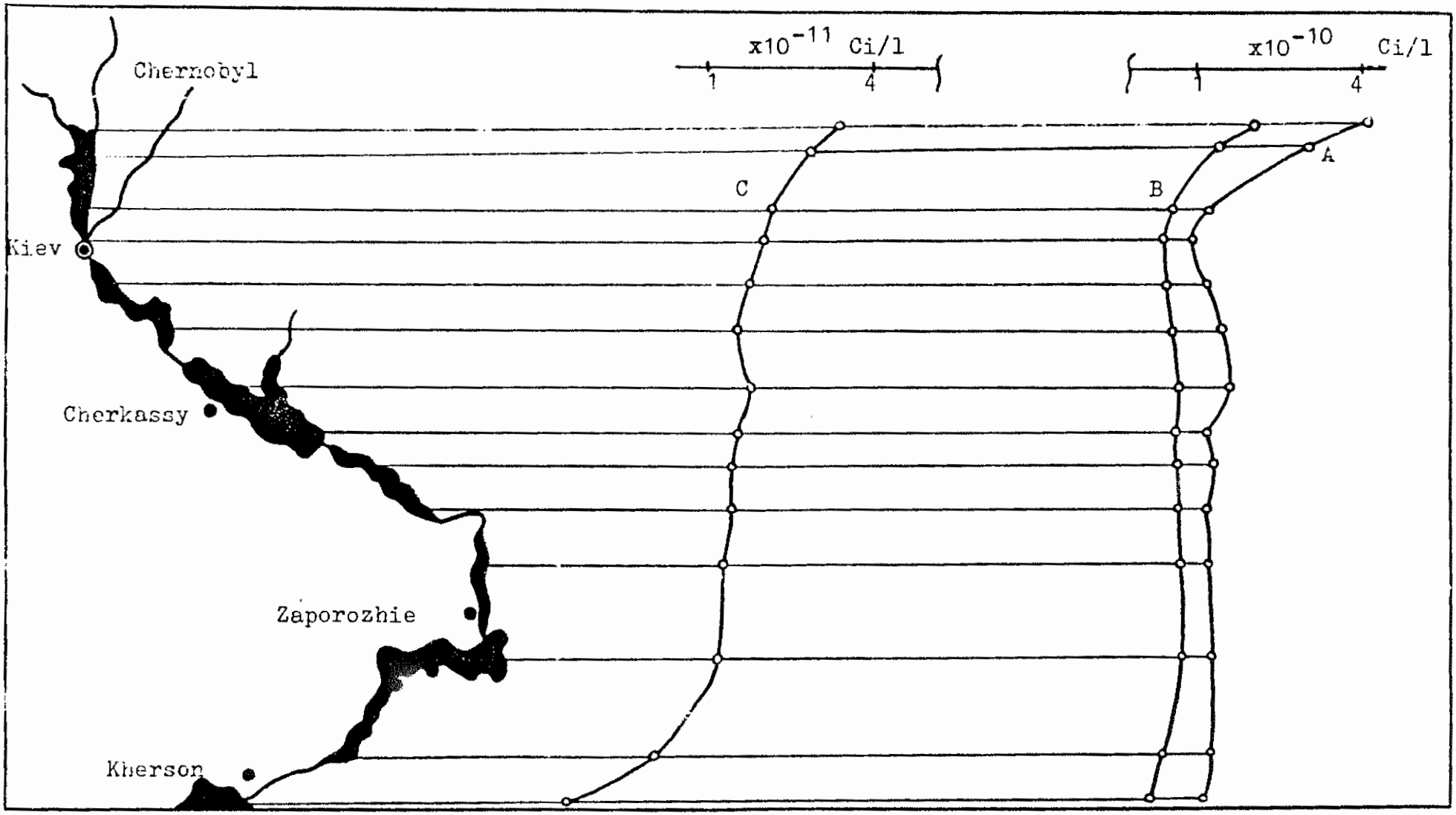

Fig.7. Gamma activity distribution in the Dnepr cascades (according to 1986-1987 results)
A - august 1986, B - october 1986, C - october 1987

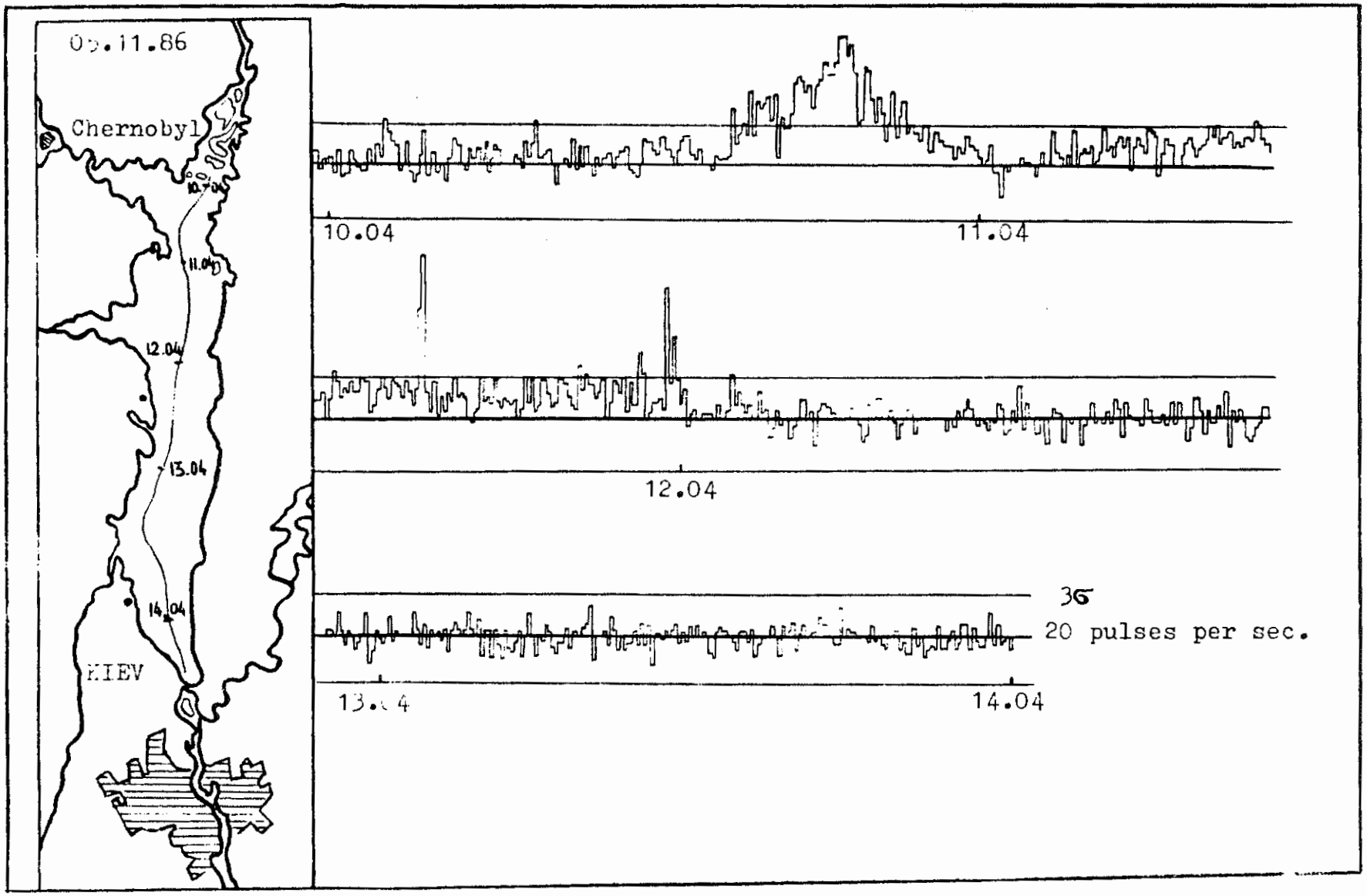

Fig.8. Microstructure of anomaly regions of the Kiev reservoir. 05.11.86

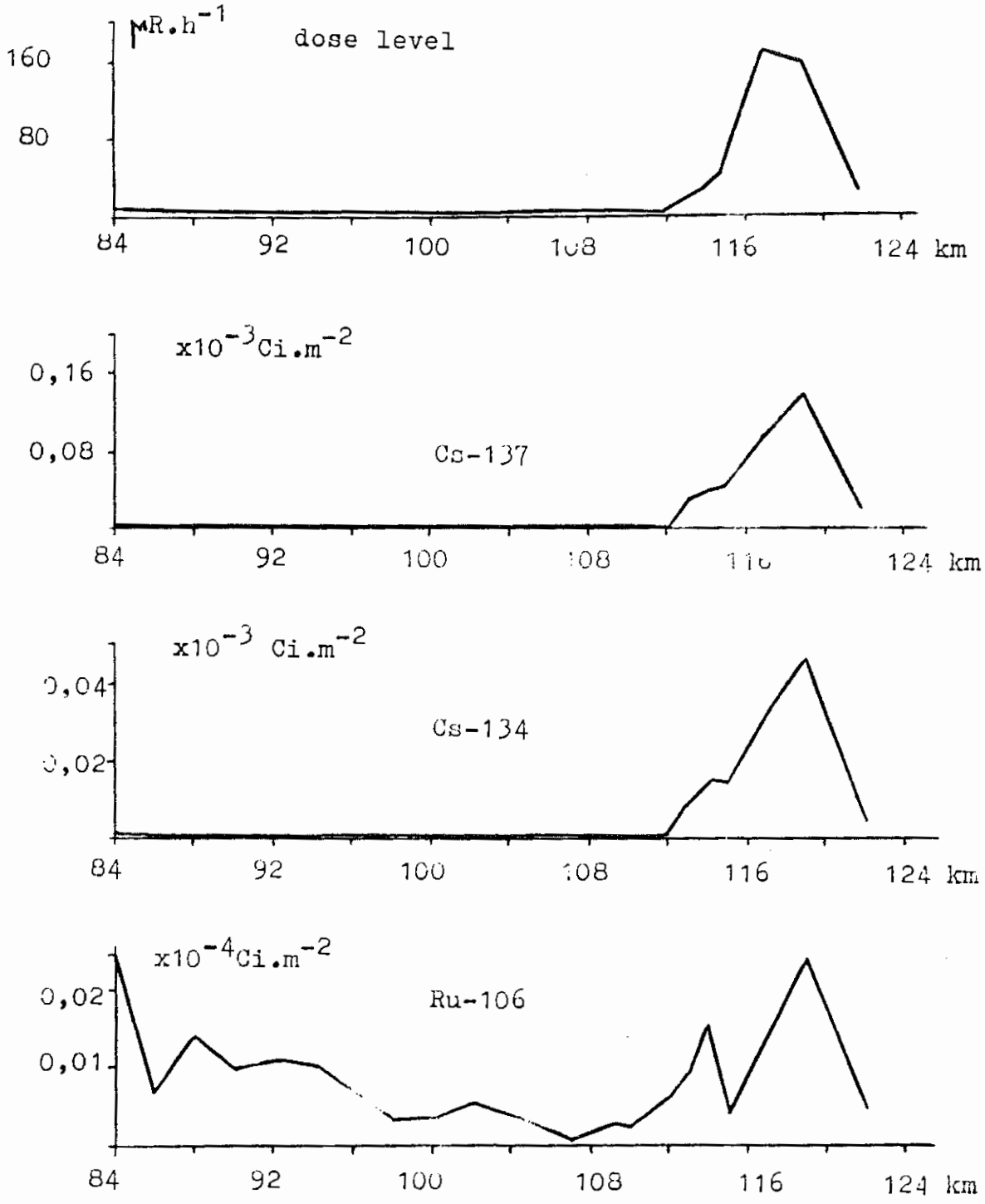

Fig.9. Results of the continuous autogamma imaging on one of the routes in the 30-km zone (August, 1987)

Radioactive Contamination of the Dnieper River and Baltic Sea Basins after the Chernobyl NPP Accident

Y.V. KUZNETSOV

ABSTRACT

The paper sets out the results of radiochemical and gamma-spectrometric analyses of the aqueous phase, suspensions and bed sediments from the Pripyat-Dnieper river network and Baltic Sea (from 1986 to 1988) employing methods devised at the V.G. Khlopin Radium Institute.

In order to study the vertical distribution of radionuclides in bed sediments and calculate their content on the bed, we adopted a layer-by-layer study method involving removal of integral cores with the aid of a specially developed sampling device; in this way we were able to establish the vertical and horizontal profiles of radionuclide distribution in sediments, and how they change in time and space. By means of selective lixiviation we estimated the resuspension ability of radionuclides in Kiev Reservoir bed sediments. Investigation of radionuclide interphase distribution between the aqueous and suspended phases, between water and sediments, plus use of ultrafiltration, made it possible to determine the forms of uptake and migration of major radionuclides in water bodies. It was shown that the overwhelming majority of radionuclides (over 10) are associated with the suspended phase in water bodies, mainly with the finely divided component. The main portion of caesium isotopes, and to a lesser degree strontium-90, was linked to the aqueous phase in water bodies, such nuclides being highly mobile. Radionuclide concentrations exceeding the permitted values under Radiation Safety Standards NRB-76/87 were not detected in the Kiev Reservoir or in the Baltic Sea.

The paper predicts - on the basis of knowledge of radionuclide forms of occurrence in water bodies and their physico-chemical behaviour - how the radioactivity in the water masses of the water bodies studied will change in time and space.

The paper details the dynamics of changes in radioactivity in the near-ground atmosphere of the Baltic region from April to June 1986 (on the basis of measuring the concentration in atmospheric aerosols of over 20 radionuclides, including caesium, plutonium and iodine isotopes).

The report ends by comparing the environmental impact of the Windscale and Chernobyl accidents.

The accident at unit four of the Chernobyl NPP released fragmented, induced radionuclides and shattered reactor fuel into the environment, subjecting not only the countryside within a 30-km radius to particularly intensive contamination, but also vast areas much further afield (via transport in the radioactive plume), including the water catchment areas of the Pripyat and Dnieper Rivers and the Kiev Reservoir. The radioactive cloud moved north, northwest and northeast, and contaminated a vast region in the northwestern USSR (Baltic Sea water catchment area and Baltic Sea proper). Although the total area of the USSR which was radioactively contaminated is considerably larger than that of the regions mentioned above, this paper deals only with these catchment areas since they were the ones studied by scientists from the Radium Institute between 1986 and 1988 (we began studying radioactive contamination of the Baltic Sea Basin in 1970 and have continued to do so on a regular annual basis up to the present time).

This report presents and discusses the results of our radiochemical and gamma-spectrometric analyses of the content of the main dose-emitting radionuclides in the liquid and solid (suspensions, bed sediments) phases of the water bodies studied. Our survey of the river system of the Dnieper and the Kiev Reservoir, started as early as the end of May 1986, lasted the whole summer and was continued in the subsequent years. This enabled us to study the changes in the radiation situation over time.

To study how the radionuclides were distributed in the aqueous phase of the water bodies we measured their content both in unfiltered water free of suspensions and in the suspended phase as well. We separated the suspensions from the aqueous phase by using nucleopore filters in a specially designed membrane device. We collected the sediments using aspecial sampler (designed by the Radium Institute) which allowed us to collect sediment cores without damaging their structure (retaining the surface layer), and we then studied how the radionuclides were distributed in the surface layers of the sediments (horizontal profile) and by depth (vertical profile).

To determine the plutonium content in the Pripyat-Dnieper-Kiev Reservoir water system we used existing data on the distribution of radionuclides in various parts of the environment around the Chernobyl NPP. From June to August 1986 the survey team from the V. G. Khlopin Radium Institute - working as part of the "Complex Expedition" arranged by the USSR State Committee for Hydrometeorology - measured the content of a number of gamma-emitting nuclides in the water, bed sediments and suspensions in the water system under study. We discovered that in July 1986 the caesium-137 volume activity in the water of the Pripyat River had hardly changed in the course of its flowing from Chernobyl to the river mouth, and amounted to some 4 000 Bq/m^3. The caesium-137 volume activity in the water of the Dnieper

around Kiev and in the Kiev Reservoir was 160-200 Bq/m^3. We also discovered that the content in the water of radionuclides such as cerium-141,144, ruthenium-103,106, zirconium-95 and niobium-95 tended to decrease as the water flowed through the Kiev Reservoir.

From these findings we obtained a rough estimate of how much plutonium was in the water system.

Table 1 gives the initial findings for plutonium-239,240 and plutonium-238 and their ratio in water samples taken from the Pripyat-Dnieper-Kiev Reservoir system between 8 and 19 July 1986, together with the results of a second series of measurements taken between 16 May and 4 June 1987. In 1986 the plutonium-239,240 volume activity in the water of the Pripyat River ranged from 0.6 to 48 Bq/m^3 (unfiltered samples). A decrease to 0.3 Bq/m^3 was observed in the surface waters of Kiev Reservoir, whereas plutonium-239,240 volume activity in the benthic waters was 27 Bq/m^3. The bulk of the plutonium (over 95%) was attached to suspensions.

Comparing these figures with the permissible concentrations (PCs) for plutonium 239,240 in drinking water, we find that the levels are several orders of magnitude below the PC_b or permissible concentration of radionuclides in water (or air) for a limited section of the population (Radiation Safety Standards NRB-76/87, where for plutonium 239,240 the PC_b in water is $2.2 \cdot 10^{-9}$ Ci/l or 81 000 Bq/m^3).

In 1987 the mean volume activity values for plutonium-239,240 in the water of the River Pripyat had decreased, whereas those in the surface waters of Kiev Reservoir had increased slightly. Fig. 1 gives the plutonium-239,240 measurements for 1986 and 1987.

Due to the small number of samples we are unable to specify the underlying processes involved in the distribution of plutonium. The scattered nature of the findings is apparently due to the different amounts of suspended matter in the various water samples, and this is particularly true of 1986 when the water samples were collected at a time of heavy river traffic and even dredging work. In order to determine the role played by suspensions in plutonium behaviour, we filtered water collected from the mouth of the Pripyat River in July and August 1986 and measured the plutonium-239,240 content in the filtrate and on the filter (Table 2). Over 90% of the plutonium contained in the water sample as a whole was attached to the suspended fraction. This confirms the earlier findings of French researchers, who found that 90% of the plutonium-239,240 was attached to suspensions extracted from samples of water from the Var and Rhône Rivers.[1] The major role played by suspensions in determining the behaviour of radionuclides in river waters applies not only to plutonium. Gamma-

spectrometric analysis of suspensions extracted from the River Pripyat has shown that some 75-97% of cerium-141,144, zirconium-95, niobium-95 and ruthenium 103,106 attaches to the suspensions. By contrast, radioactive caesium is found mainly in the soluble form (about 66%), which explains why this radionuclide cannot serve as an indicator of plutonium content in the water. This is confirmed by the fact that the plutonium-239,240/caesium-137 ratio in the water samples differs by two orders from the initial value in the damaged reactor.[2] As for cerium-144, both in the suspensions (Table 3) and the bed sediments (Table 4) we observed a correlation between its specific activities and those of plutonium-239,240. In 1987 the plutonium-239,240/cerium-144 activity ratios varied from 1.8 to $5.8 \cdot 10^{-3}$ in suspensions and from 0.4 to $2.3 \cdot 10^{-3}$ in bed sediments (mean value $1.75 \cdot 10^{-3}$). These ratios were determined at the time the samples were taken; in an extrapolation back to the time of the accident the ratio, according to our data, was found to be $7.85 \cdot 10^{-4}$, which is close to the value given in [2] for the damaged reactor.

In 1986 the plutonium-239,240 specific activities in the Pripyat River bed sediments varied considerably - from 8 to 890 Bq/kg (Table 5) - due to the work going on in this region to construct dykes and dams.

The plutonium content in Kiev Reservoir bed sediments varied from 10 to 54 Bq/kg, with higher levels (up to 340 Bq/kg) being observed in stagnant areas.

Table 4 shows the measurements of plutonium content in the bed sediments of the Pripyat and Dnieper Rivers; in May and June 1987 the specific activities of plutonium-239,240 varied from 26 to 810 Bq/kg in the Pripyat River, from 13 to 29 Bq/kg in Kiev Reservoir and from 1.6 to 26 Bq/kg in the creeks and channels near Kiev. These results are comparable with those obtained from bed sediment samples taken from this region in July and August 1986.

The attachment of plutonium-239,240 to suspended matter determines its subsequent behaviour. The deposition of suspended matter on the bed or, conversely, turbidity due to flooding and human agricultural activity, will also determine the displacement of plutonium-239,240 in the Pripyat-Dnieper-Kiev Reservoir water system.

In 1986 most of the radionuclides were located in the top 3 cm layer of the bed sediments. Specific activity for strontium-90 was up to 15 Bq/kg. Plutonium was unevenly distributed in the bed sediments (plutonium content varied from 41 to 2 520 Bq/kg in one and the same sample of sediment from the zone near where the accident occurred). The specific activities in sediments on the bed surface also varied considerably (up to 315 Bq/kg at the mouth of the Pripyat River and, on average, 22 Bq/kg in Kiev Reservoir, i.e. more or less the same as in the suspensions). In both sediments and suspensions we observed a high

positive correlation between the distribution of plutonium and cerium-144 ($r = 0.99$, $P = 0.95$, where r = correlation coefficient and P = confidence limit).

In 1986 a study of the vertical layer-by-layer distribution of radionuclides (cerium-144, zirconium-95, niobium-95, ruthenium-106, rhodium-106, caesium-137,134, strontium-90 and plutonium-239,240, inter alia) in the bed sediments revealed a sharp drop in their content below the top 3 cm layer, but as early as 1987 this high had shifted deeper into the sediment, pointing to radionuclide penetration into deeper layers.

Radioactive Contamination of the Environment in the Baltic Sea Basin

The Baltic Sea – which is a comparatively shallow and partially enclosed body of water exchanging water with the North Sea via the narrow Danish straits – is used intensively by a number of states for many economic purposes, including cooling of their nuclear power plants. This body of water has been subject to stringent ecological monitoring over the past few decades ever since it was found to be one of the worst in the world on a number of scores (e.g. pollution of the sea water with hydrocarbons). It is also monitored for radioactive contamination by the V. G. Khlopin Radium Institute in Leningrad and the Institute for Experimental Meteorology (IEM) in the town of Obninsk which since 1970 – in conjunction with scientific establishments in the GDR, the Polish People's Republic and Finland – have carried out regular studies into the processes underlying the propagation of a number of technogenic radionuclides in the Baltic Sea. This research covers not only the water systems of the Baltic Sea Basin (sea and river waters, the suspended phase, bed sediments, aquatic biota), but also the air masses in the near-surface layer of the atmosphere and atmospheric fallout, since this is one of the sources of radioactive contamination of water bodies.

The Chernobyl accident released into the atmosphere a considerable amount of different radionuclides, which air currents carried mainly west and northwest, leading to contamination of the Baltic Sea. Which radionuclides, and in which concentrations, determined the radiation situation in this body of water, what their fate will be and how quickly the Baltic Sea waters will cleanse themselves naturally are cardinal questions requiring an answer in the near future. In order to answer these questions we need to know how radioactively contaminated the Baltic Sea was before the Chernobyl accident and what were the main radionuclides involved, what is the link between the radioactive contamination in the air above the Baltic and in the basin itself, what determines the dynamics of radionuclide distribution in this body of water, and how soon can the Baltic Sea waters cleanse themselves of the dose-emitting radionuclides.

The work done by the Radium Institute from 1960 to 1985 in this field can supply some of the answers to these questions.[3-5]

Furthermore, related information culled from the extensive studies of radioactive contamination in the air and water masses in the Baltic Sea region can be used to evaluate the consequences of the Chernobyl accident as well, not only to compare data but also to make sound predictions about the future fate of the radionuclides from the fallout deposited on the Baltic Sea at the end of April 1986.

Fig. 3 contains curves showing strontium-90 and caesium-137 accumulation from 1957 to 1987, plus those for ruthenium-106 and cerium-144 in so far as they were contained in the fallout at the end of April/May 1986 and contributed significantly to the overall contamination of the earth's surface. Fig. 2 shows that prior to 1986 there was a wide spread of changes in the mean monthly volume activity of caesium-137, but that after the USA and USSR stopped carrying out nuclear tests in the atmosphere the trend is clear - natural cleansing of the atmosphere - with caesium-137 volume activity in the near-surface air falling from a peak of 2.10^3 $\mu Bq/m^3$ in 1963 to 0.2 $\mu Bq/m^3$ in 1985.

Technogenic Radionuclides in the Baltic Sea

As in the case of the air, strontium-90 and caesium-137 were also the main dose-emitting technogenic radionuclides in the water mass of the Baltic Sea from 1970 to 1985 (Fig. 4). Natural cleansing of the water bodies took place during this period via transfer of radionuclides to bed sediments and via radioactive decay. Fig. 5 shows the distribution in 1985 of caesium-137 in the surface and benthic layers of water in the Gulf of Finland and the eastern part of the Baltic Sea proper. As we can see, the caesium-137 content in the Gulf of Finland is lower than in the Baltic Sea proper, especially in the surface horizons; this shows that the water in the rivers emptying into the Gulf of Finland - such as the Neva and Narva - has a lower caesium-137 content due to sorptive removal of caesium by river suspensions and their subsequent deposition in the sea around the river mouth.

Up to 28 April 1986 the levels of artificial radionuclide contamination of the near-surface air and the levels of atmospheric fallout were the same as in 1985. As already noted, movement of air masses in the first few days after the accident carried the radioactive contaminants towards the Baltic Sea area, and precipitation during this period led to radioactive contamination of parts of the environment. The front of the radioactive cloud passed over the Scandinavian countries and the northwest of the USSR on 28 and 29 April 1986, sharply increasing the volume activity of a number of radionuclides in the near-surface air. In the following days and up to the end of 1986 the level of radioactive contamination of the atmosphere decreased due to radioactive decay of the short-lived radionuclides, and

throughout the troposphere in the northern hemisphere the fallout reaching the earth's surface also thinned out.

The intensity of fallout and accumulation of the individual radionuclides on the earth's surface after 26 April 1986 varied. Comparing the levels with those from 1961 to 1963 shows that they reached a new high for caesium-137, ruthenium-103 and ruthenium-106, while those for strontium-90, cerium-141, cerium-144 and zirconium-95 did not exceed the levels measured for fallout from 1970 to 1980.

Towards the end of 1985/beginning of 1986 radon, thoron and their decay products in the air were responsible for the volume radioactivity in the atmosphere (total natural radioactivity being ~ 1 Bq/m^3), while the contribution of technogenic radionuclides was negligible (this was determined via caesium-137 and strontium-90 content, and amounted to < 0.2 $\mu Bq/m^3$).

In the first few days after the Chernobyl accident, contamination of the atmosphere with iodine-131 was the most significant feature, accounting for up to 5% of the permissible annual iodine-131 content in the air (Radiation Safety Standards NRB-76). Subsequently this fell sharply and by 15 May did not exceed 1 000 $\mu Bq/m^3$, or 0.07% of the PC_b. Mean caesium-137 contamination of the air was 100 $\mu Bq/m^3$, i.e. 0.1% of the mean annual permissible concentration of this radionuclide in the air in populated areas. During this period the total activity of technogenic radionuclides in the air in the Leningrad area was considerably lower than its natural radioactivity from radon, thoron and their decay products (< 1 Bq/m^3). The high density of fallout inevitably increased the radioactive contamination of the Baltic Sea waters.

Initial measurements, made at the end of April/beginning of May 1986 in diametrically opposite regions of the Baltic Sea (in the eastern part of the Gulf of Finland [6] and near to the Danish straits [7]), revealed an increase in the radioactive contamination of the surface water layers in each case. At the same time increased radioactive contamination was recorded in a number of bodies of water along the Baltic coast.[8-9] These observations prompted wider studies.

In June 1986 we surveyed the waters in the Gulf of Finland and Gulf of Riga. The preliminary results obtained, which were presented at the 13th STC HELCOM Meeting [6], brought out in particular the heterogeneous nature - the "patchiness" - of the radioactive contamination, which reflected in turn the heterogeneous nature of the fallout. In these Gulfs we discovered two contamination zones between which there was an approximately tenfold difference in the volume activities of caesium isotopes, the more contaminated zone being in the eastern and central parts of the Gulf of Finland, while the less contaminated zone covered the western part of the Gulf of Finland and the Gulf of Riga. Our findings showed that the caesium isotopes predominated among the radionuclides present, the caesium-134/caesium-137 ratio being 0.5 on average for the entire body of water

surveyed, as it was in the fallout stemming from the Chernobyl accident. At the same time it was established that the strontium-90 content did not increase, remaining more or less at the global level even in the most contaminated parts of the sea, and from then on we paid particular attention to studying the distribution of caesium isotopes.

In July and August 1986 we surveyed the waters of the eastern part of the Baltic Sea proper, including the Kursky Zaliv (Courland Lagoon) and the Kaliningradsky Zaliv (Vistula Lagoon), and carried out a fresh survey of the Gulf of Finland. The findings, given in Table 6, are improvements on the caesium isotope measurements made in the Gulfs of Finland and Riga in June. All the caesium-134 and caesium-137 findings were obtained through radiochemical analysis of water samples.

The July-August survey confirmed the June observations. We saw a substantial difference in contamination levels in various parts of the Baltic Sea. In August the region of increased contamination in the Gulf of Finland could still be easily made out, but its clear-cut boundaries had become blurred, with the more contaminated region shifting westwards in the direction of the surface-water flow. Fig. 5 shows the distribution of caesium-137 in surface waters in the Gulf in June and August 1986. Given the constant caesium-134/caesium-137 ratio in the sea areas studied, we present the findings for one isotope only - caesium-137.

The eastern part of the Baltic Sea proper was the least contaminated of the bodies of water we surveyed, with a caesium-137 content in the surface waters of around 40 Bq/m^3. In the southeastern part and the Kaliningradsky Zaliv, as well as in the Bay of Danzig as a whole ^{10}, the caesium-137 content was slightly higher at 80-100 Bq/m^3. In all probability this region also hada "patch" of increased fallout, but one which was significantly less marked than that in the Gulf of Finland.

Analysis of water samples from the deep water horizons clearly revealed that caesium isotopes had penetrated to the deep layers of the sea, but the penetration rate was not the same for the various sea regions. Fig. 5a shows the distribution of caesium-137 in the surface and benthic layers of the sea throughout the regions surveyed in July and August 1986. Contamination with caesium isotopes had increased in the surface layers throughout the body of water surveyed, albeit to varying degrees. Thus, in July and August we observed, over the entire area, penetration of caesium isotopes from the surface layer to the deep ones. The rate of propagation of the contamination differed, as one would expect. By August 1986 it was only in the shallow areas of the Gulf of Finland (25-30 m) that the caesium-137 concentrations in the benthic and surface layers were comparable, whereas they had differed significantly in June (Table 6). In July and August we continued to find noticeable differences in caesium isotope content in the surface and benthic layers in very deep areas. Fig. 5 gives a general idea of caesium-137 distribution in the surface and benthic layers of the part of the Baltic Sea surveyed.

Given the increased radioactive contamination of the Baltic Sea waters in 1986, it was necessary to monitor the ingress of caesium isotopes via water from the Baltic Basin rivers. Fig. 6 contains measurements of caesium-134 and caesium-137 in the waters of a number of rivers which empty into the Baltic Sea from Soviet territory. Comparison of the results shows that rivers along the west coast of the Baltic were considerably less contaminated than rivers along the east coast - the Neva and the Narva. The waters of the Neva and its source - Lake Ladoga - contained significantly smaller concentrations of caesium-137 in comparison with the central part of the Gulf of Finland (Fig. 6). All this confirms the non-uniformity of the radioactive contamination of the water surfaces, due to the non-uniformity of the fallout, as a result of which a vast radioactive patch formed in the Gulf of Finland containing areas of differing contamination intensity. Fig. 6 contains data covering November and December 1986, showing that by this time the content of caesium-137 and caesium-134 in the Neva River and Lake Ladoga had decreased some twenty-fold as compared to the beginning of May.

From June to August 1986 - in addition to measuring the caesium isotopes - we measured the plutonium-239,240 content in the Baltic Sea waters. The findings set out in [7], relating to the waters of the southwestern Baltic Sea, show that in June 1986 the increase in plutonium-239,240 content was small as well, being no more than three to five-fold compared to 1985.

When studying radioactive contamination of the waters in the Gulf of Finland we always paid great attention to the region near the Leningrad NPP - the Koporsky Zaliv (bay) - where observations were carried out monthly in 1986. The Koporsky Zaliv, like the entire Gulf of Finland, was subjected to additional fallout, although to a somewhat lesser extent than the central part of the Gulf of Finland. The caesium-137 content in the surface waters of the Koporsky Zaliv fell from 400 Bq/m^3 in June to 190 Bq/m^3 in November 1986 (Table 6). During this period the phase distribution of caesium-137 in the Koporsky Zaliv waters was studied. The results obtained (Table 6) showed that most of the caesium-137 is found in the aqueous phase and some 2% on suspensions. Similar studies, carried out in 1982, had also shown that some 2.5-10% of the entire amount of caesium-137 in the water was attached to suspensions.[12]

The Chernobyl accident caused an increase in caesium-137 content throughout the Baltic Sea waters surveyed. However, in many regions, e.g. in the eastern part of the Baltic Sea proper, the contamination caused by this nuclide is lower than the caesium-137 content in the North Sea, whose waters are highly contaminated due to the systematic discharge of liquid radioactive wastes from nuclear reprocessing plants in Western Europe. As a result the North Sea will continue to increase the amount of caesium-137 in the Baltic Sea.

Thus, on the basis of the studies carried out from June to November 1986, we can conclude that only in the case of caesium-137 and caesium-134 is there an

additional accumulation stemming from the atmospheric transport of radionuclides released during the Chernobyl accident, and that there is no significant increase in the accumulated amount of strontium-90 and plutonium 238,239,240. The increased accumulation of caesium-137 raises the expected effective dose equivalent - for the population in the region where the highest radioactive fallout was registered - from 190 mSv (mean value for the northern hemisphere) to 191.6 mSv, i.e. by no more than 0.85% in all.

Throughout most of the Baltic Sea region the contribution from caesium-137 deposited as a result of the Chernobyl accident will be below this.

The contribution from the remaining radionuclides is negligible, given their half-life and low content (Fig. 3).

We should therefore expect that these waters will gradually cleanse themselves of caesium-137 as a result of cyclical sorptive removal of this radionuclide from the water by suspended matter. Caesium-137 accumulation in bed sediments will depend on the speed of suspension settlement and the type of bed sediment.

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Table 1: CONTENT OF PLUTONIUM-238 AND PLUTONIUM 239.240 IN SAMPLES OF UNFILTERED WATER TAKEN FROM THE RIVER PRIPYAT AND THE KIEV RESERVOIR IN 1986 AND 1987

Sample index	Sample collection point	1986				1987			
		Collection date	Pu-238 Bq/m3	Pu-239,240	Pu-238 / Pu-239,240	Collection date	Pu-238	Pu-239,240	Pu-238 / Pu-239,240
	RIVER PRIPYAT								
1.	Chernobyl town, landing stage	-	-	-	-	25.05	0.70 ±0.12	0.94 ±0.14	0.74
1*.	Chernobyl town, landing stage	17.07	33.0 ± 3.0	48.0 ± 4.0	0.69	-	-	-	-
2.	above Chernobyl town	16.07	0.5 ± 0.2	0.6 ± 0.2	0.85	-	-	-	-
3.	Chernobyl town, creek	-	-	-	-	03.06	0.17 ±0.04	0.25 ±0.04	0.68
4.	Buov 240	17.07	0.7 ± 0.2	1.2 ± 0.3	0.60	04.06	2.6 ± 0.30	4.7 ± 0.47	0.55
5.	Buov 234	17.07	17.0 ± 2.0	48.0 ± 4.0	0.36	-	-	-	-
6.	Buov 219	-	-	-	-	16.05	0.34 ±0.06	0.54 ±0.08	0.63
7.	Mouth	16.07	1.8 ± 0.3	5.1 ± 0.5	0.36	25.05	1.9 ± 0.20	3.5 ± 0.35	0.54
	KIEV RESERVOIR								
8.	Sukholuche	19.07	-	0.3 ± 0.2	-	26.05	0.7 ± 0.15	1.2 ± 0.18	0.58
8*.	Sukholuche	19.07	20.0 ± 2.0	27.0 ± 3.0	0.75	-	-	-	-
9.	Glebovka	-	-	-	-	23.05	0.35 ±0.15	0.4 - 0.06	0.87

* samples of benthic water

Table 2: DISTRIBUTION OF PLUTONIUM-239,240 IN THE SUSPENSION-FILTRATE SYSTEM
(natural water)

Sample index	Sample collection point	Collection date	Collection horizon m	Filtre type	Distribution of Plutonium-239,240 Bq/m^3		Distribution of Plutonium-239,240 %	
					Suspension	Filtrate	Suspension	Filtrate
6	R. Pripyat, month (buoy 219)	30.07.86	0	Paper 'blue band'	39.0 ± 4.0	0.4 ± 0.2	99	1
6	R. Pripyat, month (buoy 219)	13.08.86	0	Paper 'blue band'	36.0 ± 4.0	1.5 ± 0.3	96	4

Table 3: RESULTS OF MEASURING THE SPECIFIC ACTIVITY OF PLUTONIUM-239.240 IN SAMPLES OF RIVERS SUSPENSIONS FROM THE PRIPYAT AND DNIEPER (May and June 1987)

Sample index	Collection point	Volume of water l	Suspension concentrat. g/m^3	Specific activity Bq/kg suspension		Pu-239.240 / Ce-144
				Pu-237.240	Ce-144	
1.	Buoy-203	200	21	57	1.62 E 4	3.5 E -3
2.	Buoy-210-212	300	31	155	7.97 E 4	1.9 E -3
3.	Buoy-219	200	31	31	0.90 E 4	3.4 E -3
	R. PRIPYAT					
4.	Below pontoon bridge	200	42	20	0.57 E 4	3.4 E -3
5.	landing stage	155	47	19	1.06 E 4	1.8 E -3
6.	landing stage at Chernobyl	200	47	33	5.59 E 3	5.8 E -3
7.	Below pontoon bridge at entrance to quarry	200	41	238	6.34 E 4	3.8 E -3
8.	recess opposite mouth of R. Uzh	200	47	64	1.81 E 4	3.5 E -3
9.	Buoy-40. Kiev reservoir	30	32	14	4.69 E 3	3.0 E -3

Table 4: SPECIFIC ACTIVITY OF PLUTONIUM-239,240 IN SAMPLES OF BED SEDIMENTS FROM THE RIVERS PRIPYAT AND DNIEPER (May-June 1987)

Sample collection point	Specific activity Bq/kg		$\frac{\text{Pu-239,240}}{\text{Ce-144}}$
	Pu-239,240	Ce-144	
Buoy at Chernobyl creek	130 ± 13	7.26 E 4	1.8 E -3
Creek opposite mouth of R. Uzh (0-3 cm)	310 ± 30	1.66 E 5	1.9 E -3
R. Uzh (3-6 cm)	270 ± 30	1.47 E 5	1.8 E -3
R. Uzh (6-9 cm)	330 ± 30	1.97 E 5	1.7 E -3
Creek opposite mouth of R. Uzh	810 ± 80	4.22 E 5	1.9 E -3
Buoy-251 (bayou)	230 ± 20	1.85 E 5	1.7 E -3
Buoy-217	180 ± 20	7.91 E 4	2.3 E -3
Buoy-203	540 ± 50	2.58 E 5	2.1 E -3
Buoy-40	23 ± 5	1.24 E 4	1.9 E -3
Buoy-15	29 ± 6	1.58 E 4	1.8 E -3
Buoy-01	13 ± 3	7.69 E 3	1.7 E -3
R. DNIEPER Rusanovsky channel	1.6 ± 0.4	0.40 E 2	0.4 E -3
Matveyevsky creek	7.4 ± 1.4	3.52 E 3	2.1 E -3
Obolonsky creek	26 ± 6	1.43 E 4	1.8 E -3

Table 5: RESULTS OF MEASURING SPECIFIC ACTIVITY OF PLUTONIUM 239.240 IN SAMPLES OF BED SEDIMENTS TAKEN FROM THE RIVERS PRIPYAT AND DNIEPER AND THE KIEV RESERVOIR (July 1986)

Sample collection point	specific activity Bq/kg*	Sample collection point	specific activity Bq/kg*
R. PRIPYAT		KIEV RESERVOIR	
at Chernobyl	7.9 ± 0.8	Buoy 64	230 ± 35
below bridge	43 ± 4.0	Buoy 30	200 ± 20
Buoy 224	890 ± 90	Sukholuche	20 ± 2
Buoy 249	200 ± 30	Lyutezh	23 ± 4
Buoy 251	42 ± 4	Site of hydro-elec power station	47 ± 5
Mouth	140 ± 14	Near dam	340 ± 40
DNIEPER ABOVE KIEV Reservoir	9 ± 3		
R. Teterev, mouth	54 ± 8		
R. Desna, mouth	1.5 ± 0.3		
Sobache girlo	1.1 ± 0.2		
Obolonsky channel	1.7 ± 0.3		
Rusanovsky channel	32 ± 3		
Venetsianovsky chann.	20 ± 2		

* Results obtained from two parallel measurements

Table 6:
CONTENT OF CAESIUM-134 AND CAESIUM-137 IN WATER SAMPLES FROM THE EASTERN PART OF THE BALTIC SEA PROPER AND THE GULF OF FINLAND, AND ALSO FROM THE GULF OF RIGA, THE KURSKY ZALIV (Courland lagoon) AND THE KALININGRADSKY ZALIV (Vistula lagoon)

Sample index	Sample collection point		Collect. date	General water depth m	Depth at which sample taken m	Concentration Bq/m^3		Cs-134 / Cs-137
						Cs-134	Cs-137	
B-2	59°30'N	23°00'E	19.07	96	0	44 ± 7	110 ± 8	0.40
B-2	59°30'N	23°00'E	19.07	96	90	7	28 ± 3	0.25
B-4	59°00'N	21°30'E	19.07	103	0	30 ± 10	65 ± 10	0.45
B-4	59°00'N	21°30'E	19.07	103	100	10	28 ± 3	0.40
B-6	58°00'N	20°45'E	21.07	96	0	25	40 ± 11	0.62
B-6	58°00'N	20°45'E	21.07	96	90	9	23 ± 3	0.37
B-37	57°22'N	19°57'E	23.07	230	0	13	41 ± 6	0.31
B-37	57°22'N	19°57'E	23.07	230	200	6	22 ± 3	0.26
B-46	56°06'N	19°14'E	24.07	136	0	22	44 ± 7	0.50
B-46	56°06'N	19°14'E	24.07	136	100	6	36 ± 3	0.17
B-50	55°30'N	18°53'E	24.07	90	0	35 ± 10	87 ± 8	0.39
B-50	55°30'N	18°53'E	24.07	90	80	16 ± 3	34 ± 3	0.47
	GULF OF RIGA							
R-1	Central part		17.06	26	0	27 ± 5	68 ± 5	0.40
R-2	Eastern part		17.06	24	0	15	48 ± 5	0.33

Table 6: (continued)

Sample index	Sample collection point	Collect. date	General water depth m	Depth at which sample taken m	Concentration Bq/m³		Cs-134 / Cs-137
					Cs-134	Cs-137	
	GULF OF RIGA						
R-3	Eastern part	17.06	21	0	49 ± 6	79 ± 6	0.62
R-4	Southern part	18.06	25	0	23 ± 6	52 ± 5	0.44
R-5	Eastern part	18.06	35	0	36 ± 5	64 ± 6	0.56
R-6	Eastern part	18.06	30	0	40 ± 5	73 ± 5	0.54
R-7	Eastern part	17.06	20	0	61 ± 6	95 ± 6	0.64
	GULF OF FINLAND						
F-3	Nevsky Zaliv	06.08	4	0	130	330 ± 68	0.4
F-7	Nevsky Zaliv	06.08	5	0	100	130 ± 52	0.8
F-27	Koporsky Zaliv	23.06	11	0	180 ± 20	400 ± 24	0.45
F-28	Koporsky Zaliv	23.06	20	0	230 ± 20	440 ± 22	0.52
F-28	Koporsky Zaliv	23.06	20	15	245 ± 25	470 ± 24	0.52
F-31	Koporsky Zaliv	23.06	7	0	230 ± 16	400 ± 16	0.57
F-32	Koporsky Zaliv	23.06	13	0	220 ± 10	420 ± 16	0.52
F-32	Koporsky Zaliv	23.06	13	10	190 ± 19	420 ± 25	0.45
F-33	Koporsky Zaliv	23.06	9	0	195 ± 10	400 ± 16	0.49
F-34	Koporsky Zaliv	23.06	5	0	210 ± 10	400 ± 16	0.52
F-35	Koporsky Zaliv	23.06	6	0	200 ± 10	405 ± 16	0.49
F-38	Koporsky Zaliv	27.06	-	0	180 ± 10	300 ± 30	0.60

Table 6: (continued)

Sample index	Sample collection point	Collect. date	General water depth m	Depth at which sample taken m	Concentration Bq/m^3		Cs-134 / Cs-137
					Cs-134	Cs-137	
F-20	Central part	14.06	64	0	520 ± 31	970 ± 39	0.54
F-20	Central part	02.08	64	0	360 ± 58	870 ± 61	0.41
F-20	Central part	02.08	64	25	210 ± 23	490 ± 24	0.43
F-21	Central part	14.06	70	0	390 ± 23	660 ± 26	0.59
F-21	Central part	14.06	70	50	23 ± 5.7	71 ± 5.7	0.32
F-21	Central part	02.08	70	0	480 ± 62	1100 ± 77	0.44
F-21	Central part	02.08	70	50	66 ± 15	160 ± 16	0.41
F-23	Central part	14.06	84	0	475 ± 28	910 ± 27	0.52
F-23	Central part	01.08	84	0	460 ± 68	930 ± 75	0.49
F-23	Central part	01.08	84	75	23	66 ± 12	0.35
F-24	Eastern part	14.06	80	0	45 ± 5	97 ± 6	0.47
F-24	Eastern part	01.08	80	0	320 ± 60	530 ± 63	0.60
F-24	Eastern part	01.08	80	75	33	37 ± 19	0.87
F-36	Eastern part	20.06	35	0	42 ± 7	100 ± 6	0.42
F-37	Eastern part	20.06	50	0	50 ± 6	120 ± 6	0.42
	KURSKY ZALIV						
K-1	Central part	24.07	8	0	16	37 ± 8	0.42
K-2	Central part				26 ± 6	43 ± 6	0.61
	KALININGRADSKY ZALIV						
F-24	Eastern part	24.07	7	0	32 ± 5	83 ± 6	0.39
						mean	0.38

Table 7:
VOLUME ACTIVITY OF RADIONUCLIDES IN THE GROUND-LEVEL AIR NEAR LENINGRAD (28 April 1986)

Number	Radionuclides	Volume activity Bq/m
1	I-131	2.84
2	Np-239	1.16
3	I-132	0.372
4	Te-132	0.365
5	Za-140	0.358
6	Ba-140	0.311
7	Cs-137	0.286
8	Zr-95	0.279
9	Tc-99m	0.263
10	Ce-141	0.263
11	I-133	0.244
12	Mo-99	0.240
13	Ru-103	0.233
14	Nb-95	0.230
15	Ce-144	0.151
16	Cs-134	0.140
17	Ns-147	0.100
18	Te-129m	0.0537
19	Cs-136	0.0410
20	U-237	0.0355
21	Te-129	0.0341
22	Ru-106	0.0194
23	Be-7	0.00222
24	Pu-239,240	5.2 E -5
25	Pu-238	1.8 E -5

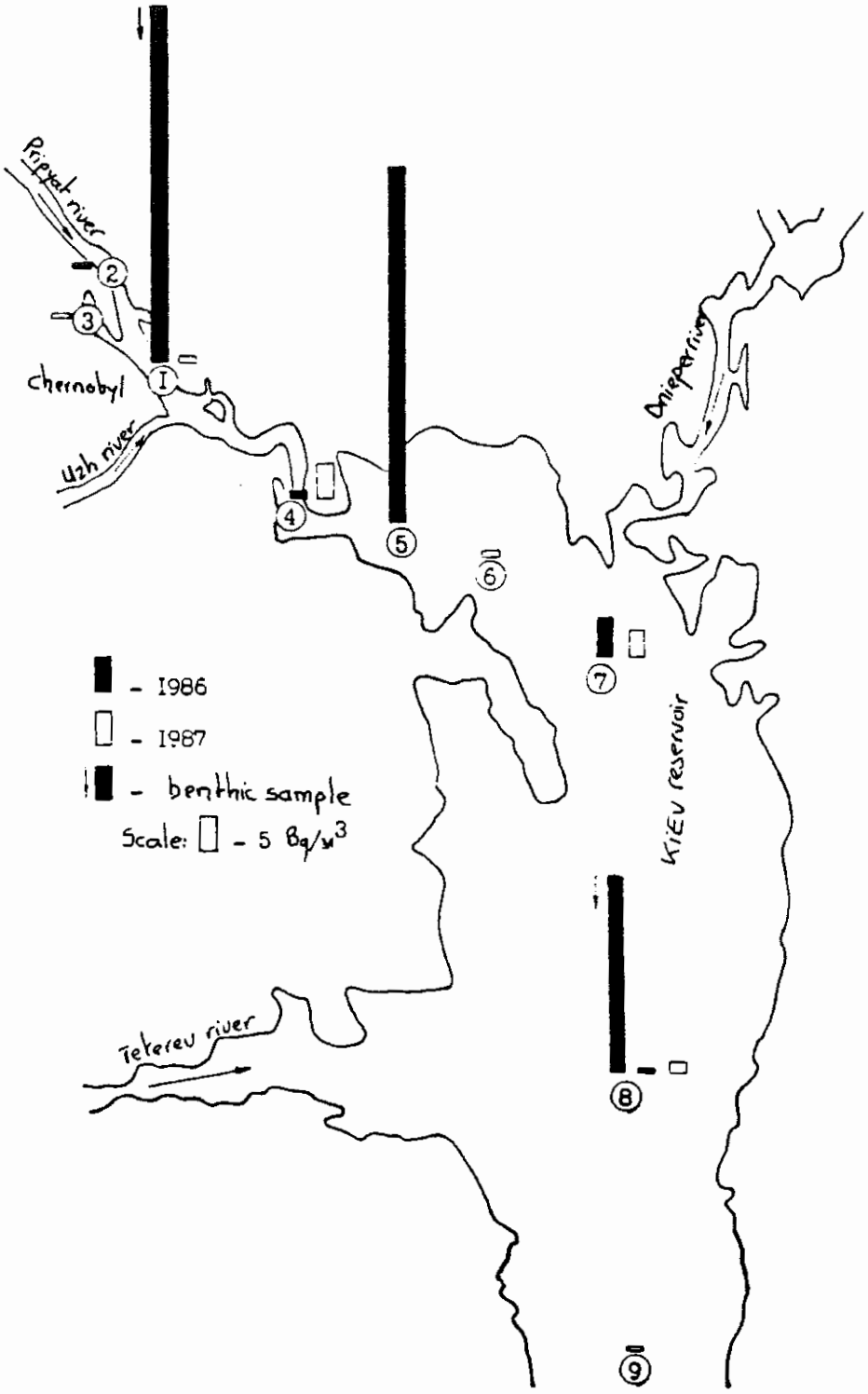


Fig.1: Sample collection sites and volume activities of Pu-239,240 (by scale) in the Pripyat-Dnieper water system in 1986 and 1987

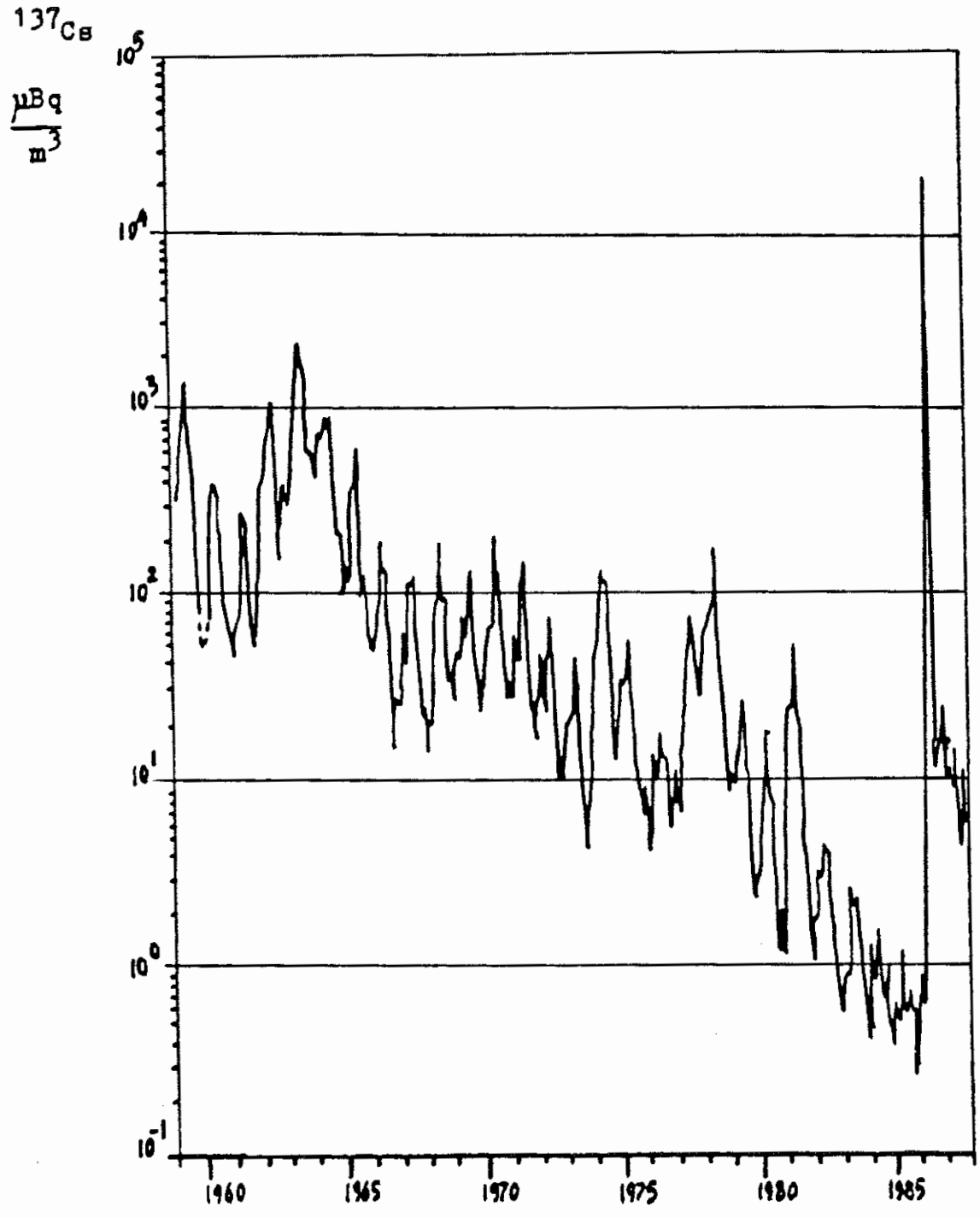


Fig.2: Change in average monthly activity of Caesium-137 in near-surface air, Leningrad, 1959-1986

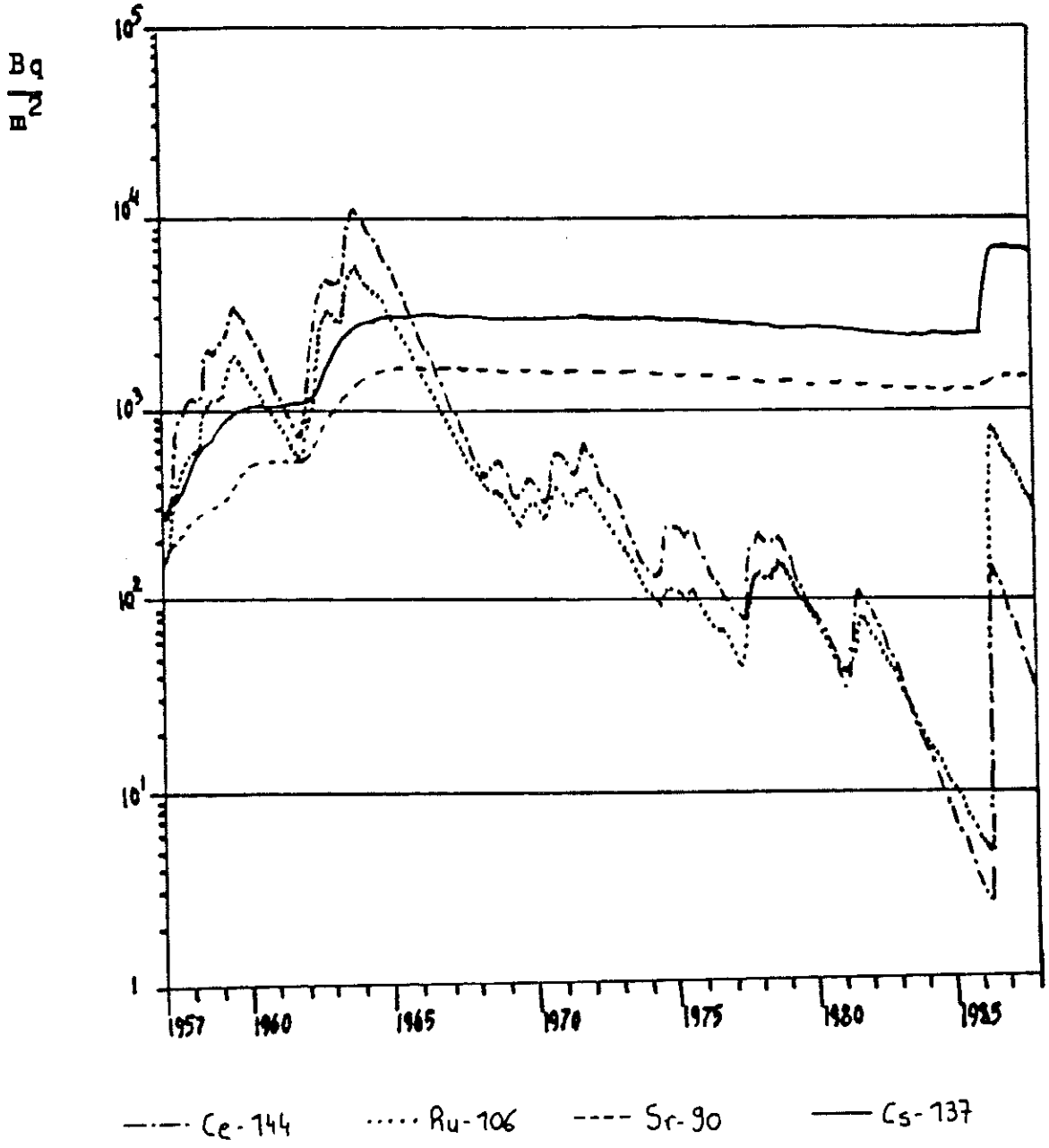


Fig.3: Accumulation of individual radionuclides on the earth's surface in the Leningrad area. 1957-1986

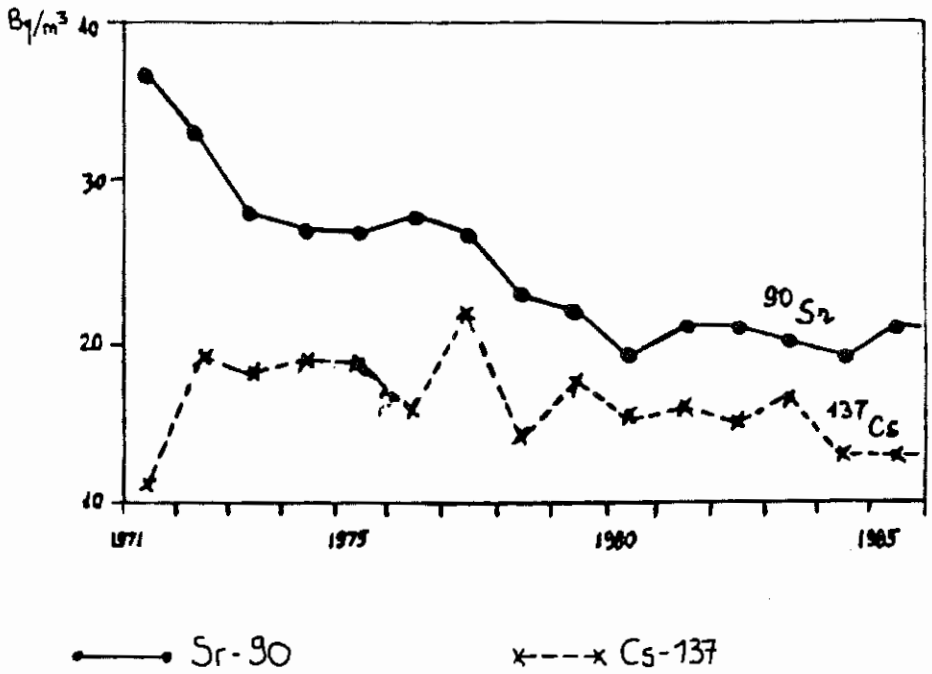
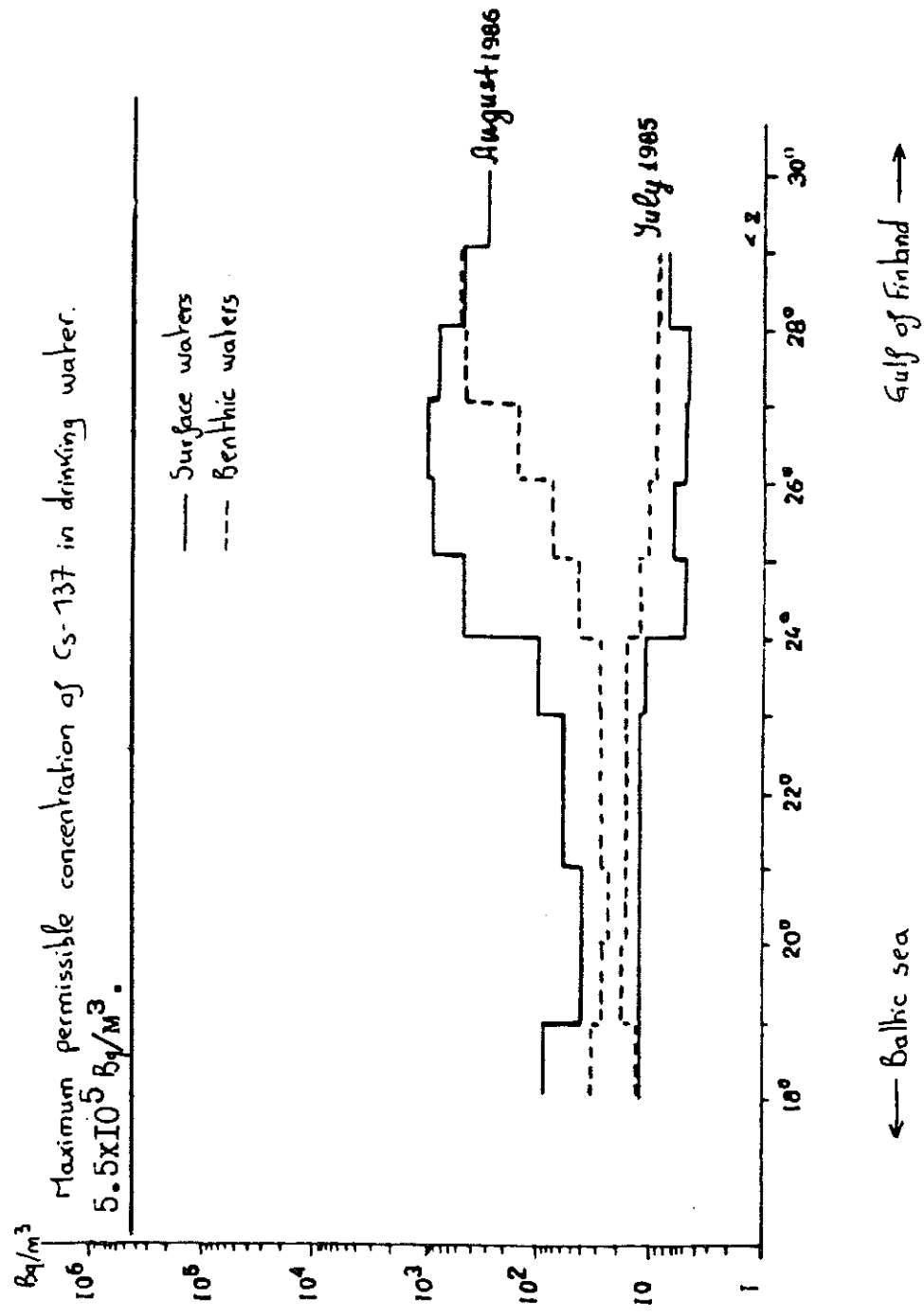


Fig.4: Radioactive contamination of the Baltic sea (1971-1985)

Fig.5: Distribution of Cs-137 in surface and deep waters of the gulf of Finland and in the eastern part of the Baltic sea proper



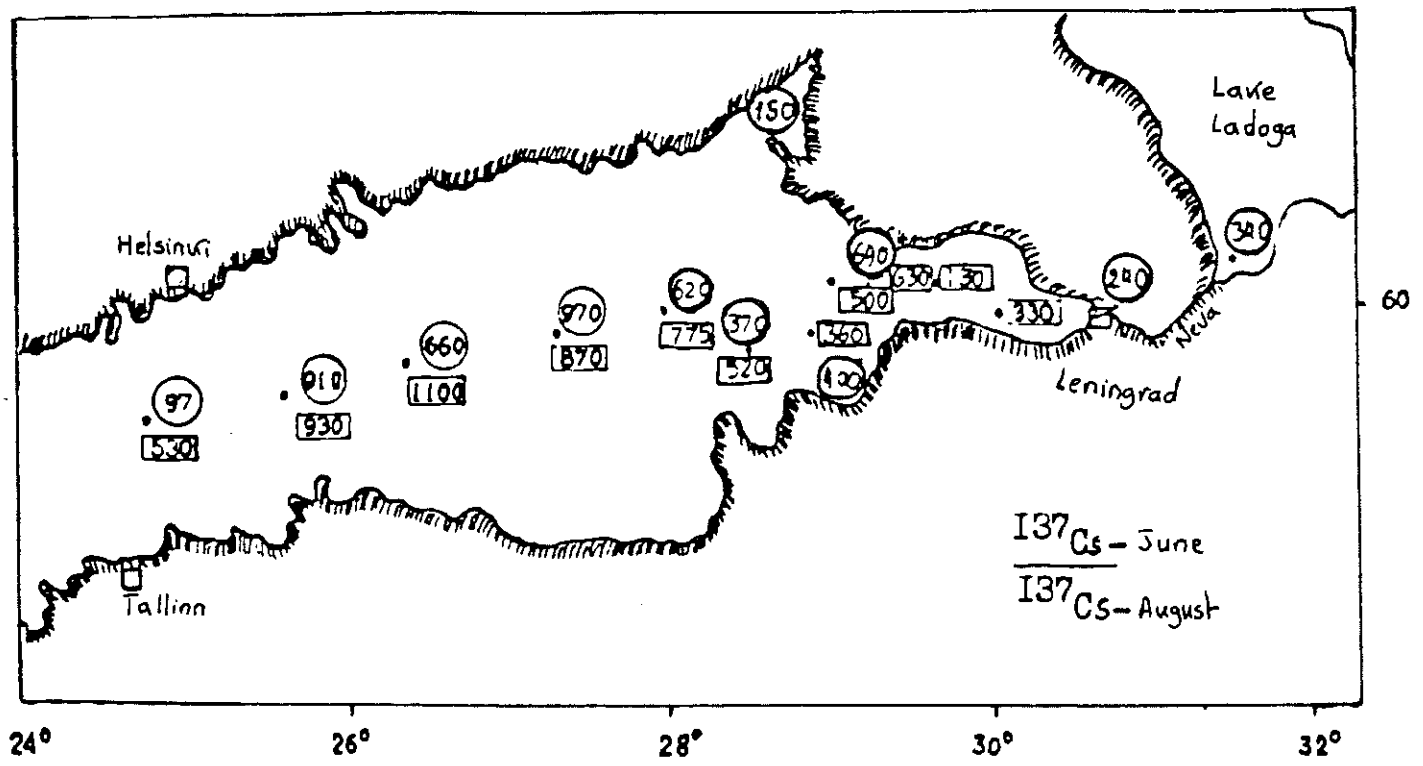


Fig.5a: Distribution of Cs-137 (Bq/m) in surface xaters of the guif of Finland (June-August 1986)

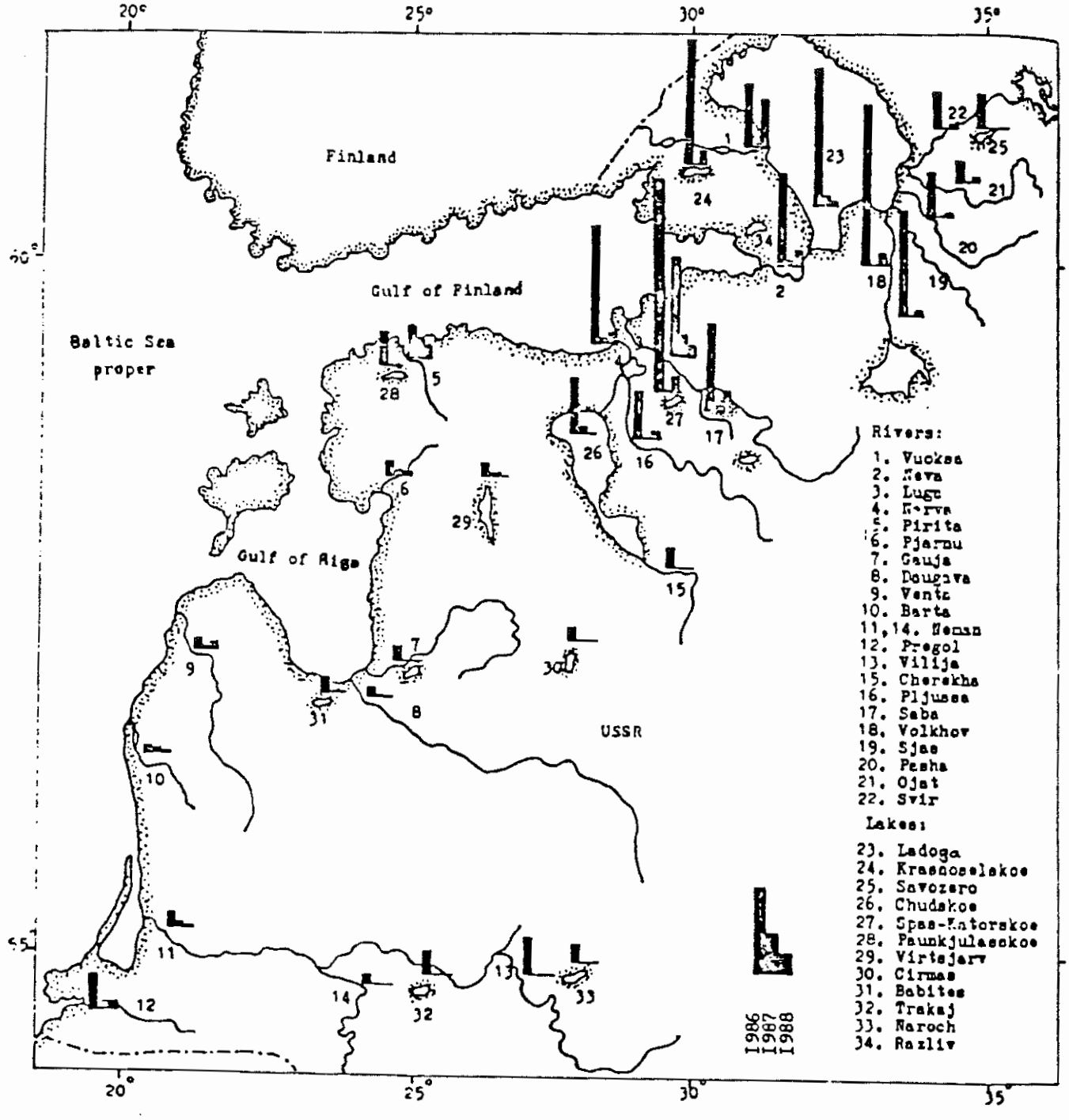


Fig.6: Distribution of Cs-137 in lakes and rivers in the Baltic area of the USSR, 1986-1988

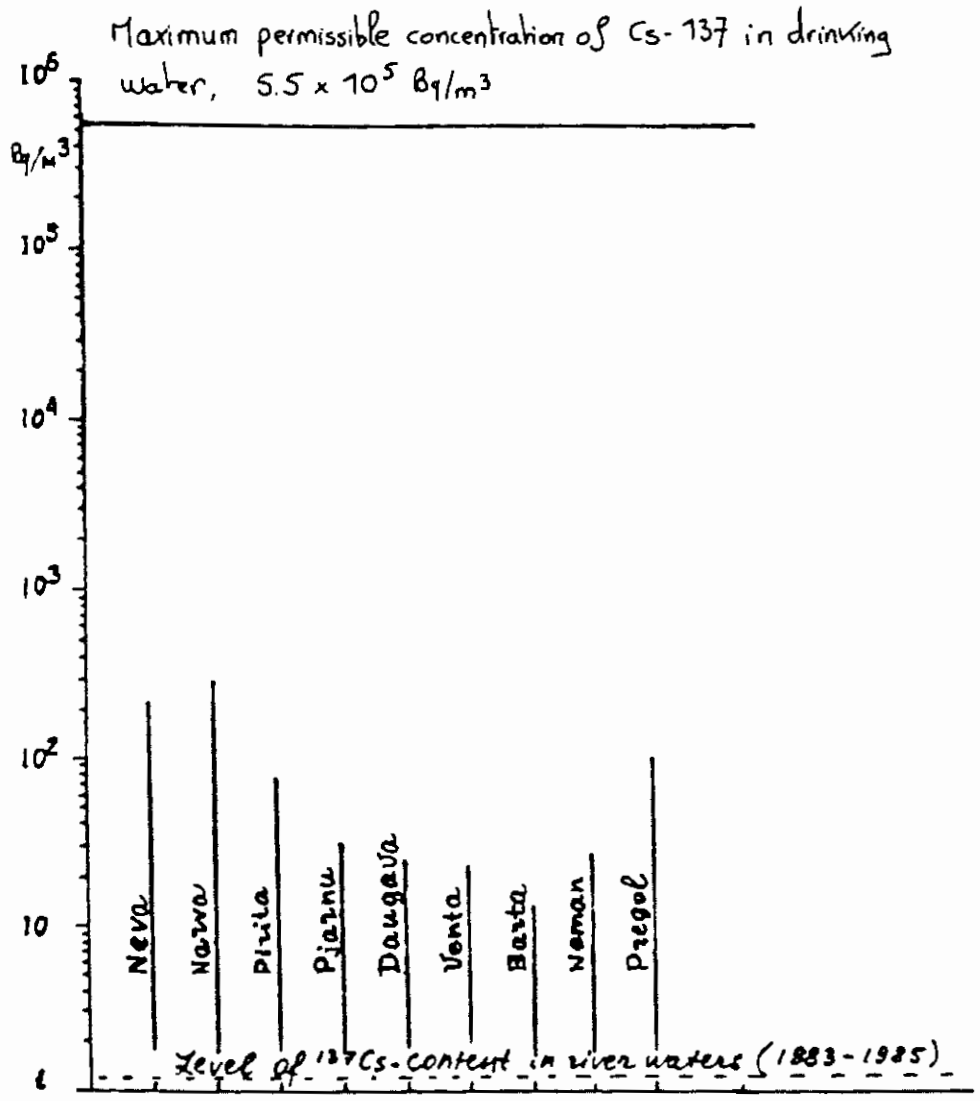


Fig.7: Content of Cs-137 (Bq/m) in May, July 1986 compared to background level and maximum permissible concentration for drinking water