



European Commission

# **Radiation protection 132**

## **MARINA II**

**Update of the MARINA Project on the radiological exposure of the European Community from radioactivity in North European marine waters**

### **Volume I**





# **Radiation Protection 132**

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## ACKNOWLEDGEMENTS

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<http://europa.eu.int/comm/environment/radprot/#studies>

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A great deal of additional information on the European Union is available on the Internet. It can be accessed through the Europa server ( <http://europa.eu.int> ).

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**Annex A Civil Nuclear Discharges into North  
European Waters**

**Annex B Environmental Data**



## Foreword

The present study, which is called MARINA II, has been undertaken for the Directorate-General for Environment of the European Commission in order to provide information on radionuclide discharges into North European marine waters and on radioactivity concentrations in the environment, and to provide an assessment of their impact. It builds on an earlier MARINA study, which considered data up to the mid-1980s.

The 1992 OSPAR Convention for the Protection of the Marine Environment of the North East Atlantic binds the following Contracting Parties: Belgium, Denmark, Finland, France, Germany, Iceland, Ireland, Luxembourg, the Netherlands, Norway, Portugal, Spain, Sweden, the United Kingdom of Great Britain and Northern Ireland and the European Community. The Convention replaces and up-dates the 1972 Oslo Convention for the Prevention of Marine Pollution by Dumping from Ships and Aircraft and the 1974 Paris Convention for the Prevention of Marine Pollution from Land-Based Sources. The definition of the North East Atlantic for the purposes of the OSPAR Convention covers the area north of the latitude of the Straits of Gibraltar ( $36^{\circ}$  N) and east of the longitude of the southern point of Greenland ( $42^{\circ}$  W) as far east as Novaya Zemlya ( $51^{\circ}$  E). The Baltic Sea (other than the Kattegat) and the Mediterranean Sea are excluded. The MARINA II Study is therefore of particular relevance to the work of the OSPAR Commission. In relation to the OSPAR Convention the major activities of The European Commission with regard to radioactive substances relate to activities agreed in discussions within the normal work of the OSPAR Radioactive Substances Committee and to consideration of ways in which the work of the European Commission and the OSPAR Commission may be arranged to assist each other's complementary aims. The OSPAR Commission has specific objectives set out in its Strategy with regard to Radioactive Substances, which provides focus for these activities. The MARINA II report is expected to contribute substantially to the implementation of the OSPAR Strategy by assisting the OSPAR Commission to address, in particular, sections 5.4 *a* and *b* of the strategy with regard to Radioactive substances and in the establishment of baselines for discharges and concentrations in the environment against which progress in implementing the Strategy can be judged, informed by an assessment of the environmental impacts.

The present document is the final report of the MARINA II study, which was conducted by NNC (UK) and its subcontractors: NRPB (UK), NRG (Netherlands), CEPN (France), Risø (Denmark), University College Dublin (Ireland), The Netherlands Institute for Fisheries Research, SPA Typhoon (Russia), CEFAS (UK), SSI (Sweden) and STUK (Finland). The progress and outcome of the study were thoroughly discussed by the Steering Committee, which included representatives of the European Commission and non-governmental organisations: the World Nuclear Forum, KIMO (Organisation of Coastal Local Authorities) and Greenpeace.

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Acting Director  
Environment and Health





European Commission

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### **Executive Summary**



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## **Executive Summary**

### **1 Scope of work**

The primary objective of the MARINA II study is to provide an input from the European Commission into the work of the OSPAR Commission in implementation of the OSPAR strategy with regard to radioactive substances and the work of the European Commission in respect of this strategy. It provides information on radioactive discharges, concentrations of radioactivity within the marine environment and an assessment of their impact on humans and marine biota. It follows an earlier MARINA I study [Commission of the European Communities, 1990], which considered data up to the mid-1980s.

The OSPAR Strategy with regard to Radioactive Substances, including waste, sets the objective of preventing pollution of the maritime area from ionising radiation through progressive and substantial reductions of discharges, emissions and losses of radioactive substances. The ultimate aim is for concentrations in the environment to be near background values for naturally occurring radioactive substances and close to zero for artificial radioactive substances. In achieving this objective, legitimate use of the sea, technical feasibility and radiological impacts on man and biota should be taken into account. As its timeframe, the Strategy further declares that, by the year 2020, the Commission will ensure that discharges, emissions and losses of radioactive substances are reduced to levels where the additional concentrations in the marine environment above historic levels, resulting from such discharges, emissions and losses, are close to zero.

The work of the MARINA II study was carried out by five groups of technical experts:

1. Group A, which collated data and made estimates of the discharges of radioactive substances and other inputs of radioactivity into North East Atlantic.
2. Group B, which analysed environmental concentrations and the radiation doses to members of critical groups of people.
3. Group C, which collated information on fishing and trade in sea products for use by Group D in collective dose assessments.
4. Group D, which modelled radionuclide transport in the North East Atlantic and assessed collective doses to the population of Europe.
5. Subgroup D\*, which assessed the impact of radioactivity on marine biota.

### **2 Conclusions**

The overall civil nuclear and other anthropogenic inputs of radioactivity into the North East Atlantic have decreased by several orders of magnitude for  $\alpha$ - and  $\beta$ -emitters and for tritium since the maximum levels were reached in the 1960s and early 1970s (Figures 1-6). Over the same time period this resulted in reductions in

radionuclide concentrations in the marine environment and consequently reductions in the individual doses to members of critical groups and in collective doses to the public.

Since the mid-1980s, the main contribution to discharges of  $\beta$ -activity into the OSPAR region is from the nuclear reprocessing plants (Sellafield and Cap de la Hague) while the discharges of  $\alpha$ -activity have been dominated by the phosphate industry and, later by oil production in the North Sea. As a result, oil production currently is the major contributor to the collective dose to the population of the European Union from industrial activities as discussed below.

### ***Main findings related to discharges from the nuclear industry:***

1. **Nuclear industry discharges** are still dominated by the reprocessing of nuclear fuel. Excluding the Chernobyl fallout in 1986, the input of  $\beta$  activity (excluding tritium, which has a very low radiotoxicity) into the OSPAR region decreased by over a factor of four from 1986 to 1991. By this date, the annual discharge had reached the same level as in the early 1950s (Figures 1 and 2). The reason was the major reduction in discharges from Sellafield and Cap de la Hague nuclear fuel reprocessing plants, which were major contributors over the years 1986-1991. Over the same period, the discharges of  $\alpha$  activity into the OSPAR region from Sellafield and Cap de la Hague decreased by a factor of three (Figures 3 and 4). Inputs of tritium have also decreased since the mid-1960s (Figure 5). However they have increased since the mid-1980s, due to the increase in reprocessing at Cap de la Hague. (Figure 6).
2. This lead to comparable reductions in the **concentrations of  $^{137}\text{Cs}$**  in the areas of highest concentrations in the Irish Sea near Sellafield (Figures 7 and 8). The increase in  $^{137}\text{Cs}$  concentration in the Baltic Sea is due to Chernobyl fallout. Outflow of water from the Baltic Sea means that concentrations of  $^{137}\text{Cs}$  in seawater from the Kattegat, the straight between Sweden and Denmark, have not declined significantly in recent years.  $^{137}\text{Cs}$  is the most widely measured radionuclide in North European waters because of its significance for radiation exposure and because it is relatively easy to measure.
3. Since 1986, the **radiological impact on the most exposed groups of populations** (effective dose to members of the critical group) in the vicinity of the major nuclear sites, such as Sellafield and Cap de la Hague was consistently and significantly below the ICRP and EU Basic Safety Standard limit of 1 mSv per year to members of the general public. The range of doses to members of the critical groups for these two sites during 1988-1999 was 0.01-0.4 mSv per year for anthropogenic radionuclides. The variation in such doses was primarily due to changes in the consumption rates of marine produce by the most exposed groups of the population.
4. Over the period 1988 to 1999 effective doses to critical groups in the **Sellafield area** show no trends but for the **Cap de la Hague** area there is a decreasing trend. For the **OSPAR region in general** the doses to critical

groups follow the same decreasing trend as the environmental radionuclide concentrations.

5. Human habits naturally play an important role in the assessment of radiation doses to critical groups. Existing assessments of radiation exposure from marine pathways to critical groups in the OSPAR region have used a variety of habit data ranging from cautious conservative assumptions to realistic data from detailed habit studies. Dose assessment is very sensitive to variation in habit data so in order to normalise the data **individual doses** have also been calculated **using normalised consumption rates** (Figure 9). In this estimate all exposure has been assumed to result from consumption of seafood based on the maximum seawater concentration of the isotopes  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{99}\text{Tc}$  and  $^{239,240}\text{Pu}$ .
6. Near **Sellafield, critical group doses** were dominated by  $^{241}\text{Am}$ ,  $\text{Pu}$  isotopes and  $^{99}\text{Tc}$ . Since 1986, the level of  $^{241}\text{Am}$  in water and marine organisms remained relatively stable due to its ingrowth from  $^{241}\text{Pu}$  and remobilisation from sediments in the Irish Sea. Sediment contamination resulted from peak discharges in the early 70s. The same remobilisation phenomenon was the basis of the relative stability in plutonium concentrations. While marine discharges of other significant radionuclides have declined since 1986, discharges and environmental concentrations of  $^{99}\text{Tc}$  increased in 1994 when the treatment of historic liquid wastes started at the Enhanced Actinide Removal Plant in Sellafield.
7. **The overall radiological impact from the nuclear industry** (collective dose rate) on the population of European Union from the OSPAR area has decreased from 280 man Sv/yr in 1978 to 14 man Sv/yr in 2000. This reduction was primarily due to decreases in the discharges of  $^{137}\text{Cs}$  and  $^{106}\text{Ru}$  (Figure 10). Collective doses are conventionally used as an input into the optimisation of radiation protection. They can also be used to compare the radiological impact of particular industrial practices and this is the main purpose of the estimation of collective doses in this study.
8. In terms of contributing to collective dose, **discharges from nuclear power generation, fuel fabrication and research reactors** were negligible compared with discharges from nuclear fuel reprocessing. The contribution to the total collective dose from marine discharges from these installations was estimated to be just 2% in 2000.

### ***Main findings related to discharges from non- nuclear industries:***

9. ‘Natural’ as opposed to ‘man-made’ radionuclides were largely present when the earth was made or result from the natural radioactive decay of such nuclides. Two major sources of the so-called ‘Naturally Occurring Radioactive Materials’ (NORM) were considered in the MARINA II study. NORM can be defined as all naturally occurring radioactive materials where human activities have increased the potential for exposure in comparison to the unaltered situation. Activity concentrations may or may not be increased.

- a. **Discharges of phosphogypsum**, which contain significant quantities of such radionuclides as  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  and smaller quantities of Uranium and Thorium isotopes. Phosphogypsum used to be discharged into the OSPAR area in the Netherlands, UK, France, Belgium/Luxembourg, Spain, Denmark, Former West Germany, Portugal and Ireland during the production of phosphoric acid by the fertiliser manufacturing industry. These discharges were largely stopped by 2000 with the introduction of the dry process, new treatment techniques, the storage of phosphogypsum ashore and the import of phosphoric acid from North Africa and the Middle East. There is no information on phosphogypsum discharges prior to 1981.
  - b. Pumping **oil and gas** from the continental shelf in the North Sea produces large quantities of contaminated water, known as 'produced water'. This results in releases into the marine environment of  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$  and  $^{210}\text{Pb}$ , which are concentrated, and made available for consumption by biota. Off-shore oil production in the North Sea, which is located mainly in the Norwegian and UK coastal waters, increased significantly from the 1970s until 1995, but has remained relatively constant since then.
10. Except for the phosphate production in Whitehaven in Cumbria, North-West England, there are **very little data on discharges** or environmental concentrations resulting **from non-nuclear industries**. MARINA II made 'best estimates' of the magnitude of such discharges and the resulting radiological impact based on the estimated normalised concentrations of radionuclides and the quantities of discharged effluents.
  11. The **overall discharge of  $\alpha$ -emitters into the OSPAR region** has remained constant since 1986 due to the discharges from the phosphate industry and the production of oil in the North Sea (Figure 4). By 1999, the estimated discharges of produced water alone contributed 90% of the discharge of  $\alpha$  activity into the OSPAR region. Since at least 1981, the discharges of phosphogypsum from the phosphate industry have dominated the **collective dose** to the population of the European Union (Figure 11). This is because of the higher radiotoxicity of the radionuclides discharged by these industries compared to that of the radionuclides that are discharged from the reprocessing plants.
  12. The **peak collective dose rate from NORM industries** occurred in 1984 and was just over 600 man Sv  $\text{y}^{-1}$ . This collective dose was almost entirely due to discharges from the phosphate industry with the important sources being discharges into Cumbrian waters from the UK and into the North Sea from the Netherlands. Discharges from the phosphate industry, particularly in the UK, were reduced in the 1990s but the phosphate industry is still a major contributor to the collective dose rate.
  13. **Discharges from the oil and gas industry**, which made a small contribution over much of the period from 1981 to 1999, have become relatively more



important. In 2000, discharges from the oil and gas industry contributed about 39% to the total collective dose rate from the NORM industries.

14. The possible **discharges from the production of phosphoric acid around the Mediterranean Sea** and the consequential impact on the population of the European Union have not been assessed as part of this study. This aspect may need to be addressed if MARINA MED [Commission of the European Communities, 1994] is updated at some point in the future.
15. It was found that discharges and collective doses resulting from the production and application of **radiopharmaceuticals** were negligible in comparison with those from either nuclear reprocessing or oil production. The same applies to discharges from **shipyards** servicing nuclear submarines in the UK, **historic dumping of wastes at sea and submarine accidents**.
16. The **Chernobyl accident** caused an additional input of radioactivity into the OSPAR area since 1986, which resulted in a small increase in collective dose rate (see Figure 11). The impact via marine pathways of earlier fallout due to the open-air testing of nuclear weapons peaked in 1964 at 43 man Sv but now is relatively stable at 7 man Sv y<sup>-1</sup>.

#### ***Overall impact of discharges:***

17. The **collective dose rates** to the population of the EU over the period 1981 to 2000 due to discharges from all sources are shown in Figure 11. At its peak, collective dose rate of about 760 man Sv y<sup>-1</sup> is around a factor of 20 less than the annual collective dose from natural radioactivity in the marine environment.
18. **If all discharges of radioactivity stopped** in 2000, the collective dose rate to the European population in 2020 would be approximately half of what it would be if the nuclear industry and the oil extraction industry continued to discharge at the present rate (Figure 12). However, the dose to individuals in the critical group close to Sellafield would be less affected by reduction in discharges because it is largely due to historic discharges. The collective dose rates can be compared with a collective dose rate to the population of the European Union from natural radionuclides in the marine environment of 17,000 man Sv and an annual collective dose from all sources of natural background radiation of 844,000 man Sv (see Figure 13).
19. The present **model for estimating collective** dose rates has been well validated for current conditions and the physical mechanisms that determine the dose rate are well understood. However, significant changes, such as global warming, would invalidate predictions into the future and there is clearly a need to continue to monitor both discharges into and concentrations of radioactivity in the marine environment throughout the OSPAR region.

### ***Impact of discharges on marine biota:***

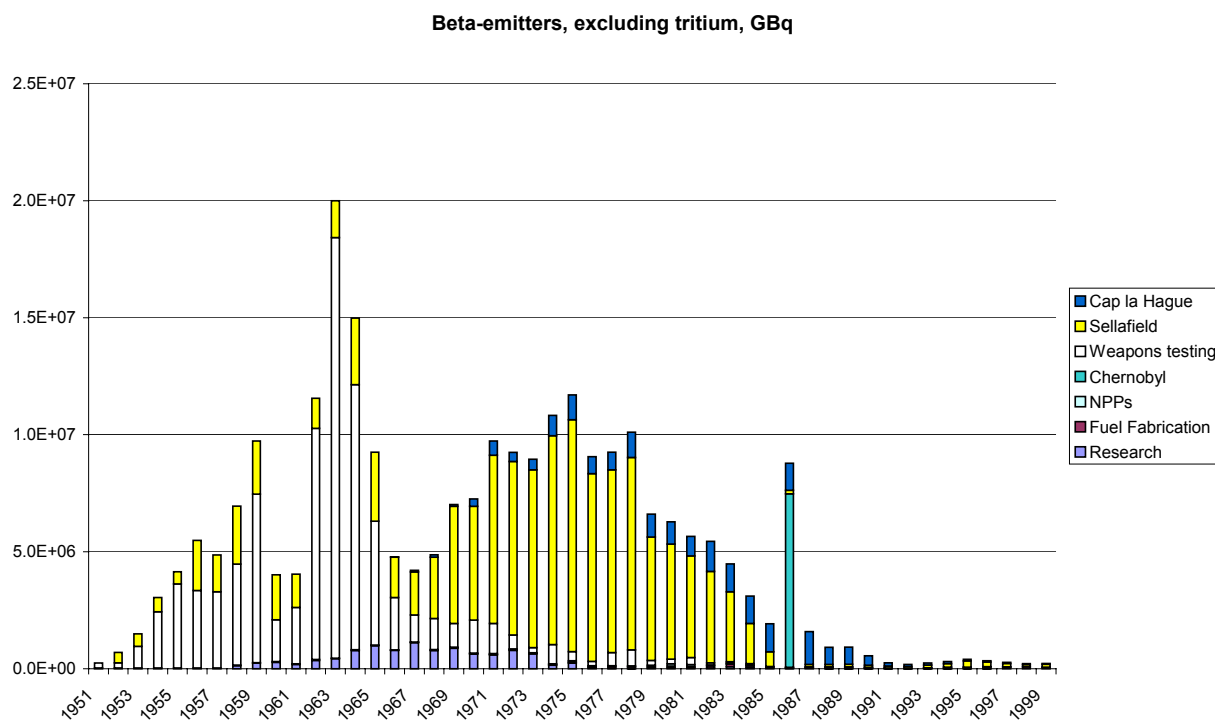
20. The methodology for determining the **impact of radioactivity on marine biota** is still under development. However, according to the available information, there is no identifiable impact on populations of marine biota from radioactive discharges (Figure 14).

## **3 References**

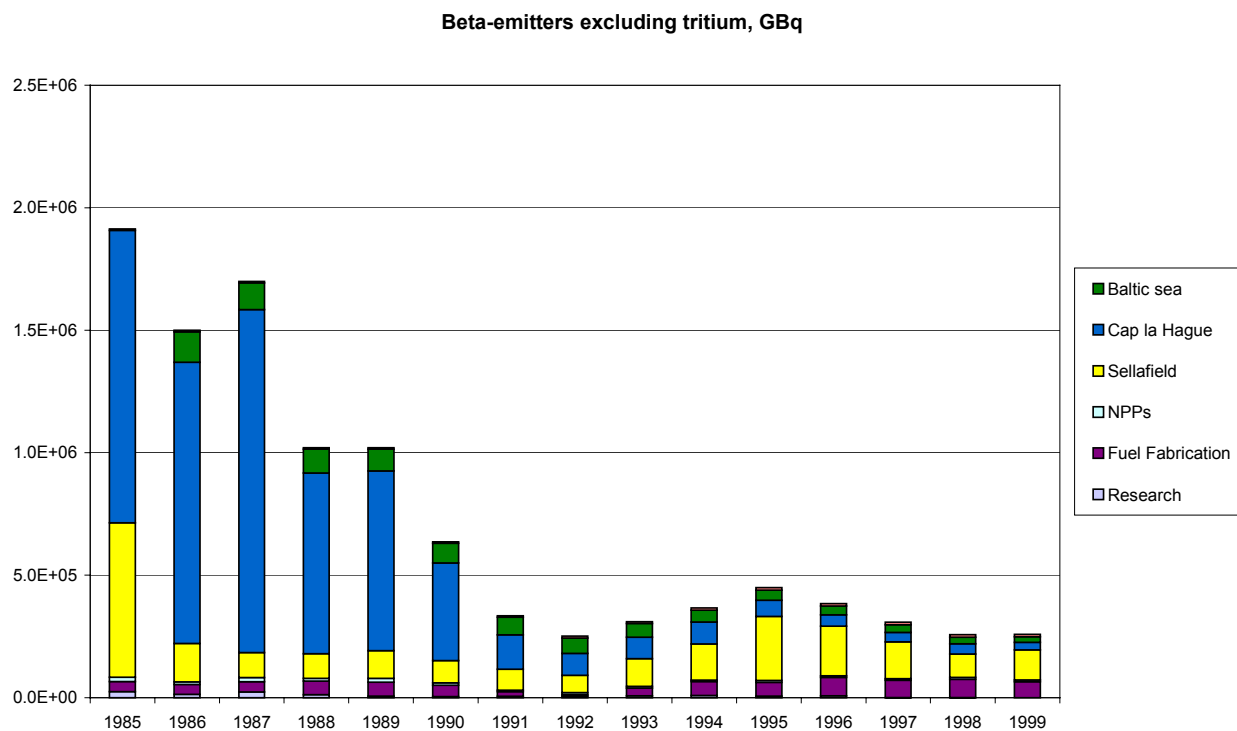
Commission of the European Communities. 'The radiological exposure of the population of the European Community from radioactivity in North European marine waters Project 'MARINA'. EUR report 12483EN (1990).

Commission of the European Communities *The radiological exposure of the population of the European Community to radioactivity in the Mediterranean Sea* Radiation Protection 70 Marina-Med Project Report EUR 15564 EN. (1994).

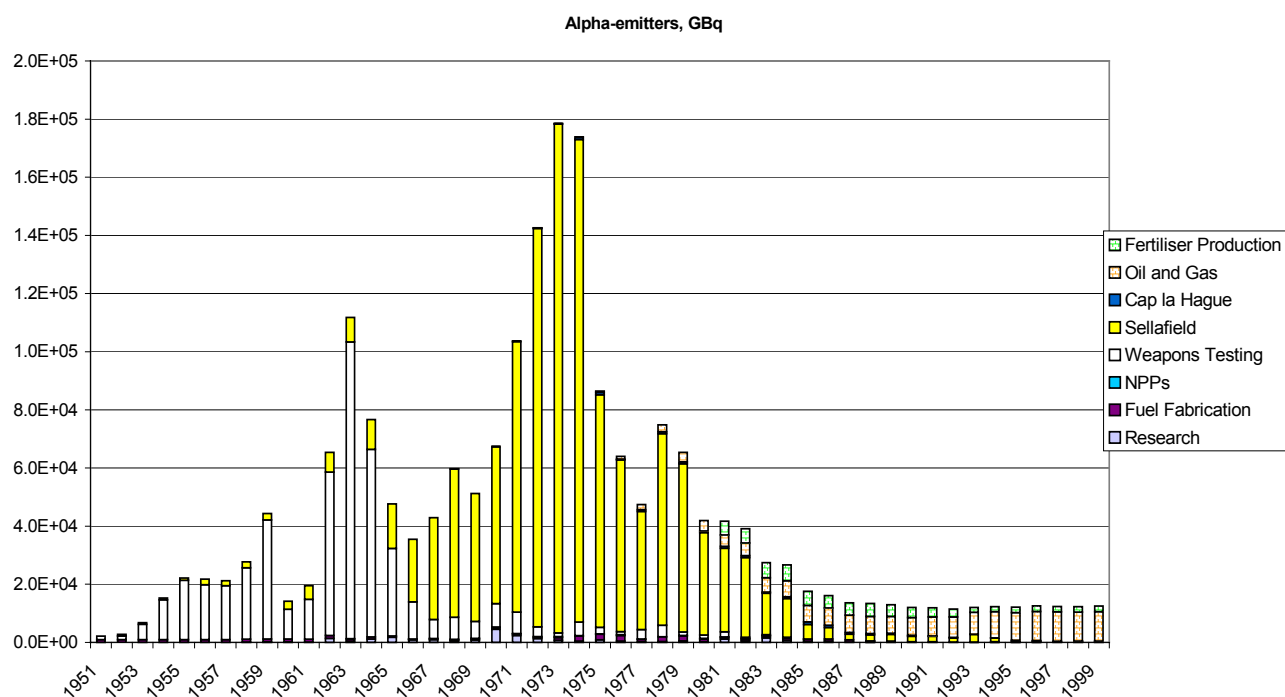
**Figure 1 Trends in overall input of  $\beta$  activity, excluding tritium, into the OSPAR area**



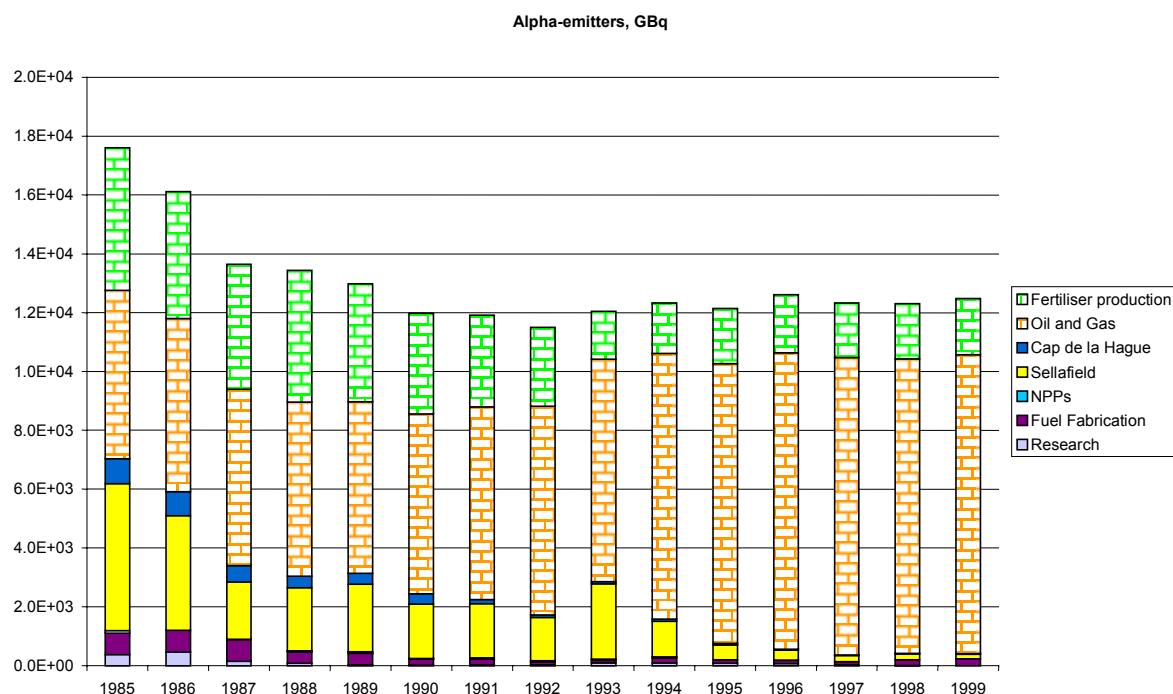
**Figure 2 Recent trends in overall input of  $\beta$  activity (excluding tritium and direct inputs from Chernobyl fallout and Mediterranean Sea) into the OSPAR area**



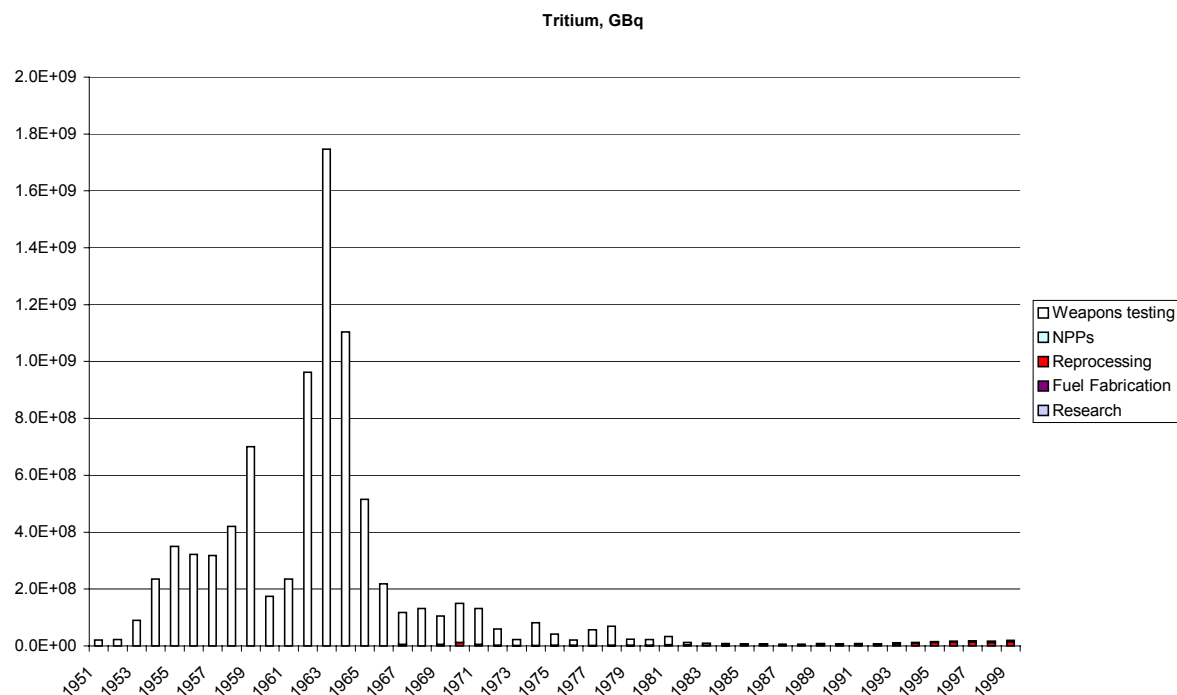
**Figure 3 Trends in overall input of  $\alpha$  activity into the OSPAR area**



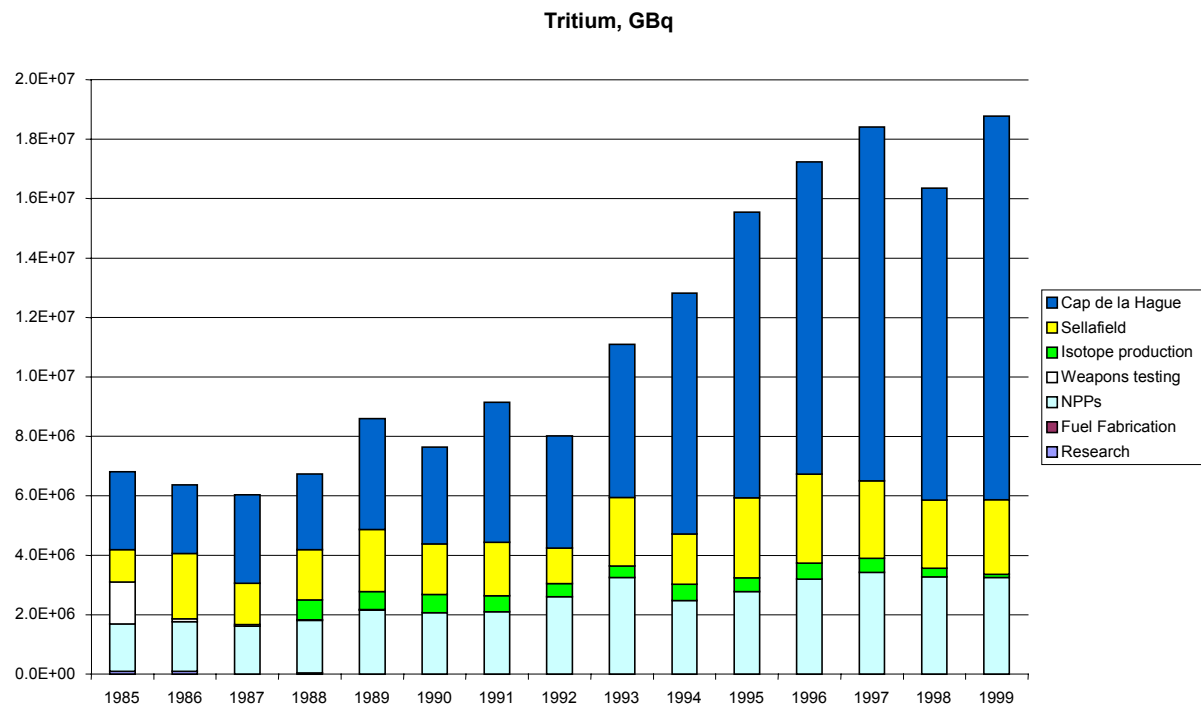
**Figure 4 Recent trends in overall input of  $\alpha$  activity into the OSPAR area**



**Figure 5 Trends in overall input of tritium into the OSPAR area**



**Figure 6 Recent trends in overall input of tritium into the OSPAR area**



**Figure 7**  $^{137}\text{Cs}$  in surface waters of European seas (1981 – 1985)

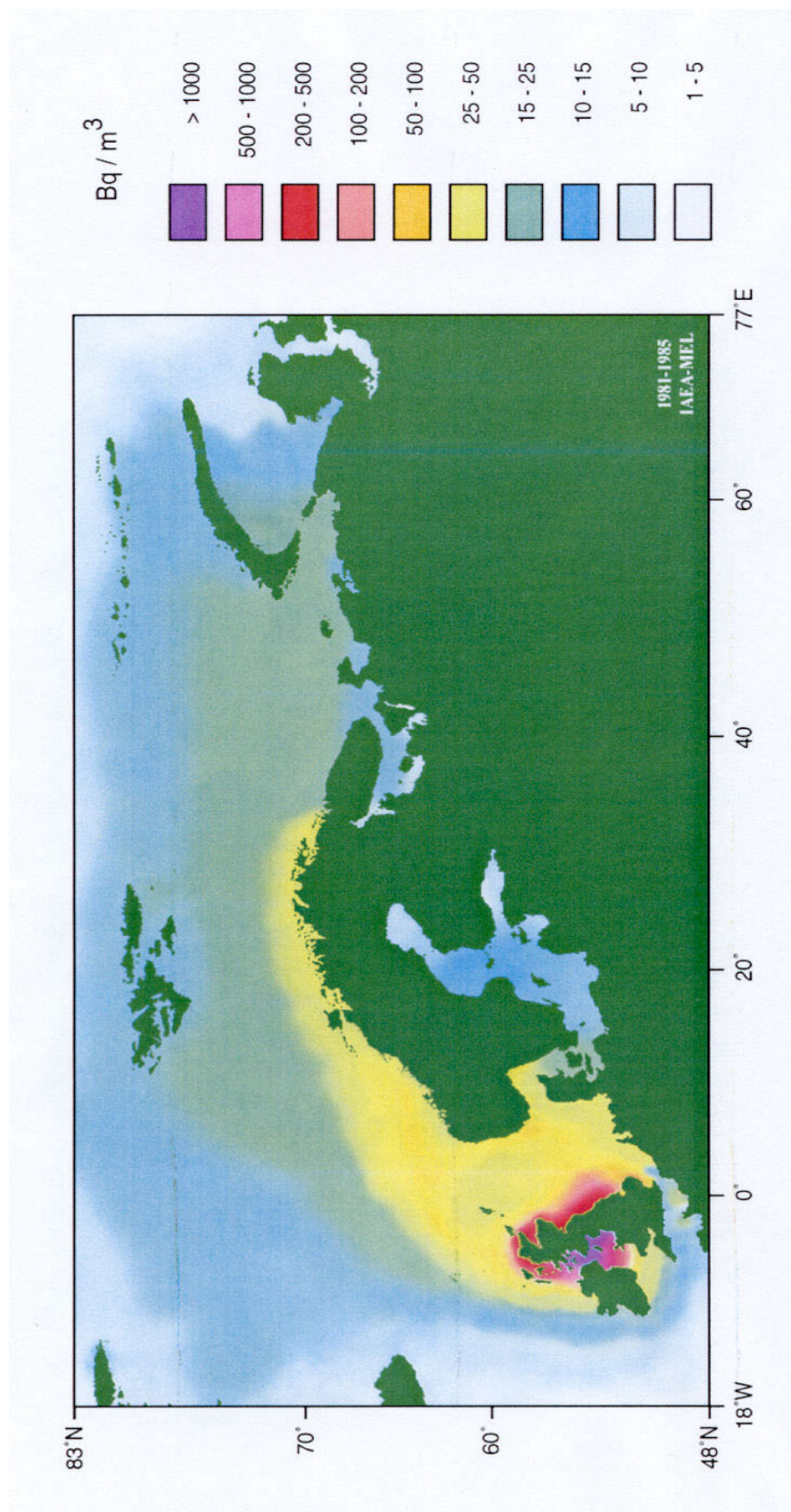
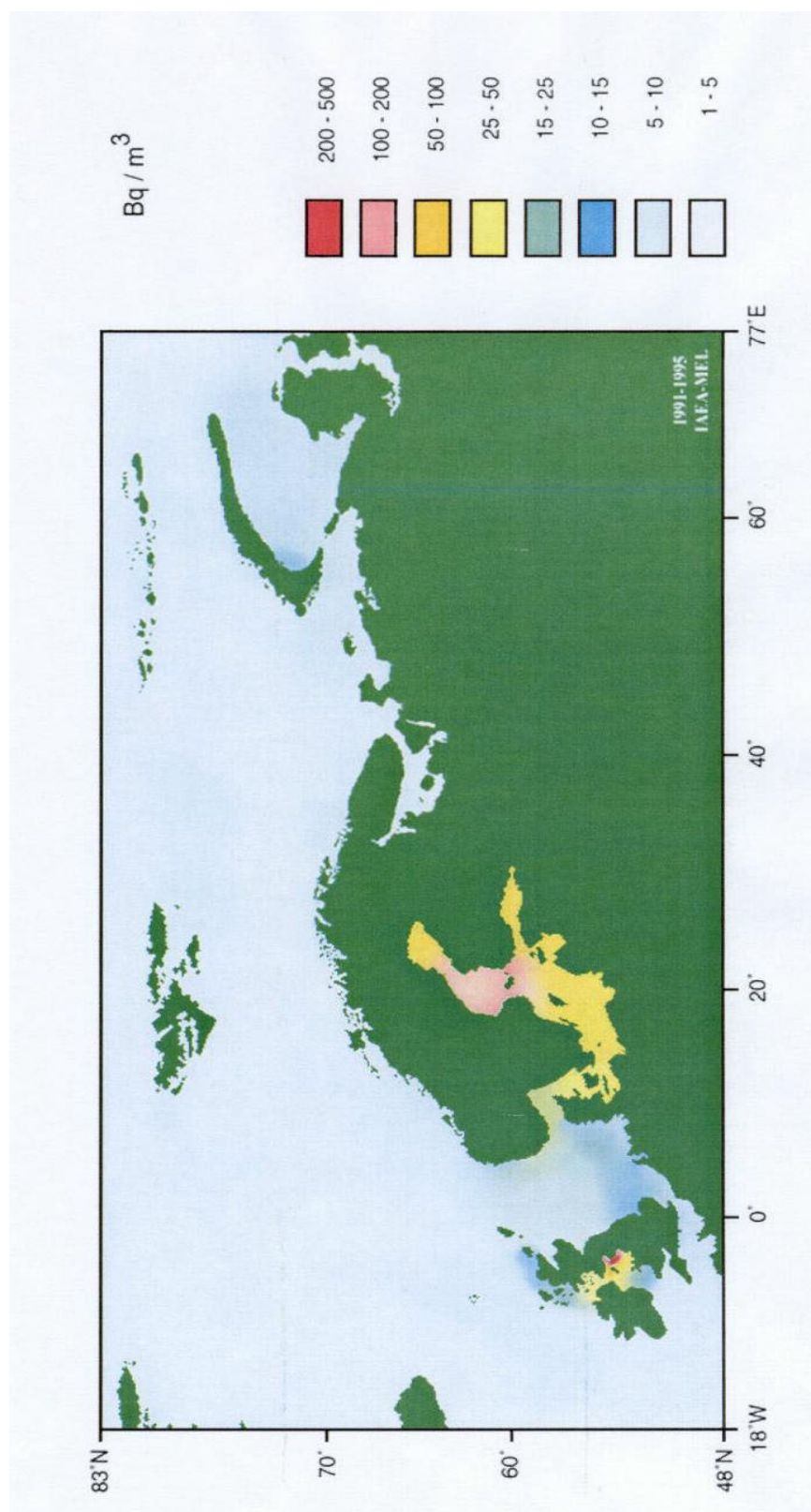
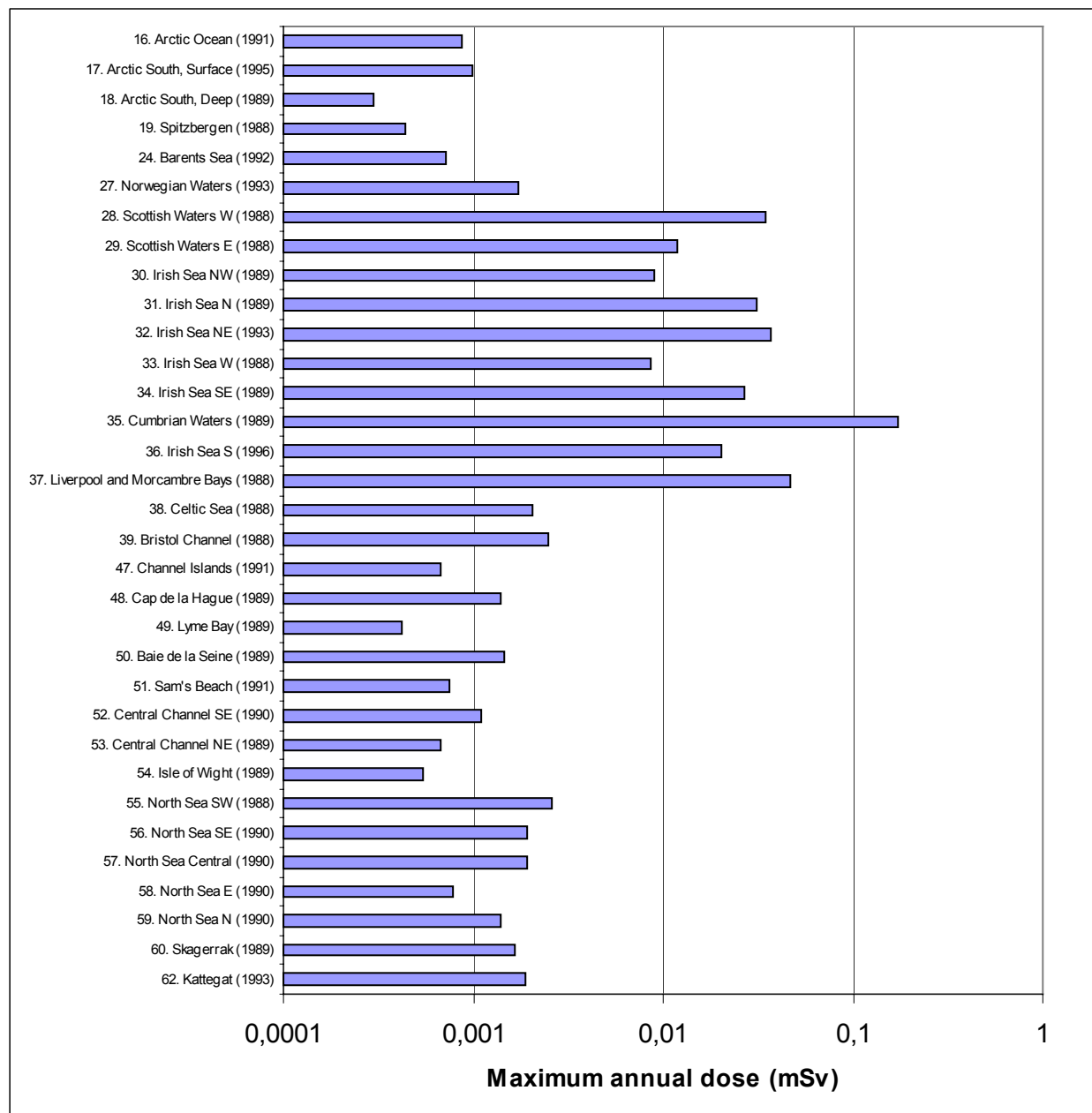


Figure 8  $^{137}\text{Cs}$  in surface waters of European seas (1991 – 1995)

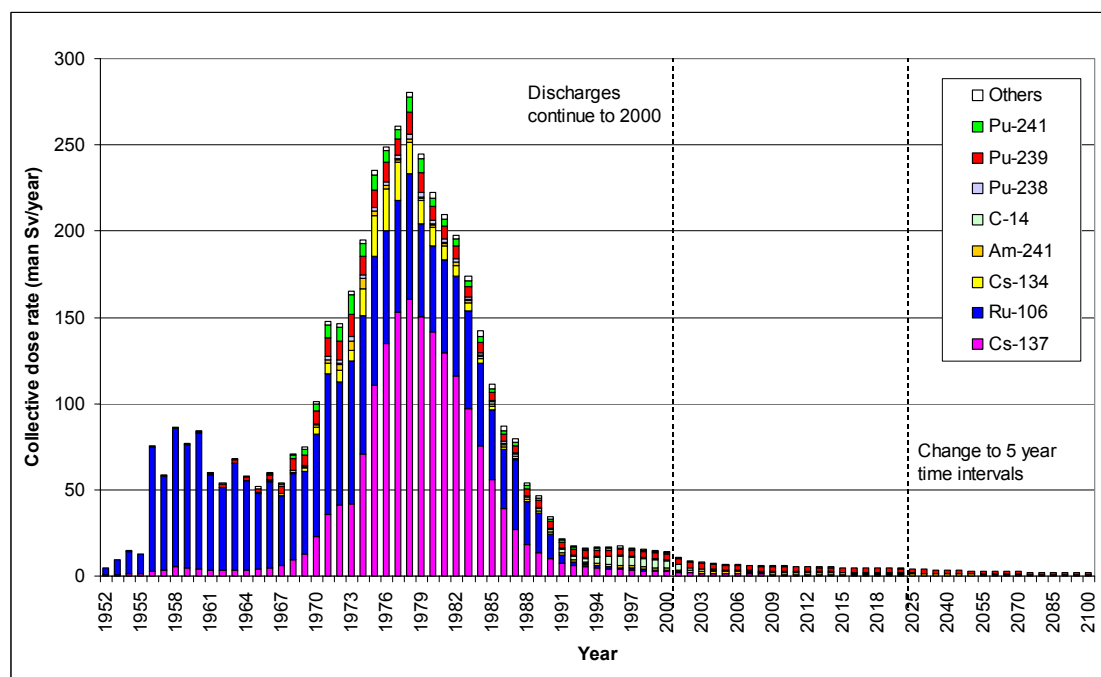


**Figure 9**      **Representative maximum annual doses in the OSPAR region from marine pathways calculated from observed concentrations of man-made radionuclides in the water for normalised consumption rates.**

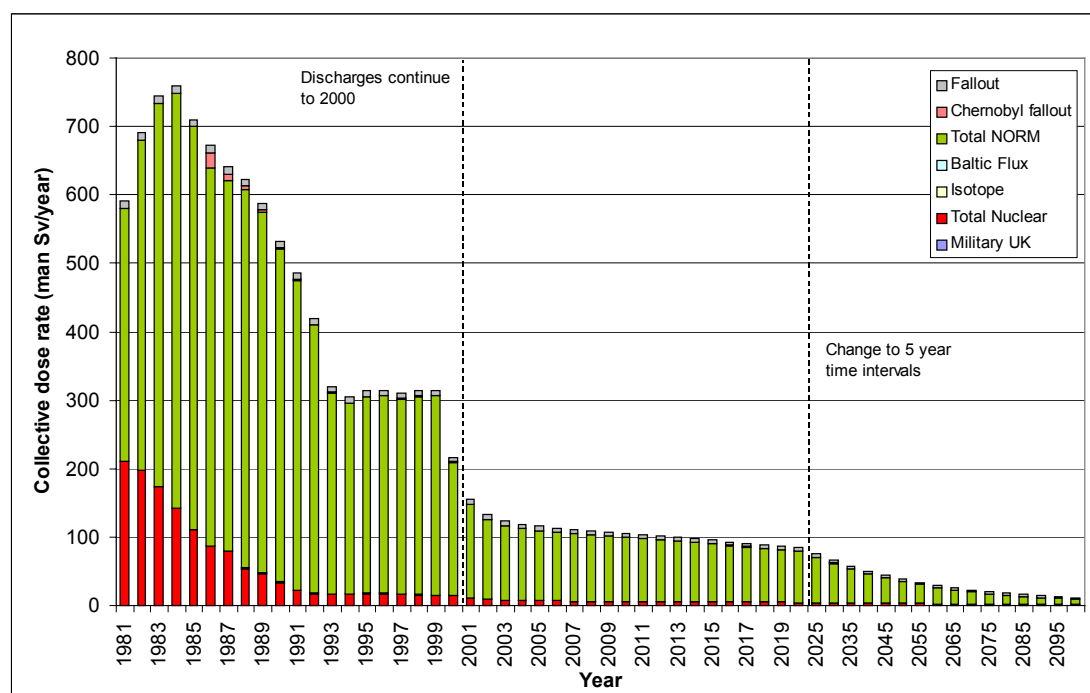




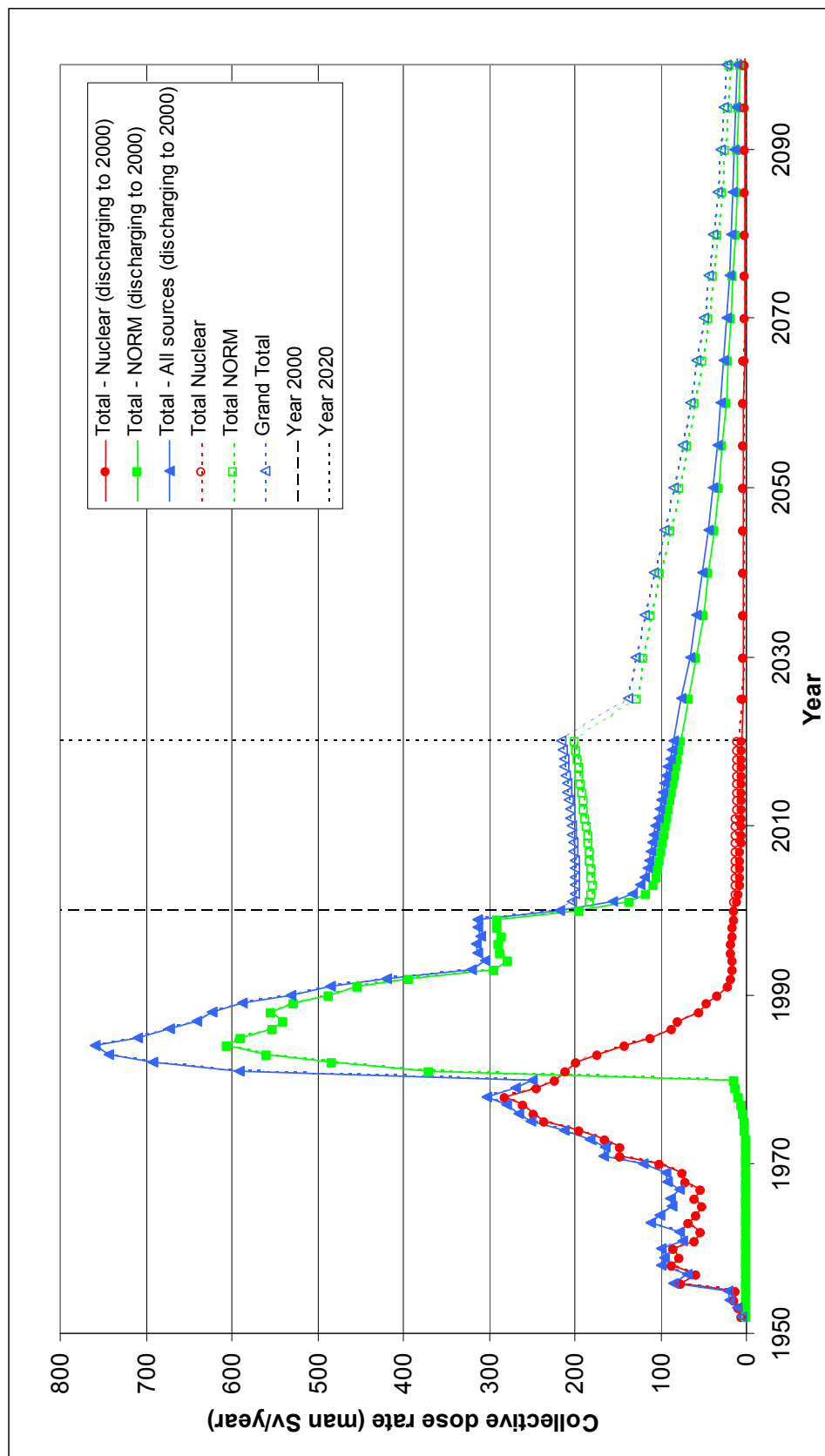
**Figure 10** Collective dose rates to the European Union population by radionuclide for nuclear sites assuming discharges continue to 2000.



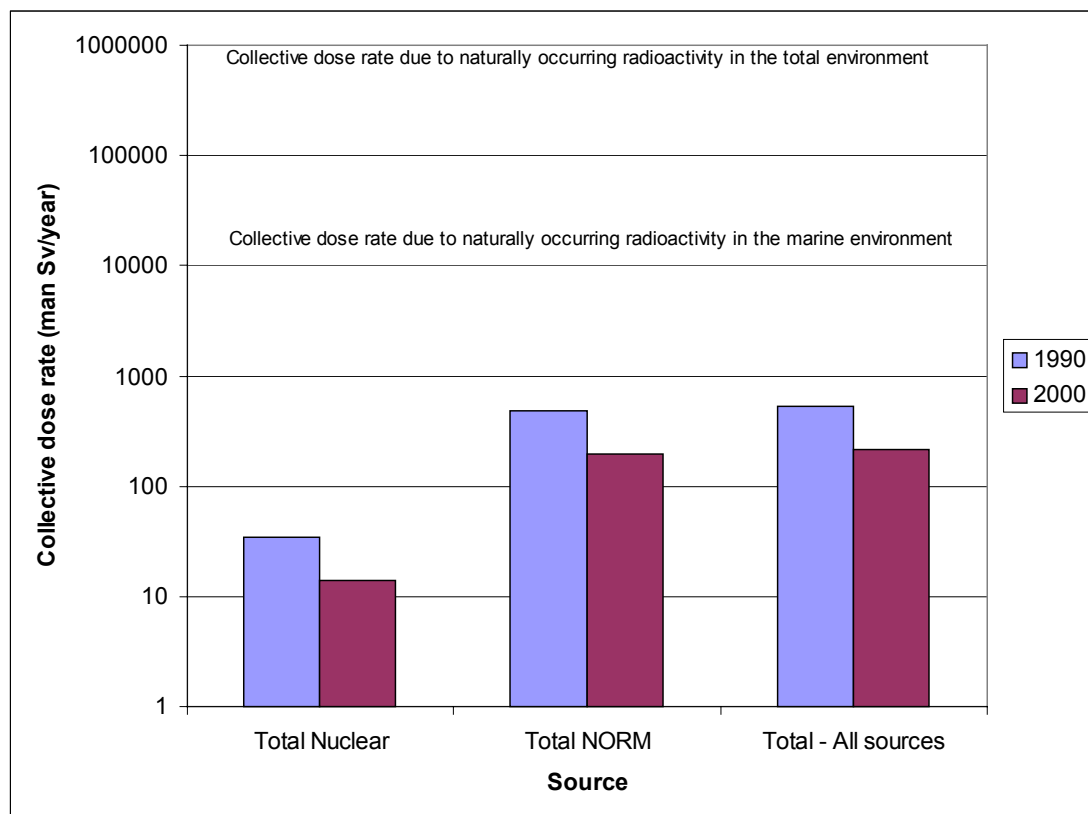
**Figure 11** Collective dose rates by source to the European Union population assuming discharges continue to 2000



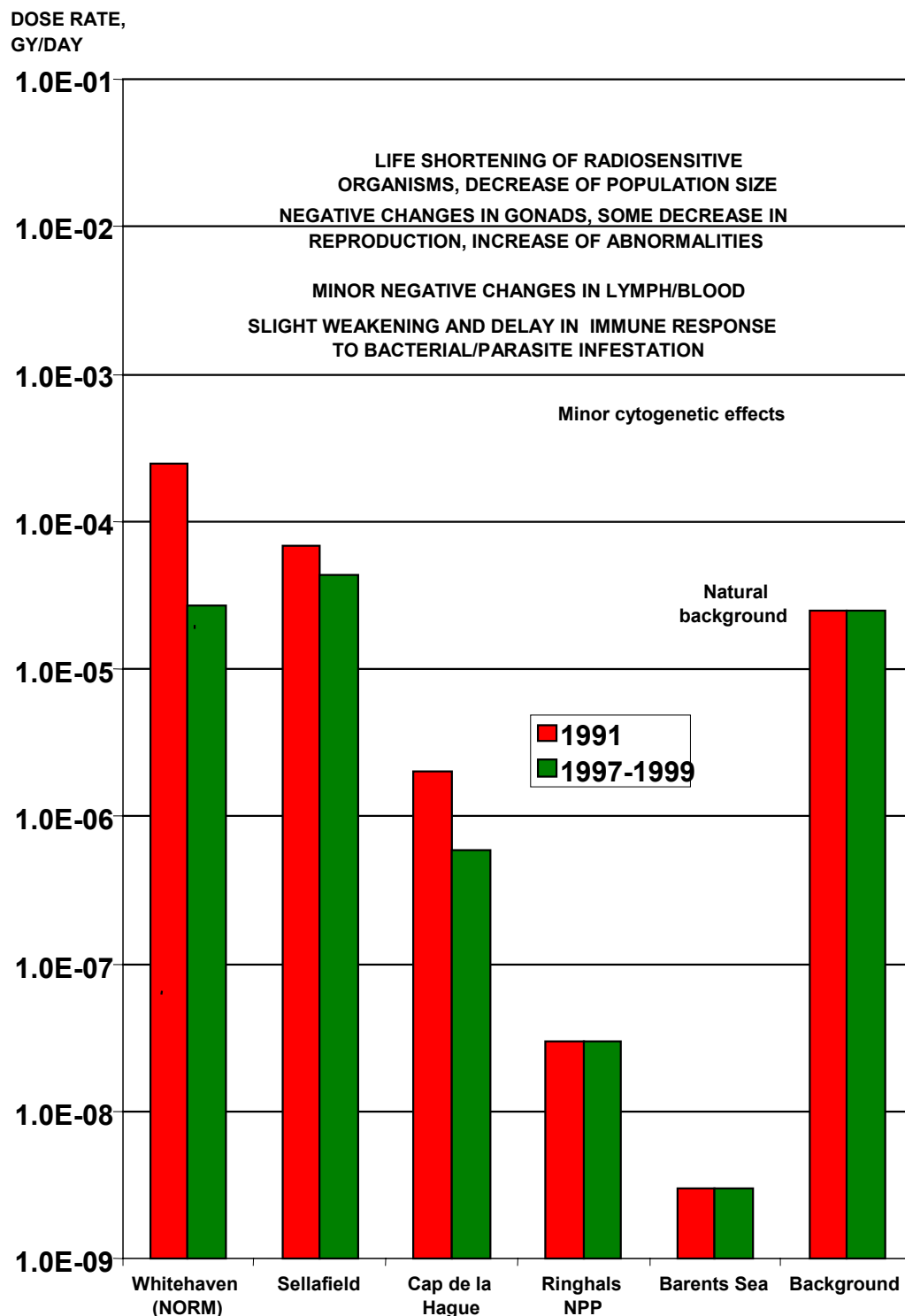
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**Figure 13**      **Collective dose rates to the European Union population from major sources compared with naturally occurring radioactivity**



**Figure 14 Dose rates to molluscs in the OSPAR region (above natural background) along the scale of radiation effects to aquatic biota**



Note. Presented are annual average values of dose rates to molluscs at different locations of the OSPAR region; values for molluscs near Ringhals NPP are upper estimates of dose rates.

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## **MARINA II**

**Update of the MARINA Project on the radiological exposure of  
the European Community from radioactivity in North European  
marine waters**

**Annex A: Civil Nuclear Discharges into North  
European Waters**





# **CIVIL NUCLEAR DISCHARGES INTO NORTH EUROPEAN WATERS**

## **Report of Working Group A**

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Albert van Weers, NRG, Netherlands

A P Bexon, NRPB, UK

S Lopicard, CEPN, France



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# 1 Introduction

The original MARINA study (MARINA I) compiled information on discharges from 44 nuclear sites in the UK, Federal Republic of Germany, the Netherlands, Belgium, France, Denmark and limited data relating to nuclear power stations in Switzerland, Finland and Sweden. Experts from the working group tabulated the data up to the year 1986 for reprocessing facilities and up to 1984 for other sites, based on the reports provided by the site operators or Governmental bodies.

The current study (MARINA II) has collated information for 64 nuclear sites, which discharge radioactive isotopes in the area covered by the OSPAR Convention (see Figure 1), including:

- 56 Nuclear Power Plant (NPP) sites,
- 2 reprocessing facilities (excluding Dounreay and WAK in Karlsruhe which are officially classed as research and development facilities)
- 4 research and development facilities
- 2 fuel fabrication plants

Overall, data were collated for 9 countries (Belgium, Denmark, France, Germany, The Netherlands, Spain, Sweden, Switzerland and United Kingdom) for the period from the end of the original MARINA dataset until the year 2000<sup>1</sup>. In such a way all historical discharges from European Nuclear Power Plants into the OSPAR area have been analysed, with the exception of the Kola NPP in North West Russia, for which no information is available. Information was also obtained on a further 12 research and 6 fuel fabrication facilities, which have not been included in the discharge database due to the shortage of consistent information and the negligible amount of activity discharged from these sites.

In addition, the current study assessed all significant marine discharges into the OSPAR area from the following sources:

- Radionuclide releases from non-nuclear industries, including isotope manufacture, oil, gas and phosphoric acid production,
- Fallout from nuclear testing and the Chernobyl accident,
- Sea dumping,
- Input into the OSPAR area from the Mediterranean and Baltic seas,
- Other sources, including accidents involving submarines, nuclear weapons or satellites and discharges associated with the administration of radionuclides for medical purposes

Publicly available information was also collated on selected defence establishments.

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<sup>1</sup> Data for reprocessing facilities are provided until the end of year 2000; for the other nuclear facilities until the end of year 1999.

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## **2 Sources of information on discharges**

### **2.1 Nuclear sites**

A list of all the nuclear sites that discharge radioactivity into the OSPAR area is provided in Table 1. This table also provides basic information on the type of the facility and OSPAR map reference (see Figure 2).

In the original MARINA study, experts from the working group tabulated data up to 1986 for reprocessing facilities and up to 1984 for other sites based on the reports provided by the site operator or Governmental bodies. These data are included in the MARINA II dataset without alterations, except for discharges from the reprocessing facilities. For these, historical discharge data have been reviewed in recent studies [Jackson et al, 2000; Groupe Radioecologie Nord Cotentin, 1999].

The Bilcom database, which was compiled by the recent EU project B4-3040/99/118894/MAR/C1, provided the main source of information on discharges from nuclear facilities other than the Sellafield and Cap de la Hague sites for the period 1987-1999 (see Appendix A). Bilcom discharge data are themselves taken from EC reports [Willemenot et al, 1999; Schnepf et al, 1996], which tabulated data reported by the EC Member States. Information for the period 1985-1986 was obtained directly from the EC report [Metairie et al, 1995].

In cases where the reported aggregate activity data required further radionuclide breakdown, discharges for individual isotopes were calculated based on the estimates of radionuclide contribution to overall plant discharges for specific reactor types in EU countries. The individual reactor type breakdown data were also compiled in the EU project B4-3040/99/118894/MAR/C1.

Recent data on discharges from Sellafield site were taken from Food Standards Agency and Scottish Environment Protection Agency annual reports [MAFF and SEPA, 1995-2000]. Unpublished data for the year 2000 were obtained directly from British Nuclear Fuels plc (BNFL).

The Nord-Cotentin study [Groupe Radioecologie Nord Cotentin, 1999] reported radionuclide discharges from the Cap de la Hague for the period 1966-1996. Earlier data were taken from the original MARINA study. Data for 1997-2000 were provided by COGEMA, who operate the reprocessing facility in Cap de la Hague.

The available discharge data for four sites in Switzerland, which discharge via rivers into the North-East Atlantic, were taken from the OSPAR reports [OSPAR 1994-2000]. The data for the period 1985-1992 were estimated using average discharge figures.

### **2.2 Other sources of information**

Data on discharges from non-nuclear industries, fallout and accidents are generally based on relatively limited measurements and have a larger inherent uncertainty associated with them (see Section 4 for further comments on uncertainty). MARINA II made maximum use of the already available estimates. However, in a number of cases, it was necessary to implement new assessments as detailed below.

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Information on discharges of Naturally Occurring Radioactive Materials (NORM) that result from the production of phosphoric acid in the fertiliser industry was derived from the OSPAR report that addressed non-nuclear industries [OSPAR, 1997], a more recent Dutch report to OSPAR [Report of the Netherlands, 1999] and a Belgian report on NORM industries [Paridaens et al, 2001]. Statistics on the production of phosphoric acid in European countries were provided by the European Fertilizer Manufacturers Association (EFMA). The analysis of these reports (see Appendix B) is the basis of the estimated discharges of radionuclides.

The first comprehensive assessment of radionuclide input into the OSPAR area as a result of oil and gas production was carried out for the current study (see Appendix C). For each of the European countries there is detailed and up-to-date information on the production rates [Oil and Gas in the Netherlands, 2000; <http://www.dbd-data.co.uk/bbonline>; Oil and Gas Activity 3<sup>rd</sup> quarter, 2000; Statistical Yearbook of Norway 2000; Oil and Gas Production in Denmark, 1998-1999]. On the other hand, there are little data on the volume of co-produced water and on the radionuclide concentrations. This information is mostly contained in unpublished and partly confidential industry data. Nevertheless the available information is sufficient to derive generic estimates of radionuclide discharges per unit production rate for both oil and gas as averaged figures over the lifetime of installations.

The radionuclide deposition from nuclear testing has been calculated using data from the UNSCEAR 2000 report [United Nations, 2000]. Only for <sup>90</sup>Sr are adequate measurements of deposition available; these originate from a global sampling network operated by the United States. Some additional data are available from other locations. There are also limited measurements of the deposition of <sup>95</sup>Zr. Information for other radionuclides is either based on the isotopic ratio against <sup>90</sup>Sr or on the calculated results. Global fallout estimates in the UNSCEAR report [United Nations, 2000] are related to areas of 10° latitude bands.

No new information on Chernobyl fallout into North East Atlantic has come to light since the original MARINA study [Commission of the European Communities, 1990]. Relatively small changes to MARINA I deposition estimates were made within the framework of the current study.

Information on sea dumping in the North East Atlantic is taken from a recent IAEA review [Rastogi et al, 1999]. No quantifiable discharges could be identified from this source. The releases had been estimated to be relatively small [Baxter et al, 1995; OECD, 1985]. According to the available information there has been no impact on the North-East Atlantic from the spent fuel dumping in the Arctic [Ali et al, 1997], [IAEA, 1999]. Similarly low levels of contamination in the Arctic are quoted for the historic dumpings in the North West of Russia and discharges from the nuclear fuel cycle facilities Mayak, Tomsk-7 and Krasnoyarsk-26 via Ob and Yenisey rivers [Norwegian-Russian Project, 1998, Strand et al, 1997, AMAP, 1997, Petrov, 1995].

An on-going IAEA study [Calmet et al, to be published] provided a comprehensive review of information on accidents at sea. [Hansen, 1980] gave an estimate of radionuclide fallout which resulted from the burn-up of an American satellite SNAP 9A.

Discharges from the isotope manufacturing facilities at Amersham and Cardiff and from the UK military establishments were obtained from the Digest of Environmental Statistics [DEFRA 2001], which collated the data from Amersham International, SEPA, MOD and MAFF.

Finally, an estimate of discharges from the administration of radionuclides for medical purposes was made using information on the amount of activity administered per unit of population in countries which are part of OSPAR drainage area [United Nations, 2000].

### **3 Analysis of the data and trends**

#### **3.1 Overview of trends**

Figures 3 to 8 show trends for radioactivity input into the OSPAR area from all significant sources. It can be seen that up until the late 1960s the main input of radionuclides was due to weapons testing. From that time on, nuclear fuel reprocessing at Sellafield, and, to a lesser extent, at Cap de la Hague have become the main sources of releases. Chernobyl fallout resulted in another significant input of  $\beta$ -emitters in 1986 (see Figure 3). Fallout of tritium from weapons testing in the early 1960s dominates the cumulative input of this radionuclide into the North-East Atlantic.

The more recent trends, excluding Chernobyl fallout, are illustrated in Figures 4, 6 and 8. Reprocessing remains the most important source of  $\beta$ -emitters, excluding tritium. However, in the last five years, with the reduction of discharges of  $\alpha$ -emitters from Sellafield and Cap de la Hague and cessation of phosphogypsum discharges by several manufacturers of fertilisers, discharges from offshore oil fields have become more important. The majority of these originate from the Norwegian and British sectors of the continental shelf.

From Figures 3 to 8 it can be seen that in the 1990s the input of activity into the North-East Atlantic has been relatively minor for all types of radionuclides. Historic inputs of  $\beta$ -emitters into the OSPAR area reached their peak in 1963 and of  $\alpha$ -emitters in 1973. Current discharges of  $\alpha$ -emitters and  $\beta$ -emitters represent only around 5 and 1% of the maximum values, respectively.

For significant sources, excluding NORM, the total integrated inputs of activity into the OSPAR area are given in Table 2. Inputs of activity from the oil, gas and the fertiliser production industries are given in Table 3.

It should be noted that although the discharges have been grouped as total alpha, total beta excluding tritium and tritium these groupings do not necessarily reflect the radiotoxic significance of these emitters. Effective dose, which is dependent on the behaviour of the radionuclides, is what is significant to health rather than simply the magnitude of  $\alpha$  and  $\beta$  discharges.

#### **3.2 Discharges from the nuclear industry and research facilities**

##### **3.2.1 Nuclear industry - summary and trends**

A summary of the discharges into the North-East Atlantic from the nuclear industry and research facilities is given in Tables 4 to 6 and Figures 9 to 15. In all cases the situation since 1984 is presented separately to allow comparison between overall discharges and recent trends since the completion of the MARINA I study. The most important data are shown in Figure 13 from which it can be seen that there was a major reduction in the discharges of both  $\alpha$ -emitters and  $\beta$ -emitters excluding tritium between the 1970s and 1980s and again between the 1980s and 1990s. However, the discharge of tritium has increased.

The MARINA I study concluded that ‘By far the main sources of radionuclide releases are the reprocessing plants’. ‘Nearly all  $\alpha$  activity discharges up until the end of 1984 arise from reprocessing plants and in particular from the reprocessing plant at Sellafield. Discharges from reprocessing plants also account for over 90% of the total  $\beta$  activity discharges and nearly 90% of these reprocessing plant discharges are from Sellafield.’ [Commission of the European Communities, 1990].

It can be seen from Table 4 that the principal contribution to the total discharge from the nuclear industry, up to the year 2000, is still from reprocessing plants (83%) followed by the nuclear power stations (13%). Excluding NORM discharges, reprocessing plants make the major contribution to all three categories of  $\alpha$ -emitters,  $\beta$ -emitters excluding tritium and tritium itself.

Of the individual sites, Table 5 and Figure 9 show that Sellafield has made the greatest contribution to the total amount of radioactivity discharged by the nuclear industry (52%) followed by Cap de la Hague (32%). When looking at radionuclide breakdown of these discharges, it transpires that Sellafield has made the greatest contribution to the discharge of  $\alpha$ -emitters (94% - see also Figure 10) and  $\beta$ -emitters excluding tritium (79% - see also Figure 11), Cap de la Hague has contributed most to the discharge of tritium (45% - see also Figure 12).

Table 6 shows that, over the 15 years from 1985 to 2000, Cap de la Hague has made the major contribution to the release of both  $\beta$ -emitters excluding tritium (64%) and tritium itself (60%), but Sellafield is still the main contributor to the discharge of  $\alpha$ -emitters (72%).

In any of the last 5 years discharges of  $\alpha$ -activity from Sellafield represented less than 0.2% of the peak discharges seen in the 1970s. Recent discharges of  $\beta$ -emitters excluding tritium were about 1% of those seen in the 1970s. Similar decreases can be observed for the reprocessing facilities in Cap de la Hague.

Dounreay, the only other single nuclear site with a noticeable contribution to overall marine discharges, was responsible for just over 4% of  $\alpha$ -activity and over 1% of  $\beta$ -activity excluding tritium discharged into the OSPAR area since 1984.

These trends reflect the commissioning of new power plants and reprocessing facilities in the 1980s and 1990s in the OSPAR area and the simultaneous commissioning of modern liquid waste treatment facilities at the reprocessing sites. Since at present it is not possible to remove tritium from liquid waste, the increased discharges of this radionuclide are linked to the increased rates of fuel reprocessing and commissioning of new nuclear reactors.

Radionuclide breakdown of  $\alpha$ -emitters and  $\beta$ -emitters excluding tritium, has not changed significantly since the original MARINA study. Processing of previously accumulated wastes at the Enhanced Actinide Removal Plant in Sellafield resulted in an increase of  $^{99}\text{Tc}$  discharges, which over the period 1993-2000 amounted to 668 TBq or 57% of the Sellafield beta discharges excluding tritium (up from 21 TBq of  $^{99}\text{Tc}$  discharges for the 5-year period prior to 1993).

### 3.2.2 Reprocessing facilities

#### Overview of French Facilities

In France, plutonium separation began as a part of the development of the nuclear weapons research program after World War II. Three plutonium producing reactors were put into operation between 1956 and 1958 at the Marcoule site in the south of France [Schneider et al, 1997]. UP1, the first full-scale reprocessing plant was completed there in 1958. The design capacity of the plant was 600 tons per year. It was shut down in 1997.

The Cap de la Hague reprocessing plant is located 20 km west from Cherbourg, at the far north-west of the Cotentin peninsula. Cogema operates two large scale reprocessing facilities, UP2 and UP3, which together produced roughly 80% of all separated plutonium in the world in 1995. The units in operation at Cap de la Hague had reprocessed a total of 1680 tons of spent fuel by 1997.

The nominal annual capacity of each of the two units is 800 metric tons of heavy metal. UP2 was started up in 1966, originally to reprocess Magnox fuel (see Table 7). Its 'nominal' capacity varied and was finally put at 400 tons per year. From 1976 onwards a new installation enabled the plant to reprocess oxide fuels of light water reactors (LWRs). Since 1994, after significant modification and expansion, the plant operates under the name UP2-800 to indicate its new nominal annual capacity.

The unit UP3 for the reprocessing of foreign light water reactor fuel started operating in 1989.

#### Trends in discharges from Cap de la Hague

Table 4 and Table 6 show that Cap de la Hague, together with Sellafield, represent the major sources of radioactive releases from the nuclear industry into the marine environment. The trends in discharges of  $\alpha$ -emitters,  $\beta$ -emitters excluding tritium and tritium from Cap de la Hague are illustrated in Figure 16, Figure 17 and Figure 18 respectively. Excluding tritium, there have been major reductions in the discharges since 1986, which is the date of the last reprocessing dataset included in MARINA I.

Effluent treatment facilities (STE in Table 7) have always been used to treat the effluent prior to release. Radioactive liquid effluents are collected and sent by a piping system to the effluent treatment facilities, where they undergo decontamination operations (settling, chemical treatment and filtration) during which the major radionuclides are precipitated into sludge.

Since 1995 a new process for effluent treatment has been operating that allows the recycling of most of the effluents produced, and so reduces the flux of effluents to be chemically treated at the effluent treatment plants.

Since the commissioning of this treatment facility, discharges of  $\alpha$ -emitters have been reduced from 810 GBq in 1986 to 21 GBq in 2000. For  $\beta$ -emitters excluding tritium, the respective figures are 1100 TBq and 34 TBq. Tritium discharges have increased

almost fivefold (from 2300 TBq to 11 000 TBq) in line with the increased rate of reprocessing.

### **Overview of UK facilities**

After France, Britain is the second largest reprocessor of power reactor spent fuel in the world. This activity is located at the Sellafield (formerly Windscale) plant in the north-west of England. BNFL's main activities at Sellafield are the reprocessing of spent Magnox and oxide fuel, waste management and decommissioning and the operation at Calder Hall of four Magnox nuclear reactors, which supply the National Grid. In the past, UK Atomic Energy Authority (UK AEA) reprocessed spent fuel from small reactors at Dounreay in the north of Scotland.

### **Thermal reactor fuel reprocessing**

Magnox fuel is reprocessed within two years of removal from the reactor to avoid excessive corrosion of the cladding. Magnox power-reactor fuel has been reprocessed in Building 205 (B205) (design capacity 1500 tons of fuel per year) on the current Sellafield site since 1964. It has served a crucial role in the British Magnox reactor programme, while also servicing fuel from the Japanese and Italian Magnox reactors. All Magnox fuel has routinely been transported to Sellafield. By the end of 1995 some 26,800 metric tons (Te) of fuel had been processed at B205 from which a total of about 59 Te of plutonium had been separated.

Oxide fuel reprocessing began at the present Sellafield site in 1969 [Berkhout, 1997] when a small Head-End Plant (HEP, B204), at which oxide fuel was prepared for feed into the B205 plant, was brought into operation. In total, 110 metric tons of fuel were processed through HEP/B205 before an accident caused the permanent closure of B204 in 1973.

Large-scale oxide fuel reprocessing got underway at Sellafield when the Thermal Oxide Reprocessing Plant (THORP) came on line in 1994. With a design capacity of 7 000 Te in 10 years (up to 1,200 tons per year), THORP has contracted to reprocess fuel from seven other countries, notably Japan. By the end of its first 10 years of operation, THORP is expected to have separated 40-50 Te of plutonium [Berkhout, 1997].

### **Fast reactor fuel reprocessing**

Fast reactor and materials test reactor fuel has been reprocessed at Dounreay in northern Scotland since July 1958 [Berkhout, 1997]. Two facilities have been operated by the UKAEA: D1204 for the materials test reactor fuel; and D1206 for fast reactor fuel. D1204 is a small facility which has processed fuel from British and non-British research reactors. D1206 began operation in 1961 and processed highly-enriched uranium fuel from the Dounreay Fast Reactor (DFR, shutdown 1977) and MOX fuel from the Prototype Fast Reactor (PFR, shutdown 1994). Both reactors were located at Dounreay. By the end of 1995 about 21 Te of PFR fuel had been reprocessed at Dounreay, containing some 4.5 Te of plutonium. Reprocessing at Dounreay was suspended in 1998 and, in 2001, the UK Government announced that it would not be restarted.

## **Trends in discharges from Sellafield**

The trends in discharges of  $\alpha$ -emitters,  $\beta$ -emitters excluding tritium and tritium from Sellafield are illustrated in Figure 19, Figure 20 and Figure 21 respectively.

As for Cap de la Hague, new data on historic discharges have become available since publication of the MARINA I study. These show some differences between the current and the previous MARINA study for the period up to 1986. For example, the 1971 peak in tritium discharges, according to the latest data [Jackson et al, 2000] (not available in MARINA I) is associated with the discharges made from Sellafield as a result of material tankered from Capenhurst in the period 1967-1972.

Fission products, including  $^{137}\text{Cs}$ , are the major radiologically-significant contributors to marine discharges. They arise, not only from reprocessing operations, but also from the fuel storage ponds, from where they were discharged directly to sea until the mid-1970s. Discharges from 1952 broadly reflected pond and reprocessing activity until the mid-1970s, when the increased storage and corrosion of Magnox fuel caused significantly greater discharges from the ponds [Hunt et al, 1997]. Until 1985, interim abatement was provided by pumping pond water through containers of zeolite clay to remove caesium.

The Salt Evaporator and Site Ion Exchange Effluent Plant (SIXEP) came on line in 1985 and greatly reduced the discharges of fission product radioactivity to the Irish Sea [BNFL, 2000].

The Segregated Effluent Treatment Plant (SETP) completed active commissioning in 1993. This plant provides fine mesh screening, hydrocyclone particle separation and sampling/sentencing facilities for low activity effluents. Effluents include arisings from THORP and Magnox reprocessing operations, the Salt Evaporator, Fuel Handling Plant, analytical services and decommissioning operations. SETP also provides neutralisation of acidic effluents.

More recently, the Enhanced Actinide Removal Plant (EARP), which completed active commissioning in 1995, continues to make significant reductions in discharges of plutonium and other actinides. However  $^{99}\text{Tc}$  discharges have increased from 6.6 TBq in 1986 to 44 TBq in 2000 due to processing of historical wastes at EARP.  $^{99}\text{Tc}$  is currently responsible for around 50% of all Sellafield discharges, excluding tritium.

From 1986 until 2000, discharges have been reduced for  $\alpha$ -emitters and  $\beta$ -emitters, excluding tritium, from 3900 to 150 GBq/y and from 160 to 84 TBq/y respectively. Discharges of tritium remained more or less constant at approximately 2200 TBq/y. These changes are in spite of the commissioning of THORP and are due to the improved treatment of liquid wastes.

## **Reprocessing in Germany**

Reprocessing began in Germany in 1971 at the WAK pilot facility at Karlsruhe [Washington Nuclear Corporation]. Plans to expand reprocessing met political

opposition and a facility under construction at Wackersdorf in Bavaria was abandoned in 1989. The federal and state governments stopped funding reprocessing research and WAK closed in 1990. Plutonium separation in Germany never exceeded 13 kg annually.

### 3.2.3 Nuclear Power Plants

There are 56 sites with Nuclear Power Plants in Belgium, France, Germany, the Netherlands, Spain, Sweden, Switzerland and United Kingdom in commercial operation as of 2000, which discharge either directly into the North-East Atlantic or into the rivers which are part of the North Atlantic drainage basin (see Figure 2 and Figure 22).

One hundred and eighty nine reactor units have generated electricity since Calder Hall, the first European power reactor, was connected to the National Grid in 1956. Thirty one reactors have been shut down and forty six reactors have been connected to the grid since 1984, which is when the MARINA I dataset ended.

In 2000, one hundred and forty six operational European reactors supplied over 821 000 GWh(e) of electric power [IAEA Information System]. France, the largest producer of nuclear power, generated 395 000 GW(e), Germany generated 159 600 GW(e), the UK generated 78 300 GW(e), Spain generated 59 305 GW(e), Sweden generated 54 800 GW(e), Belgium generated 45 400 GW(e), Switzerland generated 24 949 GW(e) and the Netherlands generated 3 699 GW(e).

Figure 23 shows that, in spite of the new nuclear power plants that have been commissioned since 1984, the improved management of liquid waste has resulted in a decrease in the discharge of  $\beta$ -emitters excluding tritium. However, commissioning of the new plants has resulted in an increase in the discharge of tritium as shown in Figure 24. The discharge of tritium is 1 000 times greater than for the other  $\beta$ -emitters and is the only discharge from the nuclear power stations that is significant compared to that from the reprocessing plants, as shown in Tables 2 to 4. Figure 25 shows that the main contribution to the discharge of  $\beta$ -emitters excluding tritium is the fission product  $^{137}\text{Cs}$ . Figure 26 shows that more recently, the short-lived activation product  $^{35}\text{S}$  has become proportionately more significant than the other activation products  $^{55}\text{Fe}$  and  $^{60}\text{Co}$ .  $^{35}\text{S}$  mainly originates from the British gas-cooled reactors. The British gas-cooled reactors also release relatively high quantities of tritium.

### 3.2.4 Fuel enrichment and fabrication facilities

The OSPAR annual reports [OSPAR 1994-2000] provide recent data on 8 fuel enrichment and fabrication facilities (Lingen, Gronau, Hanau, Karlstein in Germany; Almelo in the Netherlands; Juzbado in Spain; and Capenhurst and Springfields in the UK). In 1998, Capenhurst and Springfields were responsible for 2% and 98% of the  $\alpha$ -emitters discharge from fuel fabrication facilities respectively. In the same year, all the other fuel fabrication facilities discharged 0.1% of the total discharge of  $\alpha$ -emitters. Unlike the British facilities, they are not required to report  $\beta$ -emitters. It should be noted that the French enrichment and fuel fabrication facilities (Eurodif, Miramas, Melox and Cadarache) are not in the OSPAR drainage area. Furthermore,



there are no data on discharges from the fuel fabrication facilities outside the UK prior to 1992 or after 1998. Therefore, it has been decided to restrict the MARINA II dataset to data for Capenhurst and Springfields with only the latter site containing additional data to those that were available for the MARINA I study.

Springfields is mainly concerned with the manufacture of fuel elements for nuclear reactors and the production of uranium hexafluoride. Radioactive liquid waste arisings consist mainly of thorium and uranium and their decay products. The time dependence of discharges of  $\alpha$ -emitting radionuclides from Springfields is illustrated in Figure 27. Since 1984 the discharges of  $\alpha$ -emitters have decreased by almost a factor of 8. After 1992 discharges remained stable at around 100 GBq/yr until 1998 when discharges of  $\alpha$ -emitters from Springfields almost doubled to 200 GBq/yr.

The Springfields site also dominates the discharge of  $\beta$ -emitters from fuel fabrication facilities (although other sites do not report  $\beta$ -emitters), which in recent years have been approximately 150 TBq/yr. This is double the magnitude of discharges in the mid-1980s. The increase is due to the higher rate of uranium ore concentrate processing.

The Capenhurst site currently undertakes the enrichment of uranium and the dismantling of the old enrichment plant, with only minor arisings of tritium, uranium and its daughter products and  $^{99}\text{Tc}$  and  $^{237}\text{Np}$ .

### 3.2.5 Research facilities

In 1998 OSPAR reported data on 16 research and development facilities [OSPAR, 1994-2000], including:

- Mol in Belgium,
- Risø in Denmark,
- Geestacht, Karlsruhe, HMI Berlin, Jülich and Rossendorf in Germany,
- Campus de Sacavem in Portugal,
- Delft and Petten in the Netherlands,
- Halden and Kjeller in Norway,
- Paul Scherrer Institute in Switzerland,
- Dounreay, Harwell and Winfrith in the UK.

Only the most significant research facilities, including Dounreay, Harwell, Karlsruhe, Risø and Winfrith, were considered in MARINA I. It was found that until the end of 1984 these facilities (excluding Dounreay which was classed as a reprocessing facility in MARINA I and Winfrith which operated a reactor until 1990) contributed 0.01, 0.02 and 10.8% of  $\alpha$ -emitters,  $\beta$ -emitters and tritium respectively. The collective dose from these facilities was some 0.01% of the total. In 1998, all the research facilities, listed in the OSPAR annual report [OSPAR, 2000] discharged 0.66 TBq<sup>2</sup>, compared to 4.5-9.1 TBq in 1990-97. Dounreay released 0.58 TBq<sup>2</sup>. Research facilities, including Dounreay discharged less than 3% of the total nuclear industry discharges in any year in the 1990s. Based on the OSPAR data, in 1998 all research facilities apart from Dounreay, discharged the following percentage of the total

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<sup>2</sup> Excluding tritium

discharge from nuclear facilities: 0.3% of  $\beta$ -emitters, no  $\alpha$ -emitters and 0.08% of tritium.

Since research reactor discharge data are only available for selected years and the contribution to the overall discharges is so small, only data for Winfrith (until shut down) and Dounreay were collected in addition to those available from MARINA I. Discharges from Dounreay showed decreases from the 1980s levels (see Figure 28 and Figure 29). This is consistent with the end of reprocessing at Dounreay in the mid-1990s and the closure of the two fast reactors. Future discharges will be determined by decommissioning and remediation of the site and are likely to be small.

It should be noted that historic contamination of the marine environment near Dounreay with irradiated nuclear fuel fragments ('particles') of the size of a grain of sand resulted from three possible sources [SEPA, 2001]:

1. Discharges prior to 1973 from an unidentified source,
2. A potentially on-going release from the Shaft via groundwater (old disposal site at Dounreay),
3. The old diffuser system may contain particles which still flow through fractures and out at freshwater springs on the seabed.

Up to December 2000, 475 particles were found on the seabed and 14 ashore.  $^{137}\text{Cs}$  activity levels in these particles varied between  $10^3$  and  $10^8$  Bq. Work to estimate past releases of particles is still on-going.

### **3.3 Discharges of naturally occurring radionuclides**

#### **3.3.1 Overview**

The Earth's crust and the oceans contain a number of Naturally-Occurring Radioactive Materials (NORM). 'Natural' as opposed to 'artificial' radionuclides include those formed when the earth was made or result from the decay of such nuclides. There are also natural cosmogenic radionuclides. Natural radionuclides can be mobilised and released into marine environment by industrial processes, such as:

- Phosphate processing operations for the production of phosphoric acid for the fertiliser industry,
- Off-shore oil production, and to a lesser degree, natural gas production,
- Production of titanium dioxide pigment, mining of other ores, foundries, production of non-ferrous metals and rare earth. All these industries also involve radioactive emissions, but no data are available for them yet.

Radionuclide discharges from the phosphate industry have been shown to be relatively significant [OSPAR, 1997]. The main source of such discharges is the production of phosphoric acid from the ore using sulphuric acid by the fertiliser industry. Several natural radionuclides of the uranium series, which are contained in the ore, also appear in the produced phosphogypsum that can be discharged into surface waters, stockpiled on land or sold as a by-product. The phosphogypsum contains nearly all of the  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  and smaller quantities of uranium and thorium isotopes. It is the discharges of phosphogypsum into the sea that cause the input of radioactivity from the fertiliser industry.

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Large amounts of water are extracted and discharged into the sea during off-shore oil and gas production. This so-called 'produced' water may contain significant quantities of radionuclides, the most important of which are  $^{228}\text{Ra}$ ,  $^{226}\text{Ra}$  and  $^{210}\text{Pb}$ . Furthermore the equipment used in the production accumulates  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$  and  $^{228}\text{Th}$  contaminated scale. Removal of this scale may result in radioactive releases into the sea if it is disposed as solids off-shore or if it is dissolved so that it does not interfere with the production process.

The trends in the input of NORM into the OSPAR area can be clearly seen in the charts illustrating the total input of  $\alpha$ -activity into the North-East Atlantic (Figure 5 and Figure 6). They show a decrease in the discharges of waste from the fertiliser industry and increases in discharges from the oil and gas industries in line with production. Further information is provided in Sections 3.3.2 and 3.3.3 below.

### **3.3.2 Discharges from the phosphate industry**

According to the OSPAR report on discharges of radioactive substances by non-nuclear industries [OSPAR 1997], the following countries discharged phosphogypsum into the OSPAR area:

- Belgium-Luxembourg (discharged into the River Scheldt at Antwerp),
- Denmark (at Fredericia, Aalborg, and Kattegat),
- Former West Germany (reported discharges were very small),
- France (discharge in the Baie de la Seine),
- Ireland (Cork, stopped operating in early 1980s),
- Netherlands (discharged near Rotterdam into the Rhine estuary),
- Portugal (stored on the banks of the Tagus river),
- Spain (estuaries of the Tinto and Odiel rivers),
- UK (Whitehaven Plant in North West England; Belfast in Northern Ireland).

Over the last two decades phosphogypsum discharges have been reduced significantly as a result of the following:

1. European production of phosphoric acid has been substituted with imports, mainly from North Africa and the Middle East,
2. Technological developments led to a reduction in the use of the wet process,
3. Phosphogypsum tailings have been stored ashore,
4. Introduction of improved effluent treatment.

Substitution of phosphoric acid production with imported phosphoric acid has taken place at the UK Whitehaven plant in 1992. At the Antwerp fertiliser plant the sulphuric acid process was replaced with the nitric acid process in the beginning of 1993. The two phosphoric acid production plants near Rotterdam were both closed down in 1999 and 2000 respectively. Discharge of phosphogypsum in the Seine River was abandoned in 1990; the remaining phosphogypsum production was stockpiled on land until reported closure of the plant in 1992 [UNSCEAR, 1993].

Phosphate ore contains all radionuclides from the uranium and thorium natural decay chains. However the radionuclides from the decay chains of  $^{235}\text{U}$  and  $^{232}\text{Th}$  occur in much lower concentrations than those of the  $^{238}\text{U}$  decay chain. Only the latter are

considered in the present assessment. The  $^{238}\text{U}$  decay chain not only comprises  $^{238}\text{U}$  but  $^{234}\text{U}$  as well. In addition the short-lived  $^{234}\text{Th}$  and the long-lived  $^{230}\text{Th}$  can be assumed to occur in phosphate ore in activity concentrations equal to  $^{238}\text{U}$ . However the fate of these radionuclides in phosphoric acid production is not sufficiently documented to derive reliable estimates of their discharges with phosphogypsum. The discharges of uranium reported for the UK plant at Whitehaven [Camplin et.al, 1996] appear to be largely derived from uranium analyses of ores and products and not from the analyses of the discharged phosphogypsum. From 1992 the plant processed imported crude phosphoric acid and the associated discharges are therefore not typical for production of phosphoric acid from ore. The derived releases of the thorium isotopes at the Whitehaven plant also do not provide a sound basis for assessing releases at other sites. Uranium and thorium discharge data are not available for the Dutch and Belgian sites. It has also been reported that only a small proportion of uranium and thorium isotopes become associated with phosphogypsum compared with 80% of the  $^{226}\text{Ra}$  and practically all of the  $^{210}\text{Pb}$  and of the  $^{210}\text{Po}$  [OSPAR, 1997]. Therefore, the discharges of uranium and thorium radionuclides have not been considered in the present assessment. Hence it was decided to only consider three principal radionuclides in all cases:  $^{226}\text{Ra}$ ,  $^{210}\text{Po}$  and  $^{210}\text{Pb}$ . These radionuclides are likely to be the main cause of radiological impact.

In the absence of site-specific nuclide discharge data the estimates of radionuclide discharges are based on the following normalised figures (see Appendix B for details):

- a) a mass ratio of 1:4.5 for  $\text{P}_2\text{O}_5$  and produced phosphogypsum,
- b) discharges of 0.49, 0.47 and 0.45 GBq of  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  respectively per ktonne of phosphogypsum,
- c) Activity ratio of 0.49:0.47:0.45 for  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$ .

It was estimated that since 1981, the total discharges of the  $\alpha$ -emitters ( $^{226}\text{Ra}$  and  $^{210}\text{Po}$ ) and  $\beta$ -emitters ( $^{210}\text{Pb}$ ) amounted to 65 TBq and 32 TBq respectively. In fact, large discharges of radioactivity have been taking place since early in the beginning of the twentieth century, but there is no information on the scale of these discharges. The trend of decreasing phosphoric acid production and respective reductions in activity discharges has been observed since the early 1980s (see Figure 30). Large radionuclide discharges into the OSPAR area ended with the closure of plants in Rotterdam in 1999 and 2000.

Since 1981, the Dutch industry accounted for 43% of the total activity discharges. It was followed by the UK (18%), Belgium-Luxembourg (12%), Spain (12%) and Denmark (10%). The remaining 4% were discharged by France.

### 3.3.3 Discharges from the oil and gas industries

The Marina II assessment (see Appendix C for details) estimated discharges of activity with produced water<sup>3</sup>. It did not consider inputs of activity from the releases of solid scale or from onshore decontamination. A reference ratio of 0.33 between the volumes of produced oil and water is used as a reference average value for all oil producing platforms in the sea area of interest over their lifetime. This means that for each 1 m<sup>3</sup> of oil 3 m<sup>3</sup> of water is assumed to be co-produced. With this ratio the discharge of produced water is not likely to be overestimated. Ratios for individual platforms may differ considerably from this value. It is believed that it is likely to be optimistic, i.e. it underestimates the average water production from a platform over its lifetime. A ratio of 5 10<sup>-5</sup> is used between the water production and standard m<sup>3</sup> of produced gas, e.g. for each million m<sup>3</sup> of gas 50 m<sup>3</sup> of water is assumed to be co-produced.

The average concentration of the radionuclides <sup>226</sup>Ra and <sup>228</sup>Ra in produced water discharged from all oil producing platforms and over all years is estimated at a reference value of 10 Bq/l each. For gas production the corresponding figures are: <sup>226</sup>Ra 10 Bq l<sup>-1</sup>, <sup>210</sup>Pb 5 Bq l<sup>-1</sup> and <sup>228</sup>Ra 3 Bq l<sup>-1</sup>.

The normalised annual releases of natural radionuclides are derived from the annual production rates of oil and gas on the continental shelves of the four countries (United Kingdom, the Netherlands, Norway and Denmark), on the reference values for the ratio between produced water and oil and gas respectively and on the reference nuclide concentrations in produced water from oil and gas production. The results do not represent actual releases in specific production fields or from individual platforms but are estimates of the collective releases by all production installations over the years of the development of the oil and gas production.

Discharge trends for  $\alpha$ -emitters and  $\beta$ -emitters from the oil industry are illustrated in Figure 31. It can be seen that these discharges grew rapidly from the development of the first off-shore oil and gas fields in the early-1970s stabilising in the 1990s at around 20 TBq/y with a roughly equal split between  $\alpha$ -emitters and  $\beta$ -emitters.

By activity input, <sup>226</sup>Ra is responsible for some 50% of discharges with <sup>228</sup>Ra and <sup>210</sup>Pb contributing 25% each.

The radionuclide releases take place in the North Sea (vast majority of them from oil fields) and, in 1999, were split between the countries as follows:

- Norway – 52%
- UK – 45%
- Denmark – 2.5%
- Netherlands – 0.5%

In a follow-up of the 1997 OSPAR report on radionuclide discharges by non-nuclear industries [OSPAR, 1997] further detail on discharges was sought from Contracting

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<sup>3</sup> Produced water: water produced in the form of vapour or liquid with crude oil and natural gas. The liquid water may be free or emulsified.

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Parties. The resulting RSC (Radioactive Substances Committee) draft report prepared by the UK [OSPAR 2002] presents UK discharge data for the offshore oil and gas industries, which are several orders of magnitude lower than the MARINA II estimates.

The RSC discharge data only pertain to discharges of solids (scales) and do not include the contribution from produced water. It is very likely that the activity removed by de-scaling and then discharged is only a small fraction of the activity discharged with produced water in dissolved or finely dispersed form.

In the absence of data from monitoring of produced water, the MARINA II approach provides a more reliable estimate of total discharges than the data on discharges from descaling.

The arguments in support of the Marina II approach for UK discharges are equally valid for the Norwegian and Danish discharges, for which no figures were given in the RSC draft report.

For the Netherlands a figure of 89 GBq/y for 1989/1990 from the RSC report is in agreement with that given in Marina II however the basis of the estimates differ significantly. From the RSC data it seems that the 89 GBq/y covers estimated discharges from all offshore oil and gas production but the figure taken from the reference used only pertains to oil production. From the monitoring experience accumulated since 1995 it is clear that produced water from gas production contains  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$  and  $^{210}\text{Pb}$  in highly variable concentrations.

Nevertheless, total discharges by the Dutch offshore industry are dominated by discharges from the relatively small oil production because of the much larger volumes of produced water.

### **3.4 Atmospheric fallout**

#### **3.4.1 Nuclear tests**

The testing of nuclear weapons in the atmosphere was the most significant cause of exposure of the world population to man-made environmental sources of radiation [United Nations, 2000]. The practice continued from 1945 to 1980 by China, France, the Soviet Union, the United States and the United Kingdom, who together carried out 543 atmospheric tests.

Based on the UNSCEAR assessment [United Nations, 2000], the total global fallout in the Northern Hemisphere is estimated to equal  $1.4 \cdot 10^5$  PBq. The OSPAR 'share' of this fallout was 500 TBq for  $\alpha$ -emitters; 90 PBq for  $\beta$ -emitters excluding tritium; and  $8.5 \cdot 10^3$  PBq for tritium itself. Therefore, the testing contributed more activity into the OSPAR area than all the other sources taken together. Time dependent data for each  $10^\circ$  latitude band of the Northern Hemisphere are illustrated in Figure 32. It can be seen that deposition peaked in the early 1960s. It tailed off in the early 1980s. Integrated over time, values of activity deposition in northern hemisphere for  $10^\circ$  latitude bands are given in Table 8.

The majority of the fallout (99%) is due to tritium with the remaining activity resulting primarily from  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ . The radionuclide distribution, excluding tritium, is illustrated in Figure 33.

In the North-East Atlantic the greatest deposition occurred in the early 1960s near the coast of Spain and Portugal (40-50°N) and amounted to some 2 000 Bq/m<sup>2</sup> (excluding tritium).

### 3.4.2 Chernobyl

Following the accident at Unit 4 of the Chernobyl Nuclear Power Plant, the atmospheric release of radionuclides took place in the period April 26-May 10, 1986. During the accident, the fuel elements ruptured and the resultant explosive force of steam lifted off the cover plate of the reactor, releasing fission products to the atmosphere. A second explosion threw out fragments of burning fuel and graphite from the core and allowed air to enter the reactor, causing the graphite moderator to burst into flames. The graphite burned for nine days, causing the main release of radioactivity into the environment. Some 8 of the 140 tonnes of fuel, which contained plutonium, uranium and fission products, were ejected from the reactor along with a portion of the graphite moderator, which was also radioactive. These materials were scattered around the site. In addition, caesium and iodine were released both by the explosion and during the subsequent fire.

A total of about 12 000 PBq of radionuclides was released. Although most of the fallout was concentrated within the 5 km radius of the release, the high energy of the releases ejected radionuclides to the atmosphere and resulted in the formation of a radioactive cloud, which deposited radioactivity around the world. In the first weeks after the accident I-131 (half-life 8 days) was the most important source of exposure. In the medium to long term contamination was dominated by the fallout of caesium isotopes. Other significant radionuclides (including  $^{90}\text{Sr}$ ,  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$ ), which were less volatile, were deposited relatively close to Chernobyl. The World Health Organisation (WHO) has estimated that approximately 100 PBq of  $^{137}\text{Cs}$ , and 50 PBq of  $^{134}\text{Cs}$  were released to the environment [World Health Organisation, 1987]. A more recent assessment quoted similar values of between 74 and 85 PBq for  $^{137}\text{Cs}$  and between 44 and 54 PBq of  $^{134}\text{Cs}$  [De Cort, et al 1998]. The release of  $^{134}\text{Cs}$  was about 56% of  $^{137}\text{Cs}$ , but can be approximated as 50%.

The fallout of these isotopes in the North-East Atlantic was estimated on the basis of a limited number of  $^{134}\text{Cs}$  concentrations in seawater samples in the first months after the accident:

#### 1. North Sea and Skagerrak<sup>4</sup>

Samples were taken in the North Sea and Skagerrak by two expeditions in May-June 1986 and September 1986 [Commission of the European Communities, 1990]. Deposition can be estimated from average concentrations calculated by the MARINA I study:

Concentration:  $^{137}\text{Cs}$  – 16 Bq m<sup>-3</sup>;  $^{134}\text{Cs}$  – 8 Bq m<sup>-3</sup>

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<sup>4</sup> Relates to compartments 55-60 of the MARINA II compartmental model

Deposition:  $^{137}\text{Cs} - 2\,000\text{ Bq m}^{-2}$ ;  $^{134}\text{Cs} - 1\,000\text{ Bq m}^{-2}$

## **2. North East Atlantic Ocean 60°-75°N<sup>5</sup>**

In the summer of 1987, 75 seawater samples were collected in the Greenland, Norwegian and Barents seas, at Utsira in west Norway and from the Faroe Islands, from which deposition was estimated. [Commission of the European Communities, 1990]

Deposition:  $^{137}\text{Cs} - 500\text{ Bq m}^{-2}$ ;  $^{134}\text{Cs} - 250\text{ Bq m}^{-2}$

## **3. Irish and Scottish waters, and the English Channel<sup>6</sup>**

Estimates in MARINA I were based on the average between water concentrations, integrated over time to infinity, in the North Sea and North-East Atlantic 60°-75°N. A simple average of deposition rates results in the following estimate:

Deposition:  $^{137}\text{Cs} - 1250\text{ Bq m}^{-2}$ ;  $^{134}\text{Cs} - 625\text{ Bq m}^{-2}$

## **4. Remaining North East Atlantic Ocean 40°-60°N<sup>7</sup>**

Deposition was assumed to be 10 times lower than in the 60°-75° N band because of the low average fallout in Southern Europe. Current estimates simply assume that the average deposition in Spain is applicable (0.031 PBq [De Cort et al, 1998] over 500 000 km<sup>2</sup>).

Deposition:  $^{137}\text{Cs} - 60\text{ Bq m}^{-2}$ ;  $^{134}\text{Cs} - 30\text{ Bq m}^{-2}$

It is thus estimated that the maximum deposition took place in the North Sea (3 Bq m<sup>-2</sup>), which is comparable to the peak annual deposition values in Southern Europe estimated for atmospheric nuclear weapons tests (excluding tritium). Total caesium fallout in the OSPAR area is estimated to have been 7.4 PBq

### **3.4.3 Fallout from the burn-up of SNAP-9A satellite**

The American satellite burned up during re-entry at the height of 46 km in the Southern Hemisphere over the Indian Ocean in 1964. Three quarters of the fallout occurred in the Southern Hemisphere, but enhanced concentrations of  $^{238}\text{Pu}$  were measurable all over the world. The cumulative fallout for the Northern Hemisphere is given as  $115 \pm 30\text{ TBq}$  [Hanson, 1980].

In the North-East Atlantic the maximum  $^{238}\text{Pu}$  deposition took place near Southern Europe (about 1Bq m<sup>-2</sup>, see Table 9).

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<sup>5</sup> Relates to compartments 2,5, 16, 17, 19-27 of the MARINA II compartmental model

<sup>6</sup> Relates to compartments 28-39 and 46-54 of the MARINA II compartmental model

<sup>7</sup> Relates to compartments 8-15, 40-44 of the MARINA II compartmental model

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### 3.4.4 Other

A proportion of the atmospheric releases from nuclear installations deposits in the North-East Atlantic. The estimate of this fallout is beyond the scope of the current study.

The only other 'fallout' contamination in the North-East Atlantic resulted from the accident involving a US Air Force B-52 bomber, which crashed on the ice of an Arctic bay at Thule, Greenland in January 1968. It is reported that almost all the plutonium was recovered. However, about 1 TBq went to nearby soils and bottom sediments [Hanson, 1980]. Danish expeditions conduct regular monitoring of the site and so far have found only localised contamination [Nielsen, 2001].

## 3.5 Sea dumping

### 3.5.1 Waste dumping in North East Atlantic

An overview of sea dumping in the North-East Atlantic is provided in a recent IAEA study [Rastogi et al, 1999]. At the 2 main sites in the North-East Atlantic (46°00'N, 16°45' W and 46°15'N, 17°25'W), a total activity of more than 30 PBq was disposed of [Baxter et al, 1995]. The alpha emitting inventory at these sites would be expected to be of the order of 0.5 PBq. These dumping sites were used until 1982. Dumping in the North-East Atlantic resulted in enhanced localised concentrations of some radionuclides ( $^{14}\text{C}$ ,  $^{137}\text{Cs}$ ,  $^{238}\text{Pu}$ ,  $^{239+240}\text{Pu}$ ,  $^{241}\text{Am}$ ). It can be inferred that there is measurable leakage from the dumped waste containers at these sites. However, the highest observed activities (0.6 mBq  $^{137}\text{Cs}/\text{l}$  and 20  $\mu\text{Bq}$   $^{239+240}\text{Pu}/\text{l}$ ) are extremely small.

Estimated releases from sea dumps in the North-East Atlantic are given in the OECD study [OECD, 1985]. The estimated release rates are:

- For  $^{239}\text{Pu}$ , the release rate rises from 1 GBq/y in 1950 linearly to 10 000 GBq/y in 1970 until 1990, when it drops to 0.1 GBq/y slowly decreasing to 0.01 GBq/y over the next 10 000 years.
- For  $^{55}\text{Fe}$ , the discharges rise from 0.01 GBq/y in 1950 to 1 GBq/y in 1980, dropping back to 10 000 Bq/y in 2030.
- For  $^3\text{H}$ , releases rise from 1 GBq/y in 1950 to  $10^6$  GBq/y in 1980, dropping down to 10 TBq/y in 2 000

### 3.5.2 Waste dumping in the Arctic

Reactor dumpings in the Kara Sea were estimated to have resulted in only small releases of corrosion products [Ali et al, 1997; IAEA, 1999]. Observed concentrations in the environment at the major dumpsites in Tsilkovka, Stepovoy and Abrosimov Fjords and the Novaya Zemlya Trough show that radionuclide concentrations are generally low, similar to those observed in the open Kara Sea. There are very localized areas of raised concentrations [IAEA, 1999]. Therefore it can be assumed that there has been no impact on the OSPAR area from waste dumping in the Arctic.

Other discharges in the Arctic resulted from:

- The historic dumpings of liquid waste in North West Russia
- Discharges from the nuclear fuel cycle facilities Mayak, Tomsk-7 and Krasnoyarsk-26 via Ob and Yenisey rivers

A number of studies have been conducted in the Russian Arctic (e.g. [AMAP, 1997], [Norwegian-Russian project, 1998]). They quote only low levels of local contamination. It can thus be assumed that there has been no impact on the OSPAR area from these sources.

### **3.6 Accidents at sea**

An IAEA study [Calmet et al, to be published] analyses the latest data on accidents at sea. Only a small number of the reported accidents are located close enough to have any impact on the North East Atlantic:

1. An accident occurred onboard the Komsomolets submarine, on 7 April 1989 in the Norwegian Sea 180 km south-east of Bear Island. The radionuclide release rate was estimated as no more than 37 GBq per year [Gladkov, 1994], [Vinogradov, 1996]. The Arctic Monitoring and Assessment Programme on the other hand estimates the  $^{137}\text{Cs}$  release rate as no more than 500 GBq per year [AMAP, 1997].
2. The largest reported accidental release of liquid radioactive waste (74 TBq) occurred in 1989 during the anchorage of a submarine of the Northern Fleet in the Ara Bay. The accident led to the radioactive contamination of a sea area of about 1 km<sup>2</sup> [Petrov et al, 1995]. The radionuclide composition has been assessed, based on the radionuclide content of primary circuit coolant from a typical Russian submarine that has been decommissioned at the shipyard in Severodvinsk (see Table 10).
3. The available information on the environmental concentrations in the area surrounding the sunken Russian submarine 'Kursk' indicates that there are no enhanced levels of activity [Amundsen, 2001].
4. There are no data available for the K-8 submarine, which sunk in the Bay of Biscay, other than that it is located away from the coast at a significant depth (thousands of metres).

### **3.7 Isotope production and use**

#### **3.7.1 Production of isotopes**

Large-scale commercial manufacture of radioactive materials for use in medicine, research and industry is undertaken by Amersham International at the sites in Cardiff and Amersham (UK). Other European countries, such as Belgium, Netherlands and Denmark are involved in radioisotope production at research reactors.

Discharge trends since 1988 are illustrated in Figure 34 and Figure 35 for the UK. Total discharges have decreased from 668 TBq in 1988 to 106 TBq in 1999, but discharges excluding tritium remained at a level of between 1.2 and 1.8 TBq. It should be noted that almost 90% of discharges excluding tritium are due to releases of  $^{14}\text{C}$  at the facilities in Cardiff. This radionuclide has low radiotoxicity.

### 3.7.2 Application of radiopharmaceuticals

Radionuclides are extensively used in medicine for therapy and diagnosis. During therapeutic treatments, a sealed radionuclide source, principally  $^{60}\text{Co}$ , is placed in the vicinity of the patient's body. Such sources are returned by hospitals to the supplier or radwaste management facility and do not result in direct discharges to sea.

In diagnostic radiology, radiopharmaceuticals are used as tracers in the patient's body. A wide range of radionuclides are used for this purposes including  $^{11}\text{C}$ ,  $^{15}\text{O}$ ,  $^{13}\text{N}$ ,  $^{18}\text{F}$ ,  $^{51}\text{Cr}$ ,  $^{75}\text{Se}$ ,  $^{59}\text{Fe}$ ,  $^{57}\text{Co}$ ,  $^{58}\text{Co}$ ,  $^{67}\text{Ga}$ ,  $^{90}\text{Sr}$ ,  $^{99\text{m}}\text{Tc}$ ,  $^{111}\text{In}$ ,  $^{125}\text{I}$ ,  $^{131}\text{I}$ ,  $^{123}\text{I}$ ,  $^{133}\text{Xe}$  and  $^{201}\text{Tl}$ .

The isotopes that are used most widely in medical procedures are  $^{131}\text{I}$  and  $^{99\text{m}}\text{Tc}$ . The total world usage of  $^{131}\text{I}$  in nuclear therapy is approximately 600 TBq [United Nations, 2000]. This UNSCEAR report states that 'there is high excretion of  $^{131}\text{I}$  from patients following oral administration, but waste treatment systems with hold-up tanks are effective in reducing the amounts in liquid effluents by radioactive decay to 0.05% of the amounts administered to patients'. By the time sewage reaches the sea generally most of the  $^{131}\text{I}$  will have decayed. Assuming that OSPAR countries are responsible for one third of the global  $^{131}\text{I}$  usage, this would result in only 100 GBq being discharged in the whole of North-East Atlantic. According to UNSCEAR this can be confirmed by the very low concentrations of  $^{131}\text{I}$  measured in the surface waters and sewage systems of several countries [United Nations, 1993].

The main isotope, used in 80% of all diagnostic examinations, is  $^{99\text{m}}\text{Tc}$ , which is produced in  $^{99\text{m}}\text{Tc}$  generators from  $^{99}\text{Mo}$ .

Based on the average amount of  $^{99\text{m}}\text{Tc}$  administered per procedure and the number of procedures in developed countries, the total activity of this radionuclide administered in Western European countries annually is estimated to be about 4 000 TBq. In patients, short-lived  $^{99\text{m}}\text{Tc}$  decays to  $^{99}\text{Tc}$  (0.01 GBq for the whole of western Europe), which enters the sewage system. Assuming all of it ultimately drains into the North-East Atlantic, 0.01 GBq/y of  $^{99}\text{Tc}$  is discharged annually. This is negligible in comparison with a monthly discharge figure of up to 25 TBq of  $^{99}\text{Tc}$  from Sellafield between 1994 and 1999.

## 3.8 Discharges from the Baltic and Mediterranean Seas

### 3.8.1 Discharges into the OSPAR area from the Baltic Sea

The annual  $^{137}\text{Cs}$  flux from the Kattegat (Baltic Sea) to the Skagerrak (North-East Atlantic) is given in Figure 36. These data were extracted from the compartmental model used in the European MARINA-Balt study [Nielsen, 2000]. The model showed a net outflow of  $^{137}\text{Cs}$  from the Baltic Sea during 1950-2050 except for the years 1975-1985 when Sellafield discharges cause a net inflow. The outflow since 1986 is due to Chernobyl fallout, which affected the Baltic Sea mainly as a result of

rain washout from the contaminated plume. Maximum outflow was observed in 1986, after the Chernobyl accident, when it exceeded 120 TBq. Recent data in Figure 36 are believed to underestimate the actual discharges since the effect of remobilisation was not considered sufficiently in the MARINA-Balt study [Nielsen, 2002, personal communication]. The total net input of  $^{137}\text{Cs}$  from the Baltic Sea was estimated at 1 PBq up until the year 2000.

### 3.8.2 Discharges into the OSPAR area from the Mediterranean Sea

A recent review of all activity inputs into the Mediterranean Sea [Papucci et al, 1996] concluded that the total flow of  $^{137}\text{Cs}$  from the Atlantic through the Strait of Gibraltar amounted to 1.5 PBq in the period up to 1986. This is equivalent to the outflow of  $^{137}\text{Cs}$  from the Atlantic Ocean at a relatively small annual rate of 50 TBq. The Chernobyl accident counterbalanced the flux of caesium into the Mediterranean, so that since 1986 the net activity flux through the Strait of Gibraltar was close to zero.

Furthermore, enhanced levels of other radionuclides were observed in the Western Mediterranean in 1988-93. For example  $^{239,240}\text{Pu}$  concentrations were found to be between 7 and 30 mBq/m<sup>3</sup> [Mitchell et al, 1995]. These levels are relatively small (cf  $^{239,240}\text{Pu}$  concentrations of up to 5 000 mBq/m<sup>3</sup> in filtered water in the Irish Sea in the same period). Annual net  $^{239,240}\text{Pu}$  flux into the North East Atlantic was estimated to be 0.63 TBq.

### 3.9 Discharges from military establishments

The issue of discharges from military establishments is beyond the scope of the MARINA II study, which is dedicated to the analysis of civil nuclear discharges. However, some information on discharges from military establishments in the UK is summarised for completeness. There are no data available on similar discharges in France, which is the other nuclear power in Western Europe.

Since 1988 annual reports by the Ministry of Agriculture, Fisheries and Food and Scottish Environmental Protection Agency summarise liquid discharges of radioactivity from UK shipyards (Barrow, Devonport, Faslane and Rosyth), weapons production and testing programme (Aldermaston, Burghfield, Cardiff<sup>8</sup>, Foulness) and a training reactor in Greenwich<sup>9</sup>. Discharge trends are illustrated in Figure 37 and Figure 38. A general downward trend can be observed for discharges (excluding tritium). Over the last decade they have never exceeded 5 GBq/y with the majority originating from the three shipyards. Over 75% of the discharged radioactivity, excluding tritium, is due to  $^{60}\text{Co}$ . Tritium discharges peaked in 1990 at just over 700 GBq due to an increase in the releases from Aldermaston.

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<sup>8</sup> No longer operational

<sup>9</sup> Decommissioned

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## **4 Comments on uncertainties**

### **4.1 Nuclear industry**

1. The discharges of total  $\alpha$  and  $\beta$  activity from reprocessing plants and nuclear power plants have a comparatively low uncertainty. There is a high degree of confidence in the reported activity for the major radionuclides for which discharge limits are set by the regulators. These values are normally based on the direct analysis of samples from monitoring tanks or the outflow pipe and are verified by the appropriate Government Agency. Some of these discharges are upper estimates, because they include 'less than' data derived from analyses of effluents at the limits of detection.
2. A higher degree of uncertainty is associated with those radionuclides for which discharges were estimated based on a typical radionuclide composition (or 'fingerprint') of discharges for a particular type of facility. In particular this approach was used for NPP discharges when the activity of reported radionuclides was significantly less than the total reported activity. It is possible that in specific cases this generic approach could result in a distortion of the radionuclide composition.
3. Data on historic discharges from the nuclear cycle facilities (up to 1980s) are often based on expert estimates and knowledge of the history of plant operations. In some cases, when no discharge data are available, estimates are based on averaged values, extrapolated from the recent data. For example, the earliest available data on radionuclide discharges from Sellafield are taken to be a conservative estimate of earlier discharges. In one case, such an approach is known to have resulted in an error, when the recent estimates of tritium discharged from Sellafield in 1971 gave a figure of 5.5 PBq [Jackson et al, 2000] compared to 1.2 PBq quoted in MARINA I [Commission of the European Communities, 1990].
4. The overall scale of discharges from the nuclear industry is sufficiently accurate, because these discharges are largely dominated by reprocessing facilities in Sellafield and Cap de la Hague, for which a high proportion of actual discharge data are reported and which have subsequently been studied in depth.

### **4.2 Other sources of radioactivity in the North-East Atlantic**

1. Since the 1980s the fertiliser manufacturing and off-shore oil production industries have been significant sources of radioactive discharges. It is estimated that in 1999, the off-shore oil and gas industries accounted for 95% of all releases of  $\alpha$ -emitters. In most cases, industries that discharge NORM do not report annual releases into the marine environment to the regulatory authorities. Therefore, only approximate estimates of these discharges can be made. For the oil and gas industries, current estimates are based on information on the historic production rates that are deemed sufficiently accurate for present purposes. However, there is a large uncertainty associated with the amount of co-produced water and the radionuclide composition of the releases. These vary over the lifetime of the oil platforms and depend on geographical location. Estimated activity input from the fertiliser industry can only be regarded as a scoping

calculation, because there are few reliable data on the past production of phosphoric acid and related phosphogypsum discharges.

2. Atmospheric fallout from nuclear testing represents the most significant source of man-made activity in the environment. All-time fallout in the Northern Hemisphere from nuclear tests is estimated to be 14 000 PBq compared to 400 PBq of discharges from nuclear sites into the North-East Atlantic. This fallout is estimated based on a large number of measurements of  $^{90}\text{Sr}$  deposition over a long period of time. The measurement data agree well with expert assessments based on estimated releases and atmospheric dispersion models for individual tests. However, the reliability of assessments for other, particularly shorter-lived nuclides, is limited.
3. Fallout from Chernobyl is more recent and more concentrated, which results in the higher concentrations of  $^{137}\text{Cs}$  present locally compared to fallout from nuclear tests. Unfortunately, the assessment of fallout in the North-East Atlantic is based on a very limited number of measurements, which took place in 1986 and 1987. From a large number of land measurements it is known that spatial variability of Chernobyl contamination was significant. Although the estimate of total Chernobyl fallout into the North-East Atlantic is probably sufficiently accurate, there is a very high uncertainty associated with the reported deposition rates. This may result in a disagreement between modelled and measured concentrations of  $^{137}\text{Cs}$ .
4. There is a relatively high uncertainty associated with environmental contamination resulting from sea dumping of radioactive wastes and accidents at sea involving nuclear powered vessels. However, due to the small scale of these discharges, this uncertainty does not affect the overall situation.
5. Estimates of discharges from the Baltic and Mediterranean Seas are based on rough approximations, which do not reflect the complex exchange of activity at varying depths, as a result of localised flows. These localised flows would affect activity concentrations in localised areas. However due to the relatively low contribution of activity from the Baltic Sea and the Mediterranean, these uncertainties will not unduly distort the overall picture.

## 5 Conclusions

1. It was found that in the 1990s the input of activity into the North-East Atlantic has been relatively small compared to historic inputs. Inputs of  $\beta/\gamma$ -emitters into the OSPAR area reached their peak in 1963, as a direct result of weapons testing. Maximum discharges of  $\alpha$ -emitters were reached in 1974, resulting from reprocessing at Sellafield. Current discharges for  $\alpha$ -emitters and  $\beta$ -emitters represent around 5 and 1% of these peak values.
2. Since 1984, reprocessing at Sellafield (in the west of Britain) and Cap de la Hague (in the north of France) has been the most significant source of radioactive discharges.

From 1986 until 2000, Sellafield discharges have been reduced for  $\alpha$ -emitters and  $\beta$ -emitters, excluding tritium, from 3900 to 150 GBq/y and from 160 to 84 TBq/y respectively. Discharges of tritium remained unchanged at approximately 2200 TBq/y.

Discharges of  $\alpha$ -emitters from Cap de la Hague have been reduced from 810 GBq in 1986 to 21 GBq in 2000. For  $\beta$ -emitters excluding tritium, the respective figures are 1100 TBq/y and 34 TBq/y. Tritium discharges have increased almost fivefold (from 2.3 PBq/y to 11 PBq/y) in line with the increased rate of reprocessing.

3. The improved management of liquid waste at Nuclear Power Plants has resulted in a steady decrease in the discharges of  $\beta$ -emitters excluding tritium. However, commissioning of new plants has resulted in an increase in the discharge of tritium. The discharge of tritium (3 PBq in 1999) is 1 000 times greater than for the other  $\beta$ -emitters and is the only discharge from the nuclear power stations that is significant compared to that from the reprocessing plants.
4. In the past input of radioactivity into the North-East Atlantic from fuel manufacturing facilities has been small in comparison with that from reprocessing. Since 1984, the discharges of  $\alpha$ -emitters have decreased by almost 8 times. From 1992 until 1998, they remained constant at around 100 GBq/y. In 1998, discharges of  $\alpha$ -emitters from Springfields almost doubled to 200 GBq/y and are presently responsible for almost 50% of all discharges of this type from the nuclear industry. Discharges of  $\beta/\gamma$ -emitters from these facilities are relatively small.
5. Since 1984, the research and development facilities have provided insignificant input of activity into the OSPAR area.
6. Discharges of NORM from the fertiliser and off-shore oil and gas industries have contributed significantly to the total input of  $\alpha$ -emitters. Discharges of  $\alpha$ -emitters from the fertiliser industry were at a maximum in 1981 (~7 TBq/y), which was when the earliest data were available for most countries. They tailed off by the end of 1990s due to replacement of European production of phosphoric acid with imports, other changes in fertiliser production and storage of phosphogypsum

ashore. Discharges of  $\alpha$ -emitters from oil production were estimated to have stabilised in the second half of 1990s at 10 TBq/y. In 1999, this accounted for 95% of all releases of  $\alpha$ -emitters into the OSPAR area.

7. At present most European countries do not report discharges of radioactivity with the produced water from the oil extraction. This results in significant underestimation in the reported radioactivity discharged into the OSPAR area.
8. The fallout from weapons testing has provided the largest input of activity into the OSPAR area (8,600 PBq), almost 99% of which is tritium. Following termination of the atmospheric testing, there has been no significant fallout from this source since the beginning of 1990s. The highest levels of deposition were estimated to have taken place in the south of the OSPAR area (2 000 Bq/m<sup>2</sup>, excluding tritium).
9. A major fallout of 7.4 PBq of caesium isotopes into the OSPAR area resulted from the Chernobyl accident in 1986. The highest deposition (1 000 Bq/m<sup>2</sup>) took place in the North Sea.
10. A relatively small fallout of <sup>238</sup>Pu resulted from the burn-up of SNAP-9A satellite in the 1960s. Maximum deposition took place near Southern Europe (about 1 Bq/m<sup>2</sup>).
11. Waste dumping in the North-East Atlantic and in the Arctic has not resulted in large releases of radionuclides. The most significant releases were estimated for <sup>239</sup>Pu in the North-East Atlantic in the 1970s, when, it is believed, these releases reached 10 TBq/y.
12. Only relatively small amounts of activity have been released into the OSPAR area as a result of accidents. Releases from the Komsomolets submarine in the Norwegian Sea are calculated not to exceed 500 GBq per year.
13. The largest reported accidental release of liquid radioactive waste (74 TBq) occurred in 1989 during the anchorage of a submarine of the Northern Fleet in the Ara Bay. Other naval accidents are believed to have resulted in only minor releases.
14. Discharges from isotope manufacturing facilities in Cardiff and Amersham have remained at a relatively low level. Total discharges have decreased from 668 TBq in 1988 to 106 TBq in 1999, but annual discharges excluding tritium remained at a level of between 1.2 and 1.8 TBq.
15. Discharges from the medical application of radiopharmaceuticals into the North-East Atlantic were found to be negligible.
16. The net flux of activity from the Baltic Sea was estimated to be significant. The integrated  $\beta$ -activity flux since 1984 until 2 000 amounted to 1 PBq. There is some indication that the caesium flux from the Mediterranean Sea has been of the same order of magnitude, but there are no data to provide a reasonable estimate.



17. Recent discharge data are available for discharges from the UK military establishments. A general downward trend can be observed. Over the last decade discharges, excluding tritium, have never exceeded 5 GBq/y. Tritium discharges reached the maximum of just over 700 GBq in 1990.

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## Tables

**Table 1. Sources of nuclear site discharge data**

Site	Type	Country	OSPAR Map Reference	Source of Data
Almaraz	NPP	SPAIN	E1	Bilcom
Barsebäck	NPP	SWEDEN	S1	Bilcom
Belleville	NPP	FRANCE	F1	Bilcom
Berkeley	NPP	UNITED KINGDOM	GB1	Bilcom
Beznau	NPP	SWITZERLAND	CH1	OSPAR
Biblis	NPP	GERMANY	D1	Bilcom
Blayais	NPP	FRANCE	F2	Bilcom
Borssele	NPP	THE NETHERLANDS	NL1	Bilcom
Bradwell	NPP	UNITED KINGDOM	GB2	Bilcom
Brokdorf	NPP	GERMANY	D2	Bilcom
Brunsbüttel	NPP	GERMANY	D3	Bilcom
Cap de la Hague	Reprocessing	FRANCE	F15	Nord Cotentin study, Cogema
Capenhurst	Fuel Fabrication	UNITED KINGDOM	GB16	MAFF
Cattenom	NPP	FRANCE	F3	Bilcom
Chapelcross	NPP	UNITED KINGDOM	GB4	Bilcom
Chinon	NPP	FRANCE	F4	Bilcom
Chooz	NPP	FRANCE	F5	Bilcom
Dampierre	NPP	FRANCE	F6	Bilcom
Doel	NPP	BELGIUM	B1	Bilcom
Dodewaard	NPP	THE NETHERLANDS	NL2	Bilcom
Dounreay (site)	Research Facility	UNITED KINGDOM	GB18	Bilcom, SEPA
Dungeness	NPP	UNITED KINGDOM	GB5	Bilcom
Emsland	NPP	GERMANY	D9	Bilcom
Fessenheim	NPP	FRANCE	F7	Bilcom
Flamanville	NPP	FRANCE	F7	Bilcom
Golfech	NPP	FRANCE	F9	Bilcom
Gösgen	NPP	SWITZERLAND	CH2	OSPAR
Grafenrheinfeld	NPP	GERMANY	D4	Bilcom
Gravelines				
	NPP	FRANCE	F10	Bilcom
Grohnde	NPP	GERMANY	D5	Bilcom
Hartlepool	NPP	UNITED KINGDOM	GB6	Bilcom
Harwell	Research Facility	UNITED KINGDOM	GB19	Bilcom
Heysham	NPP	UNITED KINGDOM	GB7	Bilcom
Hinkley Point	NPP	UNITED KINGDOM	GB8	Bilcom
Hunterston	NPP	UNITED KINGDOM	GB9	Bilcom
Jose Cabrera	NPP	SPAIN	E2	Bilcom
Kahl	NPP	GERMANY	D7	Bilcom
Karlsruhe	Research Facility	GERMANY	D18	Bilcom
Krümmel/Geesthacht	NPP	GERMANY	D8	Bilcom
Leibstadt	NPP	SWITZERLAND	CH3	OSPAR
Mülheim-Kärlich	NPP	GERMANY	D10	Bilcom
Mühleberg	NPP	SWITZERLAND	CH4	OSPAR
Neckarwestheim	NPP	GERMANY	D11	Bilcom
Nogent	NPP	FRANCE	F11	Bilcom

Site	Type	Country	OSPAR Map Reference	Source of Data
Obrigheim	NPP	GERMANY	D12	Bilcom
Oldbury	NPP	UNITED KINGDOM	GB10	Bilcom
Paluel	NPP	FRANCE	F12	Bilcom
Penly	NPP	FRANCE	F13	Bilcom
Philippsburg	NPP	GERMANY	D13	Bilcom
Ringhals	NPP	SWEDEN	S2	Bilcom
Risø	Research Facility	DENMARK	DK1	Bilcom
Sellafield	Reprocessing	UNITED KINGDOM	GB15	Jackson et al, 2000, MAFF, BNFL
Sizewell	NPP	UNITED KINGDOM	GB11	Bilcom
Springfields	Fuel Fabrication	UNITED KINGDOM	GB17	MAFF
St Laurent	NPP	FRANCE	F14	Bilcom
Stade	NPP	GERMANY	D15	Bilcom
Tihange	NPP	BELGIUM	B2	Bilcom
Torness	NPP	UNITED KINGDOM	GB12	Bilcom
Trawsfynydd	NPP	UNITED KINGDOM	GB13	Bilcom
Trillo	NPP	SPAIN	E3	Bilcom
Unterweser	NPP	GERMANY	D16	Bilcom
Winfrith	NPP	UNITED KINGDOM	GB20	Bilcom
Würgassen	NPP	GERMANY	D17	Bilcom
Wylfa	NPP	UNITED KINGDOM	GB14	Bilcom

**Table 2. Summary of total<sup>1</sup> activity input into the OSPAR area, GBq**

	Reprocessing	NPPs	Research	Fuel Fabrication	Weapons Testing	Chernobyl <sup>2</sup>	Baltic Sea <sup>3</sup>	Isotope Production <sup>4</sup>
<b>Total up to 1999</b>								
<b>α</b>	1.3E+06	2.7E+03	2.6E+04	3.9E+04	4.9E+05	N/A	N/A	N/A
<b>β</b>	1.4E+08	9.0E+05	1.0E+07	2.2E+06	8.7E+07	7.4E+06	N/A	N/A
<b><sup>3</sup>H</b>	1.8E+08	4.7E+07	3.1E+06	N/A	8.5E+09	N/A	N/A	N/A
<b>1985 – 1999</b>								
<b>α</b>	2.9E+04	3.8E+01	1.6E+03	4.5E+03	9.2E+01	N/A	N/A	1.3E+00
<b>β</b>	8.7E+06	1.5E+05	1.3E+05	7.4E+05	1.6E+04	7.4E+06	1.0E+06	1.8E+04
<b><sup>3</sup>H</b>	1.2E+08	3.8E+07	2.5E+05	N/A	1.6E+06	N/A	N/A	5.6E+06

Notes: 1 – integrated annual discharges, excluding NORM. In order to estimate the current radionuclide inventory it is necessary to take account of the following physical processes: radionuclide decay and ingrowth, sedimentation, resuspension and transport beyond the OSPAR area. Only significant inputs are presented in this table

2 – Chernobyl fallout took place in 1986

3 – mainly due to Chernobyl fallout.

4 – for UK facilities only, no data prior to 1988

**Table 3. Summary of NORM<sup>1</sup> activity input into the OSPAR area, GBq**

	Fertiliser industry	Off-shore Oil and Gas production
<b>Total up to 1999</b>		
<b>α</b>	6.5E+04 <sup>2</sup>	1.5E+05
<b>β</b>	3.2E+04 <sup>2</sup>	1.5E+05
<b>1985 – 1999</b>		
<b>α</b>	4.5E+04	1.1E+05
<b>β</b>	2.2E+04	1.3E+05

Notes: 1 – integrated annual discharges. In order to estimate the current radionuclide inventory it is necessary to take account of the following physical processes: radionuclide decay and ingrowth, sedimentation, resuspension and transport beyond the OSPAR area. Only significant inputs are presented in this table

2 – no data before 1981.

**Table 4. Nuclear industry - contribution to total discharges by facility type**

Type of facility	α-emitters		β-emitters, excl <sup>3</sup> H		Tritium		Total	
	%	TBq	%	TBq	%	TBq	%	TBq
Reprocessing	95.0	1.3E+03	91.4	1.4E+05	78.0	1.8E+05	83.2	3.2E+05
Power Station	0.2	2.7E+00	0.6	9.0E+02	20.4	4.7E+04	12.5	4.8E+04
Research <sup>1</sup>	1.9	2.6E+01	6.5	1.0E+04	1.3	3.1E+03	3.4	1.3E+04
Fuel Fabrication	2.9	3.9E+01	1.4	2.2E+03			0.6	2.2E+03
<b>Total</b>	<b>100</b>	<b>1.4E+03</b>	<b>100</b>	<b>1.5E+05</b>	<b>100</b>	<b>2.3E+05</b>	<b>100</b>	<b>3.8E+05</b>

Notes: 1 – includes Dounreay



**Table 5. Nuclear industry – contribution to total discharges by site (till end 2000)**

Individual site	$\alpha$ -emitters	$\beta$ -emitters	tritium	Total
Sellafield	94.2%	79.2%	34.2%	52.0%
Cap de la Hague	0.8%	12.2%	45.2%	32.2%
Dounreay	1.9%	6.6%	0.1%	2.7%
Other	3.1%	2.0%	20.5%	13.2%
<b>Total, TBq</b>	<b>1.4E+03</b>	<b>1.6E+05</b>	<b>2.4E+05</b>	<b>4.0E+05</b>

**Table 6. Nuclear industry - contribution to total discharges by site (for period 1985-2000)**

Individual site	$\alpha$ -emitters	$\beta$ -emitters	tritium	Total
Sellafield	72.0%	25.7%	18.6%	18.9%
Cap de la Hague	10.8%	63.9%	59.6%	59.8%
Dounreay	4.4%	1.3%	0.0%	0.1%
Other	12.8%	9.0%	21.9%	21.2%
<b>Total, TBq</b>	<b>3.6E+01</b>	<b>1.1E+04</b>	<b>1.8E+05</b>	<b>1.9E+05</b>

**Table 7. History of the operating of Cap de la Hague reprocessing plant**

Year (start/stop)	Installation name	Fuel type				Effluents treating process		
		UNGG	Oxide	MOX	NR	STE2	STE3	NGE
1966/1987	UP2	X				X		
1976/...	UP2 400 (HAO)		X	X	X	X		
1989/...	UP3 (foreign fuel)		X				X	Since 1990
1994/...	UP2 800 (French EDF fuel)		X					X

UNGG: Natural uranium-Graphite-Gas

Oxide: Fuel for light water reactors (REP et REB)

MOX: Mixed oxide fuel for light water reactors

NR: Rapid neutrons

STE: Effluents treating station

NGE: New effluents treating process

**Table 8. Nuclear testing deposition in Northern Hemisphere (integrated over time)**

Latitude band, °N	0-10	10-20	20-30	30-40	40-50	50-60	60-70	70-80	80-90
<b>Deposition, Bq/m<sup>2</sup></b>	2.4E+05	3.6E+05	5.3E+05	7.0E+05	9.7E+05	8.7E+05	5.2E+05	2.0E+05	7.7E+04

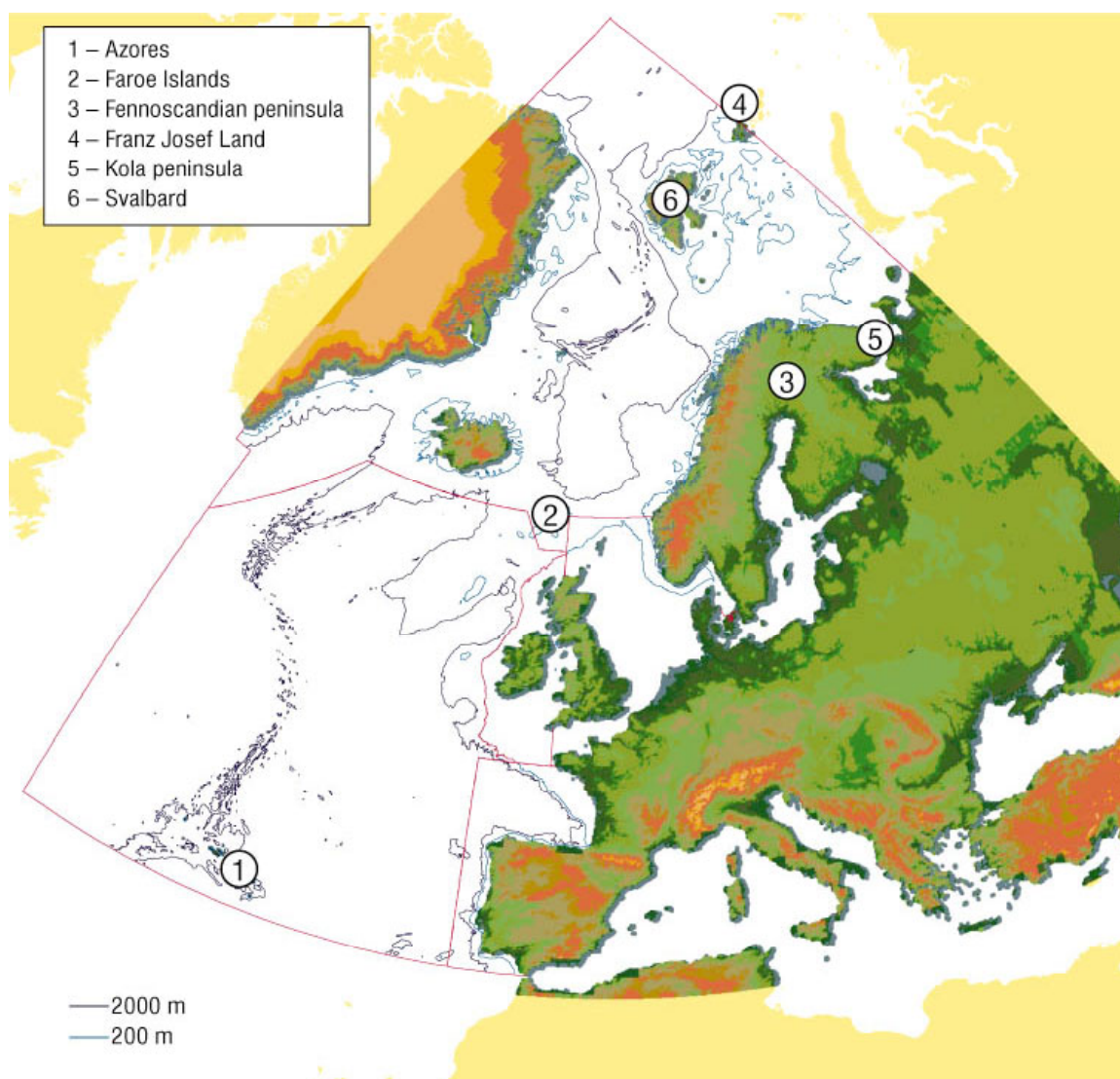
**Table 9.  $^{238}\text{Pu}$  deposition in Northern Hemisphere from burn-up of SNAP-9A satellite**

Latitude band, °N	0-10	10-20	20-30	30-40	40-50	50-60	60-70	70-80	80-90
Deposition, Bq/m <sup>2</sup>	<0.04	0.11	0.41	0.93	0.96	0.48	0.96	<0.04	<0.04

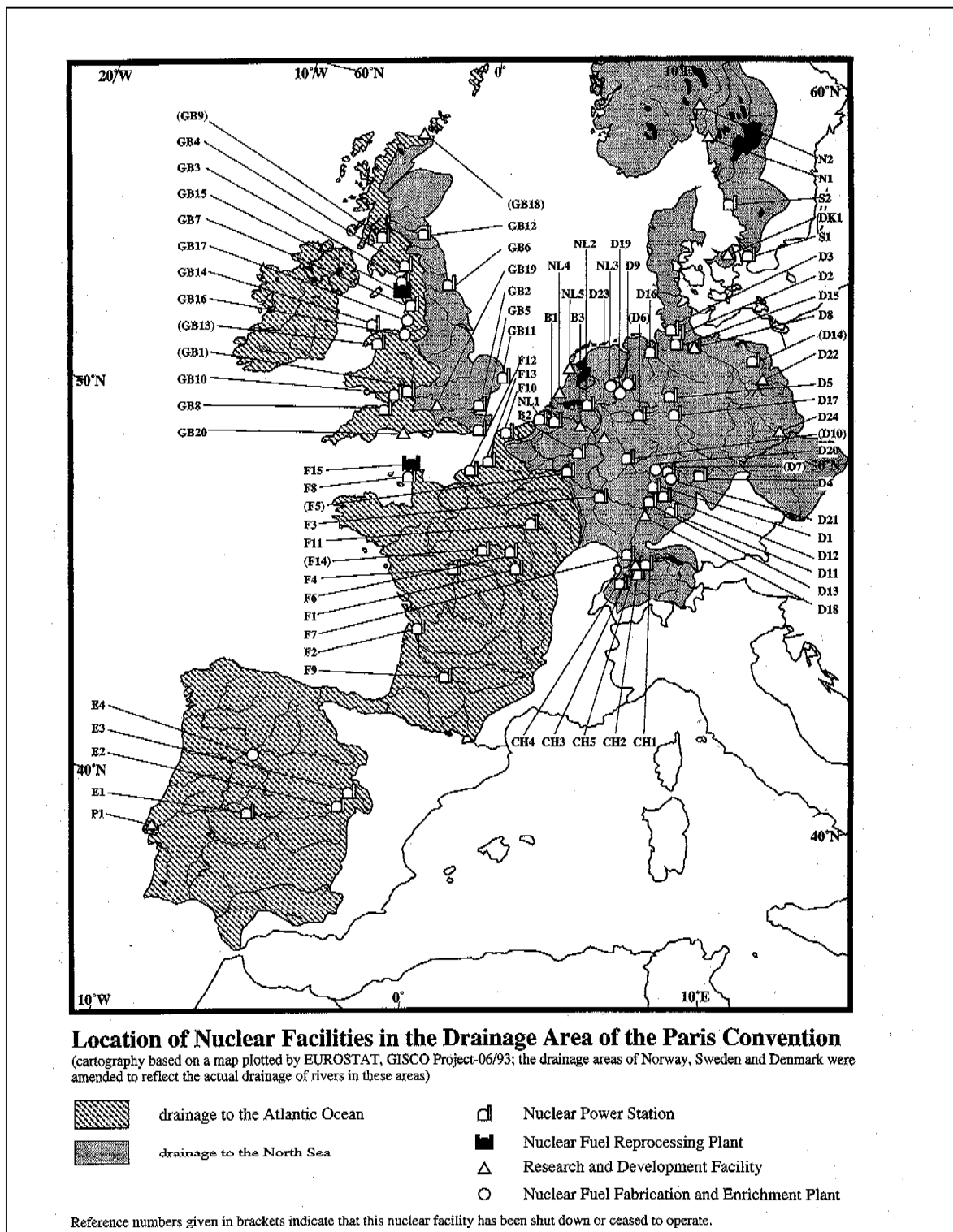
**Table 10. Estimated radionuclide composition of accidental discharge at Ara Bay**

Radionuclide	$^3\text{H}$	$^{14}\text{C}$	$^{137}\text{Cs}$	$^{55}\text{Fe}$	$^{90}\text{Sr}+^{90}\text{Y}$	$^{63}\text{Ni}$	$^{60}\text{Co}$
Activity, TBq	5.1E+01	7.6E+00	7.6E+00	3.5E+00	2.5E+00	1.5E+00	5.1E-01

## Figures

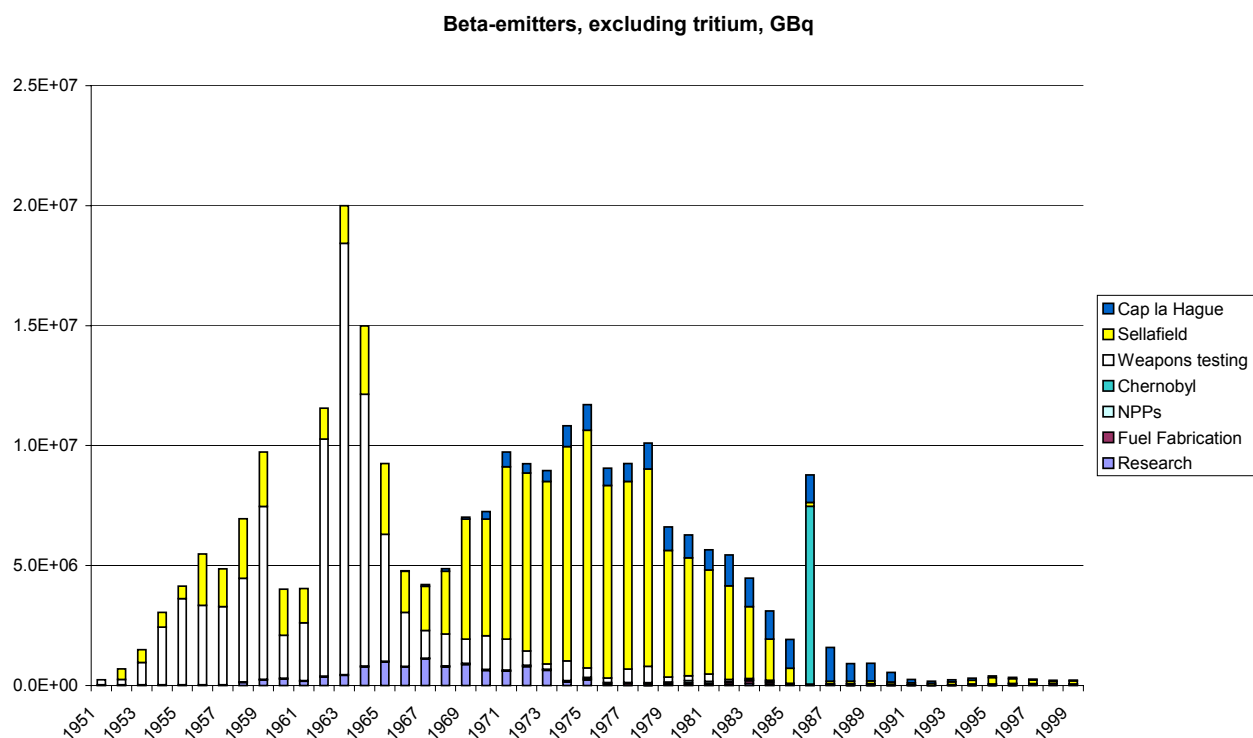


**Figure 1. OSPAR area (from OSPAR Commission 2000. Quality Status Report 2000. OSPAR Commission, London).**

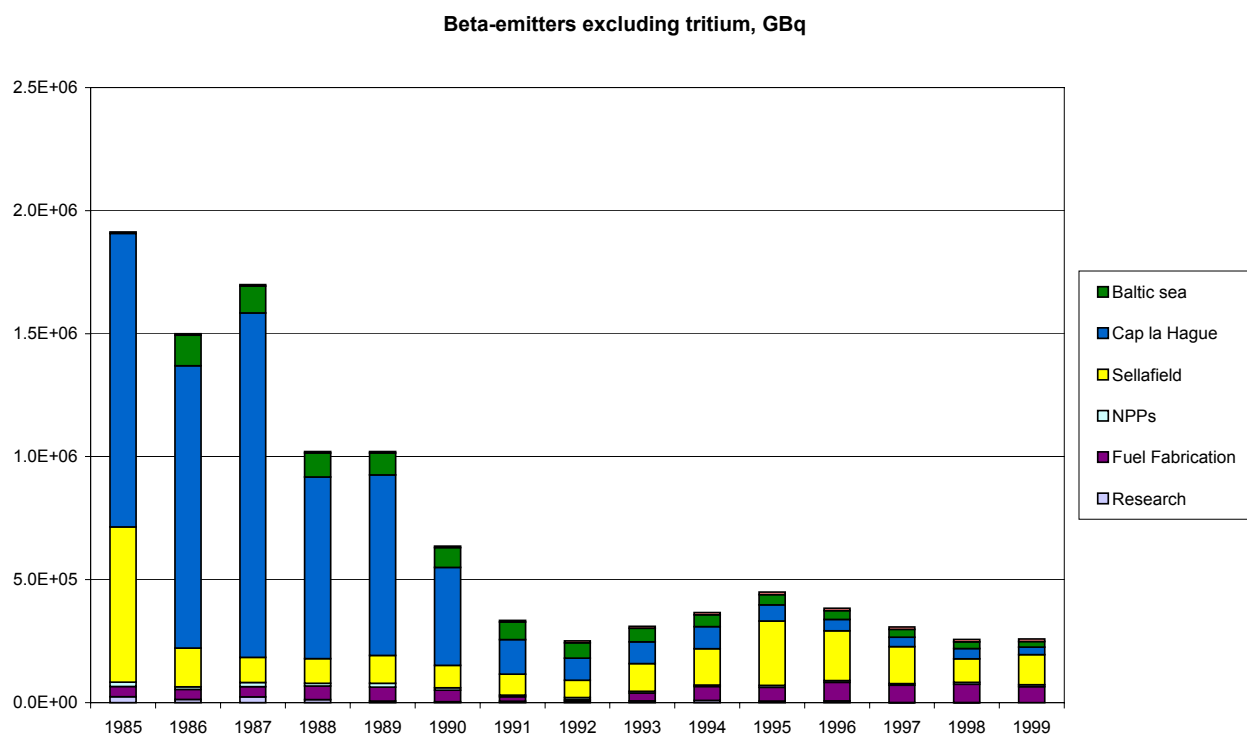


**Figure 2. Location of nuclear facilities discharging into the OSPAR area<sup>10</sup>**

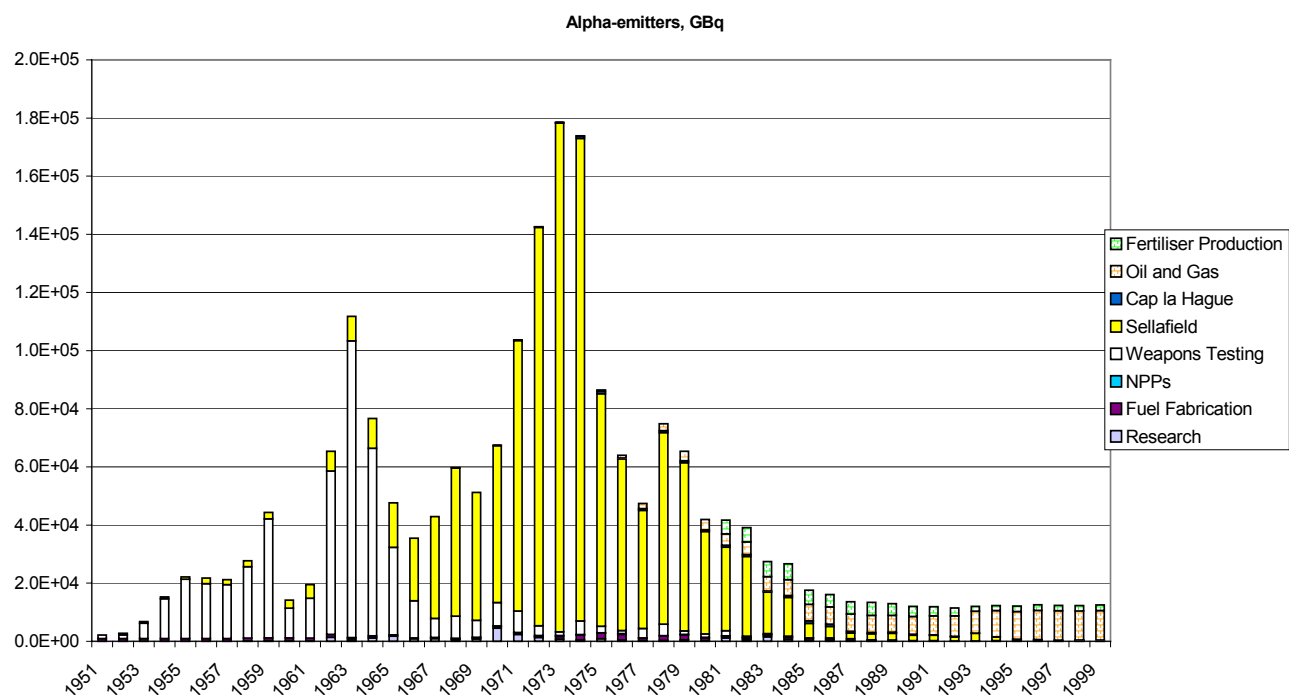
<sup>10</sup> Reference number descriptions given in Table 1.



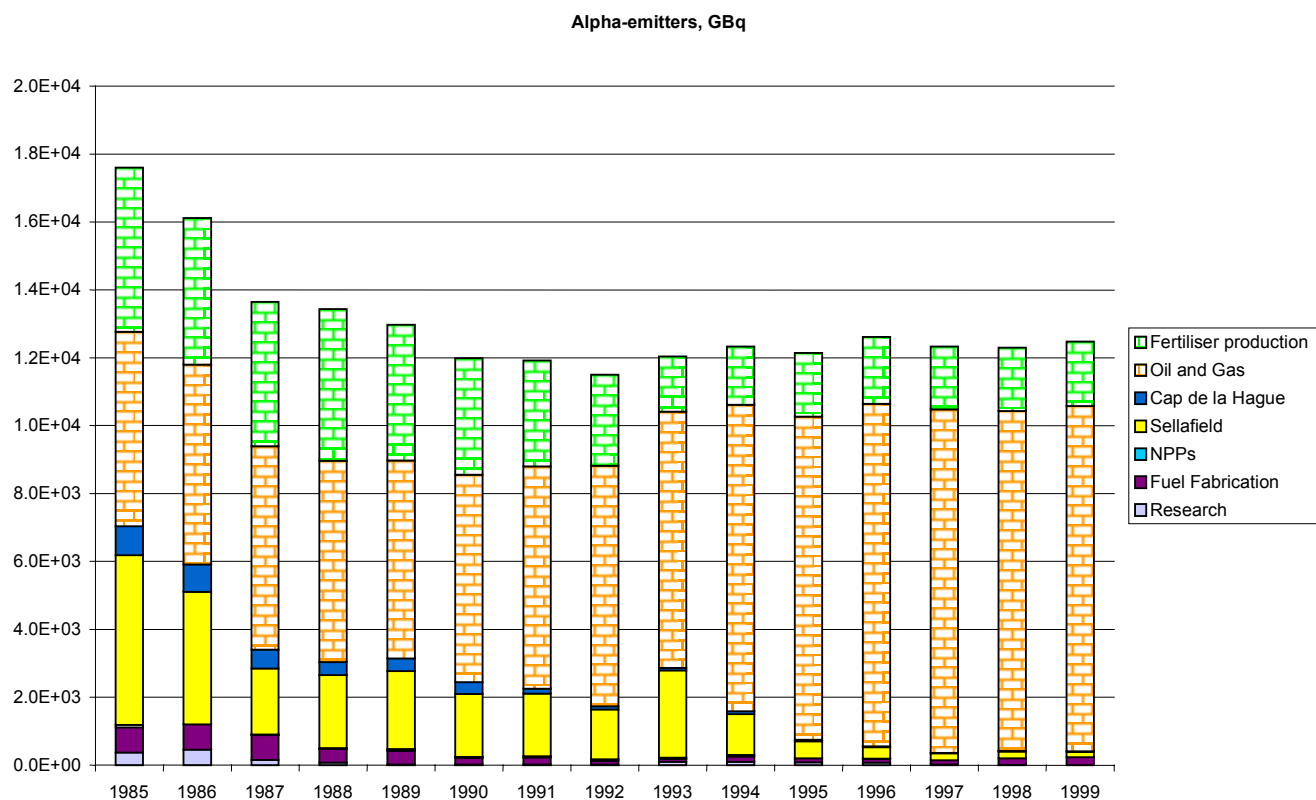
**Figure 3. Trends in overall input of  $\beta$  activity, excluding tritium, into the OSPAR area**



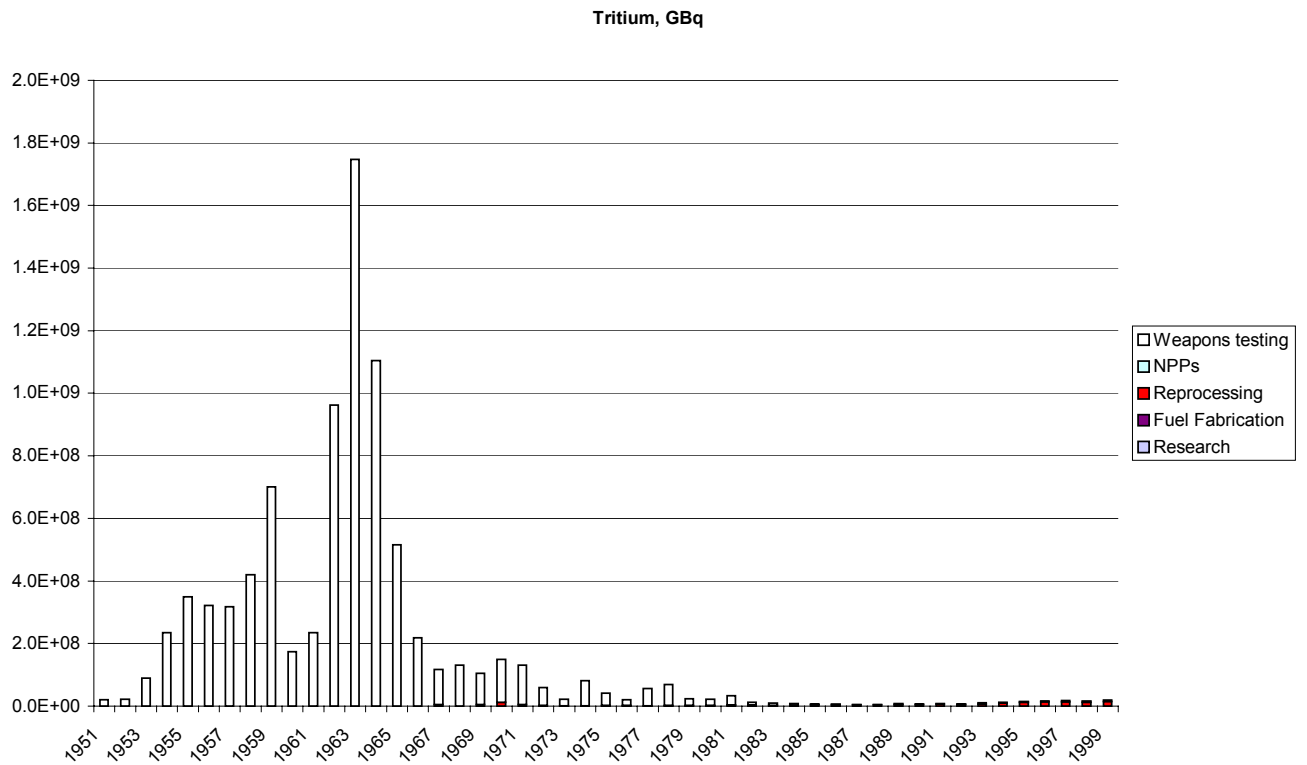
**Figure 4. Recent trends in overall input of  $\beta$  activity (excluding tritium and inputs from Chernobyl fallout and Mediterranean Sea) into the OSPAR area.**



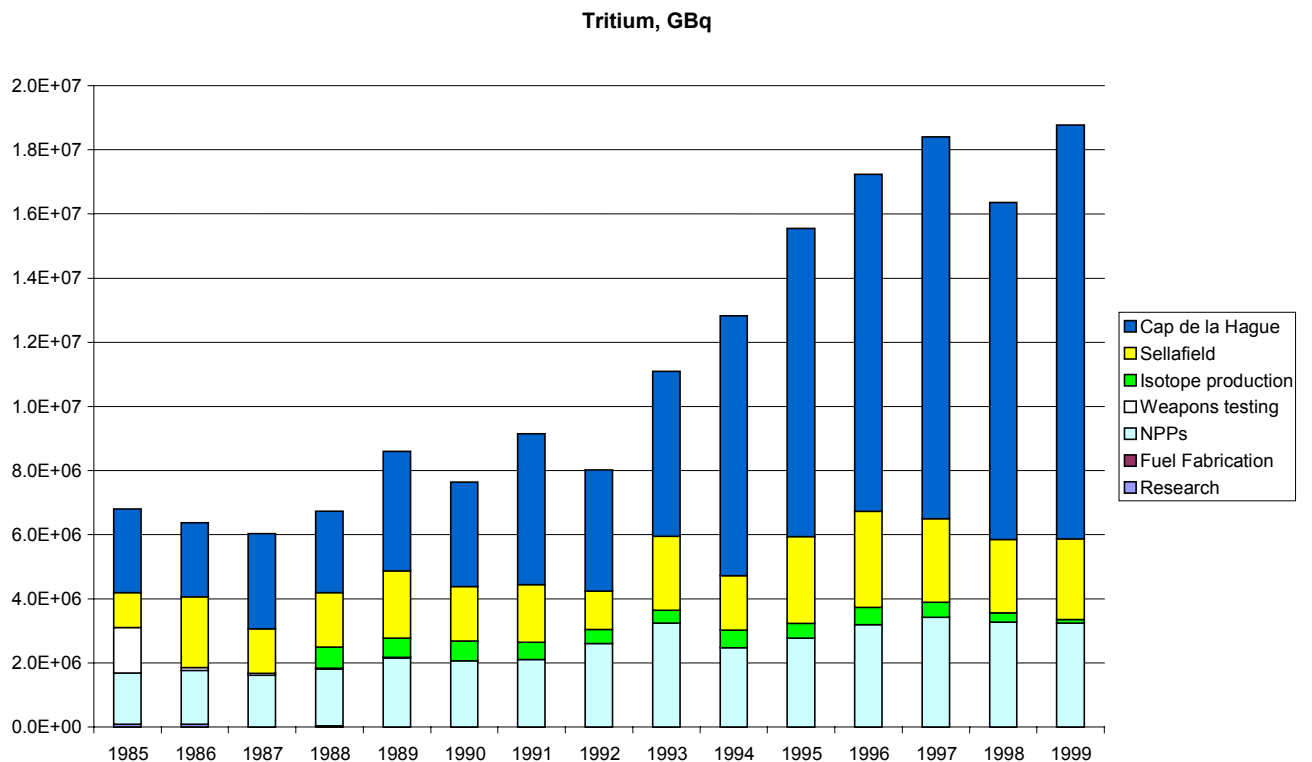
**Figure 5. Trends in overall input of  $\alpha$  activity into the OSPAR area**



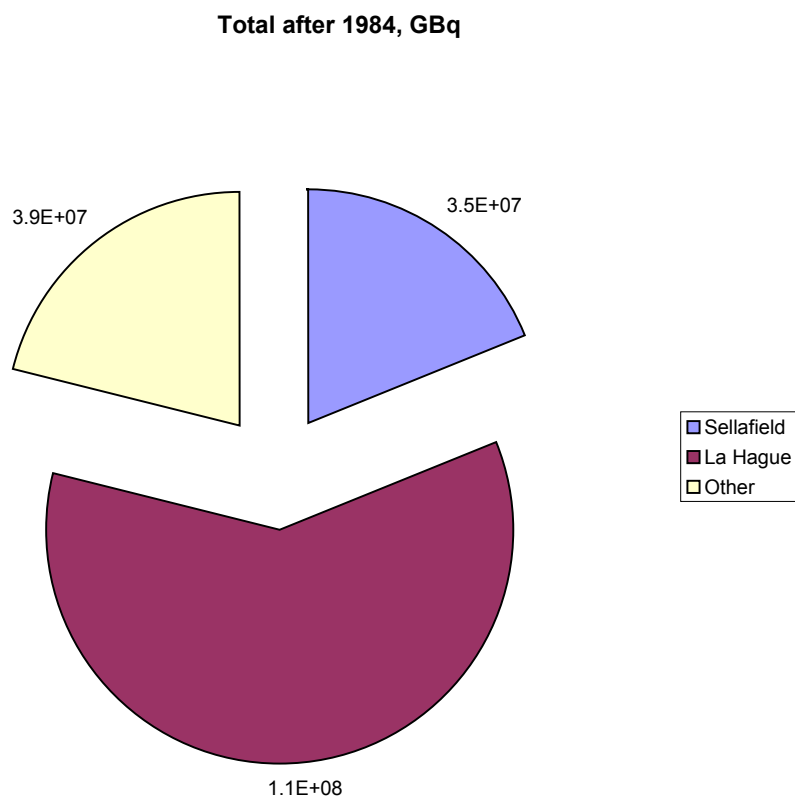
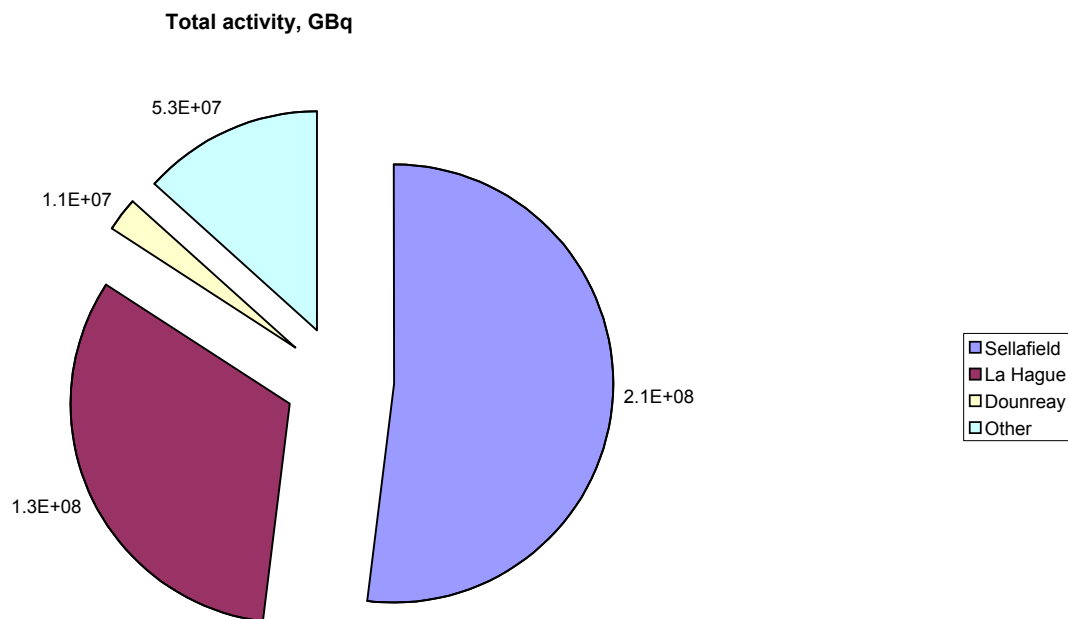
**Figure 6. Recent trends in overall input of  $\alpha$  activity into the OSPAR area**



**Figure 7. Trends in overall input of tritium into the OSPAR area**

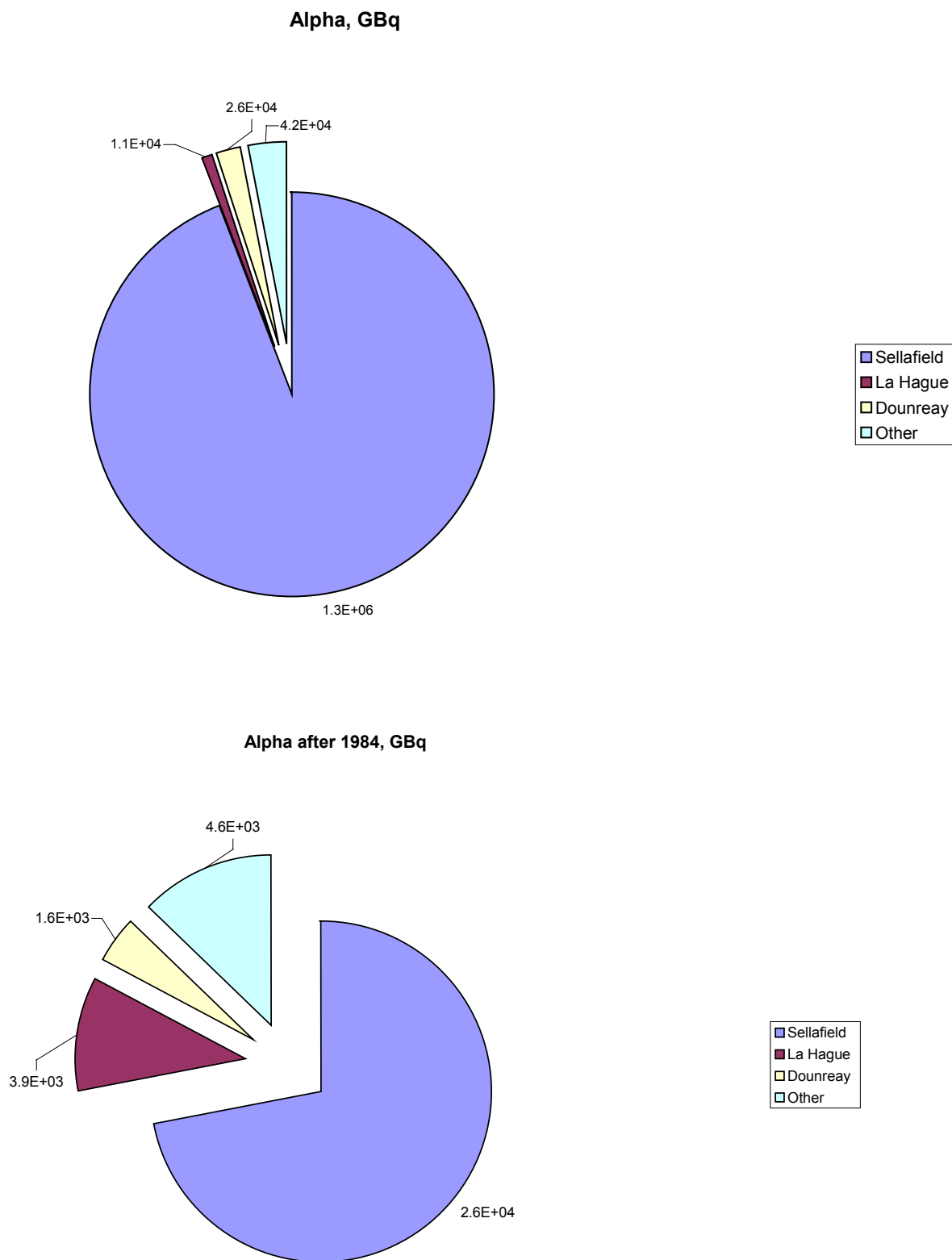


**Figure 8. Recent trends in overall input of tritium into the OSPAR area**

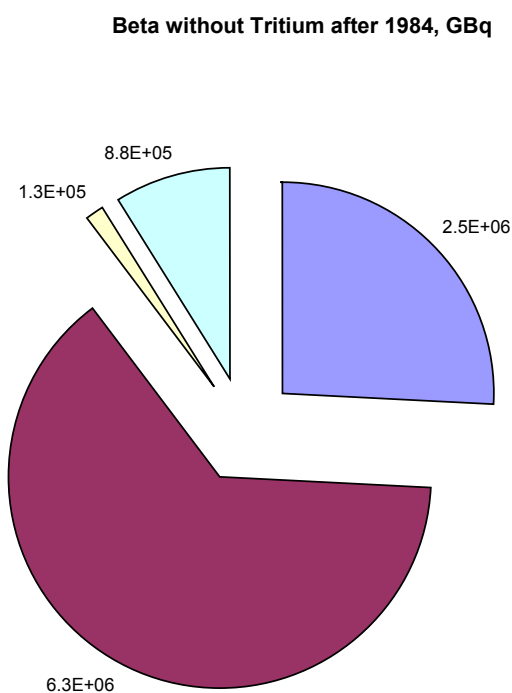
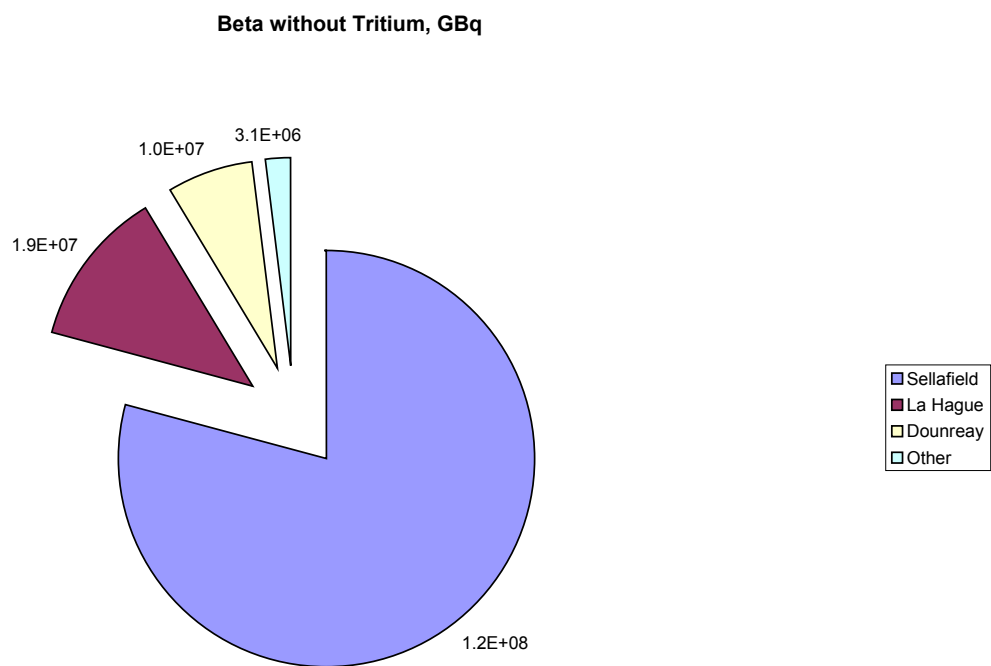


**Figure 9. Overall activity discharge into the North-East Atlantic from the nuclear facilities.**

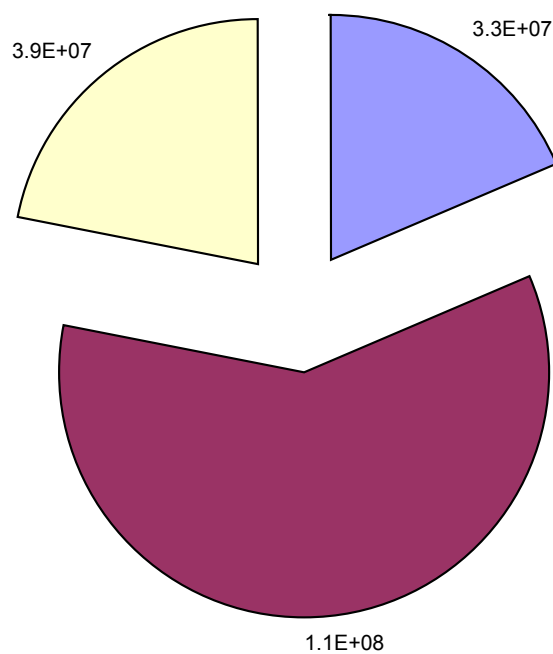
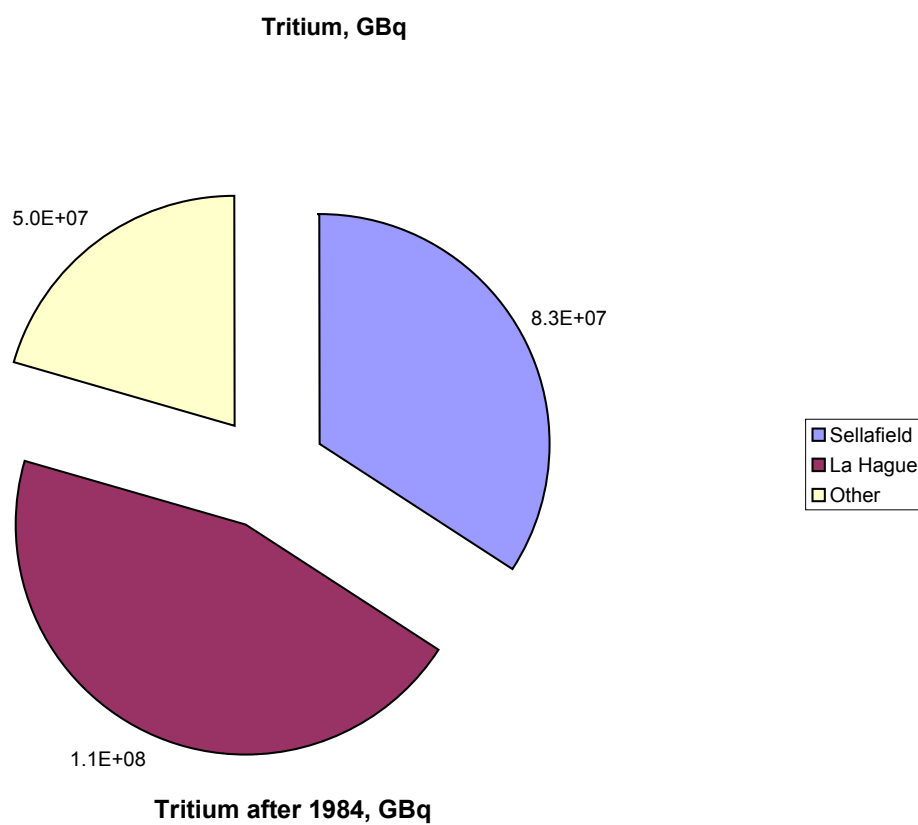




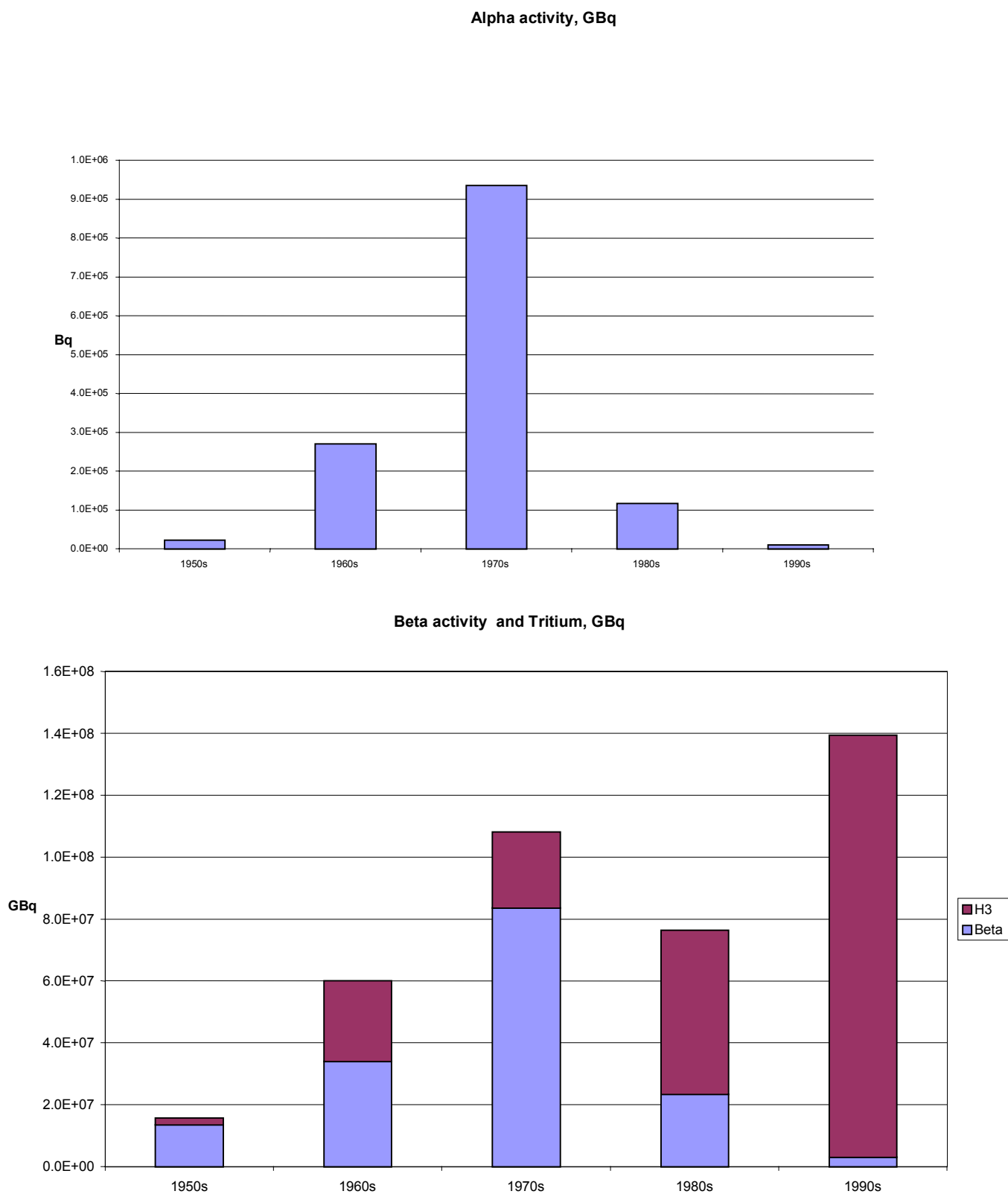
**Figure 10. Discharge of  $\alpha$ -emitters into the North-East Atlantic from the nuclear facilities.**



**Figure 11. Discharge of  $\beta$ -emitters, excluding tritium into the North-East Atlantic from the nuclear facilities.**

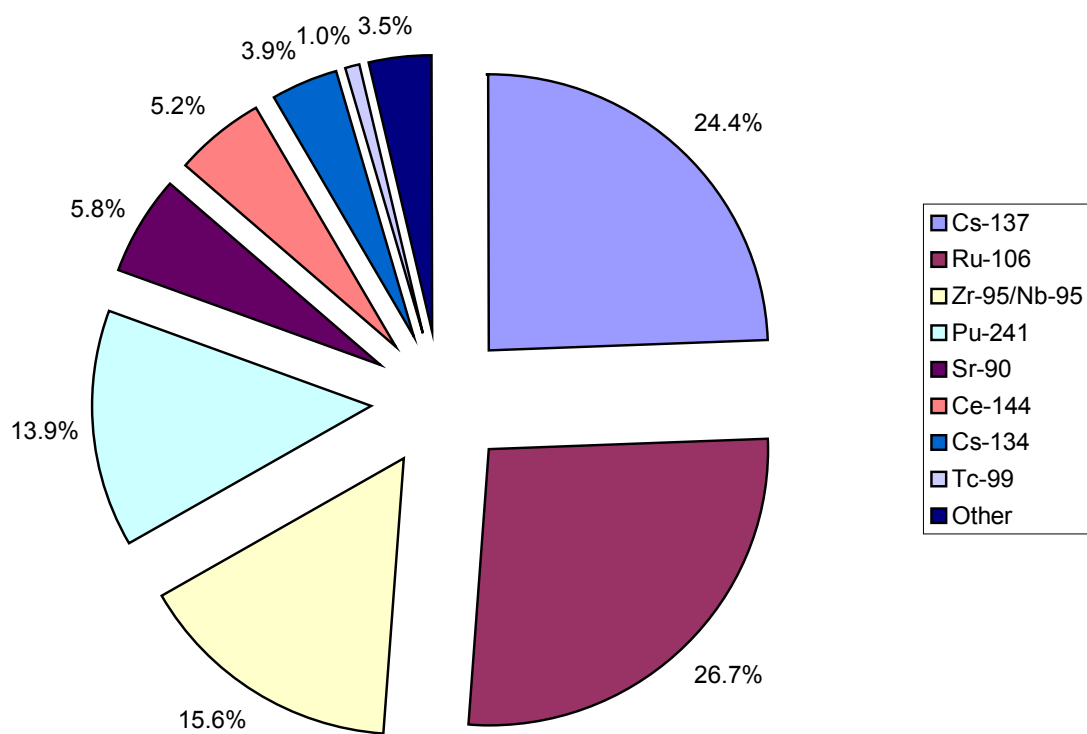


**Figure 12. Discharge of tritium into the North-East Atlantic from the nuclear facilities.**

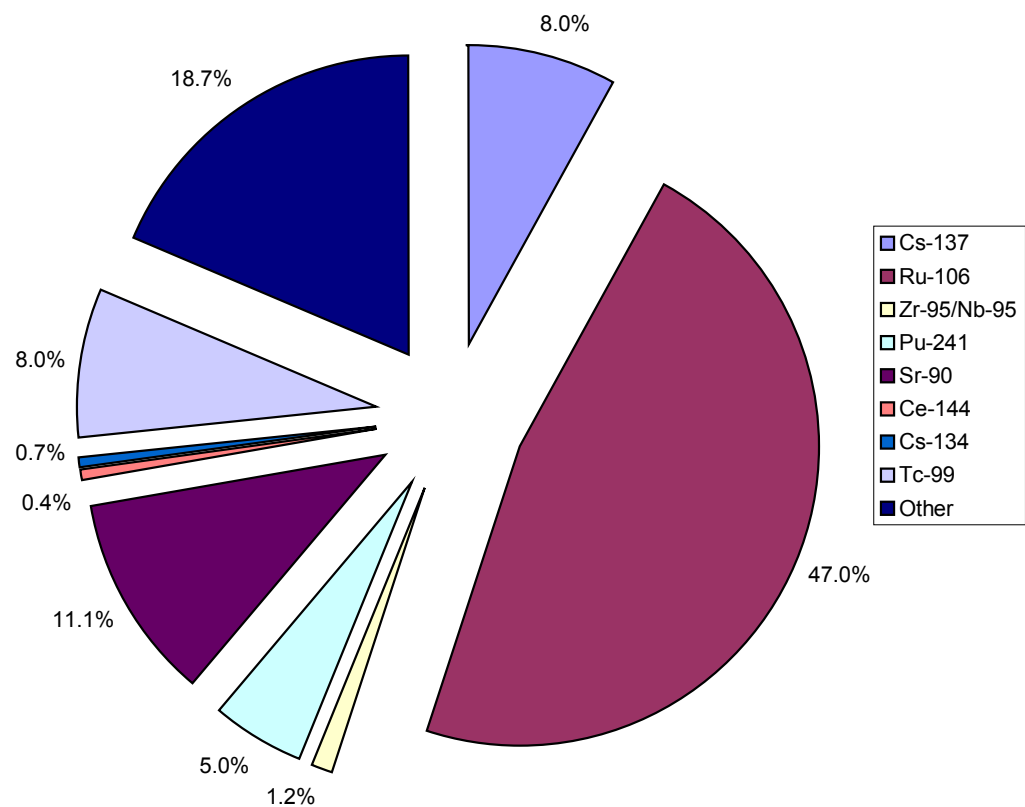


**Figure 13. Trends in discharges of  $\alpha$  and  $\beta$  activity into North East Atlantic**

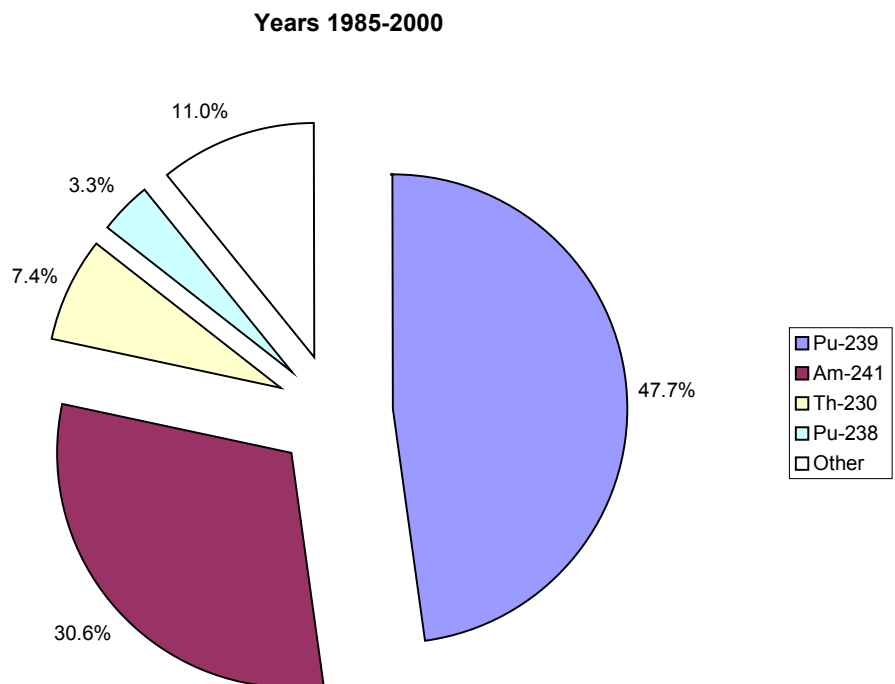
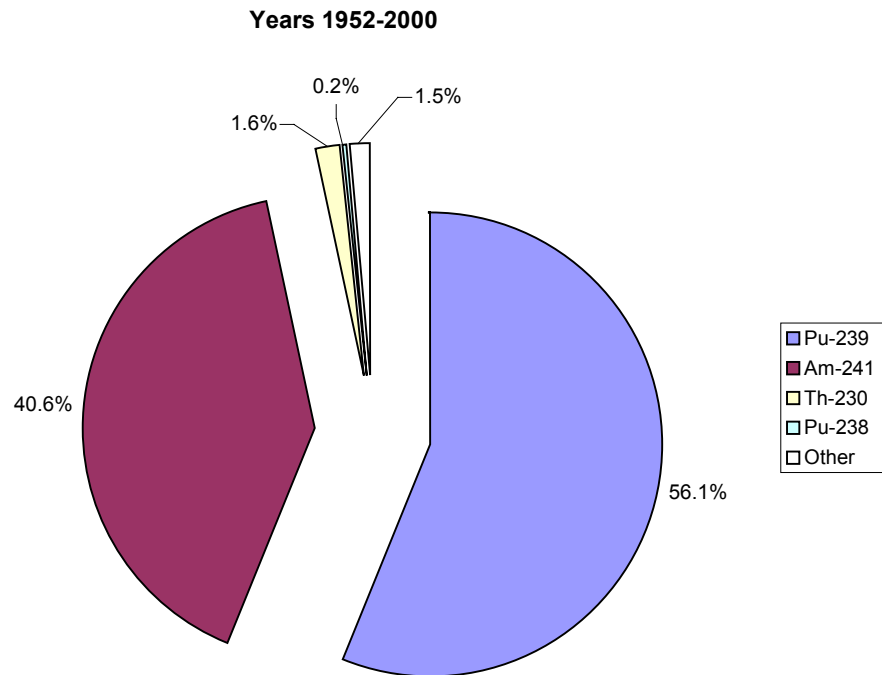
**Years 1952-2000**



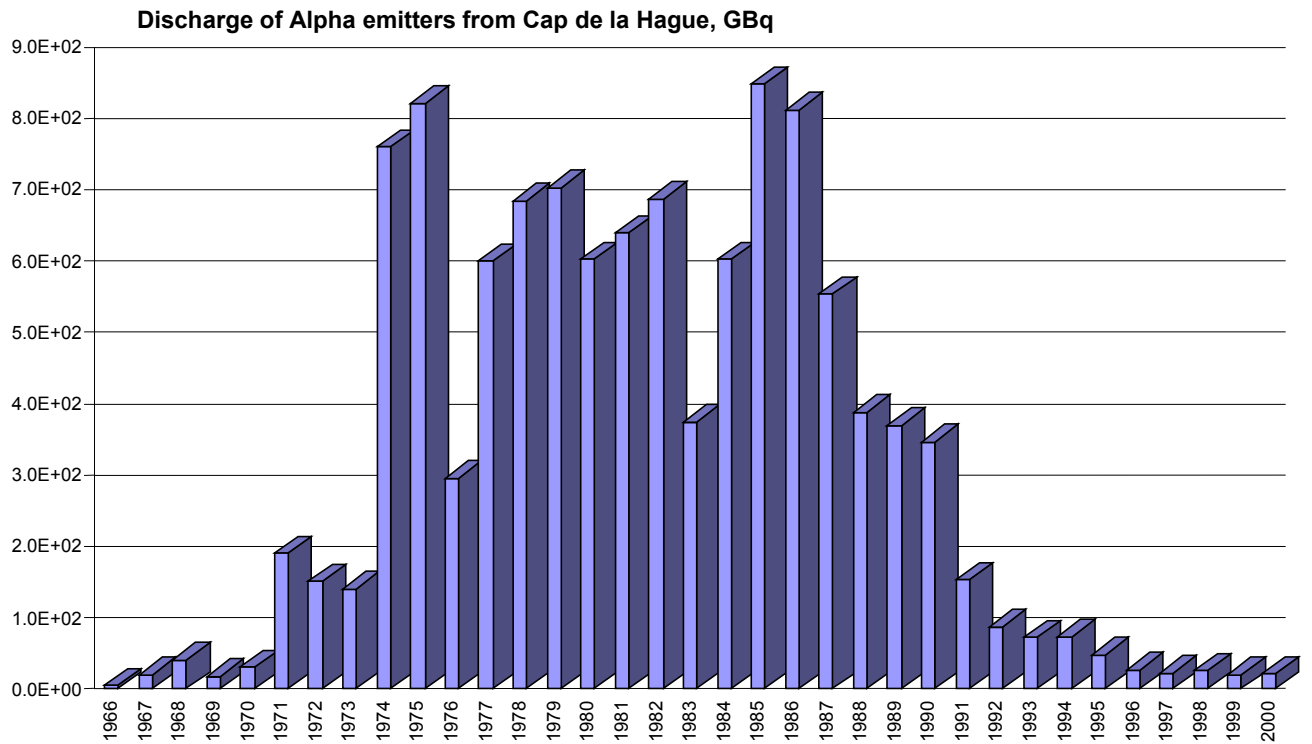
**Years 1985-2000**



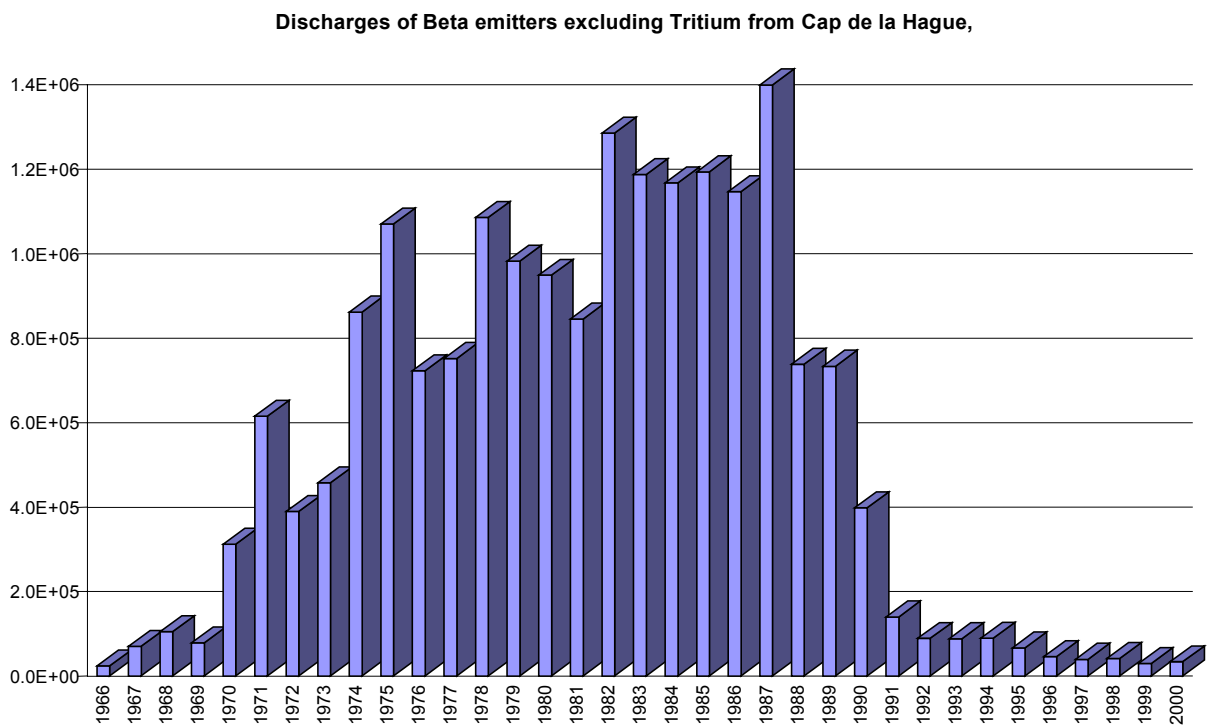
**Figure 14. Total liquid discharges of  $\beta$ -emitters, excluding tritium, from all nuclear sites (% by radionuclide)**



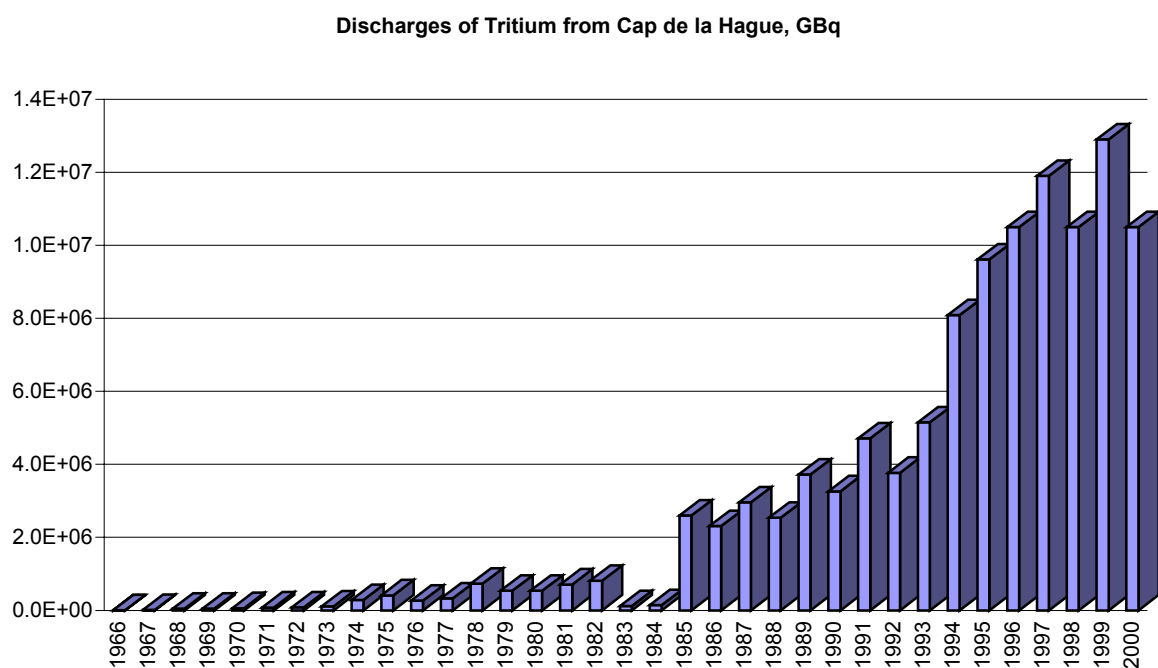
**Figure 15. Total liquid discharges of  $\alpha$ -emitters from all nuclear sites (% by radionuclide)**



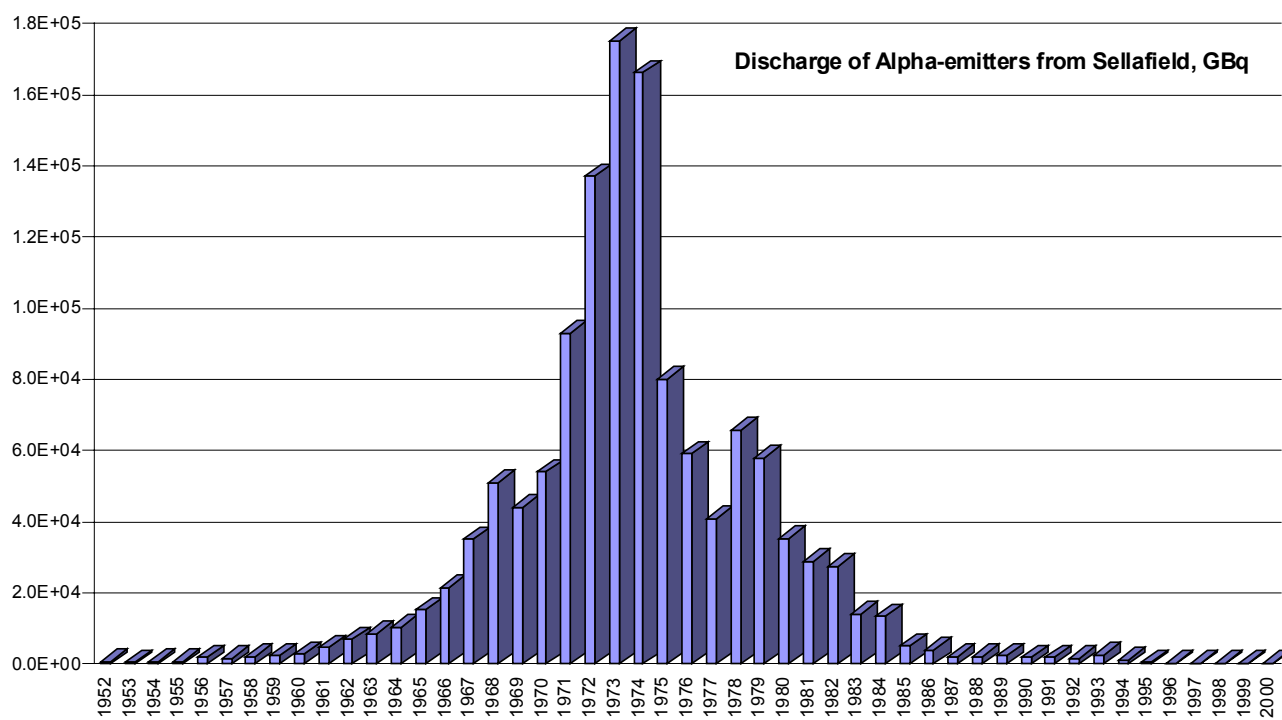
**Figure 16. Annual releases of Alpha emitters in liquid discharges from Cap de la Hague**



**Figure 17. Annual releases of Beta emitters in liquid discharges from Cap de la Hague**

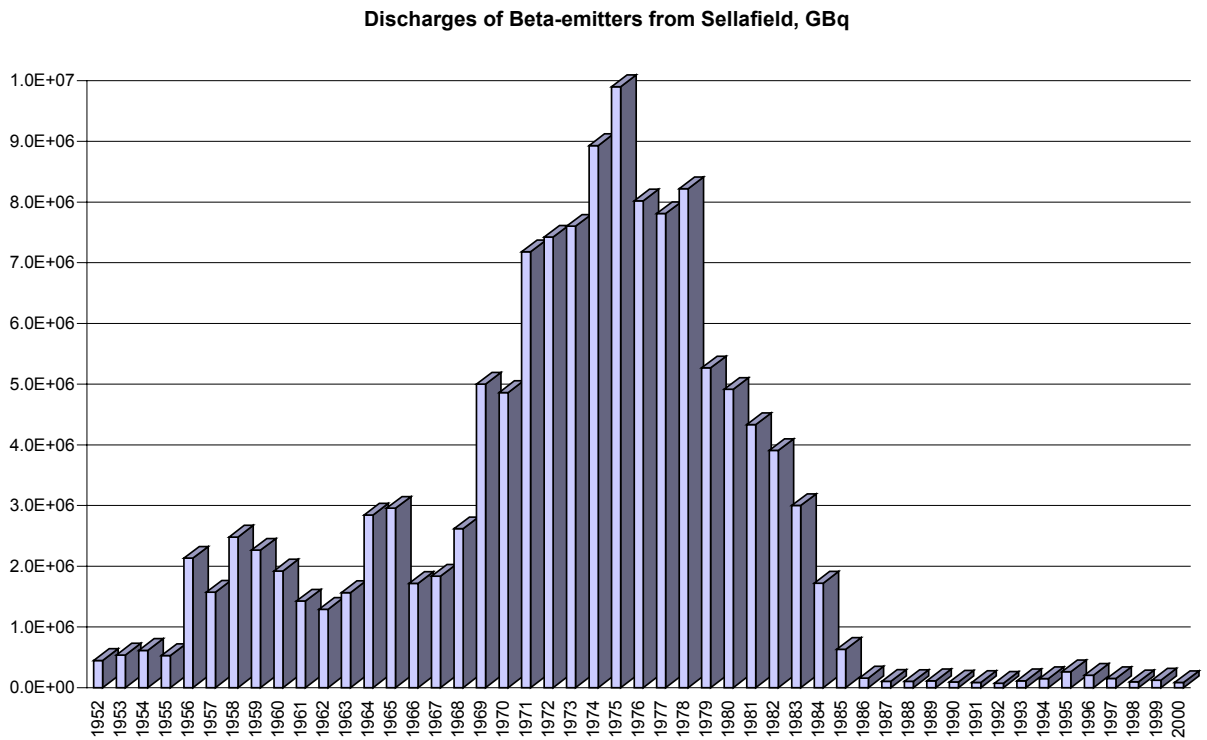


**Figure 18. Annual releases of tritium in liquid discharges from Cap de la Hague**

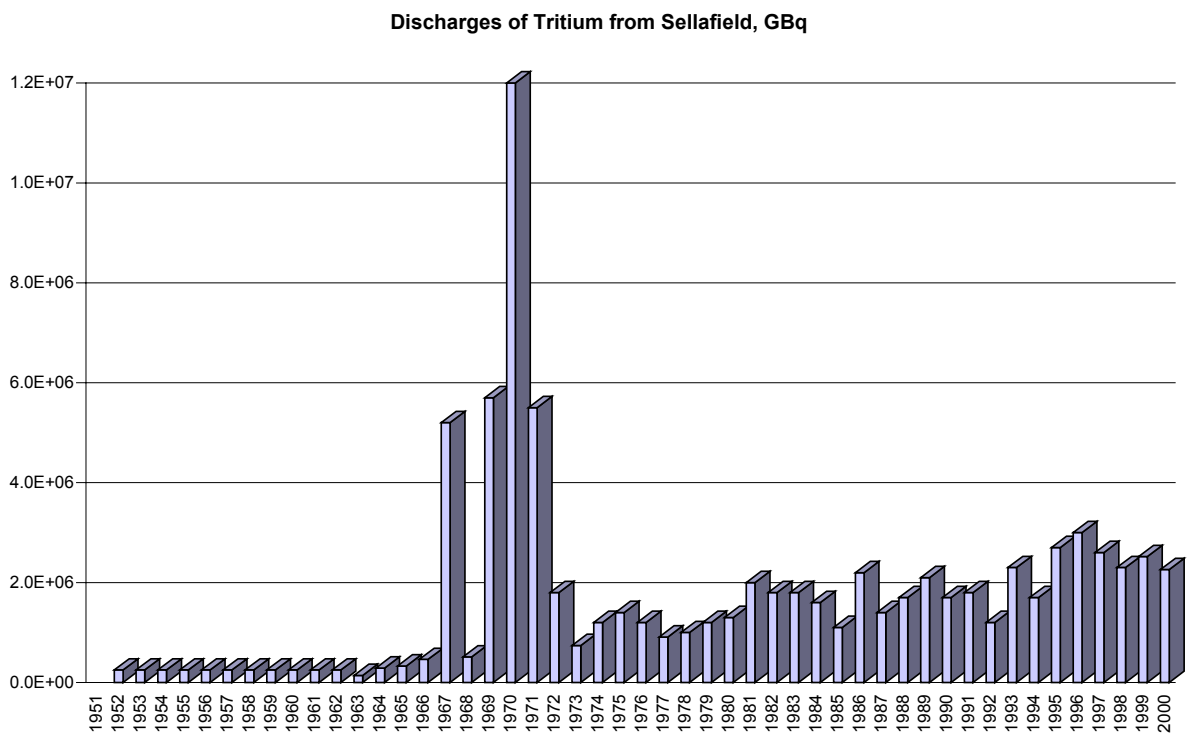


**Figure 19. Annual releases of Alpha emitters in liquid discharges from Sellafield**



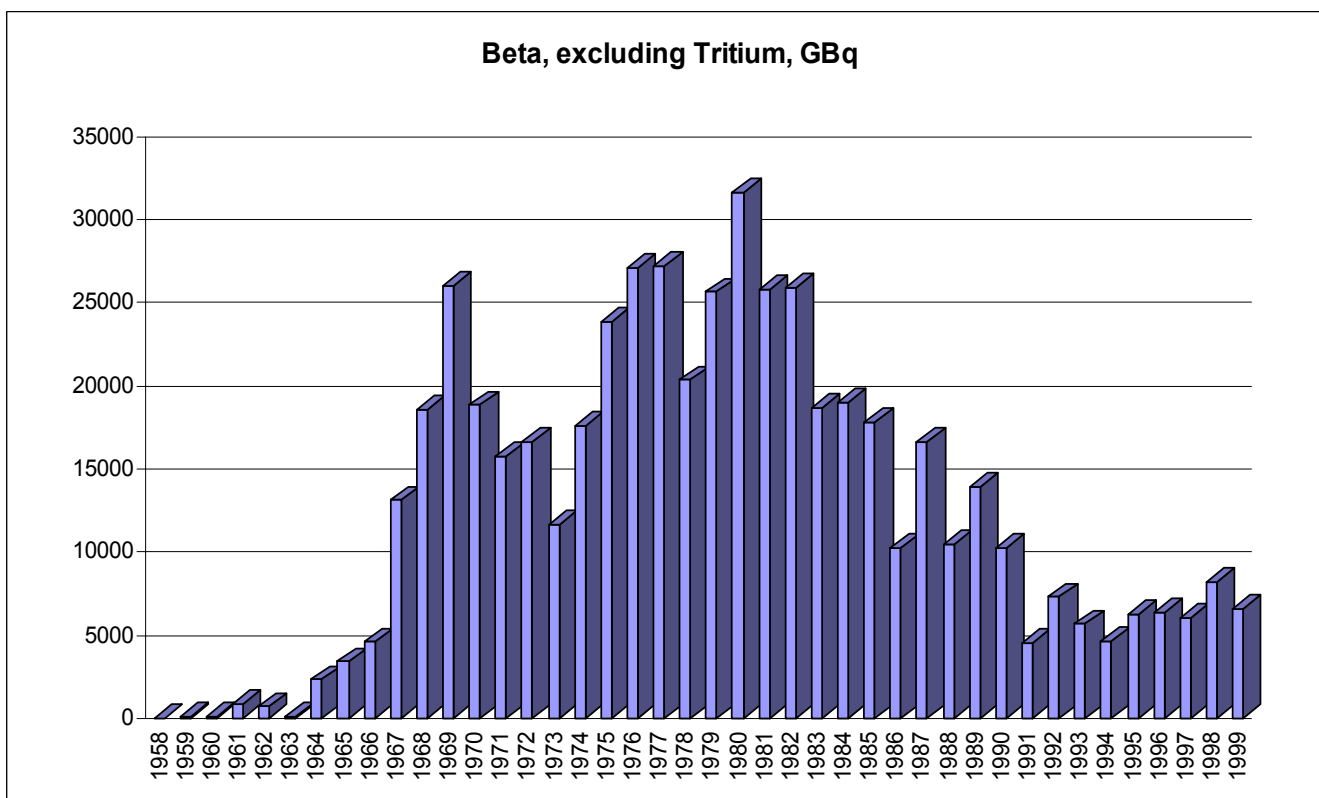


**Figure 20. Annual releases of Beta emitters in liquid discharges from Sellafield**

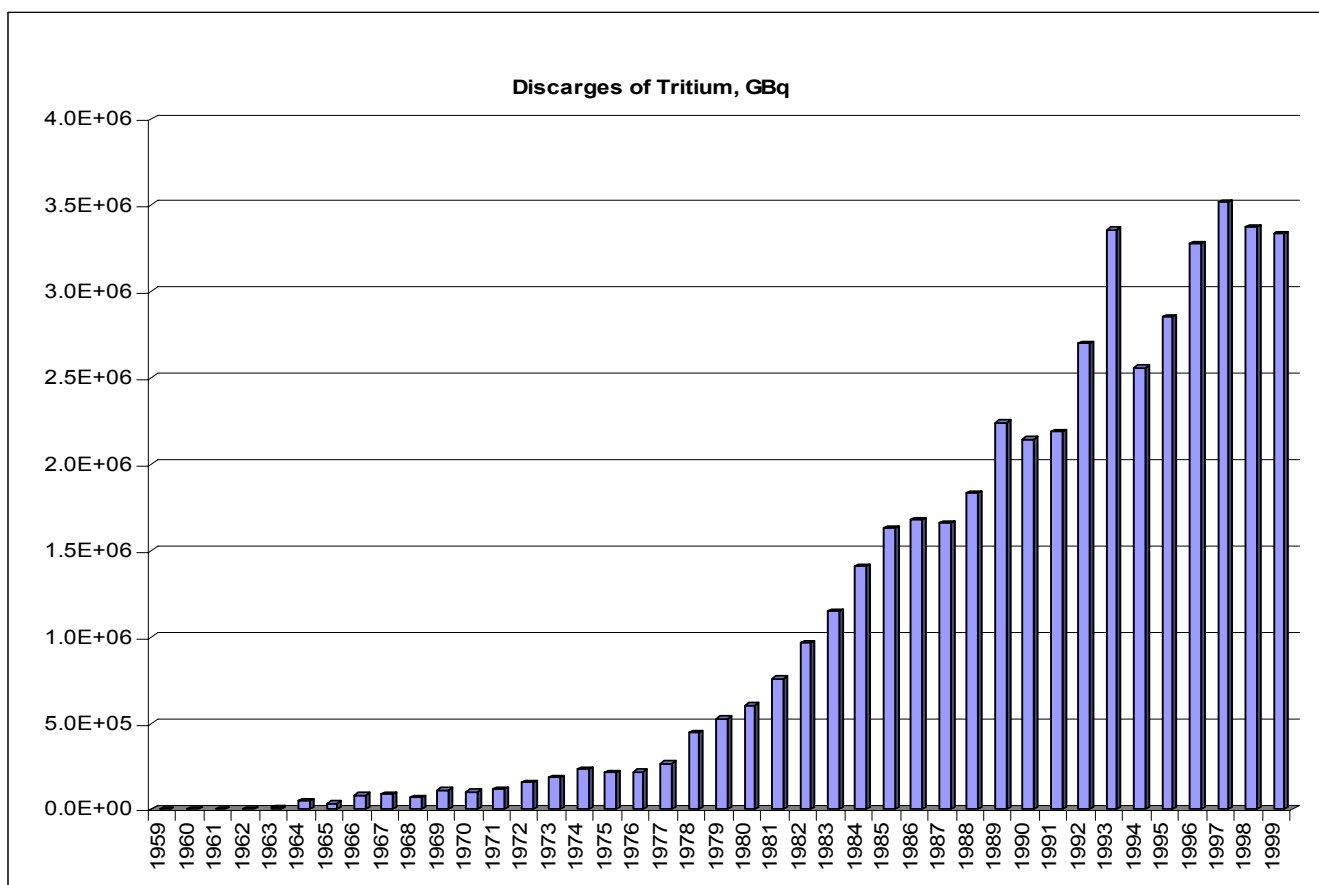


**Figure 21. Annual releases of tritium in liquid discharges from Sellafield**

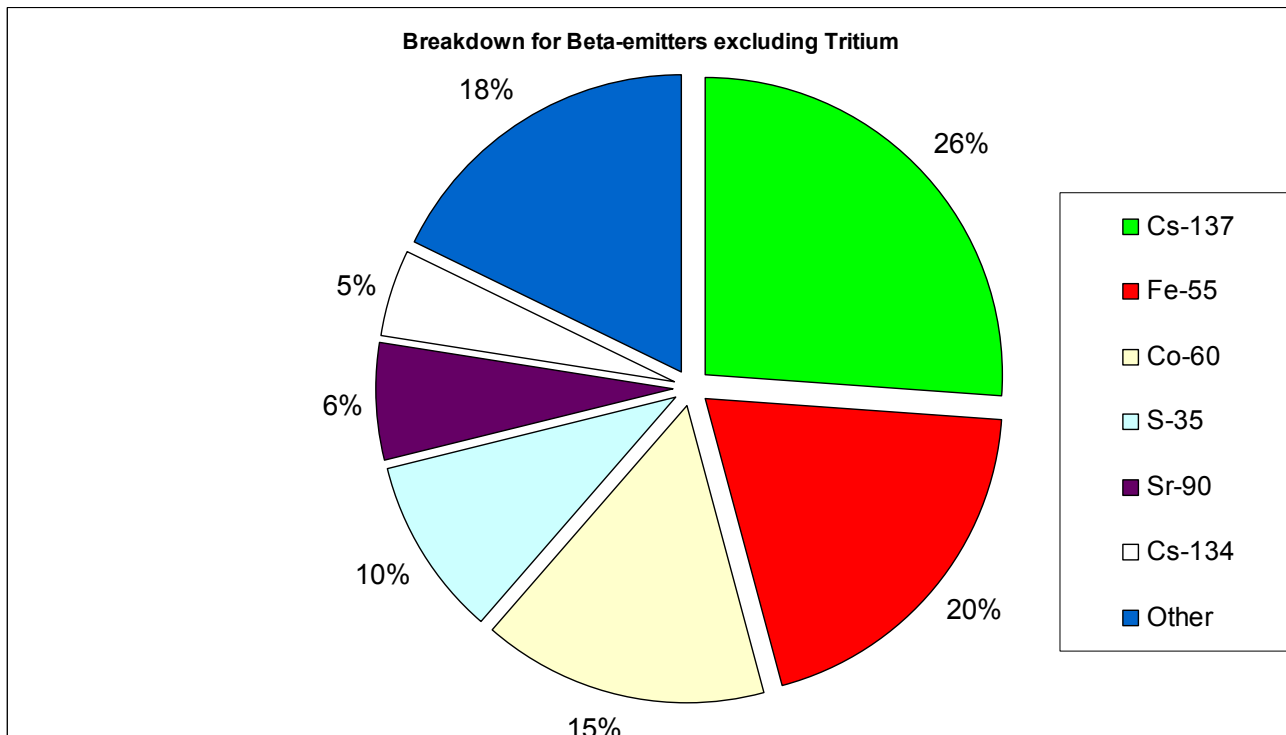




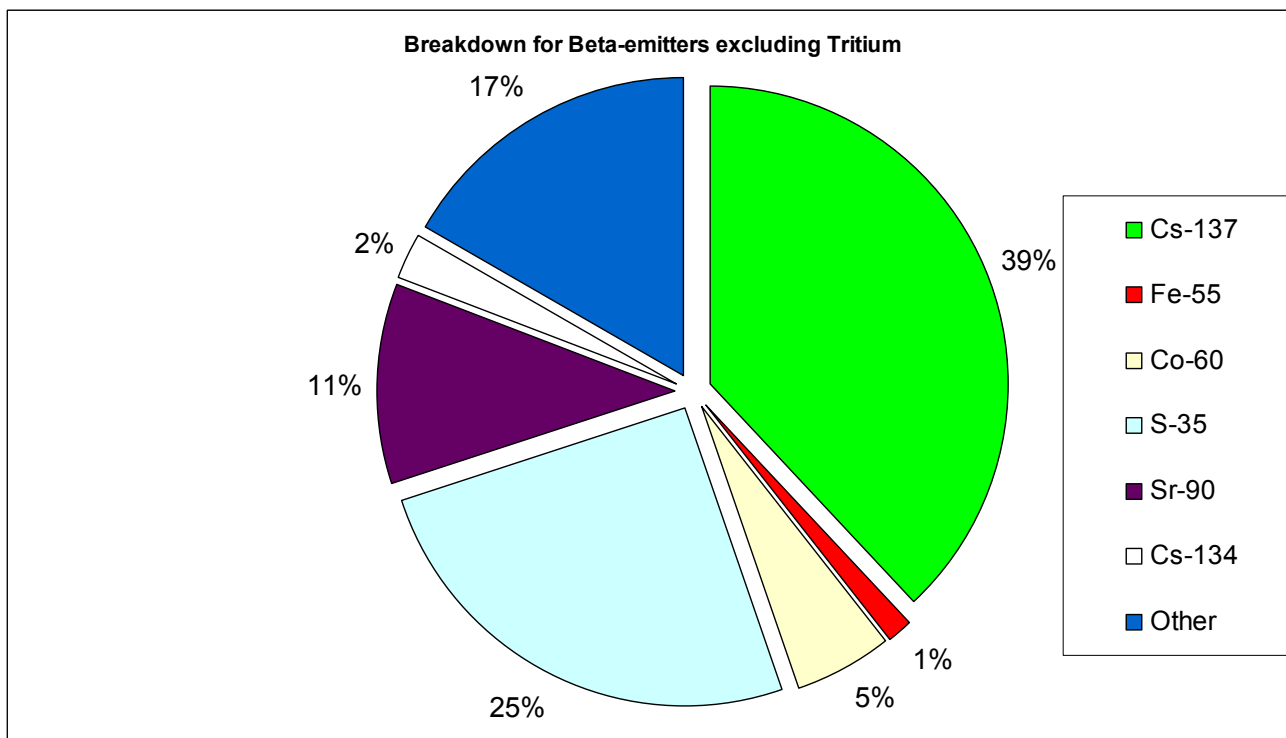
**Figure 23. Annual release of Beta emitters in liquid discharges from nuclear power plants of Contracting Parties to the OSPAR Convention**



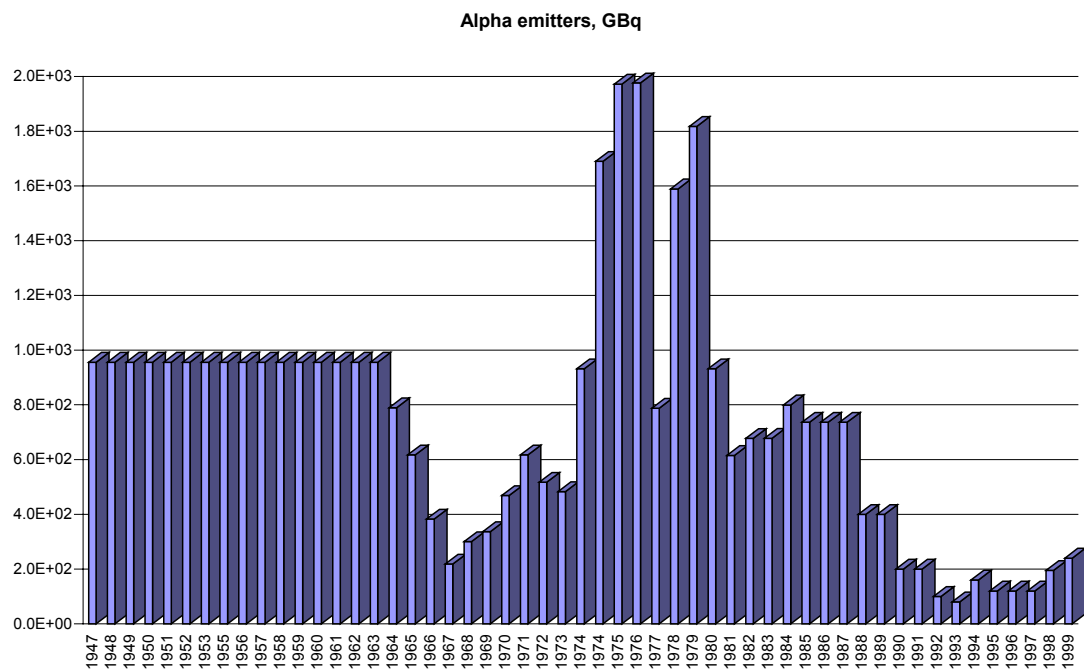
**Figure 24. Annual release of tritium in liquid discharges from nuclear power plants of Contracting Parties to the OSPAR Convention**



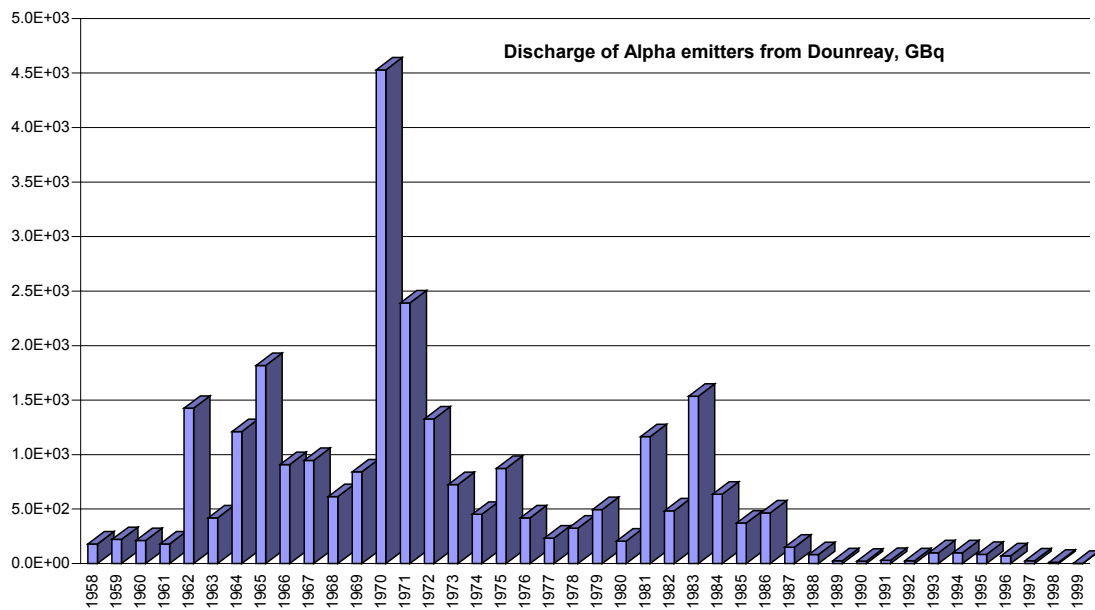
**Figure 25. Total liquid discharges of  $\beta$ -emitters from NPP sites (% by radionuclide)**



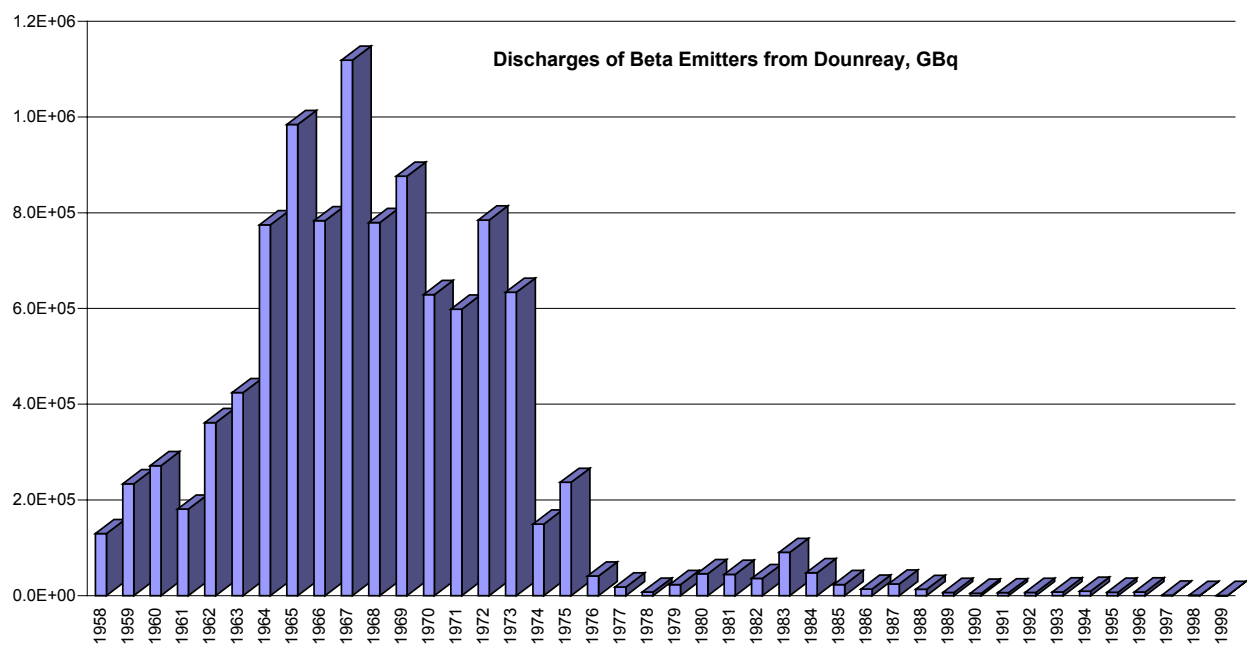
**Figure 26. Total liquid discharges of  $\beta$ -emitters from NPP since 1985 (% by radionuclide)**



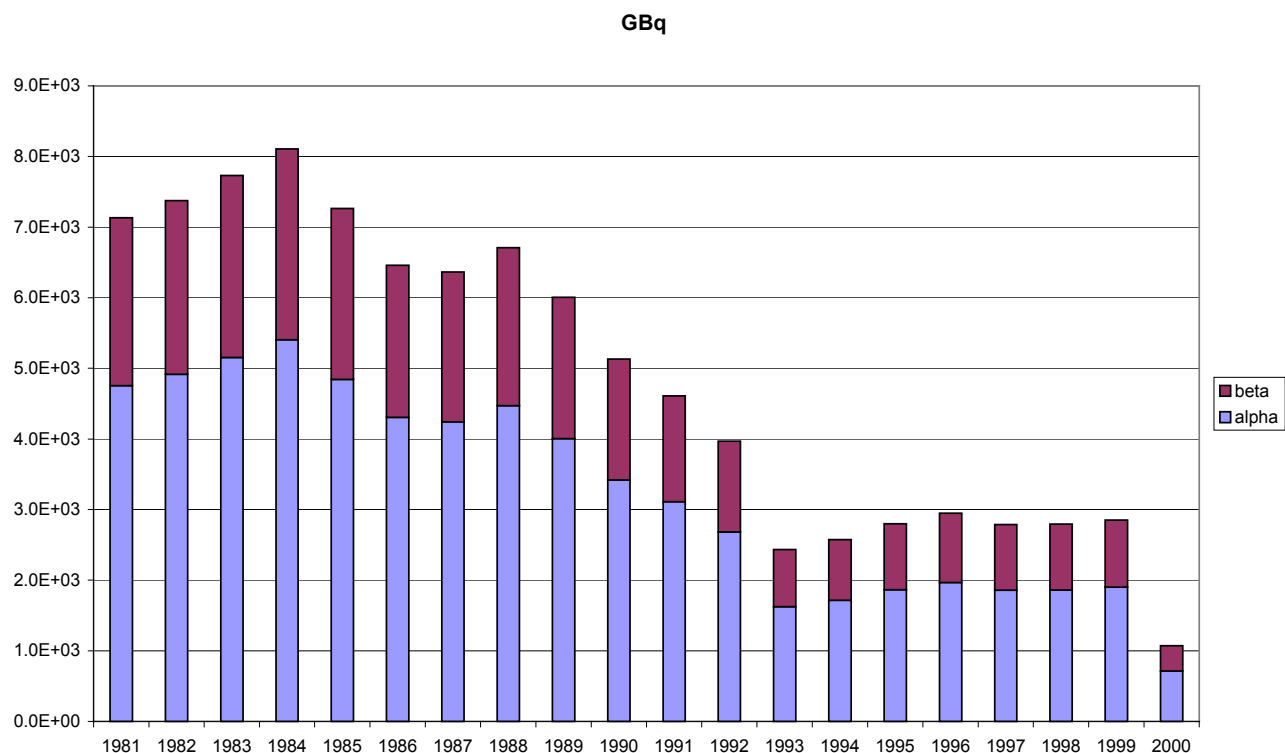
**Figure 27. Annual releases of Alpha emitters in liquid discharges from Springfields**



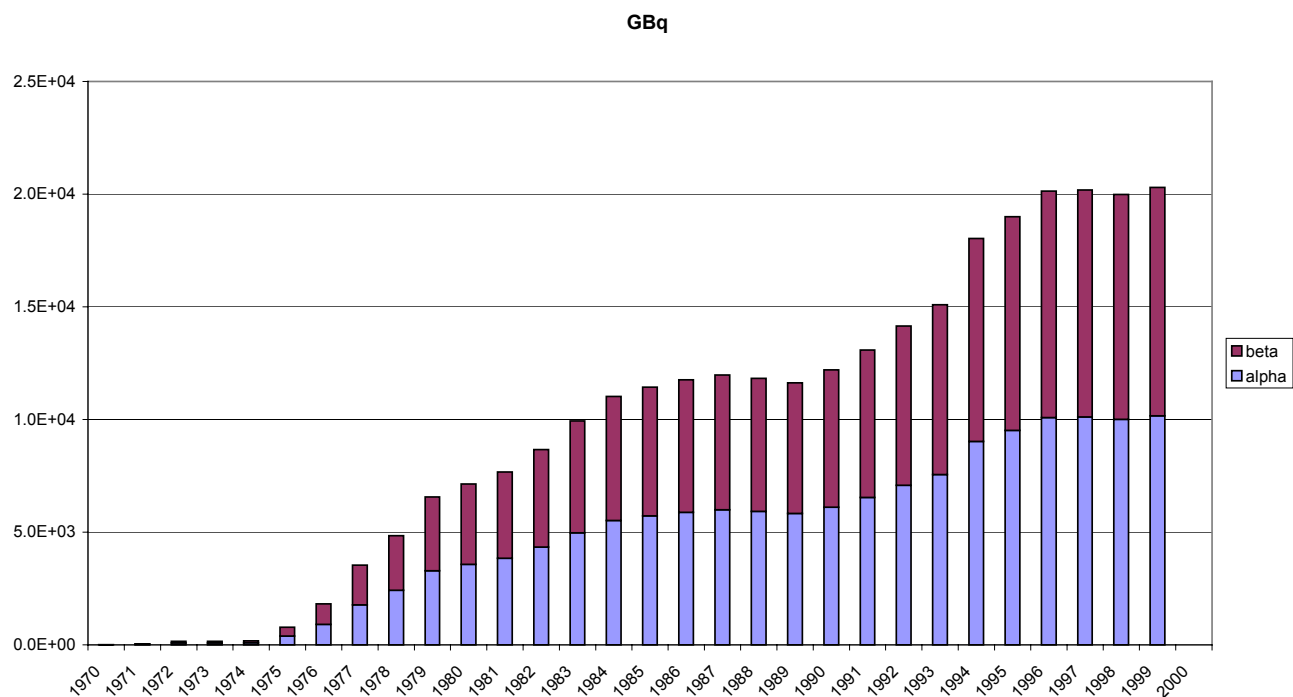
**Figure 28. Annual releases of Alpha emitters in liquid discharges from Dounreay**



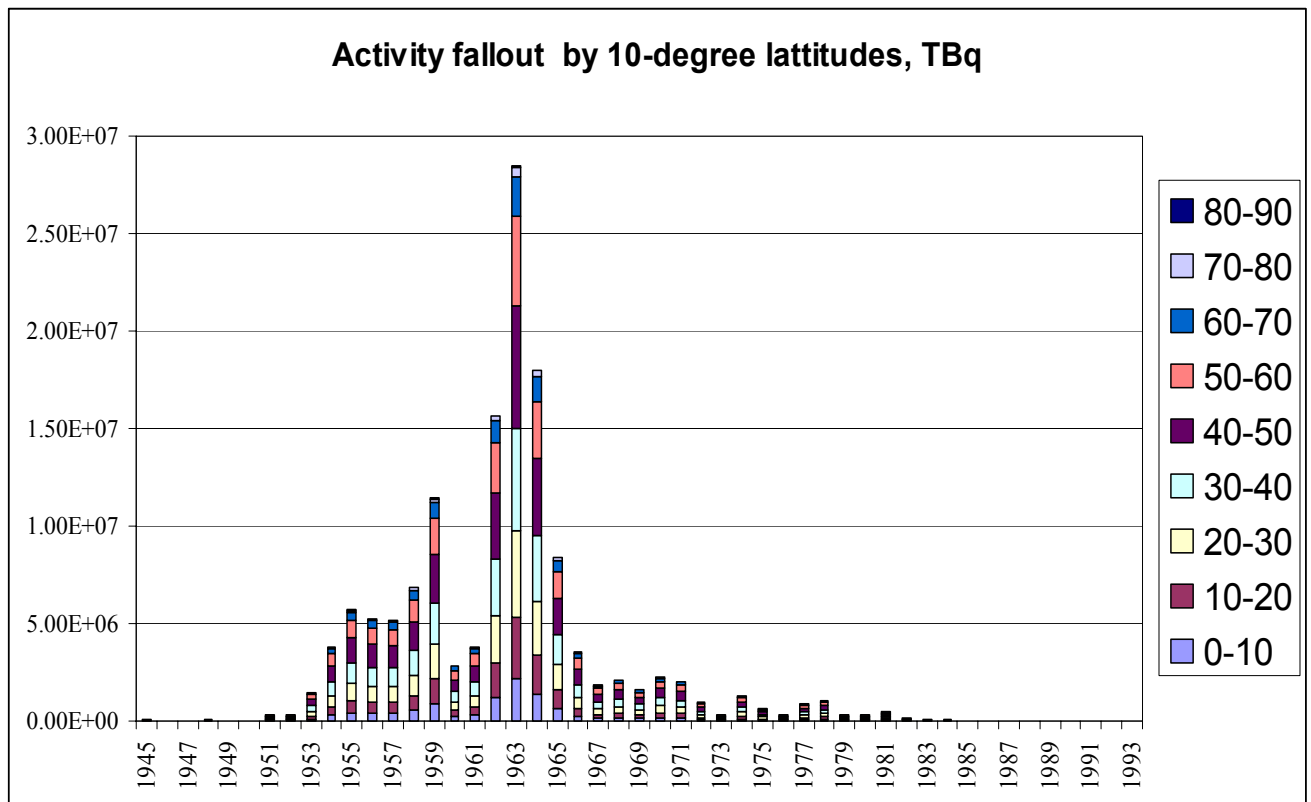
**Figure 29. Annual releases of Beta emitters in liquid discharges from Dounreay**



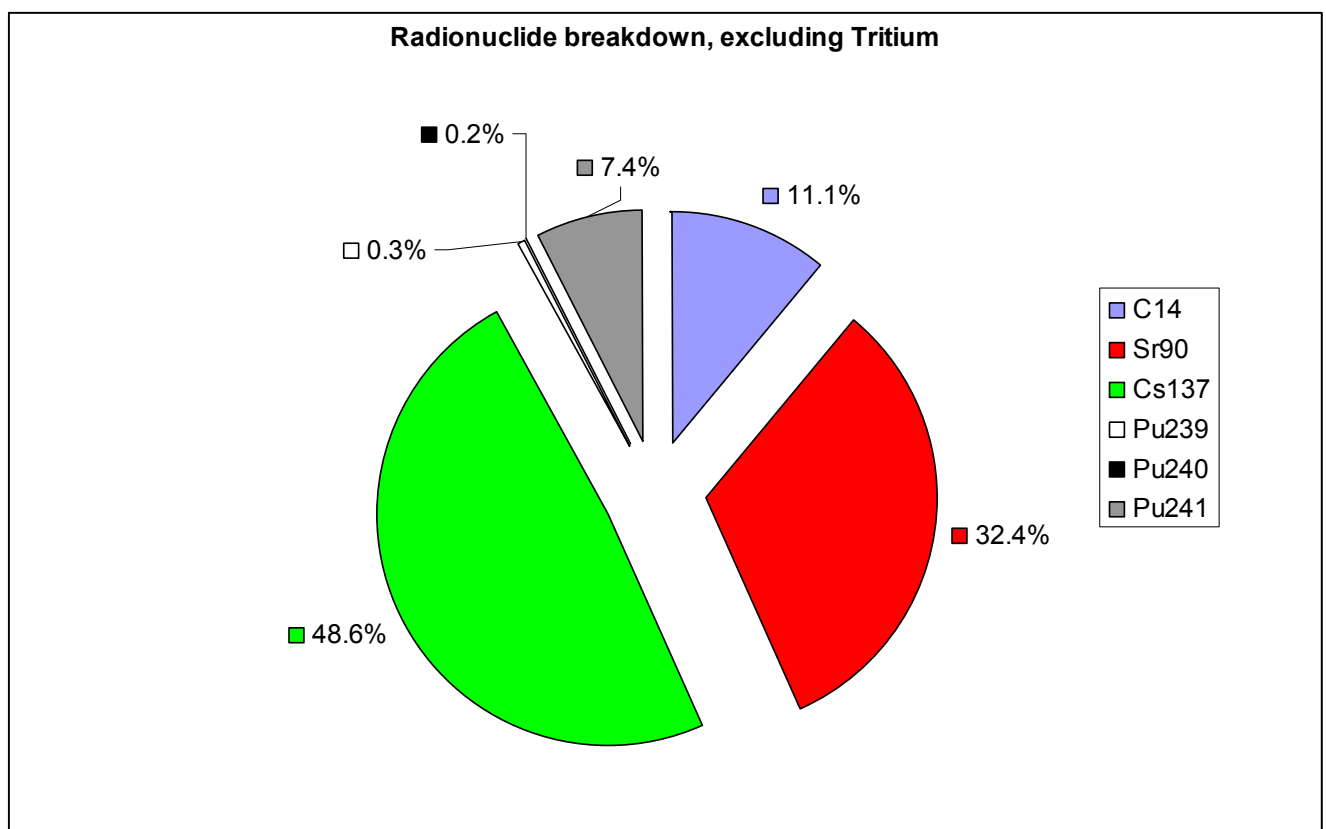
**Figure 30. Annual activity releases into the OSPAR area due to discharges of phosphogypsum from the fertiliser industry (data from 1981 onward only)**



**Figure 31. Annual activity releases into the OSPAR area due to discharges from off-shore oil and gas production**

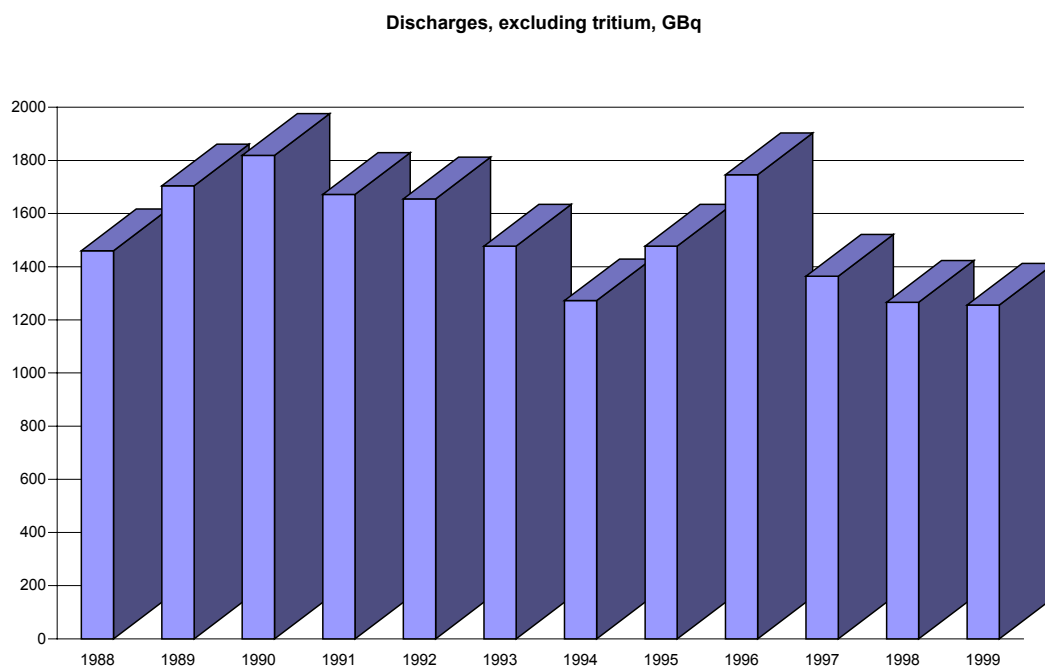


**Figure 32. Nuclear testing fallout in Northern Hemisphere**

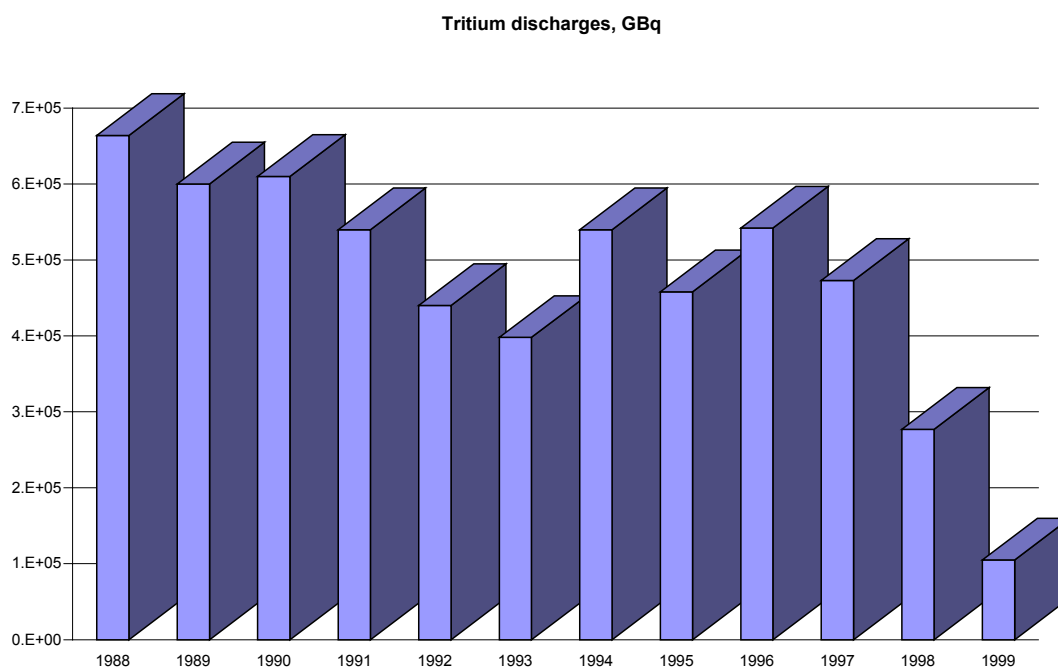


**Figure 33. Atmospheric fallout from nuclear testing (% by radionuclide)**

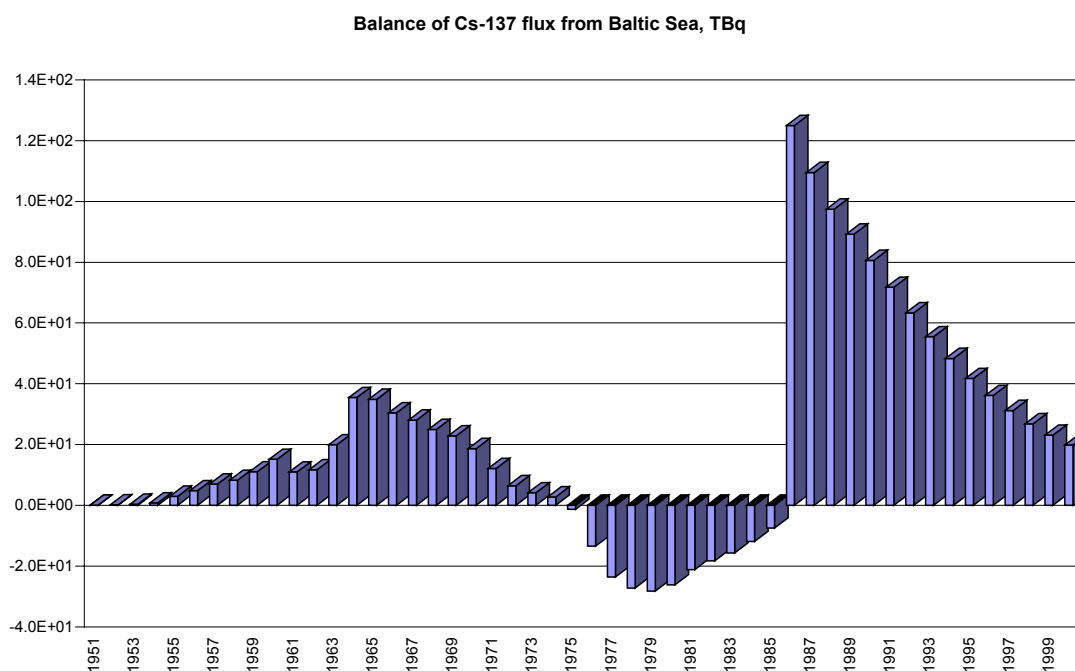




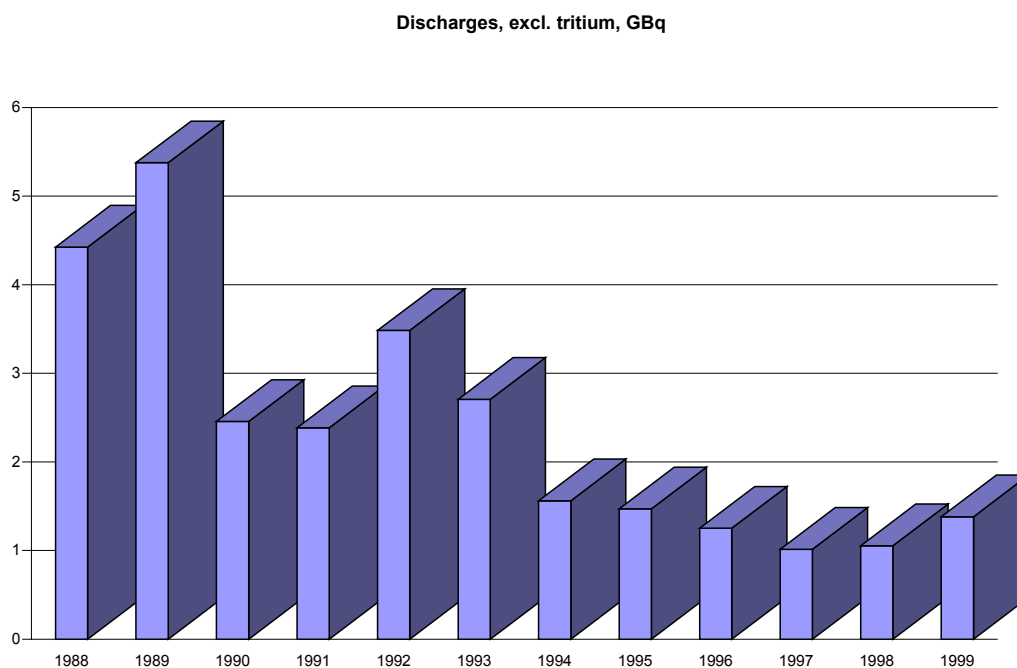
**Figure 34. Annual liquid radioactive discharges from isotope manufacturing facilities in the UK (excluding tritium)**



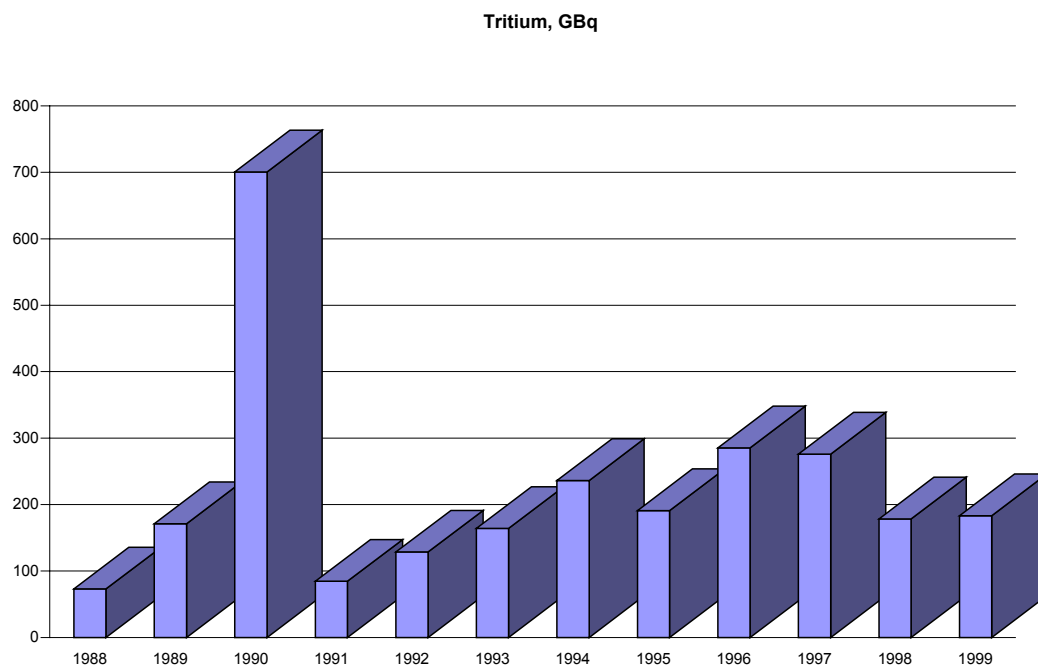
**Figure 35. Discharges of tritium from isotope manufacturing facilities in the UK**



**Figure 36. Flux of  $^{137}\text{Cs}$  from the Baltic Sea into Skagerrak**



**Figure 37. Annual liquid radioactive discharges from military establishments in the UK (excluding tritium)**



**Figure 38. Annual tritium discharges from military establishments in the UK**



## Appendix A - Annotation of the MARINA II discharge database

Data for the following Nuclear Power Stations is taken directly from the MARINA I study, covering the period from grid connection to 1984 inclusive.

Reactor	Type	Date connected
BELGIUM		
DOEL-1	PWR	28/08/74
DOEL-2	PWR	21/08/75
DOEL-3	PWR	23/06/82
DOEL-4	PWR	08/04/85
TIHANGE-1	PWR	07/03/75
TIHANGE-2	PWR	13/10/82
TIHANGE-3	PWR	15/06/85
FRANCE		
BLAYAIS-1	PWR	12/06/81
BLAYAIS-2	PWR	17/07/82
BLAYAIS-3	PWR	17/08/83
BLAYAIS-4	PWR	16/05/83
CHINON-A1	GCR	14/06/63
CHINON-A2	GCR	24/02/65
CHINON-A3	GCR	04/08/66
CHINON-B-1	PWR	30/11/82
CHINON-B-2	PWR	29/11/83
CHINON-B-3	PWR	20/10/86
CHINON-B-4	PWR	14/11/87
CHOOZ-A(ARDENNES)	PWR	03/04/67
CHOOZ-B-1	PWR	30/08/96
CHOOZ-B-2	PWR	09/04/97
DAMPIERRE-1	PWR	23/03/80
DAMPIERRE-2	PWR	10/12/80
DAMPIERRE-3	PWR	30/01/81
DAMPIERRE-4	PWR	18/08/81
FESSENHEIM-1	PWR	06/04/77
FESSENHEIM-2	PWR	07/10/77
GRAVELINES-1	PWR	13/03/80
GRAVELINES-2	PWR	26/08/80
GRAVELINES-3	PWR	12/12/80
GRAVELINES-4	PWR	14/06/81
GRAVELINES-5	PWR	28/08/84
GRAVELINES-6	PWR	01/08/85
PALUEL-1	PWR	22/06/84
PALUEL-2	PWR	14/09/84
PALUEL-3	PWR	30/09/85
PALUEL-4	PWR	11/04/86
PENLY-1	PWR	04/05/90
PENLY-2	PWR	01/02/92

Reactor	Type	Date connected
ST. LAURENT-A1	GCR	14/03/69
ST. LAURENT-A2	GCR	09/08/71
ST. LAURENT-B-1	PWR	21/01/81
ST. LAURENT-B-2	PWR	01/06/81
GERMANY		
BIBLIS-A (KWB A)	PWR	25/08/74
BIBLIS-B (KWB B)	PWR	06/04/76
BRUNSBUEITTEL (KKB)	BWR	13/07/76
GRAFENRHEINFELD	PWR	21/12/81
GREIFSWALD-1(KGR 1)	WWER	17/12/73
GREIFSWALD-2 (KGR 2)	WWER	23/12/74
GREIFSWALD-3 (KGR 3)	WWER	24/10/77
GREIFSWALD-4 (KGR 4)	WWER	03/09/79
GREIFSWALD-5 (KGR 5)	WWER	24/04/89
GROHNDE (KWG)	PWR	04/09/84
GUNDREMMINGEN-A	BWR	01/12/66
GUNDREMMINGEN-B	BWR	16/03/84
GUNDREMMINGEN-C	BWR	02/11/84
KRUEMMEL (KKK)	BWR	28/09/83
LINGEN (KWL)	BWR	01/07/68
MUELHEIM-KAERLICH	PWR	14/03/86
MZFR	PHWR	09/03/66
NECKARWESTHEIM-1	PWR	03/06/76
NECKARWESTHEIM-2	PWR	03/01/89
OBRIGHEIM (KWO)	PWR	29/10/68
PHILIPPSBURG-1 (KKP 1)	BWR	07/05/79
PHILIPPSBURG-2 (KKP 2)	PWR	17/12/84
RHEINSBERG (KKR)	PWR	06/05/66
STADE (KKS)	PWR	29/01/72
UNTERWESER (KKU)	PWR	29/09/78
KAHL	BWR	17/06/61
WUERGASSEN (KWW)	BWR	18/12/71
NETHERLANDS		
BORSSELE	PWR	04/07/73
DODEWAARD	BWR	18/10/68
SPAIN		
ALMARAZ-1	PWR	01/05/81
ALMARAZ-2	PWR	08/10/83
COFRENTES	BWR	14/10/84
JOSE CABRERA-1	PWR	14/07/68
SANTA MARIA DE	BWR	02/03/71
TRILLO-1	PWR	23/05/88
VANDELLOS-1	GCR	06/05/72
VANDELLOS-2	PWR	12/12/87
SWEDEN		
BARSEBECK-1	BWR	15/05/75
BARSEBECK-2	BWR	21/03/77

Reactor	Type	Date connected
FORSMARK-1	BWR	06/06/80
FORSMARK-2	BWR	26/01/81
FORSMARK-3	BWR	05/03/85
RINGHALS-1	BWR	14/10/74
RINGHALS-2	PWR	17/08/74
RINGHALS-3	PWR	07/09/80
RINGHALS-4	PWR	23/06/82
UK		
BERKELEY UNIT A	GCR	12/06/62
BERKELEY UNIT B	GCR	24/06/62
BRADWELL UNIT A	GCR	01/07/62
BRADWELL UNIT B	GCR	06/07/62
CHAPELCROSS UNIT A	GCR	01/02/59
CHAPELCROSS UNIT B	GCR	01/07/59
CHAPELCROSS UNIT C	GCR	01/11/59
CHAPELCROSS UNIT D	GCR	01/01/60
DOUNREAY FR	FBR	01/10/62
DUNGENESS-A UNIT A	GCR	21/09/65
DUNGENESS-A UNIT B	GCR	01/11/65
DUNGENESS-B1 UNIT A	AGR	29/12/85
DUNGENESS-B2 UNIT B	AGR	03/04/83
HARTLEPOOL-A1 UNIT A	AGR	01/08/83
HARTLEPOOL-A2 UNIT B	AGR	31/10/84
HEYSHAM-1 UNIT A	AGR	09/07/83
HEYSHAM-1 UNIT B	AGR	11/10/84
HEYSHAM-2 UNIT A	AGR	12/07/88
HEYSHAM-2 UNIT B	AGR	11/11/88
HINKLEY POINT-A UNIT	GCR	16/02/65
HINKLEY POINT-A UNIT B	GCR	19/03/65
HINKLEY POINT-B UNIT A	AGR	30/10/76
HINKLEY POINT-B UNIT B	AGR	05/02/76
HUNTERSTON-A1 UNIT A	GCR	05/02/64
HUNTERSTON-A2 UNIT B	GCR	01/06/64
HUNTERSTON-B1 UNIT A	AGR	06/02/76
HUNTERSTON-B2 UNIT B	AGR	31/03/77
OLDBURY-A UNIT A	GCR	07/11/67
OLDBURY-A UNIT B	GCR	06/04/68
PFR DOUNREAY	FBR	10/01/75
SIZEWELL-A UNIT A	GCR	21/01/66
SIZEWELL-A UNIT B	GCR	09/04/66
SIZEWELL-B	PWR	14/02/95
TORNESS UNIT A	AGR	25/05/88
TORNESS UNIT B	AGR	03/02/89
TRAWSFYNYDD UNIT A	GCR	14/01/65
TRAWSFYNYDD UNIT B	GCR	02/02/65
WINFRITH SGHWR	SGHW	01/12/67
WYLFA UNIT A	GCR	24/01/71

Reactor	Type	Date connected
WYLFA UNIT B	GCR	21/07/71

Data for the above sites for the period 1987-1999 inclusive is taken from the BILCOM database, which gives activities of radionuclides released from nuclear sites in five categories, aerosol, aqueous, dissolved gases, noble gases and iodines. Of these categories only the aqueous and dissolved gases are of interest to this study, other released activities are ignored.

The BILCOM database has a number of inconsistencies, which necessitate a degree of analysis. Some sites have reported discharges as aggregated totals of activity for alpha and beta emitters with values for significant nuclides, whereas data for other sites or for other years can contain a complete radionuclide breakdown. To determine whether the BILCOM database gives a representative nuclide breakdown of each aggregated discharge category the following methodology has been undertaken:

Firstly parent and daughter nuclides have been summed. Sub-category discharges for each specific site for each year were added, to establish if they account for more than 90 % of the aggregated discharges. Where the sum of sub-category discharges amount to more than 90% of the aggregated total, the BILCOM database nuclide breakdown has been assumed. Where the sum of sub-category discharges accounts for less than 90% of the aggregated discharge, a radionuclide composition has been calculated using the data provided by GRS, held in *breakdown.mdb*. This database gives percentage contribution values for specific radionuclides, according to the type of reactor and the country. Individual nuclide activities greater than the aggregated total for the particular site and year have been considered independently.

Data for the following sites is derived from the BILCOM database, using the methodology outlined above for the years 1987-1999 inclusive.

Reactor	Type	Date connected
BELLEVILLE-1	PWR	14/10/87
BELLEVILLE-2	PWR	06/07/88
BROKDORF (KBR)	PWR	14/10/86
CATTENOM-1	PWR	13/11/86
CATTENOM-2	PWR	17/09/87
CATTENOM-3	PWR	06/07/90
CATTENOM-4	PWR	27/05/91
EMSLAND (KKE)	PWR	19/04/88
FLAMANVILLE-1	PWR	04/12/85
FLAMANVILLE-2	PWR	18/07/86
GOLFECH-1	PWR	07/06/90
GOLFECH-2	PWR	18/06/93
NOGENT-1	PWR	21/10/87
NOGENT-2	PWR	14/12/88

A gap of two years exists between the period covered by MARINA data and the start of the BILCOM data. This period (1985 and 1986) was therefore calculated from discharge data



published by the European Commission [1]. Aggregated total activity values from the EC reports have been split, according to the GRS data, dependent on type of facility and country.

Radioactive discharges from the Swiss nuclear power stations listed below were not included in the BILCOM dataset. Data for the period 1985 to 1992 represents an average value based on the nuclides reported in the MARINA study. The average value of each nuclide reported in the MARINA data has been calculated as a proportion of the MARINA average total and multiplied by average total of both MARINA and OSPAR data. Subsequent data for 1993-1998 has been taken from OSPAR data provided in the annual reports [2].

Reactor	Type	Date connected
BEZNAU-1	PWR	17/07/69
BEZNAU-2	PWR	23/10/71
GOESGEN	PWR	02/02/79
LEIBSTADT	BWR	24/05/84
MUEHLEBERG	BWR	01/07/71

Capenhurst (1953, Enrichment Plant, UK)

Data for the period 1953-1984 is taken from the MARINA study. The radioactivity input is relatively small and data since 1984 have been omitted from this update.

Cap de la Hague (1966, NFRP, France)

Discharge data for the period 1966 to 1996 were extracted by COGEMA from the Nord-Cotentin Study the most recent discharges (1997-2000) were provided by COGEMA and correspond to results from measurements of radioactivity in effluents. Radionuclides present at levels below detection limits have been calculated based on the reprocessing rates, characteristics and transfer functions.

Sellafield (1952, NFRP, UK)

Data for the radioactive liquid discharges from Sellafield for the period 1952-1998 have been taken from the report Jackson et al 2000 [3]. Specific radionuclide activities are reported for those nuclides subject to authorisation limits. The reported Pu-alpha category has been presented as 100% <sup>239</sup>Pu. Data for 1999 has been taken from the Digest of Environmental Statistics [4]. Data for the year 2000 were supplied by BNFL, these data were found to compare closely to data published later by the FSA and SEPA[5].

Springfields (1947, Uranium Enrichment and Fuel Fabrication, UK)

Data for the period 1953-1984 were available in the MARINA study, the values were calculated using reported discharges of total alpha and total beta for 1964-1984, data prior to this period has been taken to be equal to the discharges reported in 1964.

A radionuclide breakdown of the following isotopes has been assumed for these years:

Alpha:	<sup>234</sup> U	4.9%
	<sup>235</sup> U	0.2%
	<sup>238</sup> U	4.9%
	<sup>228</sup> Th	15%
	<sup>230</sup> Th	60%
	<sup>232</sup> Th	15%
Beta:	<sup>234</sup> Th	50%

$^{234m}\text{Pa}$  50% (half life of 1.18 minutes, therefore not included in the data set)

This breakdown has also been used to disaggregate total alpha and total beta data given in the Digest of Environmental Statistics [4] for years 1988 to 1999. Data for the period 1985 to 1987 represents an average value based on the MARINA and Digest of Environmental Statistics data for each radionuclide.

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[5] FSA and SEPA Radioactivity in Food and the Environment 2000 RIFE-6 UK Food Standards Agency and the Scottish Environment Protection Agency (2000)

## Appendix B - NORM inputs from phosphoric acid production

A.W. van Weers, NRG, Petten, The Netherlands

### 1. Introduction

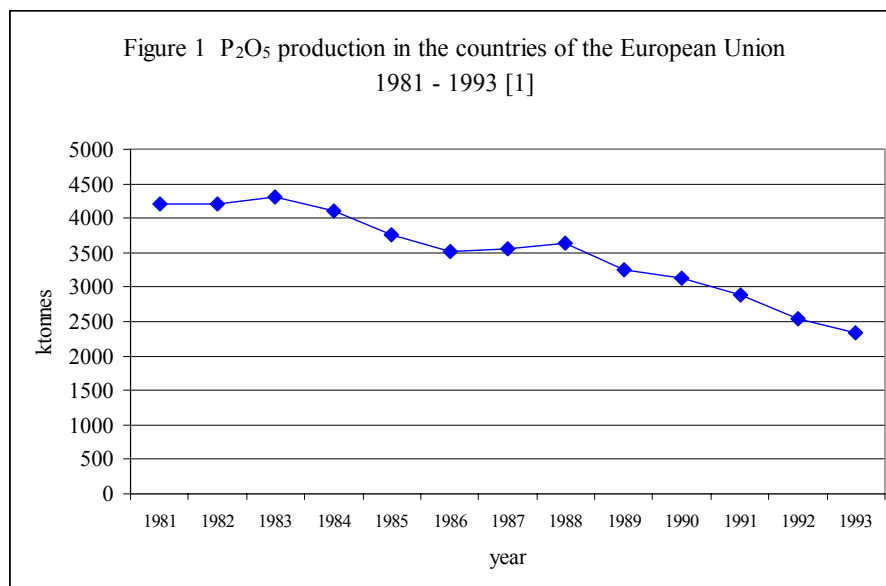
Group A of the MARINA II study collated data on the discharges of radioactive substances into the marine waters of the OSPAR region. The scope of the Group A not only covered sources of artificial radioactivity but comprised also discharges of natural radionuclides from non-nuclear industries which process naturally occurring radioactive materials (NORM). The fertiliser industry, in particular the production of phosphoric acid, had been identified in the OSPAR 1997 report as a relatively important source of input of natural radionuclides from industrial activities [1]. The present report summarises the OSPAR report and updates the information on NORM discharges by the fertiliser industry. From the currently available data normalised discharges have been derived for each relevant site in the period 1981 – 2000. Total annual inputs into the MARINA II model boxes have been calculated.

### 2. The 1997 OSPAR Report

#### 2.1 General summary

In 1997 The Oslo and Paris Commissions published a report on the discharges of radioactive substances by non-nuclear industries [1]. The report deals mainly with the phosphoric acid production from phosphate ore containing natural radionuclides. It summarises the production processes leading to discharges of natural radionuclides contained in waste phosphogypsum. It covers the period 1981 - 1993 for annual  $P_2O_5$  production and estimates annual discharges of radionuclides for 1993.

The main trend described in the report is a decrease of  $P_2O_5$  production in the countries of the European Union from 4206 ktonnes in 1981 to 2539 ktonnes in 1993 because of the closing down of production facilities and a change from phosphoric acid production to processing imported phosphoric acid. The data from the OSPAR report are shown in figure 1.



They pertain to total  $P_2O_5$  in phosphoric acid and in phosphate fertilisers. The figures therefore include production processes not involving sulphuric acid and phosphogypsum production. Consequently, discharges of phosphogypsum cannot be derived directly from the  $P_2O_5$  production data.

Phosphate ores from sedimentary origin are the main source of natural radioactivity in phosphoric acid production. Phosphate ores from volcanic origin have considerably lower radionuclide concentrations and account for a relatively small part of the ore produced. Concentrations of the dominant radionuclides (the  $^{238}U$  decay chain) in sedimentary phosphate ore from different origin differ considerably.

The natural radionuclides in the phosphate ores distribute differently between the phosphoric acid product and the phosphogypsum. The uranium isotopes appear to a large extent, but not exclusively, in the phosphoric acid while most of the radium isotopes and nearly all of the  $^{210}Pb$  and  $^{210}Po$  becomes associated with the phosphogypsum.

Discharges of natural radionuclides into surface water depend strongly on the fate of the phosphogypsum. At some plants the phosphogypsum is discharged directly as a slurry into surface waters and at others the phosphogypsum is stockpiled on land. In the latter case there may still be some input into surface water of radionuclides leached by percolating deposition.

The concentrations of radioactive and non-radioactive contaminants in phosphogypsum not only depend on the type of ore used but also on the effect of clean-up efforts by re-slurring and washing.

## 2.2 Assessment of annual discharges (1993)

The limited data available for the OSPAR 1997 report were used to derive an overall assessment of the discharges of natural radionuclides into the marine environment related to the phosphate fertiliser industry (year 1993). Table 8 from the OSPAR 1997 report is reproduced here as table 1.

Table 1. Assessment of annual discharges of natural radionuclides related to the phosphate fertiliser industry in 1993 ( $TBq \cdot a^{-1}$ ) [1].

Country	$^{226}Ra$	$^{210}Po$	$^{238}U$
Germany	0		
Belgium-Luxembourg	0.3	0.12	
Denmark	0		
Spain	0	1.1	0.4
France	0		
Norway	0		
Netherlands	1.0	1.0	
Portugal	0		
United Kingdom	0.001	0.003	0.1
Sweden	0		

It should be noted that the data have limited value as source data in the context of Marina II for one or more of the following reasons:

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- the data only cover the period 1981 to 1993,
- no annual discharge data are provided,
- production processes have been changed prior to and after 1993,
- data on  $^{210}\text{Pb}$  are lacking although it is known that this radionuclide largely ends up in the phosphogypsum,
- $^{226}\text{Ra}$  is known to be the most important radioactive constituent of phosphogypsum, nevertheless discharge data for Spain only pertain to  $^{210}\text{Po}$ ,
- For two countries out of four a figure on  $^{238}\text{U}$  discharges is presented, they pertain to quite different processes, phosphate ore and crude phosphoric acid processing respectively.

Therefore, these data cannot be used for retrospective or prospective assessment of radionuclide discharges by the phosphate industry.

### **3. Update on the OSPAR report**

#### **3.1 Introduction**

The OSPAR Commissions have not published an update on the 1997 report. However, supplementary information on  $\text{P}_2\text{O}_5$  productions in Western Europe has been made available for the purpose of MARINA II by the European Fertiliser Manufacturers Association (EFMA) [2]. The data provided comprise  $\text{P}_2\text{O}_5$  productions as phosphoric acid and as various phosphate fertilisers in a number of countries also covered in the OSPAR 1997 report. The data cover the period 1992 – 1999 but do not provide information on phosphogypsum productions nor on phosphogypsum discharges. The Netherlands have provided a report to OSPAR in September 1999 on the Implementation of the OSPAR Strategy with regard to Radioactive Substances [3]. This report provides detailed information on the discharges by the Dutch phosphoric acid production plants in the period 1993 - 1998. As in previous years the phosphogypsum from phosphoric acid production in The Netherlands was discharged in the River Rhine at Rotterdam. A study carried out very recently in Belgium provided the basis for estimates of discharges of natural radionuclides with phosphogypsum in the River Scheldt at Antwerp [4]. Efforts to update the OSPAR 1997 report with respect to discharges of natural radionuclides with phosphogypsum into the OSPAR region by other countries were not successful.

#### **3.2 General trends**

A programme of fundamental restructuring of the fertiliser industry occurred in the early 1990s when some 60 plants were closed [5]. The review predicted that 30 plants would have been closed by the end of 2000 with the loss of some 4 000 jobs. These predictions did not pertain specifically to phosphoric acid production but to the whole fertiliser industry in the European Union, Norway and Switzerland. The general trend in phosphoric acid production is illustrated by the closure of the Norwegian phosphoric acid production facility, the closure of one of the Dutch facilities by the end of 1999 and of the second one mid 2000 [3, 6]. This trend reflects the shift of phosphoric acid production from Western Europe to the countries where the phosphate ore is mined. Another important trend is the shift from phosphoric acid production to the production of phosphate fertilisers by the nitric acid and hydrochloric acid processes, which do not involve phosphogypsum production and discharges. In these processes the natural radioactivity from the phosphate ore largely goes to products and by-products.

## 4. Method of assessment of discharges

### 4.1 The period considered

The fertiliser industry in Western Europe started to develop at the end of the 19<sup>th</sup> century. Phosphoric acid production peaked in the early eighties of the 20<sup>th</sup> century and had declined considerably by the end of that century. Efforts to retrospectively assess the discharges of natural radionuclides from phosphoric acid production back to the start of this industrial activity go far beyond the scope of Group-A and will not be successful because of lack of reliable information anyhow. Therefore the assessment is restricted to the period 1981 to 2000, the period covered by the OSPAR 1997 report and the update for 1993 – 2000.

### 4.2. Radionuclides considered

Phosphate ore contains all radionuclides from the uranium and thorium natural decay chains. However the radionuclides from the decay chains of  $^{235}\text{U}$  and  $^{232}\text{Th}$  occur in much lower concentrations than those of the  $^{238}\text{U}$  decay chain. Only the latter are considered in the present assessment.

The  $^{238}\text{U}$  decay chain not only comprises  $^{238}\text{U}$  but  $^{234}\text{U}$  as well. In addition the short-lived  $^{234}\text{Th}$  and the long-lived  $^{230}\text{Th}$  can be assumed to occur in phosphate ore in activity concentrations equal to  $^{238}\text{U}$ . However the fate of these radionuclides in phosphoric acid production is not sufficiently documented to derive reliable estimates of their discharges with phosphogypsum. The discharges of uranium reported for the UK plant at Whitehaven appear to be largely derived from uranium analyses of ores and products and not from analyses of the phosphogypsum discharged [7]. From 1992 the plant processed imported crude phosphoric acid and the associated discharges are therefore not typical for production of phosphoric acid from ore. The derived releases of the thorium isotopes also do not provide a sound basis for assessing releases at other sites. Uranium and thorium discharges are not included in update figures on the Dutch and Belgian sites. Therefore, the discharges of uranium and thorium radionuclides have not been considered in the present assessment, not for the UK site nor for other sites.

### 4.3 Normalised releases

#### 4.3.1 General approach

The data available to assess discharges of natural radionuclides at the production sites of phosphoric acid range from rough estimates provided in the OSPAR 1997 report on total or annual discharges of phosphogypsum in tonnes to nuclide specific discharge data for part of the years to be considered. Consequently, for each site the discharges had to be derived on a different set of data available. The method used to arrive at estimated discharges covering the years 1981 to 2000 is based on the following approach:

- the mass ratio between  $\text{P}_2\text{O}_5$  and phosphogypsum production is set at a normalised value of 1:4.5 to derive phosphogypsum production from  $\text{P}_2\text{O}_5$  production data,
  - a normalised release of 0.49, 0.47 and 0.45 GBq/ktonne phosphogypsum derived for  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  respectively for the Dutch HAR site is applied to other sites in the absence of site specific data,
-

- a normalised activity ratio of 0.49: 0.47:0.45 between  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  for the Dutch HAR site is used in the absence of data for the latter two radionuclides,
- for sites where all produced phosphogypsum can be assumed to have been discharged the  $\text{P}_2\text{O}_5$  production figures are used to derive phosphogypsum discharges,
- for sites where only part of the produced phosphogypsum has been discharged the rough estimates of phosphogypsum discharges from the OSPAR 1997 report are used to derive estimates of annual releases.

Further details of the assessment and the results are provided below for each country and site.

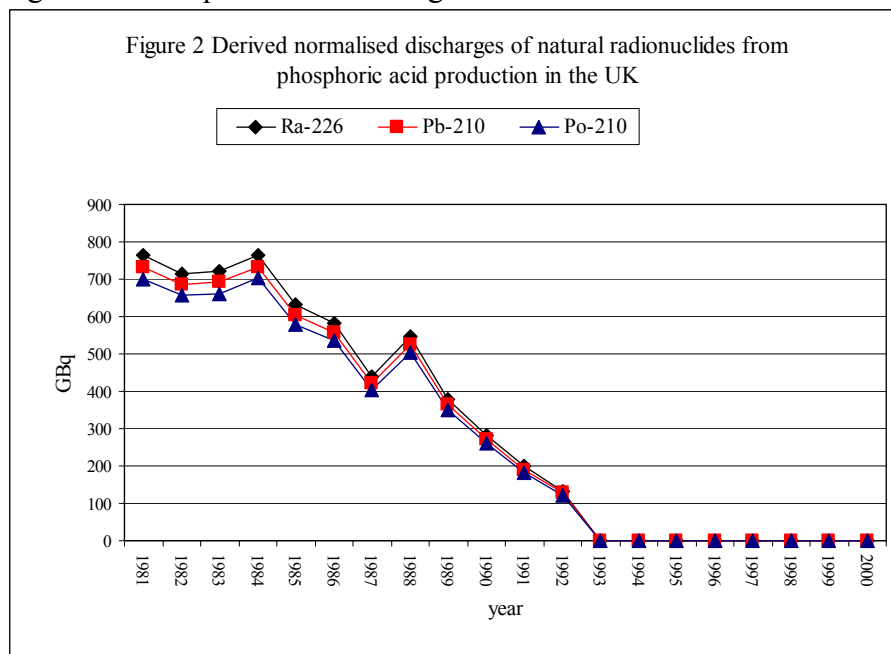
#### 4.3.2 United Kingdom

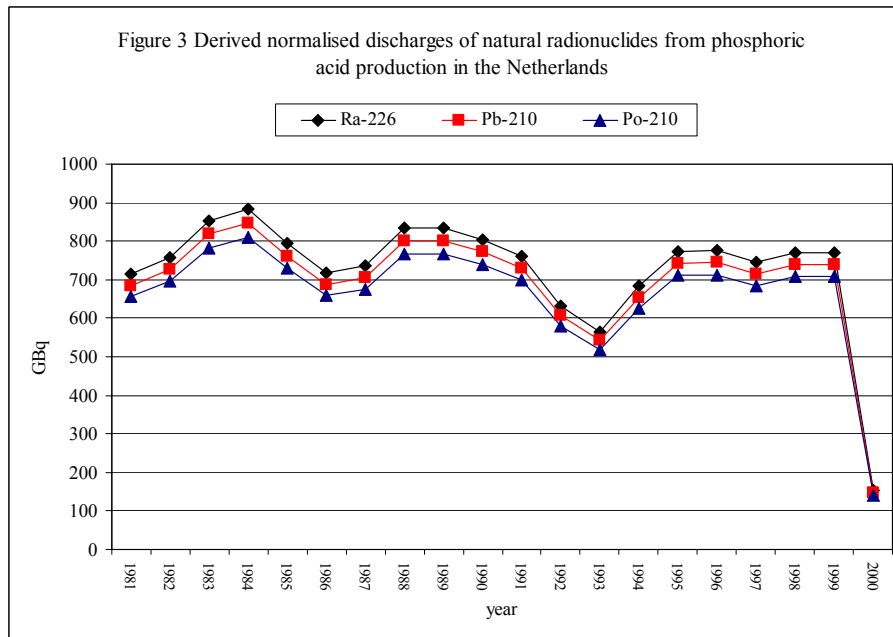
In 1992 the processing of phosphate ore at the Whitehaven site in the UK was replaced by processing of imported crude phosphoric acid. This caused a sharp drop in radionuclide releases.

The normalised releases presented in figure 2 are derived on the following basis:

- OSPAR 1997  $\text{P}_2\text{O}_5$  production data pertain to phosphoric acid only,
- mass ratio of  $\text{P}_2\text{O}_5$  to phosphogypsum 1:4.5,
- normalised releases are based on 0.49, 0.47 and 0.45 GBq/ktonne phosphogypsum for  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  respectively.

The derived total releases of  $^{226}\text{Ra}$  in the period considered are in very good agreement with the data provided in [7]. For  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  the derived normalised discharges are about 40% higher than the published discharges.





#### 4.3.3 The Netherlands

Detailed information on radionuclide discharges from the Dutch phosphoric acid production in the years 1993 to 1998 has been presented to OSPAR in September 1999 [3]. The data used pertain to  $^{236}\text{Ra}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$ .

The normalised releases presented in figure 3 are derived on the following basis:

- all phosphogypsum produced was discharged,
- phosphogypsum discharges based on OSPAR 1997 report and the Dutch September 1999 report to the OSPAR Commissions,
- normalised discharges based on 0.49, 0.47 and 0.45 GBq/ktonne phosphogypsum for  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  respectively, based on the mean values for the HAR plant 1993-1998,
- the data for the years 1999 and 2000, not in ref. [3] are based on the assumption that the plant that was closed down in 1999 discharged in 1999 the same activities as in 1998; the other plant closed mid 2000 and it was assumed that the discharges in 1999 were equal to 1998 and that they were half of these values in 2000.

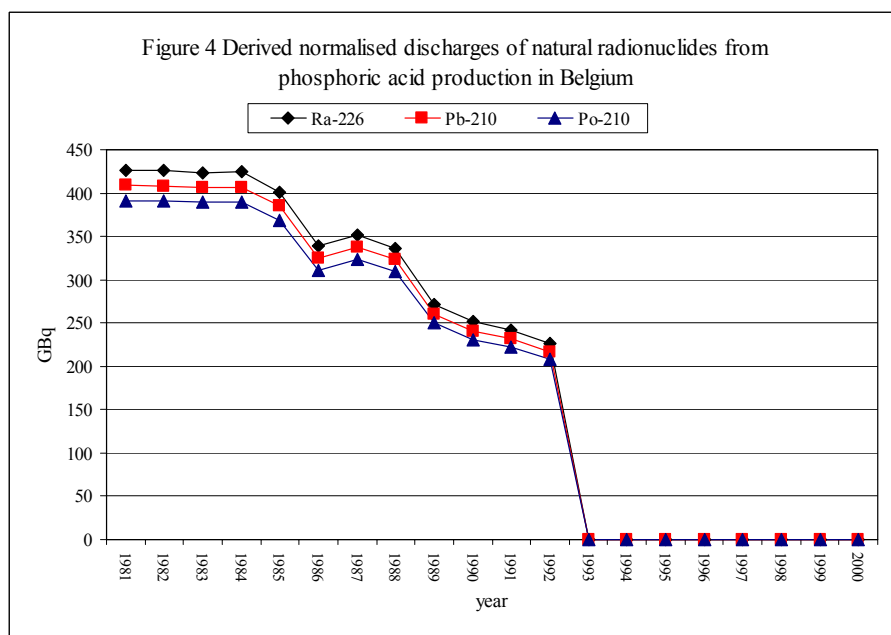
#### 4.3.4 Belgium

The past discharges of phosphogypsum were associated with phosphoric acid production in facilities located at Antwerp. Other production facilities in Belgium either stockpiled the phosphogypsum on land or used other processes for production of phosphate fertiliser from phosphate ore. At the Antwerp site the production process was completely changed in 1992 to the nitric acid process not involving production and discharge of phosphogypsum.

The normalised releases presented in figure 4 are derived on the following basis:

- phosphogypsum discharges and  $^{226}\text{Ra}$  concentrations as provided by [4]
- a normalised activity ratio of 0.49: 0.47:0.45 between  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$ .





#### 4.3.5 Denmark

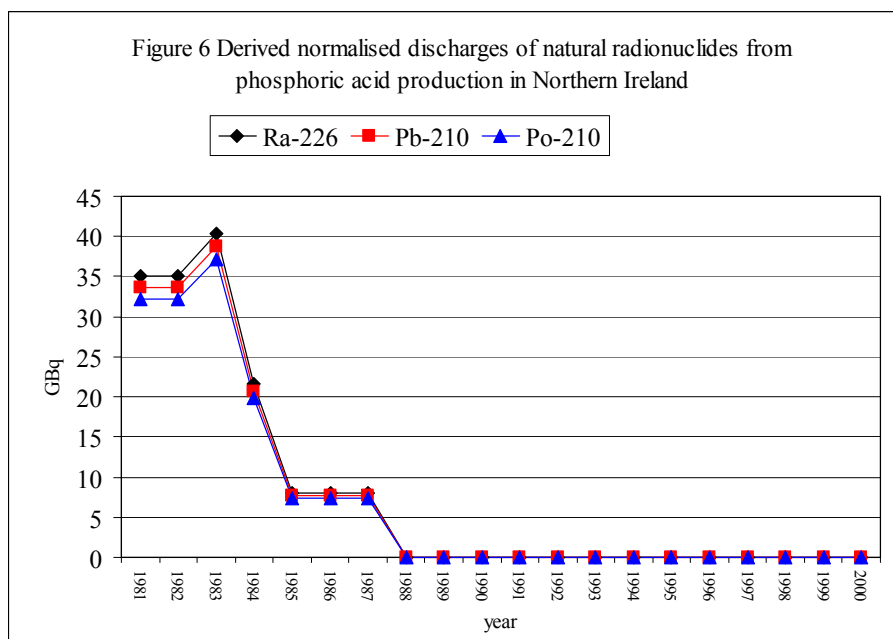
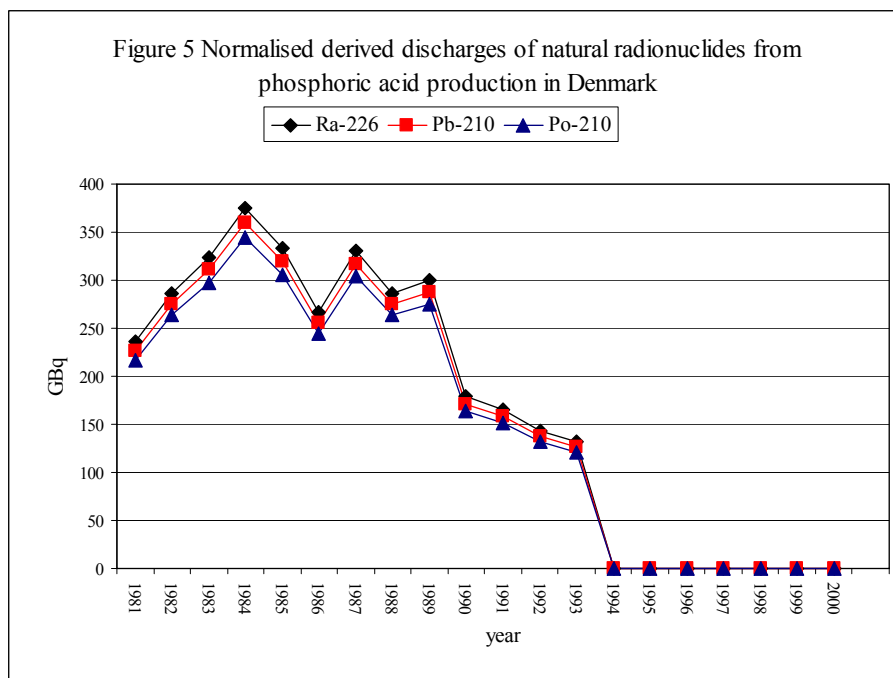
The normalised releases presented in figure 5 are derived on the following basis:

- OSPAR 1997  $P_2O_5$  production data assumed to pertain to  $P_2O_5$  as phosphoric acid only,
- mass ratio  $P_2O_5$  to phosphogypsum assumed as 1:4.5,
- normalised releases: 0.49, 0.47 and 0.45 GBq/ktonne phosphogypsum (Dutch data for HAR 1993 - 1998).

#### 4.3.6 Northern Ireland

The normalised releases presented in figure 6 are derived on the following basis:

- OSPAR 1997  $P_2O_5$  production data assumed to pertain to  $P_2O_5$  as phosphoric acid only,
- no phosphoric acid production after 1988,
- mass ratio  $P_2O_5$  to phosphogypsum assumed as 1:4.5,
- normalised releases: 0.49, 0.47 and 0.45 GBq/ktonne phosphogypsum (Dutch data for HAR 1993 - 1998).



#### 4.3.7 France

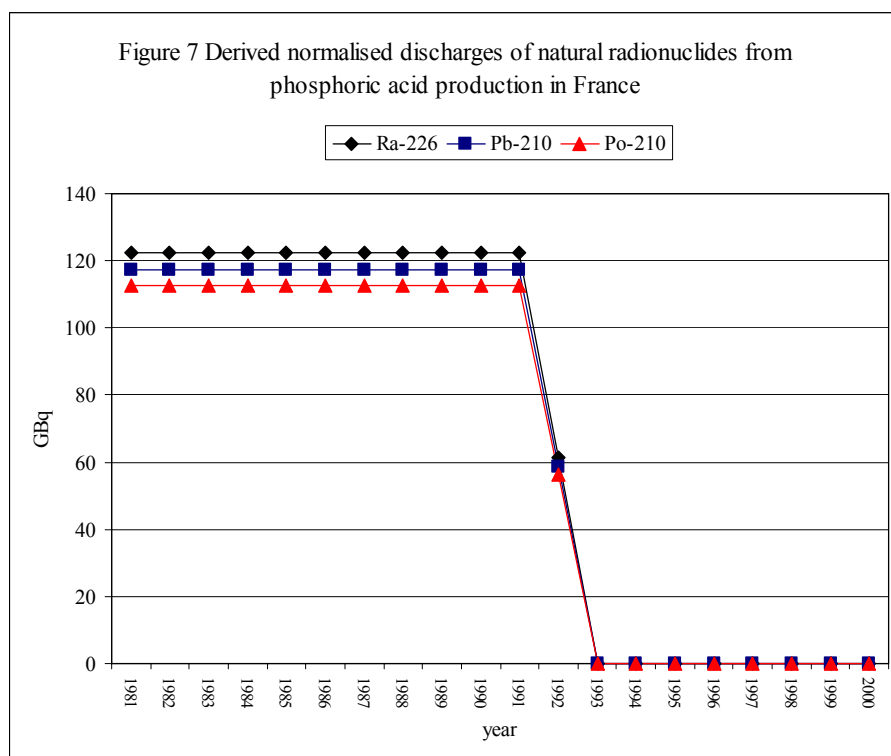
Only part of the phosphogypsum from phosphoric acid production in facilities along the Seine River has been discharged in the past. In view of the very limited data available no effort has been made to derive annual releases of natural radionuclides. The time course presented in figure 7 only reflects the end of the discharges in 1992.

The normalised releases are derived on the following basis:

- total phosphogypsum discharges in the Seine amounted to about 3 million tonnes [1],
- discharges of phosphogypsum in the Seine stopped in September 1992 [1],

- c. the 3 million tonnes phosphogypsum were assumed to have been discharged in the period 1981 – 1992[1].
- d. normalised releases: 0.49, 0.47 and 0.45 GBq/ktonne phosphogypsum (Dutch data for HAR 1993 - 1998).

Assumption c. may result in an overestimate of the annual phosphogypsum discharges in the Seine if the period of the total discharge of about 3 million tonnes was significantly longer than the 12 years assumed.

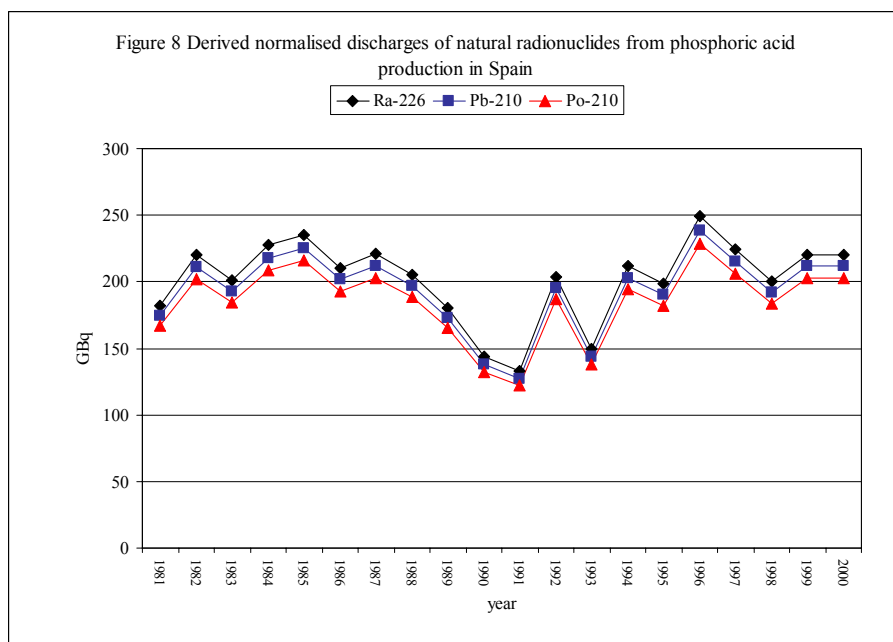


#### 4.3.8 Spain

Only part of the phosphogypsum produced in facilities bordering the rivers Odiel and Tinto in Southern Spain is discharged.

The normalised releases presented in figure 8 are derived on the following basis:

- a. 0.4 million tonnes phosphogypsum discharged annually [1],
- b. derived fraction of total phosphogypsum being discharged (0.4/1.8),
- c. annual discharges of phosphogypsum scaled on the basis of the  $P_2O_5$  production rates from the OSPAR 1997 report and EFMA 2001,
- d. normalised activity discharges on the basis of 0.49, 0.47 and 0.45 GBq/ktonne phosphogypsum (Dutch data on HAR 1993-1998),
- e. discharges assumed to have been continued up to and including 2000.

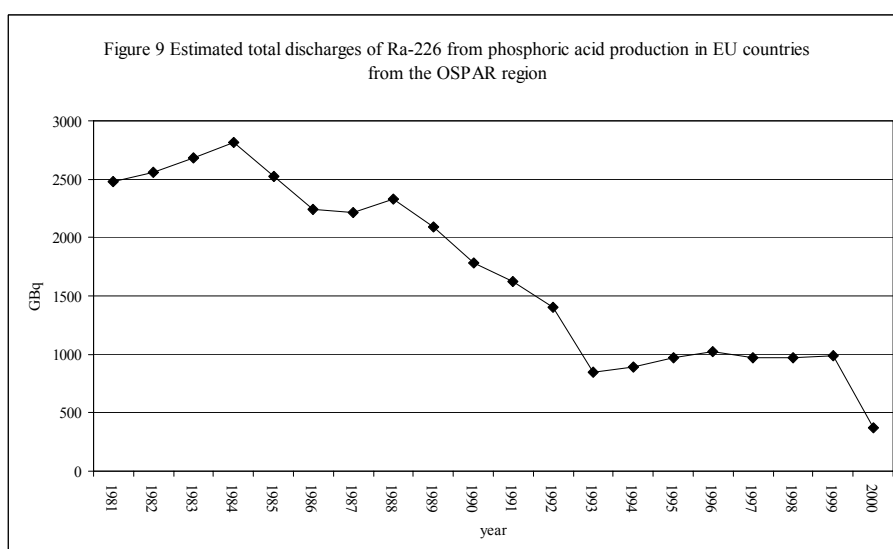


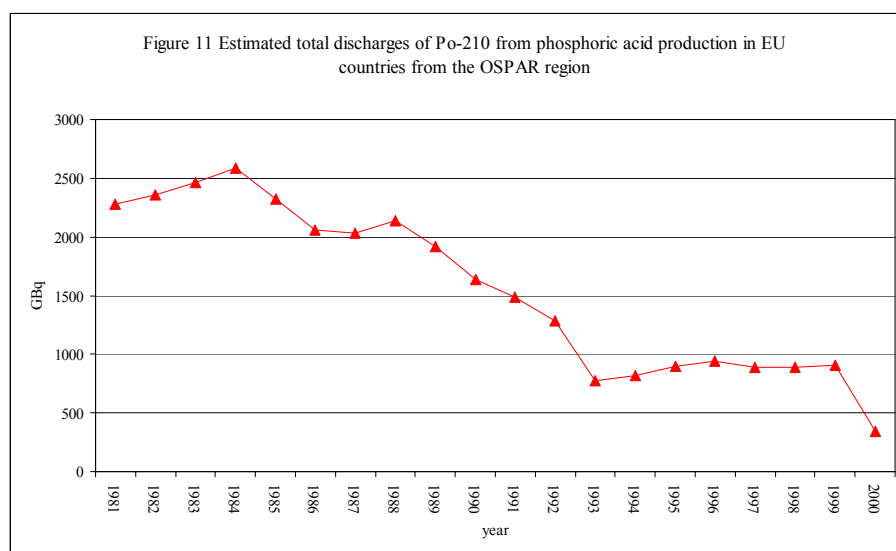
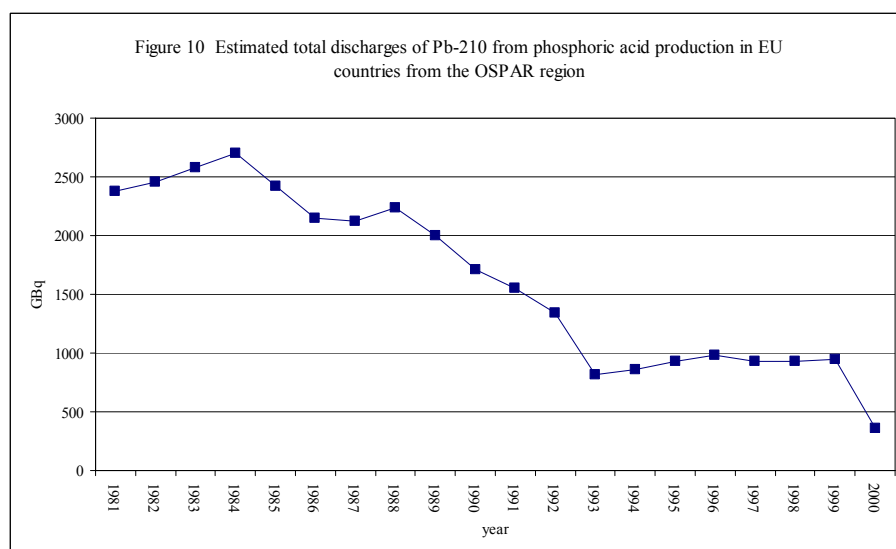
#### 4.3.9 Other countries from the OSPAR area

According to the OSPAR 1997 report phosphogypsum had not been discharged or the practice was terminated before 1981 in the following countries: Norway, Germany, Portugal, Sweden and Ireland.

### 5 Total discharges

The estimated total discharges of  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  at all sites considered are presented in figures 9 – 11. They show a considerable decrease in annual discharges after the mid-eighties. The figures do not show the further decrease in activity discharges in 2001 after the closure of the last of the two phosphoric acid production plants, which had been in operation in The Netherlands.





## 6 Annual inputs into model boxes

The annual inputs of  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  into the boxes of the MARINA II model are given in tables 1 – 6 at the end of this report. The releases are largely historical for reasons already presented. For instance, estimated annual discharges of  $^{226}\text{Ra}$  into Box 56 amounted to about 1 000 GBq in the period 1981 – 1991, declined to about 150 GBq in 2000 and are reduced to zero after the closure of the last Dutch phosphoric acid production plant in 2000.

## 7 Discussion

The data presented on the discharges of natural radionuclides from the production of phosphogypsum are only partly based on records of activity discharges. Such records either don't exist or could not be made available for the purpose of MARINA II. As a consequence, the discharge data provided are derived normalised discharges based on rather different levels of information available for the different sites. However a reasonably sound basis for the assessment of the annual discharges could be provided for the main contributors to total discharges since 1981: the sites in the Netherlands, Belgium and United Kingdom. Therefore,

the present assessment of the main inputs into the boxes of the MARINA II model can be regarded as “best estimates” at current level of information.

It should also be noted that discharges of natural radionuclides from phosphoric acid production are not limited to the radionuclides  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$ . Phosphate ore also contains  $^{234}\text{U}$ ,  $^{234}\text{Th}$  and  $^{230}\text{Th}$  in secular equilibrium with  $^{238}\text{U}$ . However, no reasonably sound basis could be found to quantify the discharges of these radionuclides with phosphogypsum or in dissolved form.

Table 1. Input into Box 30, Irish Sea NW			
	GBq	GBq	GBq
year	$^{226}\text{Ra}$	$^{210}\text{Pb}$	$^{210}\text{Po}$
1981	35	34	32
1982	35	34	32
1983	40	39	37
1984	22	21	20
1985	8	8	7
1986	8	8	7
1987	8	8	7
1988	0	0	0
1989	0	0	0
1990	0	0	0
1991	0	0	0
1992	0	0	0
1993	0	0	0
1994	0	0	0
1995	0	0	0
1996	0	0	0
1997	0	0	0
1998	0	0	0
1999	0	0	0
2000	0	0	0

Table 2. Input to Box 35, Cumbrian Waters

year	GBq <sup>226</sup> Ra	GBq <sup>210</sup> Pb	GBq <sup>210</sup> Po
1981	763	732	701
1982	714	685	656
1983	721	692	662
1984	765	734	703
1985	631	605	579
1986	582	558	535
1987	441	423	405
1988	547	525	502
1989	379	364	348
1990	282	271	259
1991	198	190	182
1992	132	127	122
1993	0	0	0
1994	0	0	0
1995	0	0	0
1996	0	0	0
1997	0	0	0
1998	0	0	0
1999	0	0	0
2000	0	0	0

Table 3. Input into Box 44, Gulf of Cadiz			
year	GBq <sup>226</sup> Ra	GBq <sup>210</sup> Pb	GBq <sup>210</sup> Po
1981	182	174	167
1982	220	211	202
1983	201	193	185
1984	227	218	209
1985	235	226	216
1986	210	202	193
1987	221	212	203
1988	205	197	189
1989	180	173	166
1990	144	138	132
1991	133	127	122
1992	203	195	187
1993	150	144	138
1994	212	203	194
1995	198	190	182
1996	249	239	229
1997	224	215	206
1998	200	192	184
1999	221	212	203
2000	221	212	203



Table 4. Input to Box 50, Bay de la Seine

year	GBq <sup>226</sup> Ra	GBq <sup>210</sup> Pb	GBq <sup>210</sup> Po
1981	123	118	113
1982	123	118	113
1983	123	118	113
1984	123	118	113
1985	123	118	113
1986	123	118	113
1987	123	118	113
1988	123	118	113
1989	123	118	113
1990	123	118	113
1991	123	118	113
1992	61	59	56
1993	0	0	0
1994	0	0	0
1995	0	0	0
1996	0	0	0
1997	0	0	0
1998	0	0	0
1999	0	0	0
2000	0	0	0

Table 5. Input to Box 56, North Sea South East			
year	GBq <sup>226</sup> Ra	GBq <sup>210</sup> Pb	GBq <sup>210</sup> Po
1981	1141	1094	1048
1982	1184	1136	1087
1983	1277	1225	1173
1984	1306	1253	1200
1985	1195	1146	1098
1986	1055	1012	969
1987	1088	1044	1000
1988	1170	1122	1074
1989	1105	1060	1015
1990	1056	1013	970
1991	1003	962	921
1992	859	824	789
1993	564	541	518
1994	683	655	627
1995	774	742	711
1996	776	744	713
1997	745	714	684
1998	771	739	708
1999	771	739	708
2000	152	146	140

Table 6. Input into Box 61, Kattegat, surface 0 - 20 m			
year	GBq <sup>226</sup> Ra	GBq <sup>210</sup> Pb	GBq <sup>210</sup> Po
1981	236	226	217
1982	287	275	263
1983	324	311	298
1984	375	360	344
1985	333	319	306
1986	267	256	245
1987	331	317	304
1988	287	275	263
1989	300	288	275
1990	179	171	164
1991	165	159	152
1992	143	137	132
1993	132	127	122
1994	0	0	0
1995	0	0	0
1996	0	0	0
1997	0	0	0
1998	0	0	0
1999	0	0	0
2000	0	0	0

## References

- [1] OSPAR 1997, Report on Discharges of Radioactive Substances by Non-Nuclear Industries. Oslo and Paris Commissions, 1997.
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## Appendix C - NORM inputs from off-shore oil and gas production

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### 1. Origin of releases

Production of oil and gas from reservoir rocks is accompanied by mobilisation of natural radionuclides which appear in water co-produced with the oil and gas [1]. The mobilised radionuclides are  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$  and  $^{210}\text{Pb}$ . The radium isotopes in produced water arise in the course of the mobilisation of the alkaline earth elements Ca, Sr, Ba and Ra; and  $^{210}\text{Pb}$  originates from mobilisation of lead. Upstream from the well, at reduced pressure and temperature, the radium isotopes may co-precipitate as mixed sulphates or carbonates of Ba, Sr and Ca and deposit as scales in production vessels and tubes.  $^{210}\text{Pb}$  deposits with stable lead as elemental lead or as lead sulphides.

$^{228}\text{Ra}$  deposited in scale gives rise to  $^{228}\text{Th}$  by radioactive decay. The activity ratio  $^{228}\text{Th}/^{228}\text{Ra}$  starts to increase from zero as the scale ages to a maximum of 1.5 after about 15 years. The maximum ratio will only be reached in scale stored for many years. After an average residence time of scale in a production installation of one year the activity ratio  $^{228}\text{Th}/^{228}\text{Ra}$  is about 0.3.

Strongly increased mobilisation of the radium isotopes is usually observed when a well, after progressive depletion, starts to co-produce brine instead of water vapour. Scale deposition is stimulated by mixing produced water with sulphate containing seawater either upstream or down the well as a result of breakthrough of sea water, injected to stimulate production. As a consequence, concentrations of NORM nuclides in produced water from oil and gas production vary considerably between wells depending on reservoir and production condition as well as over the lifetime of the same production installation. Activity concentrations of the radium isotopes and  $^{210}\text{Pb}$  in produced water range from virtually zero to about 100 Bq/l. Activity concentrations of the radium isotopes in scale range to a few hundred Bq g<sup>-1</sup> and specific activities of up to 1 000 Bq g<sup>-1</sup> have been encountered in lead deposits.

As scale deposits may strongly interfere with production capacity and safety, scale prevention and removal techniques are being applied. Scale prevention by injection of scale inhibitors into the well keeps the alkaline earth elements in solution till discharge into the sea or the re-injection of the produced water. Chemical descaling is applied to dissolve and discharge the scale. Mechanical descaling carried out offshore results in solids discharged offshore if allowed under current regulations. Onshore descaling facilities either collect the radioactive solids for on-land disposal or are authorised to discharge the scale into the sea under conditions with respect to total activity releases and maximum particle size allowed.

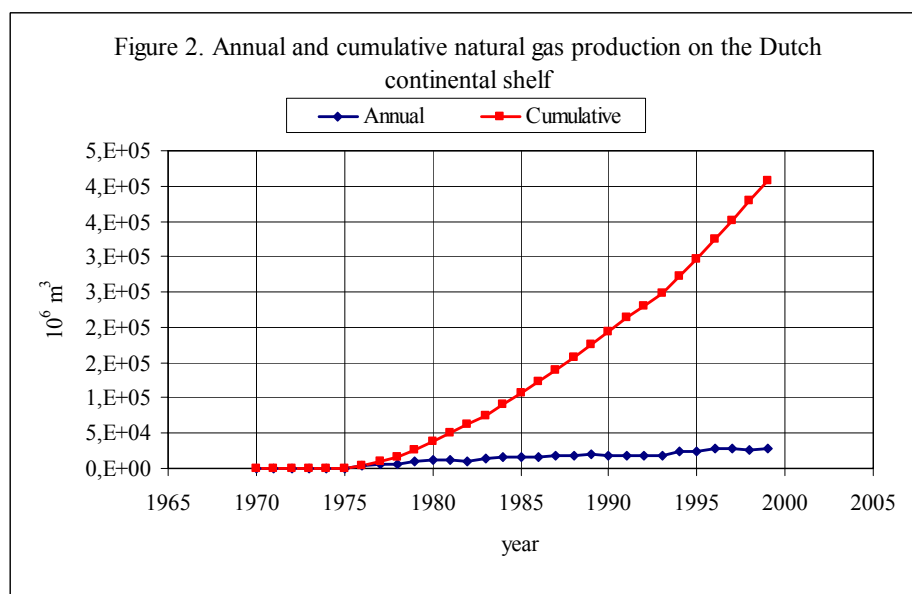
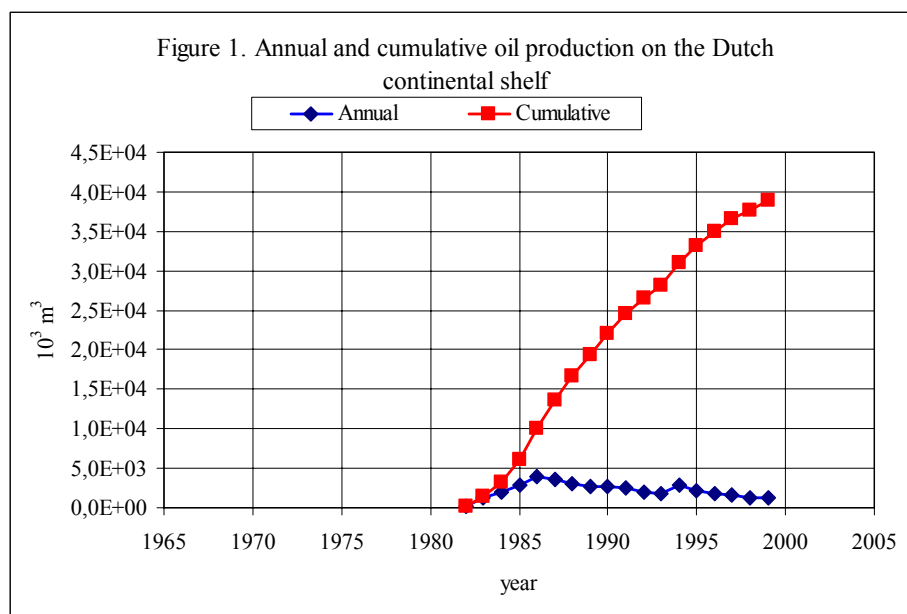
In summary the main releases of NORM from offshore oil and gas production originate from the following sources:

- produced water released offshore ( $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{210}\text{Pb}$ )
- scale from offshore mechanical descaling released offshore ( $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{228}\text{Th}$ ,  $^{210}\text{Pb}$ )
- scale from coastal descaling site discharged into the sea ( $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{228}\text{Th}$ ,  $^{210}\text{Pb}$ )

## 2. Oil and gas production data

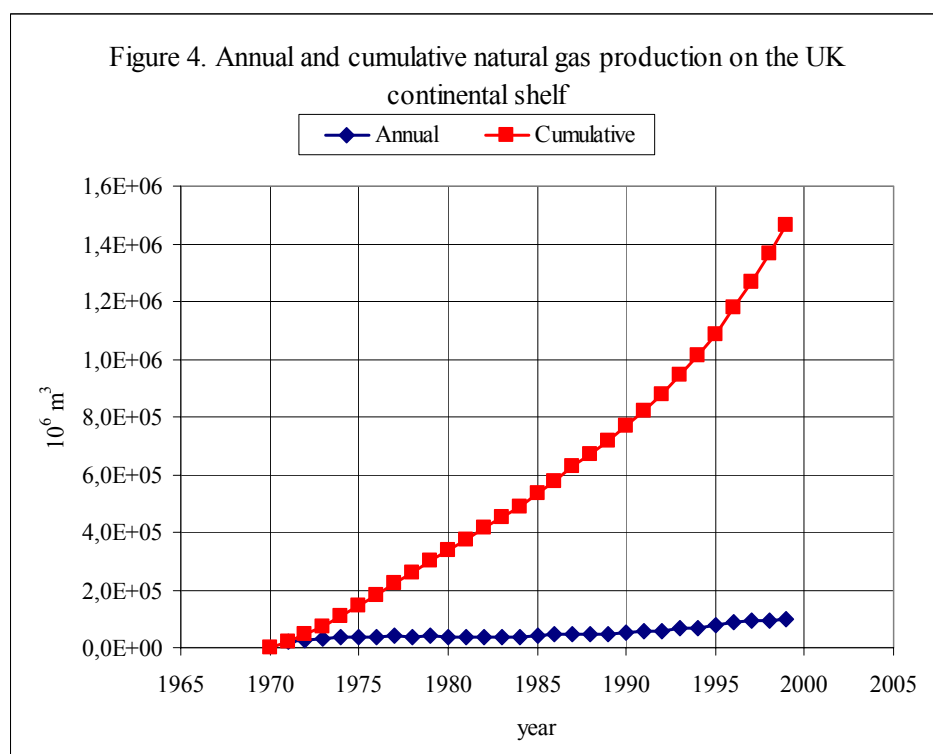
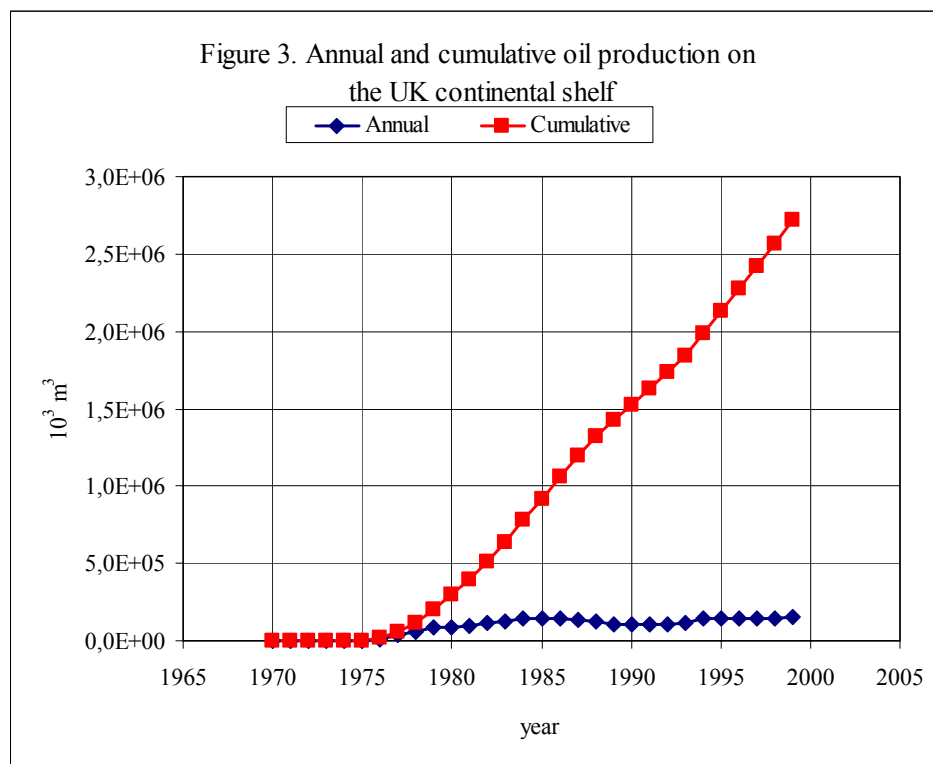
### 2.1 Dutch continental shelf

Oil and gas production on the Dutch continental shelf started in the early seventies. Annual and cumulative productions are shown in figure 1 and 2. They are based on the 2000 report by the Dutch Ministry of Economic Affairs [2].



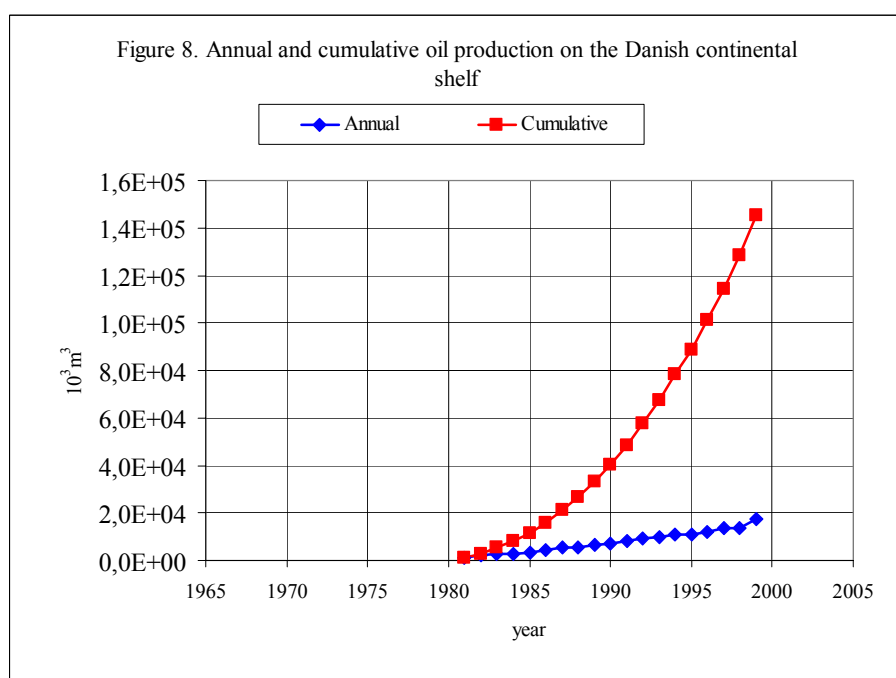
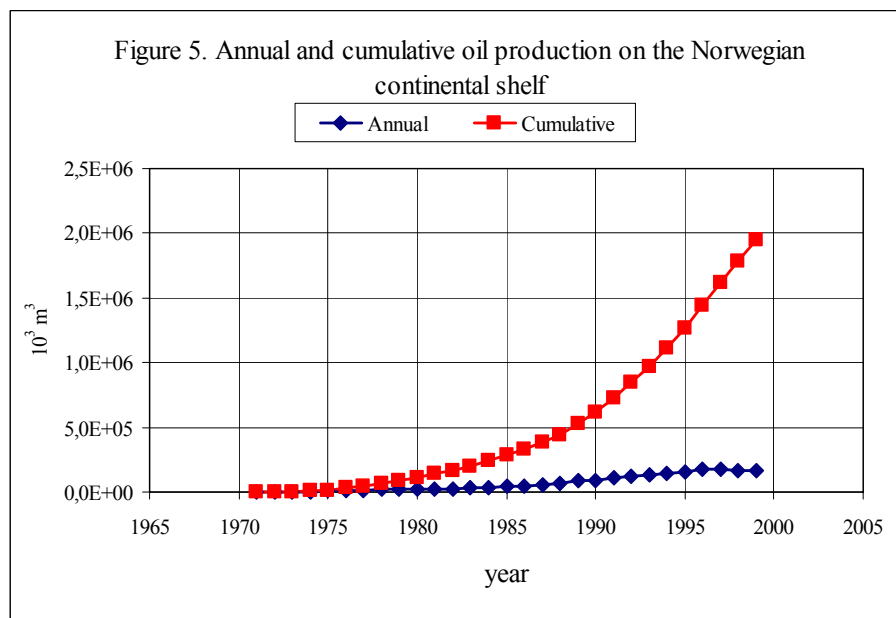
## 2.2. UK continental shelf

The development of oil and gas production on the UK part of the continental shelf is shown in figure 3 and 4. The data for oil were obtained from the so-called Brown Book [3] and the gas production data were extracted from Statistics Norway where they are presented together with productions from the Norwegian and Denmark sectors [4].



### 2.3. Norwegian continental shelf

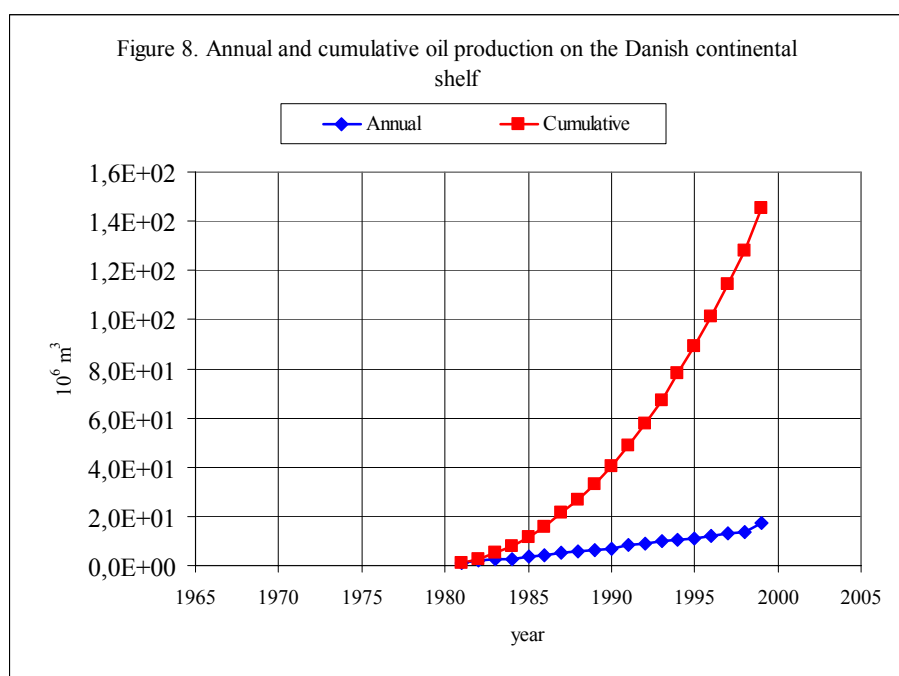
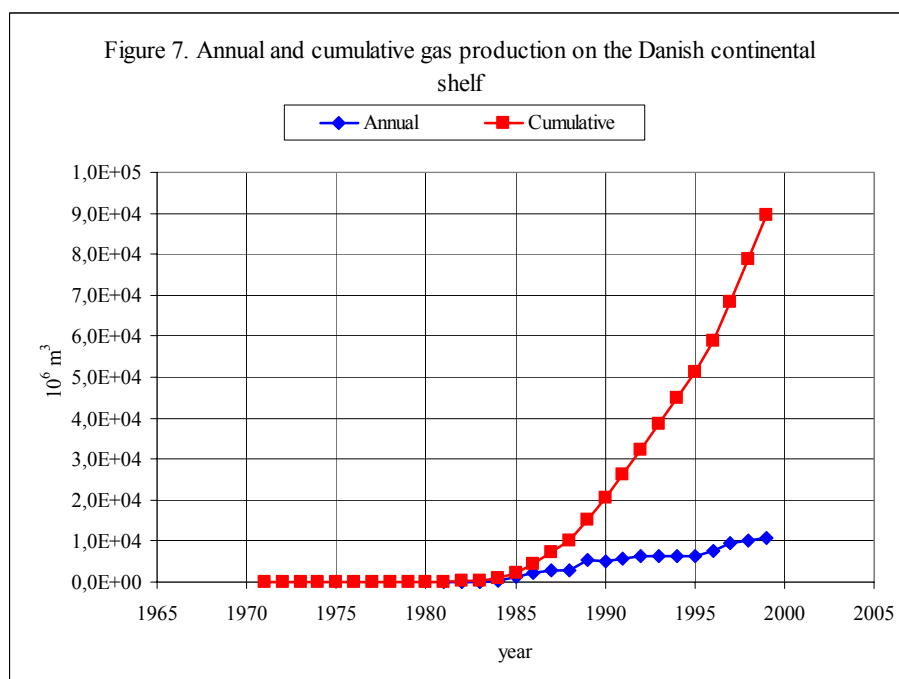
Data on the oil and gas production on the Norwegian continental shelf are presented in figure 5 and 6. They were obtained from Statistical Yearbook of Norway 2000 [5].





## 2.4. Danish continental shelf

Data on oil and natural gas production on the Danish continental shelf are presented in figures 7 and 8. They show the later start of oil and gas production on the Danish continental shelf. The data were obtained from reference [4] for gas production and reference [6] for oil production.



### **3. Derived releases of radionuclides**

#### **3.1 Introduction**

Estimates of releases of natural radionuclides from oil and gas production have by necessity to be based on limited data of actual releases. Data specifically pertaining to the sea area of interest are very seldom published and available for review. Some data on natural activity in produced water from Norwegian platforms have been presented by Strand and Lysebo [7]. A rather extensive set of unpublished and confidential data however exists in the Netherlands [8]. The latter also applies to data on co-produced water. These data can however be used to derive normalised radionuclide releases per unit of annual gas and oil production volume. In combination with figures of total annual oil and gas production on the different parts of the continental shelf in the period of interest total annual activity releases can be derived. It should be stressed that the resulting figures are still rough estimates because of the large variation in activity concentrations between all platforms and over the years for individual platforms as well as the large variations in the volume of water produced.

#### **3.2 Normalised releases of produced water**

##### **3.2.1 Oil production**

The production of large volumes of water is inherent to the production of oil. The ratio between production rates of oil and water depends on production conditions and may decrease considerably over the lifetime of a production well to a level that production rate of oil is only a small percentage of the production rate of water [8]. Virtually all of the water produced on the continental shelf of the North West Atlantic is discharged into the sea upon removal of most of the dissolved and undissolved hydrocarbons. Re-injection of produced water presently pertains only to a small fraction of the total volume produced but may increase significantly in the future because of stricter international and national limits on non-radioactive pollutants discharged with produced water [9].

For the present assessment a ratio of 0.33 between the volumes of produced oil and water is used as a reference average value for all oil producing platforms in the sea area of interest over their lifetime, e.g. for each m<sup>3</sup> of oil 3 m<sup>3</sup> of water is assumed to be co-produced. With this ratio the discharge of produced water is not likely to be overestimated. Ratios for individual platforms may differ considerably from this value in both directions. From experts comments it seems that the figure might even be a little bit optimistic, e.g. underestimating the over lifetime average water production [9], [10].

##### **3.2.2 Gas production**

Water is co-produced with natural gas to a considerably lesser extent. However, the ratio between production rates of gas and water varies even more than for oil [8]. From relatively dry gas relatively small amounts of water are condensed and separated when the gas is treated before transport. The other extreme, of relatively high water production rates, is usually associated with co-production of formation water at the end of the lifetime of the gas production well. Virtually all of the produced water is discharged into the sea.

For the present assessment a ratio of  $5 \cdot 10^{-5}$  is used between the water production and standard  $\text{m}^3$  of produced gas, e.g. for each million  $\text{m}^3$  of gas 50  $\text{m}^3$  of water is produced.

### 3.3 Normalised concentrations of radionuclides

The concentrations of the natural radionuclides  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$  and  $^{228}\text{Ra}$  in produced water from individual platforms oil and gas production wells vary between less than 0.1 Bq/l to about 200 Bq/l [7], [8]. Concentration ratios between the radionuclides in produced water also vary considerably. When a gas well starts to co-produce saline formation water from the reservoir a sharp and strong increase of the radionuclide concentrations in produced water is usually observed. Part of the radioactivity mobilised from the reservoir may be deposited as scale in the production installation and subsequently be removed and discharged into the sea [1]. Although data could be made available on limits set for such authorised discharges from oil producing platforms on the UK continental shelf, no efforts have been made in the present assessment to discriminate between discharges of radioactive solids and discharges of dissolved radionuclides in the produced water. It is assumed that the reference values of natural radionuclides in produced water from oil and gas production used for the present assessment, in combination with the normalised production rates of water, cover the discharges of both solids and dissolved radionuclides.

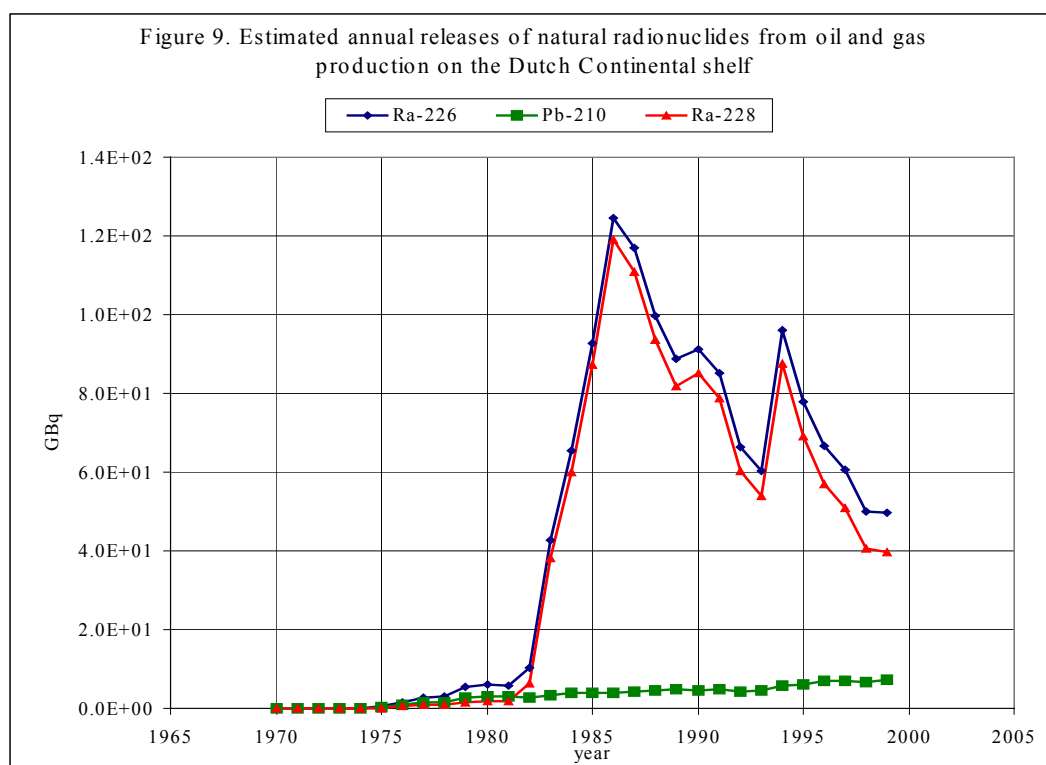
The average concentration of the radionuclides  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  in produced water discharged from all oil producing platforms and over all years is estimated at 10 Bq/l each. For gas production the corresponding figures are  $^{226}\text{Ra}$ : 10 Bq/l,  $^{210}\text{Pb}$ : 5 Bq/l and  $^{228}\text{Ra}$ : 3 Bq/l.

### 3.4 Derived normalised releases

The normalised annual releases of natural radionuclides are derived from the annual production rates of oil and gas on the continental shelves of the four countries, on the reference values for the ratio between produced water and oil and gas respectively and on the reference nuclide concentrations in produced water from oil and gas production. The results do not represent actual releases in specific production fields or from individual platforms but are estimates of the collective releases by all production installations over the years of the development of the oil and gas production. The results are presented below in figures 9 – 12.

#### 3.4.1 The Dutch continental shelf

The estimated annual releases of natural radionuclides from oil and gas production on the Dutch continental shelf are presented in figure 9. The releases of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  largely originate from oil production, even with the relative small annual productions on the Dutch part of the continental shelf. This is shown in the figure by the strong increase of the discharges when oil production started in the early eighties. The maximum derived release rate is about  $0.1 \text{ TBq y}^{-1}$  for both  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ . The derived release rates for  $^{210}\text{Pb}$  are much lower in all years.

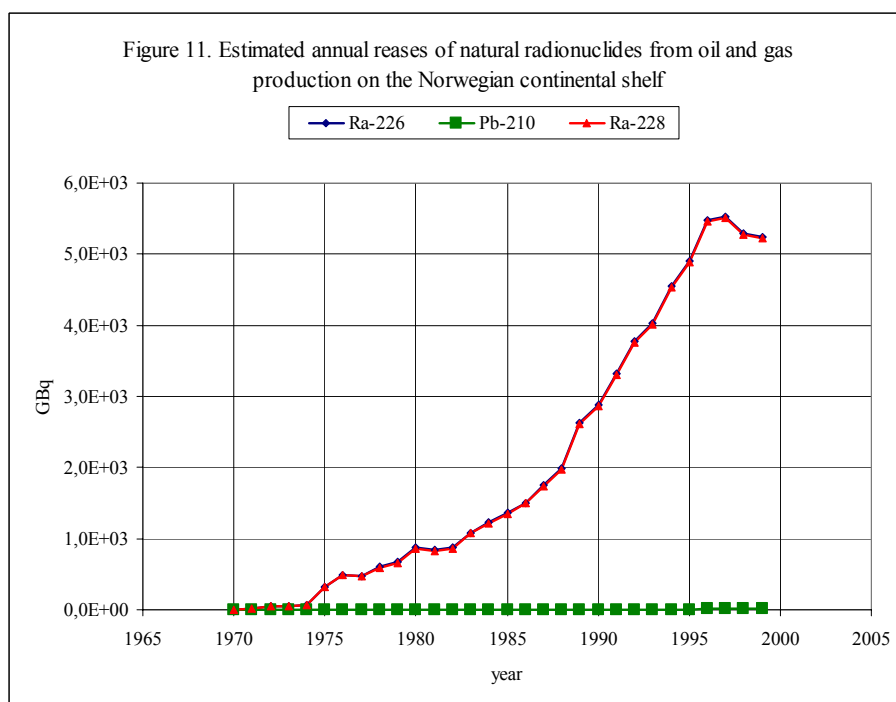
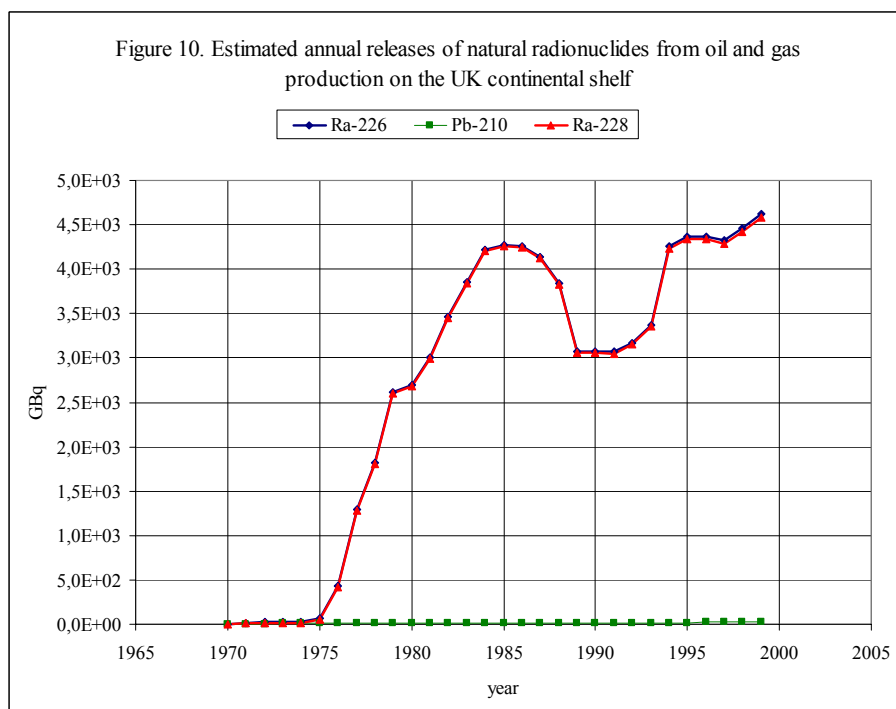


### 3.4.2 The UK continental shelf

The results for the UK continental shelf are shown in figure 10. The annual releases of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  are directly related to the trends in oil production rate which gradually increased to a peak of 140 million  $\text{m}^3$  in the mid-eighties, dropped to about 100 million  $\text{m}^3$  in 1989 – 1991 and increased to 152 million  $\text{m}^3$  in 1999. The derived annual releases of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  range between 3.0 and 4.6  $\text{TBq y}^{-1}$  between 1985 and 1999.

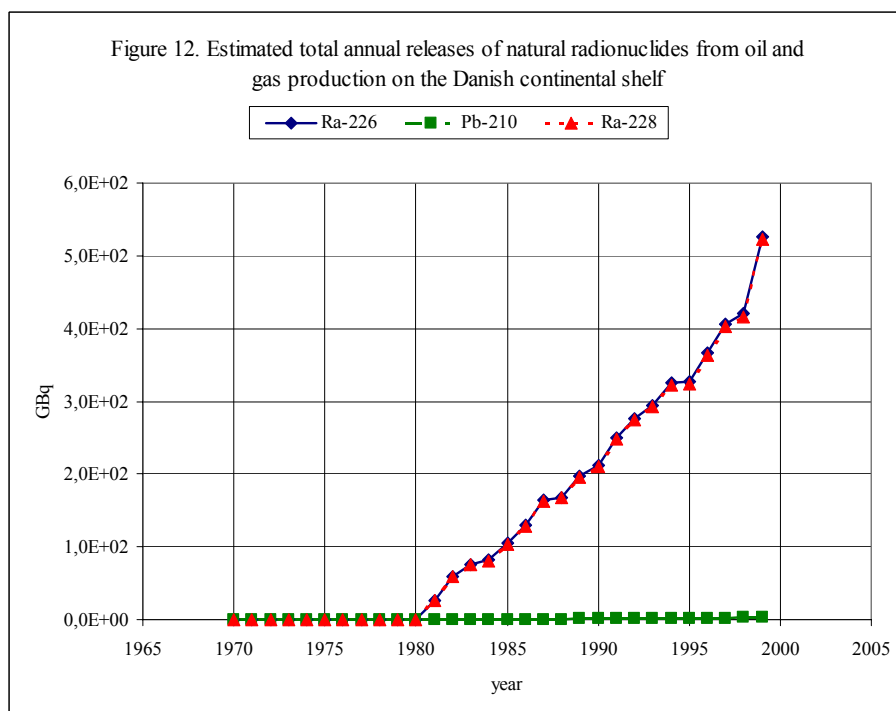
### 3.4.3 The Norwegian continental shelf

Figure 11 shows the results for the Norwegian continental shelf. Gradually increasing derived annual releases are related to the increase in oil production rate. Maximum derived annual releases of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  by all platforms of 5.5  $\text{TBq y}^{-1}$  were reached in 1997. The much lower releases of  $^{210}\text{Pb}$  are related to gas produced with a much lower water/gas production ratio.



### 3.4.4 The Danish continental shelf

The derived annual release rates of natural radionuclides from oil and gas production on the Danish continental shelf are shown in figure 12. They reflect the later start of offshore production compared with the other countries. The maximum release rate of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  of about  $0.5 \text{ TBq y}^{-1}$  was reached in 1999.



## 4. Radionuclide inputs into the boxes of the Marina II model

### 4.1 Time resolution of source data

The derived source data are not based on the actual release data. Therefore, the time dependency is derived in an attempt to approximate the actual time series data to the total releases from a large number of individual release points.

### 4.2 Geographic resolution of source data

The input from Working Group A into Working Group D requires a format compatible with the Marina II compartment model. However, each of the many individual sources of releases of natural radionuclides from oil and gas production on the continental shelf has its own history of production. At some fields the production has been started and ended within the period considered, other fields have been developed more recently and have not yet reached their production peak. Consequently the geographical distribution of productions and associated releases is changing in time and place. Therefore, the geographical distribution of annual releases can only be estimated with low resolution, e.g. for relative large boxes in the Marina II model. Fortunately, the relevant boxes of the model are relatively large. All releases from oil and gas production on the continental shelf are restricted to the boxes 55, 56, 57 and 59 of the North Sea.

### 4.3 Inputs into the boxes

Ideally the inputs in the different boxes can be derived from the registered production figures for oil and gas for all fields within each box. This would involve collection of production data for all fields and all years but would in the end still have limited use because of the fact that the release data still have to be derived on reference figures on discharges of produced water and reference nuclide concentrations. For the present purpose the same rough estimates

of the production of oil and gas within a box as a fixed fraction of the total production by a country have been used for all years. This assumed distribution is as follows:

**The Netherlands:** all releases from offshore oil and gas production in box 56.

**United Kingdom:**

Box 55: releases from 20% of national offshore gas production,

Box 57: releases from 20% of national offshore gas production and 30% of offshore oil production,

Box 59: releases from 60% of national offshore gas production and 70% of offshore oil production,

**Norway:**

Box 57: releases from 20% of national offshore gas production and 20% of offshore oil production,

Box 59: releases from 80% of national offshore gas production and 80% of offshore oil production,

**Denmark:** all releases from offshore oil and gas production in box 57

Only when calculated doses per unit input rate differ very considerably between boxes the feasibility of a more detailed assessment of the inputs should be considered.

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European Commission

## **MARINA II**

**Update of the MARINA Project on the radiological exposure of  
the European Community from radioactivity in North European  
marine waters**

### **Annex B: Environmental Data**



# **ENVIRONMENTAL DATA**

## **Report of Working Group B**

**By**

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Figure 29	Temporal variation of naturally occurring radionuclides in biota
Figure 30	$^{137}\text{Cs}$ in surface waters of European seas (1976-1980).

Figure 31  $^{137}\text{Cs}$  in surface waters of European seas (1981-1985).  
Figure 32  $^{137}\text{Cs}$  in surface waters of European seas (1986-1990).  
Figure 33  $^{137}\text{Cs}$  in surface waters of European seas (1991-1995).

# 1 Introduction

The Northern European Seas have received inputs of anthropogenic radionuclides from different sources, mainly:

- (i) Global fallout from atmospheric nuclear weapons testing,
- (ii) Liquid discharges from European nuclear reprocessing plants at Sellafield and Cap de la Hague, and
- (iii) Fallout from the accident at Chernobyl.

In addition to these sources, nuclear power plants, fuel production facilities, nuclear research facilities, and dumping of low-level radioactive waste in the deep Northeast Atlantic Ocean may contribute to the contamination of the immediate environment.

Information on radioactivity in North European waters is collated and discussed in this section. It will then be used to validate the dispersion model and to evaluate the radiation doses to members of the European population exposed to radioactivity through marine pathways. For these purposes, the main anthropogenic radionuclides and naturally occurring radionuclides in seawater, fish, shellfish, seaweed and sediment are included.

The original MARINA project (1990) examined information on marine radioactivity in Northern Europe in the period prior to 1985. The data included in this report are therefore focused on post-1986. For comparison and model validation, some longer time series of data are also included.

The geographical scope for this project is the North European waters covered by the OSPAR Convention (The Convention for the protection of the Marine Environment of the North-East Atlantic) including inputs from the Mediterranean and Baltic Seas into the North-East Atlantic. All data were compiled in the geographical sub-regions (model compartments) defined by Working Group 4 (Figure 1) and summarised by annual maxima and minima, geometric mean and average concentrations, and number of observations.

The radionuclides considered include those of importance for radiological exposure, such as  $^{210}\text{Po}$ ,  $^{137}\text{Cs}$ ,  $^{99}\text{Tc}$ ,  $^{90}\text{Sr}$ ,  $^{238}\text{Pu}$ ,  $^{239+240}\text{Pu}$  and  $^{241}\text{Am}$ , and those that are less important radiologically such as  $^{129}\text{I}$ ,  $^{60}\text{Co}$ ,  $^3\text{H}$  and  $^{106}\text{Ru}$ . Some elements (e.g. Sr, Cs and Tc) behave conservatively in seawater (i.e. are highly soluble), whereas others (e.g. Co, Pu and Am) have short residence times in the water column due to removal caused by scavenging processes.

The sources of data used here include databases, international reports and journal publications. The databases comprise the Glomard database from The Marine Environment Laboratory of the International Atomic Energy Agency, the Irish database from the University College Dublin and the Radiological Protection Institute of Ireland (RPII), the BNFL database from British Nuclear Fuels plc, the database

from the Nord-Cotentin Study and a database from the European Commission MAST-52 project. The reports are from national and international studies (e.g. IASAP, MARDOS, Nord-Cotentin, ARMARA) and laboratories (the Centre for Environment, Fisheries and Aquaculture Science, BNFL, RPII, Risø National Laboratory and Norwegian Radiation Protection Authority).

## **2 Caesium-137 in the North European Waters**

Caesium-137 has a half-life of 30 years and has been dispersed globally due to releases from the nuclear industry since the 1950's and atmospheric nuclear weapons testing in the 1960's. Caesium-137 is the most widely measured anthropogenic radionuclide in North European waters because of its abundance and ease of measurement, and it is a major component in radiation exposure from marine pathways. The main sources of this nuclide to European waters are releases from the Chernobyl accident, reprocessing discharges from Sellafield and fallout due to atmospheric nuclear weapons testing. The Irish Sea has shown the highest observed levels of  $^{137}\text{Cs}$  in north European waters due to historic discharges from the Sellafield reprocessing plant. Since 1985 discharges of  $^{137}\text{Cs}$  from Sellafield have remained about 10-20 TBq a<sup>-1</sup>, which is considerably lower than the discharges during the 1970's peaking in 1975 at 5200 TBq a<sup>-1</sup>. But the concentrations in the water in the Irish Sea are still relatively high (above 100 Bq m<sup>-3</sup> along the Cumbrian coast) due to remobilization from the seabed, which holds significant amounts of  $^{137}\text{Cs}$  from earlier releases (Cook et al. 1997, Hunt & Kershaw, 1990, Morris et al. 2000). The  $^{137}\text{Cs}$  level in the Baltic Sea is also high (above 50 Bq m<sup>-3</sup>) due to the contribution of Chernobyl  $^{137}\text{Cs}$  and the limited water exchange between the Baltic Sea and the North Sea.

### **2.1 Seawater**

The concentrations of  $^{137}\text{Cs}$  in seawater are listed in Table 1. The areas can be classified into four groups according to geographic location and concentration level of  $^{137}\text{Cs}$ :

- (i) The Arctic and North Atlantic area, which includes the Arctic Ocean, the Barents Sea, the Norwegian Sea and the North Atlantic (compartment no. 16-27).
- (ii) English Channel area, which includes the English Channel, the Bay of Biscay, the west coast of Portugal and the Celtic Sea (compartment no. 38-54).
- (iii) The North Sea area, which includes the North Sea, the Skagerrak and the Kattegat (compartment no. 55-62).
- (iv) The Irish Sea area, which includes the Irish Sea, the North Channel, the north and west Scottish waters and the west and south Wales waters (compartment no. 28-37).

In the Arctic area, lower  $^{137}\text{Cs}$  levels are observed. The concentrations of  $^{137}\text{Cs}$  in surface seawater were less than  $10 \text{ Bq m}^{-3}$  in the period 1987 to 1999, except for a higher  $^{137}\text{Cs}$  level (approx.  $10 \text{ Bq m}^{-3}$ ) in the Norwegian coastal water (compartment 27) in 1987, which was due to Chernobyl fallout. Since 1995, even lower levels of around  $5 \text{ Bq m}^{-3}$  have been observed in most of this area. There are multiple sources of  $^{137}\text{Cs}$  to this area: reprocessing discharges, fallout from the Chernobyl accident and fallout from atmospheric nuclear weapons testing. Since 1986, the Chernobyl fallout has been dominating.

Low  $^{137}\text{Cs}$  levels, less than  $10 \text{ Bq m}^{-3}$ , were observed in the English Channel area after 1987, even close to the discharge point of the Cap de la Hague reprocessing plant. A slight decline in  $^{137}\text{Cs}$  levels was observed after 1992 (Figure 2) to less than  $4 \text{ Bq m}^{-3}$  of  $^{137}\text{Cs}$  in seawater, which is similar to levels in the Arctic (compartment 16), but higher than observed in south Arctic waters (compartment 17). This occurred directly as a result of the reduction in Cap de la Hague discharges of  $^{137}\text{Cs}$ , since Chernobyl deposition had a minor impact in this area.

Slightly higher  $^{137}\text{Cs}$  levels were observed in the North Sea area (compartments 55 to 59) before 1990 than after, as a result of fallout from the Chernobyl accident. The temporal variation of  $^{137}\text{Cs}$  concentrations (Figure 2) shows a declining trend over the North Sea area in the period of interest, especially during 1986 to 1990. After 1990, there is only a slight decline, which was only observed in the eastern and northern regions (compartments 56, 58 and 59). In the Skagerrak and Kattegat, much higher and almost constant concentrations of  $15$  and  $60 \text{ Bq m}^{-3}$ , respectively, have been observed since 1989. These higher levels are due to the outflow of Chernobyl  $^{137}\text{Cs}$  from the Baltic Sea. The temporal variations of  $^{137}\text{Cs}$  concentrations in the Kattegat surface and deep water are shown in Figure 2. Before 1986, higher  $^{137}\text{Cs}$  levels were found in the deep waters, while after 1986 the higher levels have been found in surface waters. This is because the low salinity surface water in the Kattegat comes from outflowing water from the Baltic Sea, which is contaminated by Chernobyl fallout, while the high salinity water from the North Sea enters the deeper layers of the Kattegat.

A great number of data is available from the Irish Sea. A long time series of these data from 1972 to 2000 has been compiled in Table 1. Highest concentrations of  $^{137}\text{Cs}$  have been observed in this area, especially in the east Irish Sea (compartment no. 32, 35 and 37) where, up to 2001, levels above  $200 \text{ Bq m}^{-3}$  were measured. In this area, the highest levels of  $^{137}\text{Cs}$  are found in compartment 35, which is the point of discharge from the Sellafield reprocessing plant. The concentration of  $^{137}\text{Cs}$  decreases with increasing distance from the Sellafield. The temporal variation of the  $^{137}\text{Cs}$  concentrations (Figure 2) showed a rapid decline in the 1980's. Since 1990 the rate of reduction has slowed down and, especially in recent years, the  $^{137}\text{Cs}$  levels have remained almost constant. The geographical distribution of  $^{137}\text{Cs}$  and its variation with time in seawater result from the current discharges from the Sellafield reprocessing plant and the effect of previous discharges which became associated with seabed sediments and are now being remobilized into the water column. This is confirmed by the measurements of  $^{137}\text{Cs}$  in the sediment in this region, which show a decreasing trend with time (Figure 7).

Figures 30-33 show the distribution of and changes in  $^{137}\text{Cs}$  concentrations in surface waters of Northern European Seas over 5-year periods, 1976-1980, 1981-1985, 1986-1990 and 1991-1995, respectively (Povinec & Scotto, 2001). It can be seen that  $^{137}\text{Cs}$  originating from Sellafield has been transported to the Arctic Seas. Similarly, for the time intervals 1986-1990 and 1991-1995, the primary source of  $^{137}\text{Cs}$  concentration in the Baltic Sea was the Chernobyl accident. In addition,  $^{137}\text{Cs}$  concentrations have decreased markedly in Northern European waters since the middle of the 1980's as a consequence of decreased discharges from Sellafield.

Table 2 and Figure 3 compare  $^{137}\text{Cs}$  data from different sources. There is good agreement between the data sets from BNFL and MAFF+IAEA between 1993 and 1999. Before 1992, the data from BNFL are given as estimated maximum values (less-than values), which are higher than those from the IAEA database by a factor of 2. In the report from the Nord-Cotentin Study, four laboratories reported  $^{137}\text{Cs}$  levels in the coastal water from Cap de la Hague area, but only one laboratory (Commissariat A L'Energie Atomique, CEA) supplied measured values, most data from the other 3 laboratories were less-than values. Comparison of results from these laboratories shows that the lowest values are those reported by CEA. Table 2 and Figure 3 also compare the  $^{137}\text{Cs}$  results from the CEA and from the IAEA database. After 1986, good agreement was observed between the data sets, while before 1986, CEA values are higher by a factor of 2-10. This is probably due to the use of different sampling locations: CEA took samples of near-coastal water, while the results in the IAEA database include samples from offshore locations.

## 2.2 Biota

Consumption of marine organisms, particularly fish and shellfish, is the main pathway by which populations are exposed to marine radioactivity. Some sea plants are consumed but this is not common in Europe. Seaweed can be used as a bioindicator to monitor radioactivity in seawater. The data available on  $^{137}\text{Cs}$  concentrations in biota are much less abundant than for seawater. The information on concentrations of  $^{137}\text{Cs}$  in fish and shellfish over the period of 1982-1999 has been collected and sorted by model compartments according to the sampling locations. The annual maximum and minimum values, geometric mean and average as well as the number of data are tabulated (Tables 3 and 4). The  $^{137}\text{Cs}$  concentrations in fish and shellfish are not very different and not very high, which shows there is a relatively low bioaccumulation of  $^{137}\text{Cs}$  by marine biota. With the exception of the Irish Sea and the Kattegat, the concentrations of  $^{137}\text{Cs}$  in fish and shellfish are lower than  $1 \text{ Bq kg}^{-1}$  wet weight (ww) over the observation period (1988-1999), even in the Cap de la Hague area. As for seawater, the highest  $^{137}\text{Cs}$  levels in fish and shellfish occur in the Sellafield area, in the east Irish Sea. Similar levels of  $^{137}\text{Cs}$  were also observed in the Kattegat after 1992. In the east Irish Sea, a significant decline in  $^{137}\text{Cs}$  concentrations is observed over the period of reporting (Figure 4), while the concentrations in the Kattegat are increasing in this period.

Concentrations of  $^{137}\text{Cs}$  in seaweed were found to be similar to those in fish and shellfish from the same sampling location and date (Table 6). The Glomard database (IAEA) gives no information on which biota were analysed for  $^{137}\text{Cs}$ , but because no significant differences were found between  $^{137}\text{Cs}$  concentrations in fish, shellfish and

seaweed, the data from IAEA database were collected and analysed (Table 5). Good agreement was observed (within a factor of 1.5) when comparing these data with BNFL data from the east Irish Sea (compartment 35). Thus, these data can be used as the concentrations in fish or shellfish for the estimation of radiation doses.

A slight decline in the  $^{137}\text{Cs}$  concentration in seaweed samples was also observed in most locations (Figure 5). The contribution of the Chernobyl deposition was clearly seen in the samples from the west Norwegian coast. On the southwest coast, at Utsira, a higher concentration occurred in the middle of 1986, just few months after the Chernobyl accident. This took until the middle of 1997 to reach Ingøy and Indre Kibery on the northwest coast of Norway, showing an approximate transfer time of one year.

Figure 6 compares the data taken from BNFL, MAFF and RPII, and very good agreement is seen for both fish and shellfish concentrations.

### 2.3 Sediment

Caesium is known to behave conservatively (low particle reactivity and so resistant to removal from the water column) in the oceans, with a relatively low transfer to the sediments. However, where high concentrations of  $^{137}\text{Cs}$  are found in seawater, proportionally high concentrations of  $^{137}\text{Cs}$  are observed in the sediments. In the Irish Sea near Sellafield, sediment concentrations in excess of 1 kBq kg<sup>-1</sup> dry weight (dw) of  $^{137}\text{Cs}$  are observed. The emission of  $\gamma$ -rays from the decay of  $^{137}\text{Cs}$  thus contributes to the external exposure of radiation to individuals who spend time on coastlines.

Tables 7a give concentrations of  $^{137}\text{Cs}$  in sediments in different model boxes and the temporal variation. Table 7b shows the  $^{137}\text{Cs}$  inventory in the North Sea.

A significant, declining trend in  $^{137}\text{Cs}$  concentrations in the sediment from the east Irish Sea over the period 1996-2001 was observed (Figure 7). This indicates that desorption of  $^{137}\text{Cs}$  dominates the reaction between water and sediment in this period. The  $^{137}\text{Cs}$  previously discharged to the Irish Sea from the Sellafield reprocessing plant and absorbed on the sediment is remobilized to the water column. This is also why the seawater concentrations of  $^{137}\text{Cs}$  in the Irish Sea in recent years have not followed the same declining trend as the discharges from the reprocessing plant.

## 3 Technetium-99 in the North European Waters

While marine discharges of most other radionuclides have declined in recent years, discharges of  $^{99}\text{Tc}$  from the Sellafield reprocessing plant increased in 1994 due to the operation of the Enhanced Actinide Removal Plant (EARP) at Sellafield. This has prompted public attention to be focused on this radionuclide because  $^{99}\text{Tc}$  is an anthropogenic element and accumulated in seafood, especially crustaceans (lobsters) and seaweed. Furthermore,  $^{99}\text{Tc}$  is a conservative nuclide and, therefore, used as an oceanographic tracer. Information on  $^{99}\text{Tc}$  concentrations in seawater and biota are, therefore, collected and analysed in this work.

### 3.1 Seawater

An EC project MAST-52 investigated the concentrations of  $^{99}\text{Tc}$  in seawater from the English Channel to the Kattegat and the south-western Norwegian coast along the European coast in 1983-1993. The concentrations of  $^{99}\text{Tc}$  in seawater are listed in Table 8 and shown in Figure 8 according to the model boxes. The highest  $^{99}\text{Tc}$  levels were found in Gourey in the English Channel; in the investigated area, an annual average concentration of  $23 \text{ Bq m}^{-3}$  of  $^{99}\text{Tc}$  was observed in 1985.  $^{99}\text{Tc}$  levels declined up the west European coast, so that concentrations along the south Norwegian coast and in the Kattegat were at maximum  $1.6$  and  $0.7 \text{ Bq m}^{-3}$ , respectively. This is attributed to the discharge of  $^{99}\text{Tc}$  from the Cap de la Hague reprocessing plant and its transport northwards along the west European coast. A significant decline in  $^{99}\text{Tc}$  levels was observed at all locations from 1986 to 1993, resulting from a peak discharge from the Cap de la Hague reprocessing plant in 1985 followed by reduced discharges in the subsequent years.

In the Arctic and North Atlantic seawater, levels of  $0.02$ - $0.2 \text{ Bq m}^{-3}$  of  $^{99}\text{Tc}$  were observed in 1986-1994. The lowest  $^{99}\text{Tc}$  levels found were  $0.005 \text{ Bq m}^{-3}$  in 1992 in Atlantic water, which is considered to be the background  $^{99}\text{Tc}$  concentration due to fallout from the atmospheric nuclear weapons testing.

In 1986-1994, the  $^{99}\text{Tc}$  levels in the Irish Sea were not high, even in the east Irish seawater. Levels of  $5$ - $20 \text{ Bq m}^{-3}$  of  $^{99}\text{Tc}$  were observed here in 1992-1993, and  $0.2$ - $0.6 \text{ Bq m}^{-3}$  in the Scottish water. Since 1994, the  $^{99}\text{Tc}$  levels have increased significantly; in 1996,  $2000 \text{ Bq m}^{-3}$  of  $^{99}\text{Tc}$  near Sellafield,  $60 \text{ Bq m}^{-3}$  in the North Channel, and  $5$ - $10 \text{ Bq m}^{-3}$  in the northwest North Sea were observed. In 1996-1997,  $^{99}\text{Tc}$  concentrations in southwest Norwegian seawater were  $1$ - $6 \text{ Bq m}^{-3}$  of  $^{99}\text{Tc}$ , while in 1993,  $^{99}\text{Tc}$  concentrations in this area were only around  $0.4 \text{ Bq m}^{-3}$ . This is attributed to the increase in  $^{99}\text{Tc}$  discharges from the Sellafield reprocessing plant since 1994. Figure 8 shows the temporal variation of  $^{99}\text{Tc}$  concentrations in seawater from different locations.

Figure 9 compares the analytical results of  $^{99}\text{Tc}$  in seawater by three laboratories: Risø, Bundesamt für Seeschifffahrt und Hydrographie (BSH) and Commissariat A L'Énergie Atomique (CEA), their data agree well.

### 3.2 Biota

Technetium-99 is accumulated by marine organisms with concentration factors as high as  $10^5$  for seaweed (dry-weight basis). Doses to man from  $^{99}\text{Tc}$  arise through consumption of shellfish. The ranges and mean concentrations of  $^{99}\text{Tc}$  in shellfish and fish are listed in Tables 9-11. The mean level of  $^{99}\text{Tc}$  in shellfish is about two orders of magnitude higher than that in fish, even in 1993, concentrations as high as  $30$ - $50 \text{ Bq kg}^{-1}$  ww  $^{99}\text{Tc}$  were observed in the Sellafield area in the Irish Sea. In shellfish, the highest  $^{99}\text{Tc}$  concentration was found in lobster.

As for seawater,  $^{99}\text{Tc}$  levels in shellfish started to increase in 1994 in the Irish Sea and Scottish waters, but the highest concentrations occurred in 1997 in the Irish Sea and in



1998 in the Scottish water, which was 1-2 years after the maximum concentrations in the seawater. Since then, a declining trend has been observed (Figure 10).

Besides shellfish,  $^{99}\text{Tc}$  is highly accumulated by seaweed, such as *Fucus vesiculosus*, which is therefore used as a very sensitive bioindicator to monitor  $^{99}\text{Tc}$  in the water. The same temporal variation of  $^{99}\text{Tc}$  levels in seawater and seaweed were observed in the sampling locations in Goury, Utsira, Klint as well as Sellafield (Figure 11). This figure also shows the varying levels of  $^{99}\text{Tc}$  in seaweed from other locations in the North Sea and along the Norwegian coast.

## **4 Strontium-90 in North European Waters**

$^{90}\text{Sr}$  is a major anthropogenic radionuclide released from reprocessing plants and nuclear weapons testing. However, there are fewer data available on  $^{90}\text{Sr}$  in marine systems than  $^{137}\text{Cs}$ , mainly due to the relative difficulty of measuring  $^{90}\text{Sr}$ , a pure beta emitter. The annual means and ranges of  $^{90}\text{Sr}$  concentrations in European waters are listed in Tables 12 to 14.

### **4.1 Seawater**

The distribution of  $^{90}\text{Sr}$  in seawater is similar to  $^{137}\text{Cs}$ . The lowest levels were observed in the Arctic and North Atlantic water, where the annual mean values were less than  $3 \text{ Bq m}^{-3}$  in the North Atlantic (Compartment 17) and less than  $9 \text{ Bq m}^{-3}$  in the Arctic (compartment 16) during 1977-1995. The concentrations of  $^{90}\text{Sr}$  in the English Channel are slightly higher than those observed in the Arctic; at a level of  $2\text{-}10 \text{ Bq m}^{-3}$  between 1987 and 1997, but lower than those observed in the North Sea, where  $5\text{-}30 \text{ Bq m}^{-3}$  of  $^{90}\text{Sr}$  was observed in the same period. The highest levels of  $^{90}\text{Sr}$  are reported in the Irish Sea by BNFL; the annual means in the east Irish Sea (compartment 35) range between  $200\text{-}400 \text{ Bq m}^{-3}$ , but most of these data are 'less than values'.

The temporal variations of  $^{90}\text{Sr}$  level in European waters are shown in Figure 12. In the Arctic and English Channel, no significant change of  $^{90}\text{Sr}$  concentrations was observed in the investigation period, whereas in the North sea, a clear decline of  $^{90}\text{Sr}$  level was observed from  $\sim 30 \text{ Bq m}^{-3}$  in 1984 to  $\sim 5 \text{ Bq m}^{-3}$  in 1997. This results from the reduced discharges from the reprocessing plant at Sellafield.

### **4.2 Biota**

Concentrations of  $^{90}\text{Sr}$  in fish and shellfish collected in the Irish Sea have been measured by MAFF and BNFL respectively. The  $^{90}\text{Sr}$  concentration in fish is the lowest, followed by crustaceans and the highest level was observed in molluscs, especially in winkles. BNFL reported higher concentrations of  $^{90}\text{Sr}$  before 1992, but most of these are 'less than values'. In general, there is a slight decline in biota  $^{90}\text{Sr}$  concentrations in the Irish Sea during 1992-2001 (Figure 13). There is good agreement between the data from BNFL and MAFF (Table 13).

## 5 Plutonium Isotopes in North European Waters

Plutonium isotopes are important anthropogenic radionuclides. They can contribute significantly to a radiation dose when consumed, because of their high Sv/Bq dose factor. Due to the high particle reactivity of plutonium (it is readily removed from the water column to the sediment), most of the Pu discharged from the Sellafield reprocessing plant is associated with Irish Sea sediments, and very little has been transferred out of the Irish Sea. However, a recent investigation suggested that some of this sediment-bound plutonium could be remobilised (Cook et al. 1997).

Most of plutonium isotopes are alpha emitters, of these  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  emit alpha-particles with very similar energies, that cannot be clearly resolved from each other in alpha spectrometry, which is the most common method for measuring plutonium isotopes. As a result of this,  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  activities are mostly reported as one summed value. In this work, data on the most important plutonium isotopes,  $^{238}\text{Pu}$  and  $^{239+240}\text{Pu}$ , were therefore collected and analysed.

### 5.1 Seawater

The annual means and ranges of  $^{239+240}\text{Pu}$  concentrations in European waters are listed in Table 15. The highest level was observed in the east Irish Sea (compartment 35) at a level of  $3\text{--}9\text{ Bq m}^{-3}$  in 1988-1989, and  $\sim 0.6\text{ Bq m}^{-3}$  in 1995. In the west Irish Sea (compartment 33) and North Channel (compartment 28), it decreases to  $0.03\text{--}0.25\text{ Bq m}^{-3}$ .

Figure 14 shows the distribution of dissolved  $^{239+240}\text{Pu}$  in the surface water of the Irish Sea from 1988 to 1996; the highest concentration was found in the coastal water nearest Sellafield, and the level rapidly decreases with increasing distance from the discharge point. In addition,  $^{239+240}\text{Pu}$  concentrations in seawater declined from 1988 to 1996. This is due to the reduction in discharges of these nuclides in this period. But the reduction in discharges is counteracted by remobilisation of plutonium isotopes from the sediments, similar to the situation for radiocaesium. The  $^{239+240}\text{Pu}$  concentrations in the English Channel and south North Seas are similar and are an order of magnitude lower than in the Irish Sea, at a level of  $0.003\text{--}0.04\text{ Bq m}^{-3}$  from 1989-1997. Less than  $0.01\text{ Bq m}^{-3}$  of  $^{239+240}\text{Pu}$  was observed in the northeast North Sea and Skagerrak. A slight decline in  $^{239+240}\text{Pu}$  concentrations was also observed in locations outside Irish Sea.

### 5.2 Biota

The concentrations of  $^{239+240}\text{Pu}$  in fish and shellfish are given in Tables 16, 17 and 19. It can be seen that the concentrations of  $^{239+240}\text{Pu}$  in shellfish are almost 2 orders of magnitude higher than those in fish. In shellfish, the level in molluscs is more than 10 times higher than that in crustaceans. As seen for seawater, the highest level was observed in the east Irish Sea, especially in the Sellafield water (compartment 35), where  $3\text{--}7\text{ Bq kg}^{-1}\text{ ww}$  of  $^{239+240}\text{Pu}$  was measured during 1988-1999, while in the North Sea, concentrations were only  $\sim 0.01\text{ Bq kg}^{-1}\text{ ww}$ . No significant trends in the  $^{239+240}\text{Pu}$  concentrations in biota can be seen over the investigation period (Figure 15).

The concentrations of  $^{238}\text{Pu}$  are lower than  $^{239+240}\text{Pu}$ , but a similar variation and distribution is seen.

### 5.3 Sediment

The concentrations of  $^{239+240}\text{Pu}$  and  $^{238}\text{Pu}$  in sediments are given in Tables 18 and 20. The observations are similar to those for seawater and biota; a high concentration was observed in the east Irish Sea, at a level of 500-1000 Bq  $^{239+240}\text{Pu}$  kg<sup>-1</sup> dw, and 100-220 Bq  $^{238}\text{Pu}$  kg<sup>-1</sup> dw. In the west Scottish water, they are lower at 5-12 Bq  $^{239+240}\text{Pu}$  kg<sup>-1</sup> dw and 0.8-2.4 Bq  $^{238}\text{Pu}$  kg<sup>-1</sup> dw. The levels in the Barents Sea are almost 3 orders of magnitude lower than those in the Irish Sea, only 1.2 Bq  $^{239+240}\text{Pu}$  kg<sup>-1</sup> dw and 0.06 Bq  $^{238}\text{Pu}$  kg<sup>-1</sup> dw.

Figure 17 shows the temporal variation of plutonium isotopes in sediment from the east Irish Sea, both  $^{239+240}\text{Pu}$  and  $^{238}\text{Pu}$  show a declining trend over the period of 1988-2000. The distribution of  $^{239+240}\text{Pu}$  in surface sediment in the Irish Sea collected in 1983, 1988 and 1995 is shown in Figure 18, movement of plutonium away from the discharge point can be observed over this period. This indicates the remobilisation of plutonium in the sediment occurred.

Table 21 and Figure 18 show the inventories of  $^{239+240}\text{Pu}$  and  $^{238}\text{Pu}$  in the sediment in the North Sea, a higher inventory was observed in the coastal area than in the middle of North Sea.

## 6 Other Anthropogenic Radionuclides

The reprocessing of spent fuel elements and nuclear weapons testing release many additional radionuclides into the environment. In this work, the data for some of these, including  $^{241}\text{Am}$  as another transuranium element,  $^{60}\text{Co}$  as an activation production, and  $^3\text{H}$ ,  $^{106}\text{Ru}$  and  $^{129}\text{I}$  as fission products, are collected and analysed.

### 6.1 Americium-241

Americium-241 is another important alpha-emitting nuclide in terms of radiation doses from marine radioactivity, due to a high dose factor. It is an important contributor to the doses received by the critical group in the Sellafield area of the east Irish Sea. Figure 19 shows the distribution of the dissolved  $^{241}\text{Am}$  in the surface water of the Irish Sea. The distribution is the same as for plutonium and other radionuclides, with the highest levels occurring in the east Irish Sea, especially in the Sellafield coastal water. However, the difference in the concentrations of  $^{241}\text{Am}$  in the east Irish Sea and other locations in the Irish Sea are not as large as for plutonium. This is related to the higher solubility (or conservation) of  $^{241}\text{Am}$  in seawater compared to plutonium. The concentration of  $^{241}\text{Am}$  in shellfish (Table 22 and 23) ranges from 8 to 17 Bq kg<sup>-1</sup> ww in the east Irish Sea, which is higher than that for  $^{239}\text{Pu}$  by a factor of 2. The concentrations of  $^{241}\text{Am}$  in fish are much lower than those in shellfish, and the level in crustaceans is lower than those in molluscs, which is a similar observation to that for Pu. No clear temporal variation of  $^{241}\text{Am}$  concentrations in biota was observed in most locations over the investigation period, except for a decline in the east Irish Sea (Figure 20).

The concentration of  $^{241}\text{Am}$  in east Irish Sea sediment was observed to decline over time, which indicates a similar remobilisation of  $^{241}\text{Am}$  to that for plutonium in the Irish Sea. However, the reduction of  $^{241}\text{Am}$  levels by remobilisation is counteracted by build-up due to decay of  $^{241}\text{Pu}$  in the sediments. The inventory of  $^{241}\text{Am}$  in the North Sea sediment is shown in Figure 20. As for plutonium, higher inventories occur in the coastal areas and the inventory of  $^{241}\text{Am}$  is similar to that for  $^{239+240}\text{Pu}$  in this area.

## **6.2 Cobalt-60**

The level and temporal variation of  $^{60}\text{Co}$  concentrations in the shellfish are shown in Figure 21. Higher levels are observed in the east Irish Sea compared with those observed in north Irish Sea and in the North Sea.

The lowest concentrations of  $^{60}\text{Co}$  in the Irish Sea were measured in 1993-1994. In 1989-1993, a decline in the  $^{60}\text{Co}$  concentration of shellfish was observed, while after 1994, the concentration of  $^{60}\text{Co}$  increased continuously. In the North Sea, the level of  $^{60}\text{Co}$  decreased from  $\sim 2 \text{ Bq kg}^{-1} \text{ ww}$  in 1988 to  $0.5 \text{ Bq kg}^{-1} \text{ ww}$  in 1998.

## **6.3 Tritium, Ruthenium-106 and Iodine-129**

Much more tritium ( $^3\text{H}$ ) is released to the environment by nuclear activities than other anthropogenic radionuclides, but it represents less of a radiation risk as it has a low dose factor and undergoes no specific bioconcentration. The concentrations of  $^3\text{H}$  in seawater are given in Table 24. In the North Sea, its activity concentration is 2 orders of magnitude higher than that of  $^{137}\text{Cs}$ . The distribution of  $^3\text{H}$  in the Irish Sea (Figure 23) shows a clear contribution from the reprocessing plant at Sellafield, but the levels in the west Irish Sea are not higher than those observed in the North Sea. Recent measurements in the UK, noticeably in the Cardiff area, have shown elevated levels of organic bound tritium, which is still under investigation.

The temporal variation of  $^3\text{H}$  in North Sea water shows an increasing trend during 1988-1998, and higher levels of  $^3\text{H}$  were observed in European coastal water than in central North Sea water. No clear temporal variation of  $^3\text{H}$  in biota was observed in the Irish Sea. The depth profiles of  $^3\text{H}$  in the water columns of the Arctic and Atlantic oceans are also shown in Figure 22. It should be noted that  $^3\text{H}$  and  $^{14}\text{C}$  are naturally occurring radionuclides produced by the interaction of cosmic-ray particles with the atmosphere. Concentrations of naturally occurring  $^3\text{H}$  in surface waters measured before nuclear explosions began, were found to be in the range  $200\text{-}900 \text{ Bq m}^{-3}$  for continental waters (UNSCEAR, 1982).

The temporal variation of  $^{106}\text{Ru}$  concentrations in shellfish and sediment in the east Irish Sea are shown in Figure 24. A declining trend was observed in all materials over the period of 1985-2001.

There has been an increase in the discharges of  $^{129}\text{I}$  from the reprocessing plants at Cap de la Hague and Sellafield since 1991. However, the radiological risk from these

discharges is insignificant at the present levels. The highly conservative behaviour of  $^{129}\text{I}$  in the ocean makes it a useful oceanographic tracer for modelling oceanic flow.

Figure 25 shows the temporal variation of  $^{129}\text{I}/^{127}\text{I}$  ratios (as an indicator of the  $^{129}\text{I}$  level) in seaweed samples from different locations.  $^{129}\text{I}$  concentrations have increased since 1992-1993 along the southwest coast of Norway and in the Kattegat. Figure 26 shows the distribution of  $^{129}\text{I}$  concentrations in seawater around Denmark, concentrations up to  $5 \times 10^{-11} \text{ g/l}$  (or  $0.3 \text{ Bq m}^{-3}$ ) of  $^{129}\text{I}$  were observed on the west coast of Denmark, 2 orders of magnitude higher than found in the Baltic Sea.

The mean and ranges of  $^{129}\text{I}$  concentrations in fish and seaweed samples in the Irish Sea are given in Table 25. Concentrations as high as  $0.16 \text{ Bq } ^{129}\text{I kg}^{-1} \text{ ww}$  were observed in cod, while in the fucus samples,  $3\text{-}5 \text{ Bq kg}^{-1} \text{ dw}$  was measured in 2000-2001, which is about 5-10 times higher than that observed in Klint in the Kattegat in 1999.

Figure 27 shows the distribution of  $^{129}\text{I}$  in seawater along the Norwegian coast, a decreasing level of  $^{129}\text{I}$  was observed in a northerly direction along the Norwegian coast. Two depth profiles of  $^{129}\text{I}$  in the water column of the Greenland Sea are shown in Figure 28. They are similar to the profiles of  $^{137}\text{Cs}$  and  $^3\text{H}$ , with the highest levels occurring in the surface water, and concentrations decreasing with depth.

## 7 Naturally Occurring Radioactive Material (NORM)

Naturally occurring radionuclides, including  $^{40}\text{K}$  and nuclides from the uranium and thorium decay chains such as  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$ , are found throughout the seas and they are the dominant sources of radiation doses through ingestion of seafood. Polonium-210 is the most important nuclide in this group because it is found in relatively high concentrations in shellfish and, as an alpha-emitter, has a high dose factor. Collective doses to man from  $^{210}\text{Po}$  through marine pathways are about two orders of magnitude higher than from anthropogenic radionuclides.

$^{210}\text{Po}$  is a radionuclide which associates readily with sediment particles. The mean concentration of  $^{210}\text{Po}$  in global seawater is about  $1 \text{ Bq m}^{-3}$ , whereas the concentrations in crustaceans and molluscs are  $20 \text{ Bq kg}^{-1}$  and  $30 \text{ Bq kg}^{-1}$ , respectively. In some species, such as *Mytilus edulis*, much higher levels have been observed,  $100\text{-}300 \text{ Bq kg}^{-1}$  have been reported in European waters.

The presence of industrial activity, such as the phosphate industry, can result in enhanced concentrations of naturally occurring radionuclides in the marine environment. The Albright & Wilson plant at Whitehaven, Cumbria, UK has discharged waste arising from the production of phosphoric acid into the Irish Sea. This waste contains elevated levels of naturally occurring radionuclides. Enhanced levels of NORM have been observed in the Irish Sea.

Tables 26 and 27 list the concentration of NORM,  $^{210}\text{Po}$ ,  $^{210}\text{Pb}$ ,  $^{226}\text{Ra}$ ,  $^{228}\text{Th}$ ,  $^{230}\text{Th}$ ,  $^{232}\text{Th}$ ,  $^{234}\text{U}$ ,  $^{235}\text{U}$  and  $^{238}\text{U}$  in fish and shellfish collected in the Irish Sea, and Figure 29 shows their temporal variations. The concentrations of NORM in winkles (molluscs) are much higher than other shellfish, and the NORM level in fish is much lower than

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in shellfish. Of the various naturally occurring radionuclides,  $^{210}\text{Po}$  concentrations in biota are the highest. For thorium, no clear temporal variation was observed during 1991-1999. A slight decline in uranium and  $^{210}\text{Pb}$  concentrations occurs from 1991 to 1994, while a significant temporal variation of  $^{210}\text{Po}$  concentrations in fish and shellfish was observed. Before 1993, the annual mean  $^{210}\text{Po}$  concentrations in winkles collected from Parton in the east Irish Sea were higher than  $100 \text{ Bq kg}^{-1} \text{ ww}$ , from 1993 to 1999 they were lower than  $50 \text{ Bq kg}^{-1} \text{ ww}$ , and during 1994-1999 they were almost constant at about  $20\text{-}35 \text{ Bq kg}^{-1} \text{ ww}$ . This change is attributed to the reduction in  $^{210}\text{Po}$  discharges after 1993 due to the cessation of the use of phosphate ore in the plant. Tables 31 and 32 summarise the concentrations of  $^{210}\text{Po}$  found in shellfish in European waters.

Rollo et al. (1992) and McDonald et al. (1992) collected marine biota from around the UK in 1987-1990 and measured their NORM concentrations. The results are listed in Table 27, and a clearly enhanced level of NORM was observed along the Cumbrian coast. The  $^{210}\text{Po}$  concentrations at Saltom Bay, the closest sampling location to the Albright & Wilson phosphate processing plant, were enhanced by a factor of about 10 when compared with the areas remote to Whitehaven. However, the levels of NORM are not significantly different to samples collected in locations adjacent to other UK phosphate processing plants, aluminium, tin and lead smelts, titanium dioxide and steel plants. Seaweed (*Fucus vesiculosus*) samples were also collected and analysed for NORM; the concentrations of  $^{210}\text{Po}$  were generally lower than found in shellfish, while uranium isotopes are at higher concentrations in *Fucus* samples.

Besides the Albright & Wilson plant, large amounts of waste with high NORM levels have been released into the rivers and coastal water of Europe, particularly in the Netherlands and France. Koster et al. (1992) measured NORM in water and shellfish from the Dutch water and North Sea (Table 28). An enhanced NORM level was observed in the water from Nieuwe Waterweg and Vlissingen. The Schelde mouth also showed an enhanced level of NORM, this was due to the emission of phosphogypsum between Antwerp and the Dutch border. The concentrations of  $^{210}\text{Po}$  in mussels and shrimps collected in these locations are higher than those from Oosterschelde, where the lowest  $^{210}\text{Po}$  level was observed in water, but are not different to those from North Sea, Danish waters and Irish coast.

In France, certain factories near Rouen released phosphate gypsum into river Seine until 1974. Germain et al. (1992) collected a time series of mussels and seaweed samples from the Seine estuary and some 'background' locations, which are situated to the east (Wimereux) and to the west of the Seine estuary (Pointe de Moulard), the concentrations of  $^{210}\text{Po}$  were determined (Figures 29 and 30). A large variation in  $^{210}\text{Po}$  concentration in mussels ( $90\text{-}700 \text{ Bq kg}^{-1} \text{ dw}$ ) was observed from time to time, but the mean levels of  $^{210}\text{Po}$  ( $180\text{-}260 \text{ Bq kg}^{-1} \text{ dw}$ ) in the Seine estuary and the 'background' location (Wimereux) are not significantly different. The mean level in the Seine estuary is also not different to those observed on the Irish coast and in Danish waters. A less marked temporal variation of  $^{210}\text{Po}$  levels was observed in *Fucus* samples, but the mean activity of  $^{210}\text{Po}$  in *Fucus* from the Seine estuary ( $14\text{-}15 \text{ Bq kg}^{-1} \text{ dw}$ ) is higher than that observed in the 'background' locations in Wimereux ( $9.2 \text{ Bq kg}^{-1} \text{ dw}$ ) and Point de Moulard ( $4.6 \text{ Bq kg}^{-1} \text{ dw}$ ).

In southwest Spain, phosphate wastes containing NORM have also been discharged into estuaries, but no NORM data in biota is available. The results of NORM concentrations in sediment collected around UK are listed in Table 27.

## **8 Conclusions**

The marine environment in the OSPAR region shows a general trend of decreasing concentrations of anthropogenic radionuclides during 1980 to 1999 with the following exceptions.

- Irish Sea – remobilisation of plutonium isotopes and  $^{137}\text{Cs}$  from seabed sediments, build-up of  $^{241}\text{Am}$  in sediments and increased discharges of  $^{99}\text{Tc}$  from Sellafield since 1994.
- Kattegat – no clear declining trend of  $^{137}\text{Cs}$  concentrations due to outflow of Chernobyl fallout from the Baltic Sea.

The environmental data on naturally occurring radionuclides in the marine environment are very scarce compared to environmental data on anthropogenic radionuclides.

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**Table 1** Concentrations of  $^{137}\text{Cs}$  ( $\text{Bq m}^{-3}$ ) in North European seawater

	1972	1973	1974	1975	1976	1977	1978	1979	1980	1981	1982	1983	1984	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1998	1999
Com-16-S																											
Max												7.00	5.52	10.70		10.90	10.10	8.70	9.70	16.30	3.30	7.30	10.50	4.90	10.70		
Min												4.30	1.17	0.98		6.00	3.90	8.70	2.50	4.30	3.30	7.30	3.00	0.40	5.20		
Geomean												5.91	2.65	5.19		7.52	6.81	8.70	5.91	10.55	3.30	7.30	7.42	2.35	7.71		
Average												5.96	3.06	5.54		7.64	7.02	8.70	6.19	11.11	3.30	7.30	7.63	3.08	8.03		
Number												16	8	62		11	34	1	83	16	1	1	28	22	4		
Com-16-D																											
Max													1.67	4.85			6.50		5.70					3.50			
Min													1.17	0.85			2.50		1.07					0.81			
Geomean													1.43	1.63			3.44		2.08					1.58			
Average													1.45	1.94			3.83		2.45					1.81			
Number													3	22			3		12					10			
Com-17																											
Max		9.99	14.80	11.40	18.40	14.90	18.10	18.60	14.90	29.20	4.85	10.20	8.31	14.20	27.30	12.60	8.10	8.21	8.50	5.60	4.90	2.30	2.27	3.77	2.57	2.11	1.82
Min		2.59	1.66	3.37	1.52	4.44	2.70	1.72	1.69	0.80	3.11	1.47	1.10	0.98	2.50	1.17	1.22	2.36	2.10	1.50	0.70	1.47	2.21	2.04	2.36	2.11	1.35
Geomean		5.34	5.83	5.63	7.07	6.27	6.84	5.38	4.93	6.69	3.64	2.97	3.21	4.77	9.25	5.73	2.84	4.81	3.51	3.09	2.81	1.84	2.24	2.70	2.46	2.11	1.60
Average		5.53	6.30	6.04	9.17	7.26	7.53	6.31	5.93	8.17	3.68	3.16	3.49	5.15	12.38	6.62	3.23	5.23	3.76	3.29	3.09	1.89	2.24	2.74	2.47	2.11	1.61
Number		41	50	16	10	4	45	70	44	105	7	118	34	447	11	30	17	14	44	11	29	2	2	114	2	1	3
Com-18																											
Max		5.92	2.26	5.51			6.40	3.25		2.52		1.88	1.70	4.85		1.70	2.33	5.60	2.70					3.50			
Min		1.74	2.26	0.63			3.07	0.06		0.52		1.88	0.01	0.26		1.08	1.14	5.60	1.67					0.70			
Geomean		3.43	2.26	1.94			4.61	0.83		1.17		1.88	0.46	1.00		1.35	1.75	5.60	1.99					1.24			
Average		3.67	2.26	2.75			4.82	1.05		1.31		1.88	0.83	1.21		1.39	1.83	5.60	2.04					1.37			
Number		9	1	3			3	19		11		1	8	92		2	3	1	3					57			
Com-19																											
Max			11.40	8.66			14.30	21.10	19.30	11.00	14.00	11.60	22.00	14.10	12.40		8.21	4.30				5.50	3.80				
Min			3.48	5.14			13.90	4.80	2.31	5.00	6.00	1.05	1.45	6.90	6.90		4.34	3.88				3.00	1.64				
Geomean			6.89	6.67			14.10	10.75	12.95	8.30	9.46	5.34	6.81	10.31	9.67		6.66	4.12				4.32	2.97				
Average			7.23	6.90			14.10	12.28	13.67	8.57	9.77	6.03	7.29	10.64	9.79		6.79	4.12				4.39	3.01				
Number			14	2			2	6	40	9	57	47	135	7	8		7	6				14	60				

**Table 1 (cont'd)**

	1972	1973	1974	1975	1976	1977	1978	1979	1980	1981	1982	1983	1984	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1998	1999
Com-19d																											
Min									3.00				0.50	0.32										0.42			
Max									7.00				2.68	5.48										1.97			
Geomean									4.71				1.04	1.12										1.12			
Average									4.90				1.14	1.63										1.19			
Number									5				14	22										24			
Com-23(surf)																											
Max																		9.12	7.69		10.80	9.50	7.10	4.51	3.40		
Min																		3.20	3.79		5.30	3.90	5.30	2.40	2.50		
Geomean																		5.90	6.17		6.93	6.09	6.19	3.33	2.89		
Average																		6.09	6.36		7.25	6.70	6.23	3.38	2.90		
Number																		33	5		4	2	4	56	5		
Com-27S																											
Max																41.00		16.30	12.50		12.60	31.90	10.50	8.56			
Min																16.00		0.81	3.54		4.40	1.38	2.50	2.46			
Geomean																31.51		4.99	7.43		7.58	6.17	5.51	3.67			
Average																33.30		6.02	7.88		7.77	7.79	5.93	3.82			
Number																9		78	13		30	25	43	54			
Com-27D																											
Max																		0.98									
Min																		0.55									
Geomean																		0.72									
Average																		0.74									
Number																		5									
Com-28																											
Max	1090	1220	1460	2970	3360	4820	2190	2230	1680	2370	3590	2410	1410	769	479	1080	612	219				430	340	300	160	76	400
Min	28	6	6	5	8	7	5	2	3	2	6	1	2	3	4	1	1	1				26	18	14	6.9	8.7	250
Geomean	347	155	219	391	411	1040	386	414	421	581	480	169	143	84	85	41	36	13		20	50	95	73	57	37	35	316
Average	471	292	374	819	977	1284	754	756	672	949	753	382	318	166	130	77	62	25				178	140	119	71	50	325
Number	119	138	138	166	152	167	167	167	166	112	133	147	167	167	95	167	167	167				3	3	3	3	3	2

Table 1 (cont'd)

	1972	1973	1974	1975	1976	1977	1978	1979	1980	1981	1982	1983	1984	1985	1986	1987	1988	1989	1990	1991	1993	1994	1995	1996	1997	1998	1999
Com-29																											
Max		258	110	135	312	789	524	374	262	129	346	627	3640	410	195	414	200	137	14			21	15	11	13	7	9
Min		6	61	14	13	130	104	73	91	61	78	33	21	31	21	12	9	4	4			21	15	11	9	3	4
Geomean		69	84	55	160	431	296	242	145	89	191	144	86	105	72	37	27	15	11	10	10	21	15	11	11	5	6
Average		83	86	64	183	475	327	267	154	92	204	168	166	119	79	50	32	18	11			21	15	11	11	5	6
Number		50	7	33	24	19	19	8	11	8	33	32	50	33	49	39	57	43	27			1	1	2	9	8	8
Com-30																											
Max	983	603	1800	2640	2660	4040	4410	2150	3640	1410	1530	961	852	442	428	214	158	166									
Min	352	348	441	1640	1730	947	1420	0	432	710	541	457	460	92	149	71	59	32									
Geomean	627	481	794	1884	2223	2014	1859	764	1190	973	964	732	601	291	281	141	108	67									
Average	641	487	871	1912	2239	2125	1937	1079	1313	1006	1012	748	619	298	288	146	110	72									
Number	33	36	55	12	16	46	33	55	43	10	27	17	18	86	24	32	62	37									
Com-31																											
Max	2900	12000	23800	7000	19600	17800	7990	9250	4390	10200	5080	3090	1440	1240	866	412	367	575					66	46	56	39	60
Min	1010	688	1140	2500	3380	1870	1170	1370	944	3510	721	961	460	158	233	113	91	81					66	46	56	39	60
Geomean	1598	2157	2763	4523	10242	4357	2700	4052	2596	5028	1813	1919	721	388	392	262	145	166		100	100		66	46	56	39	60
Average	1754	3839	4567	4820	12270	5482	3332	4664	2807	5753	2056	2210	762	475	416	276	158	195					66	46	56	39	60
Number	18	26	24	8	8	48	17	21	38	6	24	5	22	33	77	64	26	31					1	1	1	1	1
Com-32																											
Max	7570	13600	57300	37600	47100	26000	38800	14400	14000	20200	16200	6530	6310	8790	1300	924	466	594				690	620	530	380	330	300
Min	1460	1010	2240	7000	6630	2540	3000	0	2080	1810	697	857	695	503	259	157	133	130				110	130	48	87	43	120
Geomean	2804	2904	8091	20782	20654	8754	7092	3655	4911	4784	3795	2661	1678	1556	568	371	256	296		250	250	307	292	183	189	118	189
Average	3088	4174	13194	26233	23680	10054	10151	6853	5825	6645	4977	3183	2005	2029	603	390	275	318				384	344	255	215	152	198
Number	32	50	56	6	48	50	16	44	52	22	46	34	40	44	84	74	52	34				5	5	5	5	6	6
Com-33																											
Max	699	769	1910	1930	3050	2950	2100	1250	1370	2120	1200	919	872	350	365	252	160	155	84			29	14	27	17	19	30
Min	175	370	316	862	1390	372	311	145	113	839	167	322	200	0	0	35	24	13	69			29	14	27	17	19	30
Geomean	441	508	678	1407	2194	1239	997	571	595	1085	504	535	403	74	59	124	72	79	75	50	50	29	14	27	17	19	30
Average	465	518	735	1437	2239	1333	1093	653	732	1121	581	564	437	143	143	138	77	83	75			29	14	27	17	19	30
Number	51	47	69	22	34	88	64	41	57	23	75	30	49	104	130	82	91	88	33			1	1	1	1	1	1

Table 1 (cont'd)

	1972	1973	1974	1975	1976	1977	1978	1979	1980	1981	1982	1983	1984	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1998	1999
Com-34																											
Max	5370	14200	42800	46900	29400	27400	14500	20000	13100	4700	3910	6140	1870	4020	855	514	424	427				68	44	140	84	46	80
Min	399	330	306	1550	3420	795	768	92	478	933	513	372	447	100	140	97	57	81				54	42	93	73	21	60
Geomean	1195	1915	3944	7302	7007	2505	3362	2660	1855	2084	1128	1677	966	699	368	260	184	164		100	100	61	43	114	78	31	69
Average	1559	2912	6746	11216	8330	4413	4498	4429	2899	2344	1418	2323	1056	1003	400	273	200	178				61	43	117	79	34	70
Number	78	88	83	44	50	87	76	74	79	32	74	34	52	50	74	72	78	54				2	2	2	2	2	2
Com-35																											
Max	90600	52500	204000	208000	115000	128000	93600	63300	131000	58800	142000	118000	23600	19400	2380	1490	950	1660				330	310	240	270	200	240
Min	427	1100	1450	1370	0	1390	1720	817	1460	4450	160	2130	1650	852	435	287	170	266				240	59	190	190	140	150
Geomean	7617	6184	20121	26853	32154	20447	12159	9833	15090	16616	15275	12716	4814	3065	785	559	396	524		250	250	288	180	215	216	169	179
Average	8575	9165	31893	50879	30187	25470	18849	19639	23077	19141	15396	15265	5358	4034	859	594	464	566				290	212.3	216	218	170	182
Number	198	198	166	118	180	198	164	114	112	80	152	122	112	96	120	94	122	108				4	4	5	5	5	5
Com-36																											
Max	466	484	902	1070	4780	1350	1390	371	697	846	656	291	317	110	244	170	120	323				37	14	41	37		
Min	14	14	15	20	34	19	24	20	13	23	3	9	8	5	0	11	5	7				37	14	41	37		
Geomean	77	90	89	183	352	122	109	72	53	161	32	36	40	16	20	31	22	41		25	25	37	14	41	37		
Average	115	138	174	257	523	239	200	98	93	232	60	54	64	22	53	39	32	55				37	14	41	37		
Number	94	119	155	114	119	120	136	101	113	98	130	112	104	99	94	39	87	84				1	1	1	1		
Com-37																											
Max	4470	5540	14500	13000	54400	28700	14500	28300	73800	25200	10700	5760	5440	7090	1620	907	868	761				390	380	310	270	230	210
Min	636	806	542	3970	3140	1450	0	1210	965	1460	82	1330	556	446	359	157	140	203				220	130	230	180	110	120
Geomean	1694	2726	5030	8080	10178	6052	3546	5452	4048	6300	2981	3310	2264	1535	760	497	391	385		250	250	271	230	253	225	154	176
Average	1897	3124	7666	8657	13824	7334	5609	7366	6695	7438	3924	3553	2497	1839	804	527	430	418				278	248	255	228	160	180
Number	42	40	40	24	26	79	48	79	78	62	70	70	72	81	78	72	75	50				4	4	4	4	4	4
Com-38																											
Max	21	29	44	79	298	98	69	59	84	79	26	31	10	8	9	12	38	18	9								
Min	11	4	4	6	8	6	7	4	4	7	5	2	3	2	5	10	5	4	9								
Geomean	13	9	13	29	58	26	21	15	12	31	9	8	6	5	6	11	10	5	9								
Average	14	10	15	37	91	31	24	19	17	40	10	9	6	5	6	11	13	6	9								
Number	19	41	83	47	41	58	57	50	32	30	31	46	31	26	20	6	11	11	2								



**Table 1 (cont'd)**

	1972	1973	1974	1975	1976	1977	1978	1979	1980	1981	1982	1983	1984	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1998	1999
Com-39																											
Max	20	85	40	76	206	395	280	146	188	952	1090	113	265	184	78	88	46	32									
Min	17	7	7	8	9	7	9	11	8	7	7	4	6	2	6	11	5	4									
Geomean	18	12	17	28	91	49	48	37	37	55	41	23	42	25	22	31	16	12									
Average	18	17	19	34	108	71	63	47	51	82	79	31	57	41	27	35	18	14									
Number	3	14	39	30	20	47	47	32	39	41	29	32	27	33	24	28	40	9									
Com-46																											
Max		8.14	15.70	10.00	10.50	17.00	8.21	9.27	12.90	31.00	6.65	13.80	4.50		5.60	7.61	6.80	4.90	3.70	4.78	4.45						
Min		5.18	9.03	10.00	10.50	8.55	8.21	4.93	7.80	31.00	5.17	3.38	3.76		3.55	7.61	4.36	2.76	2.96	4.78	4.45						
Geomean		6.73	10.54	10.00	10.50	11.32	8.21	6.35	9.78	31.00	5.76	5.16	4.11		4.34	7.61	5.59	3.76	3.31	4.78	4.45						
Average		6.85	10.71	10.00	10.50	11.85	8.21	6.60	10.00	31.00	5.79	6.01	4.13		4.42	7.61	5.69	3.84	3.33	4.78	4.45						
Number		6	7	1	1	3	1	3	3	1	4	5	2		3	1	4	4	2	1	1						
Com-47																											
Max		22.20	22.40					25.40	14.20	26.80	46.50	33.90	23.60	17.60	14.20	9.26	11.10	10.80	7.77	12.40	4.41		2.80	2.40			
Min		14.80	17.60					9.80	5.46	13.20	8.54	5.58	7.42	4.30	4.26	4.22	4.02	4.06	4.44	5.69	3.23		2.80	2.40			
Geomean		18.13	19.86					15.61	9.53	18.93	19.25	13.71	13.91	9.63	8.52	5.94	6.28	6.17	5.67	7.85	3.77		2.80	2.40			
Average		18.38	20.00					16.24	9.96	19.77	21.22	14.93	14.56	10.31	9.06	6.11	6.55	6.46	5.77	8.31	3.82		2.80	2.40			
Number		6	2					9	9	7	15	15	12	11	12	11	13	14	12	3	2		1	2			
Com-48																											
Max		20.70	81.40	66.60	60.70	46.10	40.60	44.00	17.40	25.70	56.10	9.45	22.90	17.20	14.70	6.94	10.10	16.80	11.70	9.19	6.70	3.51	3.70				
Min		5.18	30.10	19.00	16.60	11.50	8.18	6.83	5.67	7.09	5.50	2.92	3.21	3.79	3.94	4.25	3.39	4.12	1.11	3.40	3.60	3.29	2.70				
Geomean		10.99	42.03	35.57	31.74	21.00	12.58	16.55	9.77	15.02	11.61	5.12	8.36	6.73	6.64	5.03	5.78	7.22	4.68	4.30	5.02	3.40	3.03				
Average		13.07	47.27	42.80	38.65	24.23	16.54	19.99	10.21	17.13	15.77	5.53	9.86	7.52	7.44	5.13	6.08	8.16	5.37	4.47	5.19	3.40	3.05				
Number		6	3	2	2	6	4	6	10	3	10	8	8	9	5	4	12	12	19	16	3	2	4				
Com-49																											
Max		21.10										10.90		6.17	10.80	7.49	4.39	7.85									
Min		8.51										10.50		5.03	4.43	6.60	2.24	2.00									
Geomean		13.73										10.70		5.59	6.25	7.03	3.14	4.51									
Average		14.25										10.70		5.61	6.78	7.05	3.32	5.23									
Number		6										2		3	3	2	2	3									

Table 1 (cont'd)

	1972	1973	1974	1975	1976	1977	1978	1979	1980	1981	1982	1983	1984	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1998	1999
Com-50																											
Max		35.50	66.20	38.90	30.00	24.30	21.60	44.20	11.20	27.60	28.50	26.20	19.40	7.85	14.50	7.22	11.50	17.71	12.20	13.40	7.70	6.43	4.90	4.90			
Min		18.10	33.00	20.10	30.00	17.80	9.43	12.10	6.05	11.80	13.10	6.63	12.70	4.29	5.06	3.95	3.16	8.40	6.80	5.74	5.66	3.46	3.50	4.50			
Geomean		26.90	49.77	28.50	30.00	20.80	13.12	23.13	8.20	18.37	22.48	11.65	16.73	5.54	8.14	5.51	6.16	11.87	9.95	8.25	6.88	4.49	4.05	4.70			
Average		27.80	51.40	29.53	30.00	21.05	14.04	28.15	8.42	19.01	23.10	13.38	17.03	5.65	8.97	5.68	6.68	12.41	10.13	8.45	6.92	4.67	4.08	4.70			
Number		9	6	3	1	2	3	2	4	7	7	4	3	6	3	3	6	10	22	35	6	5	5	2			
Com-51																											
Max																		10.66	12.21	13.80	7.90						
Min																		10.66	5.18	4.94	4.20						
Geomean																		10.66	8.53	8.59	6.42						
Average																		10.66	8.71	8.77	6.52						
Number																		1	19	43	17						
Com-52																											
Max		23.70	287.0	49.20		20.20	16.10	18.30	8.98				10.30				8.00	13.20	12.60	12.10	6.60	4.77					
Min		23.70	62.2	25.90		20.20	16.10	18.30	8.98				10.30				8.00	5.98	7.97	4.07	3.30	3.27					
Geomean		23.70	133.6	32.84		20.20	16.10	18.30	8.98				10.30				8.00	8.56	10.33	6.15	4.61	3.73					
Average		23.70	174.6	34.30		20.20	16.10	18.30	8.98				10.30				8.00	9.04	10.51	6.59	4.74	3.79					
Number		1	2	3		1	1	1	1				1				1	3	10	10	6	3					
Com-53																											
Max		22.60		72.50	38.50	39.90	23.10	13.20	12.70	15.50	12.90	12.50	18.60	13.80	9.15	7.59	10.80	12.30	11.70	9.50	4.92	7.12	4.20	3.40			
Min		20.70		21.80	38.50	14.40	10.70	11.40	6.66	12.40	8.95	8.50	7.60	4.48	8.28	5.59	4.87	4.44	5.50	3.52	3.80	3.63	3.20	3.40			
Geomean		21.45		36.51	38.50	20.59	14.18	12.27	9.56	13.86	10.35	10.78	13.24	8.15	8.70	6.51	8.14	7.16	7.66	5.87	4.40	5.08	3.60	3.40			
Average		21.47		41.70	38.50	23.17	14.88	12.30	9.85	13.95	10.42	10.93	13.78	8.70	8.72	6.59	8.49	7.77	7.91	6.10	4.42	5.38	3.63	3.40			
Number		3		3	1	3	4	2	4	2	6	3	5	6	2	2	4	5	19	33	6	2	1	1			
Com-54																											
Max				69.2	43.7		12.0	119.0		159.0	138.0	34.3	26.7	29.0			6.9	10.1					4.2	3.8			
Min				69.20	43.70		12.00	11.30		13.90	11.50	7.74	12.40	5.45			5.63	10.11					4.20	3.80			
Geomean				69.20	43.70		12.00	24.96		35.62	24.01	14.73	19.27	13.78			6.22	10.11					4.20	3.80			
Average				69.20	43.70		12.00	32.90		47.51	30.93	15.91	19.77	15.16			6.26	10.11					4.20	3.80			
Number				1	1		1	9		9	14	12	11	16			2	1					1	1			

**Table 1 (cont'd)**

	1972	1973	1974	1975	1976	1977	1978	1979	1980	1981	1982	1983	1984	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1998	1999
Com-55																											
Max.	40.7	49.6	88.1	77.7	66.6	514.0	351.0	372.0	708.0	8870	309.0	244.0	459.0	962.0	174.0	95.7	40.4	39.7	29.0	25.0	18.5		10.8	18.7	13.8	11.4	
Min.	39.6	20.0	34.6	15.9	39.2	1.1	10.2	13.1	8.6	18	11.8	14.1	9.4	5.0	11.5	4.2	5.3	8.3	5.6	4.4	5.0		3.8	3.4	3.0	3.5	
Geomean	40.2	25.4	49.4	45.8	52.2	92.2	147.0	147.5	138.9	385	83.3	74.7	80.8	64.9	71.8	26.8	15.3	23.2	9.9	9.0	8.5		7.7	6.7	6.7	6.9	
Average	40.2	27.2	52.9	47.8	53.1	172.3	200.5	172.8	225.1	1004	114.6	104.7	112.1	106.6	82.9	39.7	18.9	25.5	11.1	10.0	9.3		8.3	7.8	7.8	7.6	
Number	3	5	4	32	8	30	39	29	39	39	39	39	39	39	24	37	29	23	52	58	17		7	7	7	16	
Com-56																											
Max				91.4	52.5	30.90	29.70	26.80	14.70	24.30	28.20	24.80	23.30	13.10	23.10	12.00	8.90	9.17	20.09	17.90	14.95		5.20		5.30	4.50	
Min				27.2	37.7	30.90	12.60	18.80	10.30	17.10	16.10	19.80	6.60	13.10	23.10	10.20	8.90	7.51	5.18	3.70	5.52		4.40		4.30	3.90	
Geomean				49.45	44.59	30.90	20.08	21.64	12.34	20.38	21.68	21.60	14.12	13.10	23.10	11.06	8.90	8.26	8.38	8.31	7.14		4.82		4.69	4.23	
Average				50.88	44.77	30.90	20.87	21.90	12.47	20.70	22.02	21.68	14.65	13.10	23.10	11.10	8.90	8.27	8.82	8.62	7.46		4.82		4.70	4.24	
Number				46	13	1	6	3	3	2	6	4	17	1	1	2	1	6	46	65	22		9		10	19	
Com-57																											
Max										119.0	86.0	59.5	36.1	32.0	26.9	20.6	13.2	11.7	18.1	13.0	12.8	10.9	8.4				
Min.										25.50	10.20	5.92	4.49	5.20	6.74	5.32	6.00	5.20	4.03	4.50	3.40	7.60	8.10				
Geomean										55.09	29.62	18.77	12.73	12.90	13.45	10.47	8.90	7.80	8.54	7.65	6.60	9.10	8.25				
Average										86.22	50.03	26.43	21.18	16.77	16.07	13.69	8.68	9.21	10.62	9.16	9.27	9.25	8.25				
Number										41	116	89	95	111	211	53	6	14	33	26	24	2	2				
Com-58																											
Max										53.00	26.70	36.11	20.35	19.06	11.50	7.79		9.40	9.40	7.00							
Min.										13.30	9.70	8.10	6.66	4.67	6.05	6.72		4.21	4.50	4.00							
Geomean										25.13	14.21	13.04	10.48	9.69	8.51	7.24		6.18	6.53	5.02							
Average										27.38	14.98	13.73	10.72	9.91	8.60	7.25		6.30	6.66	5.06							
number										22	8	29	87	173	41	6		31	26	20							
Com-59																											
Max										105.0	45.1	42.0	29.0	27.5	26.0			15.1	22.6	10.9	3.5	6.3					
Min										29.60	4.51	1.44	4.74	6.10	12.40			2.30	2.10	2.80	2.80	2.32					
Geomean										57.25	21.46	16.43	12.68	16.66	18.22			6.16	4.79	5.24	3.08	3.15					
Average										60.68	25.95	19.73	14.31	17.61	18.59			6.91	5.90	6.07	3.10	3.46					
Number										25	14	71	39	76	24			14	39	7	4	4					

**Table 1 (cont'd)**

	1972	1973	1974	1975	1976	1977	1978	1979	1980	1981	1982	1983	1984	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1998	1999
Com-60																											
Max.										60.90	40.30	37.70	24.60	51.17	21.64	48.70	25.50	28.20	26.70								
Min.										24.70	12.60	6.06	5.10	5.53	10.46	7.40	7.50	7.30	8.30								
Geomean										37.35	24.31	15.62	12.95	14.51	14.99	14.41	15.79	14.65	14.70								
Average										38.92	25.55	17.52	14.24	16.82	15.44	19.26	17.87	16.72	16.27								
Number										12	24	32	49	71	7	7	6	9	6								
Com-61																											
Average		30	33	29	37	36	43	61	62	68	50	36	36	29	98	69	73	62	70	61	53	67	65	50			
Number		6	5	4	7	8	13	11	20	20	13	12	5	6	28	9	17	19	6	9	16	14	16	11			
Com-62																											
Average		32	40	31	47	51	76	96	105	91	77	64	55	45	76	49	35	26	22	24	29	35	17	27			
Number		5	3	9	16	13	22	18	30	18	10	8	13	16	25	4	11	11	14	11	20	32	7	13			

**Table 2 Comparison of  $^{137}\text{Cs}$  ( $\text{Bq m}^{-3}$ ) concentrations in seawater measured by different laboratories (compartment 35 and 33 in Irish Sea measured by BNFL, RPII and MAFF + others)**

		1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001
BNFL data (Compartment 35)																	
	Max	8300	3500	6100	9600	3000	2700	1600	840	1500	530	492	1280	789	715	374	251
	Min	1200	520	1100	1100	440	570	293	200	250	120	199	172	173	125	133	185
	Geomean	1859	1682	1764	1793	1592	1436	611	495	357	219	317	320	279	235	226	209
	Average	1994	1759	2000	1960	1683	1561	651	521	390	237	339	399	306	260	235	210
	Number	47	64	65	60	74	46	43	50	27	44	5	16	16	20	18	5
* 1986-1991, most of data are lower than the LD																	
IAEA-Database & MAFF (Compartment 35)																	
	Max	2380	1490	950	1660					330	310	240	270	200	240		
	Min	435	287	170	266					240	59	190	190	140	150		
	Geomean	785	559	396	524		250		250	288	180	215	216	169	179		
	Average	859	594	464	566					290	212.25	216	218	170	182		
	Number	120	94	122	108					4	4	5	5	5	5		
RPII data (com-33)																	
		1984	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
	Max	490.0	350.0	220.0	214.0	160.0	124.0	84.0	97.0	73.0	80.0	60.0	50.0	28.0	42.0	31.0	32.0
	Min	210.0	180.0	120.0	84.0	43.0	63.0	68.0	39.0	55.0	25.0	20.0	4.0	12.0	14.0	9.0	22.0
	Geomean	330.1	222.9	166.9	142.1	81.8	93.4	74.6	63.6	65.8	51.5	38.9	26.5	19.0	23.7	20.0	27.8
	Average	341.7	230.0	169.1	146.3	88.3	94.6	74.8	65.5	65.9	54.4	40.9	32.3	19.5	25.2	21.0	28.0
	Number	6	5	11	12	15	14	14	14	14	14	14	8	12	15	13	9
IAEA-Database & MAFF (Com-33)																	
	Max	872.0	350.0	365.0	252.0	160.0	155.0	84.0				29.0	14.0	27.0	17.0	19.0	30.0
	Min	200.0	0.2	0.1	35.1	23.6	13.0	69.0				29.0	14.0	27.0	17.0	19.0	30.0
	Geomean	403.3	73.7	58.9	124.5	72.2	79.5	74.7	50.0		50.0	29.0	14.0	27.0	17.0	19.0	30.0
	Average	436.7	142.8	143.4	138.0	77.4	83.2	74.8				29.0	14.0	27.0	17.0	19.0	30.0
	Number	49	104	130	82	91	88	33				1	1	1	1	1	1

**Table 2 (cont'd) (compartment 47 and 48 in English Channel by CEA and others)**

		1981	1982	1983	1984	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995
Com-47 (IAEA database)																
	Max	26.80	46.50	33.90	23.60	17.60	14.20	9.26	11.10	10.80	7.77	12.40	4.41		2.80	2.40
	Min	13.20	8.54	5.58	7.42	4.30	4.26	4.22	4.02	4.06	4.44	5.69	3.23		2.80	2.40
	Geomean	18.93	19.25	13.71	13.91	9.63	8.52	5.94	6.28	6.17	5.67	7.85	3.77		2.80	2.40
	Average	19.77	21.22	14.93	14.56	10.31	9.06	6.11	6.55	6.46	5.77	8.31	3.82		2.80	2.40
	Number	7	15	15	12	11	12	11	13	14	12	3	2		1	2
Com-48 (IAEA database)																
	Max	25.70	56.10	9.45	22.90	17.20	14.70	6.94	10.10	16.80	11.70	9.19	6.70	3.51	3.70	
	Min	7.09	5.50	2.92	3.21	3.79	3.94	4.25	3.39	4.12	1.11	3.40	3.60	3.29	2.70	
	Geomean	15.02	11.61	5.12	8.36	6.73	6.64	5.03	5.78	7.22	4.68	4.30	5.02	3.40	3.03	
	Average	17.13	15.77	5.53	9.86	7.52	7.44	5.13	6.08	8.16	5.37	4.47	5.19	3.40	3.05	
	Number	3	10	8	8	9	5	4	12	12	19	16	3	2	4	
CEA data																
	Cap de la Hague(49°44.8N;1°55.7W) 48				Jardeheu(49°44.0N;1°51.1W)48				Jobourg(49°44.0N;1°51.1W) 47				Le Rozel(49°27.9N;1°52.2W)47			
Year	Average	s	max.	>LD/N	Average	s	max.	>LD/N	Average	s	max.	>LD/N	Average	s	max.	>LD/N
1981	49.12	21.76	75.85	4/4												
1982	30.09	2.99	33.30	3/3	110.45	126.58	300.07	4/4	49.64	13.86	72.52	6/6				
1983	36.78	20.43	72.89	5/5	58.71	20.90	86.58	6/6	77.33	77.97	132.46	2/2				
1984	18.87	13.38	29.60	4/4	38.48	8.41	47.73	4/4	30.16	2.06	32.19	4/4	37.59	17.08	54.76	5/5
1985	27.66	11.55	43.29	4/4	32.47	10.86	47.36	4/4	58.95	21.33	75.11	3/3	24.42	8.93	32.56	4/4
1986	8.05	2.52	10.73	4/4	23.40	9.11	36.63	4/4	19.33	5.51	26.64	4/4	16.10	9.16	22.57	2/2
1987	16.56	6.88	25.90	4/4	14.89	4.86	20.72	4/4	9.99	2.28	13.32	4/4	7.86	0.35	8.14	4/4
1988	15.39	11.39	31.08	5/5	9.07	2.81	11.84	4/4	11.01	4.28	15.54	4/4	11.66	2.21	14.06	4/4
1989	13.32	3.66	17.02	4/4	9.62	1.98	12.58	4/4	15.63	4.57	21.46	4/4	16.47	4.89	20.72	4/4
1990	8.05	2.35	10.73	4/4	12.83	0.43	13.32	3/3	15.66	6.26	22.57	3/3	12.21	4.18	18.13	4/4
1991	6.10	1.01	7.40	4/4	14.73	6.10	22.57	5/5	11.47	1.60	13.69	4/4	7.49	4.29	12.95	4/4
1992	6.48	1.74	8.76	4/4	8.42	1.52	10.36	4/4	5.94	1.71	8.21	4/4	5.16	0.61	5.76	4/4
1993	19.98	16.44	40.10	4/4	6.27	0.83	7.08	4/4	24.73	31.78	61.40	3/3	25.24	34.70	65.30	3/3
1994	4.61	0.72	5.42	4/4	9.24	5.95	17.80	4/4	9.21	6.30	18.60	4/4	8.24	4.21	12.50	4/4
1995	3.72	1.01	4.97	4/4	6.88	2.80	10.90	4/4	6.36	2.04	8.86	4/4	5.16	0.50	5.58	5/5
1996					5.49	1.47	7.48	4/4	3.86	1.34	5.08	4/4	4.09	1.18	5.76	4/4

**Table 3a** Concentrations of  $^{137}\text{Cs}(\text{Bq kg}^{-1} \text{ ww})$  in fish (by MAFF)

		1988	1989	1991	1992	1993	1994	1995	1996	1997	1998	1999
Com-17												
	Max				0.30	0.24	0.25	0.32	0.31	0.31	0.30	0.19
	Min				0.28	0.22	0.15	0.26	0.21	0.23	0.17	0.19
	Geomean	0.4	0.3	0.22	0.29	0.23	0.19	0.29	0.26	0.27	0.23	0.19
	Average				0.29	0.23	0.20	0.29	0.26	0.27	0.24	0.19
	Number				2	2	2	2	2	2	2	2
Com-24												
	Max				0.44	0.50	0.52	0.47	0.43	0.32	0.31	0.29
	Min				0.44	0.50	0.45	0.47	0.43	0.32	0.31	0.29
	Geomean	1.1			0.44	0.50	0.48	0.47	0.43	0.32	0.31	0.29
	Average				0.44	0.50	0.49	0.47	0.43	0.32	0.31	0.29
	Number				1	1	2	1	1	1	1	1
Com-27												
	Max				0.64	0.58	0.48	0.32		0.34	0.30	
	Min				0.64	0.50	0.38	0.32		0.32	0.26	
	Geomean	2.00	1.20	0.97	0.64	0.54	0.43	0.32		0.33	0.28	
	Average				0.64	0.54	0.43	0.32		0.33	0.28	
	Number				1	2	2	1		2	2	
Com-28												
	Max	18.00	10.00	11.00	9.60	11.00	2.20	1.80	0.85	0.76	3.60	4.90
	Min	1.70	1.30	0.24	0.27	0.17	0.37	0.29	0.28	0.19	0.27	0.16
	Geomean	8.70	5.16	2.40	1.39	1.51	0.79	0.73	0.61	0.44	0.91	0.81
	Average	8.70	5.16	2.40	2.78	3.17	0.94	0.87	0.65	0.48	1.44	1.51
	Number	10	11	13	11	10	7	6	6	7	10	10
Com-29												
	Max				1.00	1.00	13.00	7.30	6.80	4.10	1.00	0.77
	Min				1.00	1.00	1.20	0.95	1.10	1.00	1.00	0.10
	Geomean	3.50	1.10	0.81	1.00	1.00	4.04	3.05	3.11	2.29	1.00	0.28
	Average				1.00	1.00	5.42	3.81	3.61	2.57	1.00	0.44
	Number				1	1	6	6	7	6	1	2
Com-31												
	Max				4.10	3.90	5.40	5.00	4.20	4.00	2.20	3.20
	Min				4.10	3.90	5.00	5.00	4.20	3.40	2.20	2.80
	Geomean	16.00	13.00	6.80	4.10	3.90	5.20	5.00	4.20	3.69	2.20	2.99
	Average				4.10	3.90	5.20	5.00	4.20	3.70	2.20	3.00
	Number				1	1	2	1	1	2	1	2
Com-32												
	Max	91.00	69.00	64.00	53.00	45.00	43.00	28.00	29.00	28.00	7.10	26.00
	Min	0.90	1.10	0.42	0.48	0.45	0.48	0.85	1.20	0.83	0.31	0.26
	Geomean	11.38	9.42	6.14	7.25	7.86	5.46	4.28	6.45	3.82	2.13	2.60
	Average	11.38	9.42	6.14	22.83	23.15	17.13	10.72	12.63	10.41	3.46	13.13
	Number	3	3	3	3	3	3	3	3	3	5	2
Com-33												
	Max	16.00	9.60	3.00	4.00	4.70	5.20	2.00	3.10	1.30	3.00	1.00
	Min	3.60	2.80	1.70	2.30	4.50	2.80	1.80	2.00	0.95	1.30	0.52
	Geomean	7.59	5.18	2.26	3.03	4.60	3.82	1.90	2.49	1.11	1.97	0.72
	Average	7.59	5.18	2.26	3.15	4.60	4.00	1.90	2.55	1.13	2.15	0.76
	Number	2	2	2	2	2	2	2	2	2	2	2

**Table 3a (cont'd)**

		1988	1989	1991	1992	1993	1994	1995	1996	1997	1998	1999
Com-34												
	Max	25.00	9.10	7.00	6.90	6.80	6.00	3.60	3.00	2.60	2.60	3.60
	Min	8.40	5.00	2.50	4.80	4.50	3.50	2.40	1.40	2.60	2.30	1.10
	Geomean	11.56	7.34	4.37	5.96	5.84	4.93	2.82	2.10	2.60	2.45	1.99
	Average	11.56	7.34	4.37	6.03	5.93	5.07	2.87	2.20	2.60	2.45	2.35
	Number	4	3	4	3	3	3	3	3	2	2	2
Com-35												
	Max	88.00	86.00	38.00	48.00	31.00	31.00	33.00	20.00	22.00	24.00	17.00
	Min	4.20	0.80	6.70	0.63	2.20	2.70	0.33	0.64	3.10	2.70	2.70
	Geomean	28.23	19.82	21.70	16.82	15.72	12.72	9.19	7.50	9.18	9.08	8.91
	Average	28.23	19.82	21.70	20.10	17.62	14.35	12.51	9.82	10.21	10.04	9.72
	Number	22	22	21	21	17	15	16	19	16	15	17
Com-37												
	Max	130.00	68.00	39.00	40.00	46.00	36.00	480.00	110.00	39.00	180.00	17.00
	Min	12.00	11.00	4.70	0.58	0.46	0.48	0.57	0.27	0.55	0.43	0.27
	Geomean	30.31	24.10	17.42	11.02	9.74	9.74	8.96	9.34	9.03	7.95	6.58
	Average	30.31	24.10	17.42	15.86	17.39	14.89	49.76	18.90	12.70	20.25	8.81
	Number	7	9	8	13	12	10	12	13	12	14	12
Com-38												
	Max					0.37					0.59	0.40
	Min					0.37					0.48	0.30
	Geomean					0.37					0.52	0.35
	Average					0.37					0.53	0.35
	Number					1					3	2
Com-46												
	Max									0.29	0.50	0.35
	Min									0.29	0.42	0.16
	Geomean									0.29	0.46	0.27
	Average									0.29	0.46	0.28
	Number									1	2	3
Com-48												
	Max			0.98	0.72	0.98	0.50	0.52	0.51	0.38	0.33	0.33
	Min			0.28	0.36	0.12	0.18	0.30	0.16	0.12	0.14	0.10
	Geomean			0.52	0.53	0.42	0.36	0.39	0.28	0.21	0.23	0.21
	Average			0.52	0.55	0.57	0.39	0.40	0.32	0.25	0.25	0.24
	Number			7	4	3	4	3	3	2	3	3
Com-55												
	Max	2.40	1.90	1.10	0.72	1.10	0.67	0.69	0.78	0.79	0.58	0.49
	Min	0.50	0.50	0.81	0.30	0.32	0.26	0.31	0.38	0.39	0.24	0.22
	Geomean	1.45	1.09	0.94	0.54	0.69	0.48	0.48	0.54	0.53	0.39	0.34
	Average	1.45	1.09	0.94	0.56	0.74	0.52	0.50	0.57	0.55	0.41	0.36
	Number	5	5	3	5	5	3	3	4	4	5	5
Com-57												
	Max	4.90	2.40	2.10	5.00	5.00	4.00	3.00	4.00	4.00	5.00	5.00
	Min	1.20	1.30	0.37	0.30	0.12	0.18	0.30	0.16	0.12	0.14	0.10
	Geomean	2.65	1.71	0.93	0.79	0.77	0.60	0.64	0.62	0.51	0.55	0.48
	Average	2.65	1.71	0.93	1.33	1.29	1.04	0.96	1.05	0.83	1.04	1.06
	Number	6	5	6	10	10	10	10	10	13	13	13



**Table 3a (cont'd)**

		1988	1989	1991	1992	1993	1994	1995	1996	1997	1998	1999
Com-59												
	Max	3.20	2.20	0.89	0.96	1.10	0.62	0.72	1.20	0.85	0.58	0.44
	Min	0.40	0.40	0.19	0.25	0.22	0.55	0.41	0.49	0.36	0.32	0.15
	Geomean	1.55	1.13	0.55	0.60	0.55	0.58	0.54	0.63	0.52	0.44	0.32
	Average	1.55	1.13	0.55	0.64	0.61	0.58	0.55	0.69	0.55	0.46	0.34
	Number	8	8	6	7	7	4	4	4	4	4	5
Com-60												
	Max			18.00	1.10	3.10	1.10	1.30	0.66	0.82	0.55	0.66
	Min			11.00	1.00	1.20	0.66	0.76	0.51	0.48	0.44	0.42
	Geomean			14.07	1.05	1.93	0.85	0.99	0.58	0.63	0.49	0.53
	Average			14.07	1.05	2.15	0.88	1.03	0.59	0.65	0.50	0.54
	Number			2	2	2	2	2	2	2	2	2

**Table 3b      Concentrations of  $^{137}\text{Cs}$  (Bq kg<sup>-1</sup> ww) in fish (by RPII)**

		1982	1983	1984	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
Com-17																			
	Max		5.20	5.00	3.00	3.20	3.10	2.10	0.90	0.80	0.80	0.50		0.80	0.50	0.60	1.60	1.40	0.30
	Min		4.30	1.80	1.20	1.00	1.30	0.20	0.20	0.20	0.10	0.50		0.30	0.30	0.10	0.20	0.40	0.10
	Geomean		4.73	3.00	1.94	1.94	2.07	0.63	0.55	0.39	0.34	0.50		0.46	0.37	0.31	0.33	0.65	0.22
	Average		4.75	3.40	2.05	2.13	2.20	0.90	0.63	0.45	0.44	0.50		0.50	0.38	0.38	0.50	0.77	0.24
	Number		2	2	4	4	4	6	6	6	5	1		3	4	4	5	3	5
Com-28																			
	Max		17.7	20.2	17.2	9.2	5.9	3.5	2.8	2.1	1.1	1.2	1	1	0.8	0.9	0.4	0.8	0.5
	Min		1.60	4.10	1.90	0.70	1.40	0.30	0.20	0.30	0.20	0.10	0.30	0.20	0.50	0.20	0.40	0.40	0.20
	Geomean		5.21	10.18	5.43	3.27	2.66	1.27	0.86	0.95	0.55	0.28	0.51	0.46	0.61	0.47	0.40	0.62	0.31
	Average		7.43	11.90	7.98	4.34	3.04	1.72	1.20	1.14	0.65	0.46	0.55	0.54	0.63	0.54	0.40	0.64	0.33
	Number		4.00	4.00	4.00	5.00	5.00	6.00	6.00	5.00	6.00	5.00	6.00	5.00	4.00	5.00	1.00	5.00	6.00
Com-33																			
	Max	102.00	97.10	83.00	38.30	33.60	14.50	11.80	7.40	6.70	7.80	5.50	5.80	4.10	2.70	1.70	2.20	2.00	1.90
	Min	28.70	10.00	21.70	5.90	5.00	4.50	1.90	2.20	1.20	0.20	0.30	0.50	0.50	0.30	0.30	0.20	0.20	0.20
	Geomean	64.23	37.95	39.19	18.87	15.85	9.66	5.43	4.95	3.80	3.03	2.08	2.31	1.83	1.19	0.76	0.78	0.66	0.53
	Average	68.72	46.46	44.80	22.23	17.87	10.38	5.95	5.26	4.21	3.88	2.77	2.84	2.31	1.43	0.90	0.90	0.82	0.69
	Number	11	10	9	12	12	10	16	15	15	15	12	11	12	11	12	12	12	12
Com-38																			
	Max		17.30	23.50	23.90	8.70	4.50	3.60	11.10	3.40	4.00	2.00	1.80	1.10	1.50	1.10	0.90	0.90	1.10
	Min		14.20	11.80	2.10	6.40	2.40	0.20	0.20	0.60	0.20	0.30	0.50	0.30	0.30	0.20	0.20	0.20	0.20
	Geomean		16.14	17.73	9.68	7.12	3.57	1.69	1.45	1.38	0.93	0.75	0.96	0.66	0.58	0.43	0.34	0.32	0.38
	Average		16.20	18.47	13.18	7.18	3.68	2.11	2.37	1.71	1.30	0.90	1.04	0.71	0.71	0.48	0.38	0.38	0.45
	Number		3	3	4	4	4	11	12	8	9	7	7	7	7	10	10	10	8

**Table 4 Concentrations of  $^{137}\text{Cs}$  ( $\text{Bq kg}^{-1}$  ww) in shellfish (by MAFF)**

		1992	1993	1994	1995	1996	1997	1998	1999
Com-28									
	Max	3.20	2.60	0.57	1.20	1.50	0.57	1.50	0.59
	Min	0.79	0.52	0.57	1.20	0.39	0.57	0.37	0.59
	Geomean	1.45	1.25	0.57	1.20	0.82	0.57	0.90	0.59
	Average	1.73	1.48	0.57	1.20	0.94	0.57	1.02	0.59
	Number	3	4	1	1	5	1	4	1
Com-29									
	Max			2.20	1.90		1.50		1.20
	Min			0.63	0.60		0.46		0.40
	Geomean			1.00	0.98		0.77		0.75
	Average			0.19	1.11		0.87		0.83
	Number			3	3		4		3
Com-31									
	Max	1.20	1.60	0.76	0.47	0.69	0.85	0.52	0.54
	Min	0.63	1.10	0.57	0.44	0.13	0.85	0.52	0.54
	Geomean	0.87	1.33	0.66	0.45	0.30	0.85	0.52	0.54
	Average	0.92	1.35	0.67	0.46	0.41	0.85	0.52	0.54
	Number	2	2	2	2	2	1	1	1
Com-33									
	Max	21.00	15.00	13.00	12.00	12.00	7.90	8.80	7.00
	Min	9.50	5.60	2.50	2.20	2.20	2.30	1.90	1.40
	Geomean	14.41	9.79	5.60	5.08	4.16	4.10	3.84	2.86
	Average	15.17	10.43	6.70	6.13	4.92	4.43	4.24	3.33
	Number	3	4	7	7	6	7	8	8
Com-34									
	Max	1.40	1.40	0.79	0.72	0.56	0.61	0.51	0.61
	Min	0.74	1.10	0.55	0.60	0.42	0.48	0.24	0.34
	Geomean	1.02	1.24	0.66	0.66	0.50	0.54	0.39	0.46
	Average	1.07	1.25	0.67	0.66	0.50	0.55	0.41	0.47
	Number	2	2	2	2	3	2	3	3
Com-35									
	Max	27.00	27.00	23.00	22.00	17.00	17.00	18.00	15.00
	Min	2.60	3.10	1.90	2.30	2.10	1.80	1.70	1.70
	Geomean	9.82	9.51	7.07	7.85	7.00	5.52	5.60	5.11
	Average	11.75	11.52	8.77	10.32	8.23	6.71	7.30	6.28
	Number	28	29	25	25	24	24	28	28
Com-36									
	Max					3.20	7.60	4.00	3.60
	Min					3.20	4.20	3.00	3.60
	Geomean					3.20	5.65	3.46	3.60
	Average					3.20	5.90	3.50	3.60
	Number					1	2	2	1
Com-37									
	Max	16.00	15.00	12.00	10.00	8.20	15.00	17.00	9.70
	Min	2.50	3.40	1.20	1.80	2.40	1.50	1.10	1.30
	Geomean	5.62	5.97	3.62	3.66	4.09	3.85	3.44	3.54
	Average	6.88	6.89	4.57	4.12	4.39	4.74	4.73	4.21
	Number	8	7	7	10	9	10	10	10

**Table 4 (cont'd)**

		1992	1993	1994	1995	1996	1997	1998	1999
Com-38									
	Value	2.60	2.50	2.10	1.60	2.20	1.70	1.40	1.10
Com-55	Max	0.30	0.31	0.22	0.26	0.27	0.25	0.15	0.20
	Min	0.17	0.11	0.12	0.06	0.12	0.17	0.11	0.13
	Geomean	0.23	0.16	0.15	0.12	0.18	0.21	0.13	0.16
	Average	0.24	0.18	0.16	0.16	0.19	0.21	0.13	0.17
	Number	2	3	3	2	3	2	2	2
Com-57	Value	0.13	0.31	0.10		0.15	0.19		
Com-59	Value	0.41	0.28	0.39	0.19	0.29	0.31	0.25	0.21

**Table 5** Concentrations of  $^{137}\text{Cs}$  ( $\text{Bq kg}^{-1}$  ww) in biota (IAEA database)

		1985	1986	1987	1988	1989	1990
Com-16							
	Max		1.62				
	Min		0.58				
	Geomean		0.92				
	Average		1.01				
	Number		3				
Com-17							
	Max	35.20	29.40		3.60	11.60	69.00
	Min	1.20	0.70		0.10	0.20	0.20
	Geomean	7.43	5.28		0.95	1.32	2.02
	Average	12.31	8.02		1.35	2.35	8.00
	Number	18	25		35	42	53
Com-27	Max	3.40	77.00	22.00	10.50	5.20	3.20
	Min	3.40	47.00	14.50	7.20	4.30	2.90
	Geomean	3.40	60.16	17.86	8.69	4.73	3.05
	Average	3.40	62.00	18.25	8.85	4.75	3.05
	Number	1	2	2	2	2	2
Com-30							
	Max					4.70	
	Min					1.40	
	Geomean					2.48	
	Average					2.72	
	Number					6	
Com-31							
	Max					26.60	
	Min					8.60	
	Geomean					14.23	
	Average					15.93	
	Number					3	
Com-32							
	Max					26.20	
	Min					11.60	
	Geomean					16.79	
	Average					17.44	
	Number					5	
Com-33							
	Max	34.50	26.00		8.70	6.70	12.00
	Min	6.90	15.50		0.50	1.50	0.70
	Geomean	15.21	18.79		3.59	3.16	3.02
	Average	18.87	19.10		4.38	3.61	3.82
	Number	6	5		12	11	13

**Table 5 (cont'd)**

Com-34		1986	1987	1988	1989	1990	1991
	Max					10.00	
	Min					3.40	
	Geomean					4.97	
	Average					5.67	
	Number					3	
Com-35							
	Max					54.80	
	Min					6.80	
	Geomean					19.33	
	Average					23.02	
	Number					20	
Com-36							
	Max	38.30	33.60		11.80	8.10	6.70
	Min	5.90	5.00		0.20	0.20	0.20
	Geomean	17.11	11.41		3.23	3.76	2.37
	Average	20.22	13.82		4.82	4.82	3.15
	Number	9	11		17	15	17
Com-37							
	Max						54.00
	Min						4.50
	Geomean						14.38
	Average						19.24
	Number						13
Com-55							
	Max						0.60
	Min						0.30
	Geomean						0.45
	Average						0.47
	Number						3
Com-57							
	Max				1.38	1.90	
	Min				0.32	0.04	
	Geomean				0.86	0.69	
	Average				0.96	0.87	
	Number				7	19	
Com-58							
	Max	8.23	22.90	16.80	17.10	7.60	2.78
	Min	3.52	1.25	0.36	1.09	0.41	0.68
	Geomean	5.45	3.83	2.98	4.07	1.39	1.54
	Average	5.70	6.83	5.71	5.45	2.17	1.65
	Number	8	6	9	11	6	8
Com-59							
	Max						1.40
	Min						0.40
	Geomean						0.97
	Average						1.05
	Number						8

**Table 6a** Concentrations of  $^{137}\text{Cs}$  (Bq kg<sup>-1</sup> dw) in seaweed (Fucus ves. by RPII

		1982	1983	1984	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
Com-17																			
	Max	0.8	1.5		0.4	12.7	1.1	0.5	1	0.1				0.2	0.1				0.1
	Min	0.2	0.3		0.2	0.2	1.1	0.5	1	0.1				0.2	0.1				0.1
	Geomean	0.283	0.671		0.252	1.223	1.1	0.5	1	0.1				0.2	0.1				0.1
	Average	0.35	0.9		0.267	3.875	1.1	0.5	1	0.1				0.2	0.1				0.1
	Number	4	2		3	4	1	1	1	1				2	1				1
Com-28																			
	Max		15.4	8.1	0.2	9.2	0.7	0.8	0.3	0.3	0.2	0.2	0.3	0.8	0.2	0.1			
	Min		15.4	8.1	0.2	0.2	0.7	0.8	0.3	0.3	0.2	0.2	0.3	0.3	0.2	0.1		0.1	0.2
	Geomean		15.4	8.1	0.2	2.875	0.7	0.8	0.3	0.3	0.2	0.2	0.3	0.49	0.2	0.1		0.1	0.2
	Average		15.4	8.1	0.2	4.92	0.7	0.8	0.3	0.3	0.2	0.2	0.3	0.55	0.2	0.1		0.1	0.2
	Number		1	1	2	5	1	1	1	1	1	1	1	2	1	1		1	1
Com-30																			
	Geomean	58				10													
	Number	1				1													
Com-33																			
	Max	35.0	42.4	25.1	11.0	8.4	6.6	4.3	4.3	6.5	4.4	3.5	5.3	2.4	1.4	1.2	1.6	1.0	1.1
	Min	18.6	19.4	16.2	7.4	7.4	5.1	2.9	2.9	2.9	2.6	2.6	2.5	1.2	0.8	0.9	0.9	0.7	0.9
	Geomean	25.9	27.9	21.4	9.0	7.9	5.9	3.8	3.9	4.3	3.5	2.9	3.6	1.7	1.1	1.0	1.1	0.9	1.0
	Average	26.5	29.7	21.6	9.2	7.9	5.9	3.8	3.9	4.5	3.6	3.0	3.8	1.8	1.2	1.0	1.2	0.9	1.0
	Number	5	4	6	2	2	3	4	4	4	3	3	3	4	3	3	4	3	3
Com-36																			
	Max	7.9	5.5	16.9	2.8	7.4		1.3	1.1	1.4	2.0	1.5	1.7	0.6	0.4	0.2	0.3	0.2	0.1
	Min	4.8	1.4	4.1	0.8	7.4		1.3	1.1	1.4	2.0	1.5	1.7	0.4	0.4	0.2	0.3	0.2	0.1
	Geomean	5.7	3.1	8.3	1.5	7.4	4.5	1.3	1.1	1.4	2.0	1.5	1.7	0.5	0.4	0.2	0.3	0.2	0.1
	Average	5.9	3.6	10.5	1.8	7.4		1.3	1.1	1.4	2.0	1.5	1.7	0.5	0.4	0.2	0.3	0.2	0.1
	Number	3	4	2	2	1		1	1	1	1	1	1	2	1	1	1	1	1
Com-38																			
	Max	8.5	0.9		0.4		0.6	0.5	0.3	0.4	0.2	0.9	0.4	0.3	0.2	0.1	0.1	0.1	0.2
	Min	1.3	0.5		0.3		0.5	0.3	0.2	0.4	0.2	0.3	0.4	0.2	0.1	0.1	0.1	0.1	0.1
	Geomean	3.3	0.7		0.3		0.5	0.4	0.2	0.4	0.2	0.5	0.4	0.2	0.1	0.1	0.1	0.1	0.2
	Average	4.9	0.7		0.3		0.6	0.4	0.3	0.4	0.2	0.6	0.4	0.3	0.2	0.1	0.1	0.1	0.2
	Number	2	2		3		2	2	2	1	1	2	2	2	2	2	1	2	3

**Table 6b**      **Concentrations of  $^{137}\text{Cs}$  (Bq kg<sup>-1</sup> dw) in seaweed (Fucus ves. By MAFF and BNFL)**

		1988	1989	1991	1992	1993	1994	1995	1996	1997	1998	1999
MAFF data												
Com-28												
	Max	4.10	4.50	1.30	3.20	2.10	1.70	1.10	0.91	1.10	1.40	1.70
	Min	1.10	0.80	0.96	0.31	0.60	0.29	0.42	0.26	0.32	0.13	0.16
	Geomean	2.01	1.53	1.07	0.99	1.08	0.69	0.66	0.49	0.56	0.52	0.65
	Average	2.33	2.10	1.08	1.49	1.23	0.89	0.72	0.59	0.66	0.71	0.95
	Number	3	3	3	3	3	3	3	2	4	5	3
Com-32												
	Max	24.00	26.00	18.00	13.00	14.00	12.00	10.00	8.00	7.80	6.30	6.70
	Min	18.00	15.00	10.00	11.00	8.50	7.10	6.30	4.90	6.90	4.30	0.24
	Geomean	20.78	19.75	12.93	11.96	10.91	9.23	7.94	6.26	7.34	5.20	2.20
	Average	21.00	20.50	13.33	12.00	11.25	9.55	8.15	6.45	7.35	5.30	4.51
	Number	2	2	3	2	2	2	2	2	2	2	3
Com-36												
	Max	1.40	2.20	0.75	1.70	1.70	0.65	0.38	0.52	0.61	0.48	0.45
	Min	1.40	0.30	0.31	0.36	0.40	0.36	0.23	0.22	0.29	0.22	0.14
	Geomean	1.40	0.81	0.45	0.67	0.87	0.48	0.29	0.30	0.40	0.36	0.25
	Average	1.40	1.25	0.48	0.85	1.02	0.49	0.30	0.32	0.42	0.38	0.28
	Number	1	2	3	3	3	3	3	3	3	3	3
Com-35												
	Max	28.00	34.00	22.00	17.00	14.00	14.00	13.00	13.00	13.00	9.30	9.20
	Min	18.00	23.00	7.70	13.00	13.00	9.70	9.90	7.80	7.40	5.30	7.50
	Geomean	22.45	27.96	15.40	14.87	13.49	11.65	11.34	10.07	9.81	7.02	8.31
	Average	23.00	28.50	16.28	15.00	13.50	11.85	11.45	10.40	10.20	7.30	8.35
	Number	2	2	6	2	2	2	2	2	2	2	2
BNFL data												
Com-35												
		1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001
	Max	17.00	20.00	20.00	16.00		19.70	17.80	11.10	11.60	10.30	6.97
	Min	13.00	12.00	11.00	8.10		3.62	1.69	2.39	2.22	1.65	2.39
	Geomean	14.87	16.16	15.37	11.35		8.14	6.57	6.09	5.44	5.09	5.14
	Average	15.00	16.40	15.80	11.62		9.00	7.21	6.46	5.77	5.55	5.42
	Number	2	5	5	5		26	32	32	33	32	10



**Table 7a** Concentrations of  $^{137}\text{Cs}$  ( $\text{Bq kg}^{-1}$  dw) in sediment

		1988	1989	1991	1992	1993	1994	1995	1996	1997	1998	1999
Com-28												
	Max	230.0	240.0	170.0	160.0	180.0	53.0	120.0	110.0	100.0	79.0	85.0
	Min	18.0	14.0	1.6	1.1	1.9	1.5	1.1	0.9	0.9	1.1	1.2
	Geomean	64.4	56.9	30.6	28.3	28.5	10.9	15.5	18.7	24.5	16.6	15.2
	Average	104.5	99.7	67.9	57.5	52.2	21.2	35.9	38.5	45.1	32.0	33.6
	Number	6	6	11	10	8	5	8	8	7	8	7
Com-31												
	Value				670	590	450	470	390	470		
Com-32												
	Max	1900.0	1300.0	1100.0	1000.0	800.0	680.0	550.0	640.0	580.0	510.0	750.0
	Min	520.0	390.0	190.0	200.0	170.0	78.0	260.0	220.0	130.0	48.0	65.0
	Geomean	790.1	596.3	411.1	500.8	316.5	336.4	327.2	320.4	212.1	165.1	187.0
	Average	891.7	653.3	464.4	577.1	350.0	378.9	340.0	338.6	234.0	229.4	249.3
	Number	6	6	9	7	12	11	7	7	10	7	7
Com-33												
	Value			250	250	270	220	180	150	170	160	
Com-34												
	Max			310.0	700.0	360.0	370.0	260.0	230.0	220.0	9.9	21.0
	Min			310.0	230.0	16.0	14.0	13.0	55.0	77.0	9.9	21.0
	Geomean			310.0	364.2	103.2	111.8	79.7	96.9	123.1	9.9	21.0
	Average			310.0	410.0	183.5	218.0	141.0	119.0	135.7	9.9	21.0
	Number			1	3	4	3	3	3	3	1	1
Com-35												
	Max	2100.0	1900.0	1400.0	4500.0	5900.0	2500.0	3400.0	1800.0	830.0	840.0	630.0
	Min	120.0	110.0	87.0	19.0	85.0	72.0	45.0	57.0	56.0	83.0	84.0
	Geomean	555.6	462.6	503.3	479.0	458.2	348.3	311.3	337.7	303.7	262.5	223.9
	Average	794.0	644.2	689.3	922.2	790.1	558.9	545.4	498.8	398.5	336.9	284.3
	Number	10	12	18	22	23	23	22	22	17	11	9
Com-36												
	Value	61	41	44	32	34	30	8.7	8.2	7.5		

**Table 7a (cont'd)**

		1988	1989	1991	1992	1993	1994	1995	1996	1997	1998	1999
Com-37												
	Max	1300.0	1200.0	1400.0	2100.0	1800.0	1700.0	1300.0	1300.0	730.0		
	Min	21.0	17.0	14.0	13.0	12.0	10.0	7.9	9.3	11.0		
	Geomean	205.4	210.1	299.4	207.8	287.3	181.4	132.7	187.9	151.4		
	Average	508.7	488.7	497.3	489.5	575.4	443.4	343.2	383.6	287.4		
	Number	6	6	12	14	10	9	9	9	8		

**Table 7b**      **Inventory of  $^{137}\text{Cs}$  in North Sea sediment cores (kBq/m<sup>2</sup>)**

Compartment	57	58	59	60
Max	7.60	6.00	8.80	14.60
Min	0.70	4.70	0.20	4.30
Geomean	2.61	5.31	1.63	6.84
Average	3.26	5.35	2.59	7.62
Number	11	2	12	9

**Table 8                      Concentrations of <sup>99</sup>Tc in seawater (Bq m<sup>-3</sup>)**

		1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
Com-28												
	Max				2.80		15.40	32.00	4.30			
	Min				0.10		0.10	1.80	1.60			
	Geomean				0.40		0.85	8.71	2.65			
	Average				0.62		3.06	11.56	2.80			
	Number				9.00		14.00	15.00	9.00			
Com-28 (Larne)												
	Average						8.1	29	28	36	280	24
Com-29												
	Max	0.48				0.20	2.50			10.00		5.70
	Min	0.24				0.10	0.60			0.60		2.30
	Geomean	0.37				0.14	1.21			2.80		3.56
	Average	0.39				0.15	1.40			3.87		3.78
	Number	3.00				6.00	4.00			13		4
Com-30												
	Average				2.5		30	45	60			
	SD				1		5	20	20			
Com-31												
	Average				7.5		45					
	SD				2.5		15					
Com-32												
	Average				20		200	190	350			
	SD				5		50	100	150			
Com-33												
	Average				1.5			23	23	43	22	22
	SD				0.5			13	10	21	11	10
Com-34												
	Average				3.5		8.5					
	SD				1.5		5					
Com-35												
	Average				25		95	200	1200	500	110	160
	SD				5		30	50	400	100	50	40
Com-37												
	Average				10		12					
	SD				5		6					
Com-38												
	Max					0.20					0.44	0.40
	Min					0.10					0.40	0.30
	Geomean					0.12					0.42	0.35
	Average					0.13					0.42	0.35
	Number					6					2	2
Com-39												
	Max		0.41	0.44	0.67	0.20					0.40	0.40
	Min		0.13	0.16	0.10	0.10					0.30	0.30
	Geomean		0.22	0.28	0.28	0.14					0.32	0.33
	Average		0.26	0.29	0.32	0.15					0.32	0.33
	Number		4	12	8	4					5	3

**Table 8 (cont'd)**

		1987	1988	1989	1990	1991	1992	1993	1994
Com-16									
	Max	0.147	0.110		0.063				0.220
	Min	0.041	0.024		0.019				0.050
	Geomean	0.072	0.050		0.034				0.106
	Average	0.077	0.054		0.036				0.118
	Number	11	46		21				6
Com-17									
	Max	0.107			0.072				0.090
	Min	0.026			0.011				0.020
	Geomean	0.063			0.024				0.039
	Average	0.070			0.027				0.044
	Number	6			14				5
Com-23,24									
	Max								0.080
	Min								0.020
	Geomean								0.041
	Average								0.044
	Number								0.016
Com-27									
	Max								0.140
	Min								0.060
	Geomean								0.083
	Average								0.088
	Number								6

Table 8 (cont'd)

	1983	1984	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
Com-47																	
Max							0.93	0.53	0.47	1.44							
Min							0.93	0.53	0.03	0.71							
Geomean							0.93	0.53	0.13	0.90							
Average							0.93	0.53	0.25	0.96							
Number							1	1	2	3							
Com-48																	
Max	19.60	26.00	54.00	35.00	20.00	8.30	11.60	11.20	4.46	1.71	0.98						
Min	4.80	7.40	6.80	6.30	2.00	0.90	2.00	0.09	0.16	0.06	0.37						
Geomean	12.72	13.68	19.23	11.34	7.84	2.95	4.82	2.59	1.99	0.79	0.53						
Average	13.68	15.54	22.91	13.29	9.00	3.61	5.24	4.37	2.28	0.97	0.56						
Number	11	7	12	10	12	9	12	11	26	27	10						
Com-50																	
Max							4.92	3.69	4.50	2.28	0.29						
Min							3.63	0.52	0.58	0.30	0.29						
Geomean							4.30	1.83	1.29	1.03	0.29						
Average							4.33	2.28	1.55	1.31	0.29						
Number							3	4	12	9	1						
Com-51																	
Max							4.34	3.64	3.20	2.96	0.78						
Min							0.42	1.52	0.83	0.63	0.24						
Geomean							0.99	2.33	1.86	1.24	0.49						
Average							1.76	2.42	1.99	1.39	0.54						
Number							3	6	18	15	4						
Com-52																	
Max							4.91	4.91	3.20	1.85	0.45						
Min							0.90	0.90	0.50	0.07	0.14						
Geomean							2.38	2.38	1.19	0.45	0.23						
Average							2.95	2.95	1.50	0.79	0.27						
Number							3	3	9	7	3						

Table 8 (cont'd)

	1983	1984	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
Com-53																	
Max									0.35	1.29	0.16						
Min									0.35	0.06	0.16						
Geomean									0.35	0.26	0.16						
Average									0.35	0.50	0.16						
Number									1	5	1						
Com-54																	
Max								1.83	1.26								
Min								0.57	0.24								
Geomean								1.05	0.61								
Average								1.13	0.68								
Number								6	9								
Com-55																	
Max							2.80	3.94	2.84	2.57	0.30	0.60		3.00	7.10		4.30
Min							0.59	0.18	0.22	0.01	0.08	0.10		3.00	6.50		1.00
Geomean							1.50	1.16	0.87	0.26	0.17	0.24		3.00	6.79		2.07
Average							1.73	1.39	1.02	0.47	0.19	0.35		3.00	6.80		2.65
Number							4	18	49	34	8	2		1	2		2
Com-56																	
Max					5.00	5.50	3.09	4.47	3.82	2.76	0.74			0.30	0.10		
Min					2.60	2.30	1.24	0.42	0.36	0.08	0.17			0.10	0.10		
Geomean					3.65	3.74	1.88	1.92	1.56	0.77	0.41			0.17	0.10		
Average					3.75	3.83	1.95	2.17	1.74	0.91	0.46			0.20	0.10		
Number					20	22	30	33	60	54	19			2	1		
Com-57																	
Max							2.60	2.93	2.22	1.08	0.45	1.20		10.00	6.80		
Min							0.31	0.45	0.18	0.09	0.10	0.20		0.70	1.80		
Geomean							0.72	0.98	0.61	0.30	0.18	0.47		2.81	4.13		
Average							0.91	1.32	0.75	0.34	0.21	0.53		3.87	4.49		
Number							9	4	31	24	19	9		6	16		

Table 8 (cont'd)

	1983	1984	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
Com-58																	
Max						4.40	3.65	3.33	3.09	1.43	0.58			0.35	1.40		2.90
Min						1.78	0.68	0.55	0.31	0.11	0.37			0.35	0.40		0.20
Geomean						2.76	1.81	1.62	1.33	0.62	0.48			0.35	0.53		0.61
Average						2.94	1.93	1.79	1.45	0.72	0.49			0.35	0.60		0.87
Number						6	34	24	98	36	10			1	6		7
Com-59																	
Max				2.17	1.71	1.39	1.39	1.26	0.74	0.45	0.32			6.50	3.40		
Min				1.02	0.62	0.73	0.11	0.31	0.02	0.34	0.27			0.90	0.70		
Geomean				1.59	1.16	1.08	0.60	0.61	0.35	0.39	0.29			2.02	2.06		
Average				1.63	1.21	1.10	0.67	0.65	0.40	0.39	0.29			2.87	2.33		
Number				7	6	14	25	36	31	12	4			11	6		
Com-60																	
Max							2.88	2.31	1.44	1.02	0.49				1.90		
Min							0.10	0.10	0.25	0.35	0.49				0.80		
Geomean							1.18	0.71	0.50	0.67	0.49				1.40		
Average							1.50	0.94	0.55	0.72	0.49				1.48		
Number							11	18	26	10	1				4		
Com-61																	
Max		2.70	2.40	2.01	0.81	0.85	1.62	1.53	0.95	0.88	0.89						
Min		0.54	0.58	0.41	0.54	0.41	0.16	0.21	0.10	0.14	0.13						
Geomean		1.06	0.97	0.85	0.68	0.61	0.59	0.43	0.38	0.36	0.39						
Average		1.00	1.13	1.05	0.70	0.61	0.65	0.48	0.41	0.40	0.40						
Number		11	9	9	5	11	40	31	54	28	12						
Com-62																	
Max							1.21	1.21	1.21	0.88	0.49						
Min							0.21	0.21	0.21	0.17	0.17						
Geomean							0.42	0.42	0.42	0.37	0.27						
Average							0.50	0.50	0.50	0.40	0.29						
Number							17	17	17	17	8						



**Table 9      Concentrations of <sup>99</sup>Tc in seawater, biota and sediment near Sellafield**

	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001
Tc-Water Bq/m <sup>3</sup>															
Max									451	37700	5140	5640	1550	4370	168
Min									30	395	231	17.6	54.1	42.3	21.3
Geomean									104.3	2060.2	512.5	193.5	171.4	196.8	75.0
Average									158.0	6783.4	739.5	435.3	249.2	377.9	90.4
Number									17	9	30	34	43	45	10
Tc-Fish Bq/kg															
Max								34.8	68	679	505	139	15	27.9	26.4
Min								0.61	0.79	0.81	1.40	1.03	0.77	0.84	1.28
Geomean								1.67	5.12	19.82	21.10	7.54	2.58	3.54	2.55
Average								3.31	8.64	48.62	54.22	15.01	3.69	5.80	3.97
Number								64	47	48	43	68	57	55	15
Tc-crustaceans Bq/kg															
Max		150	310	160	320	150	210	2300	9100	36000	51600	25800	7520	5750	464
Min		35	3.1	45	25	1.3	0.86	0.96	1.1	25.4	6.9	1.65	2.16	14.6	450
Geomean		70.40	70.15	77.64	76.44	13.53	15.25	34.48	363.99	1128.74	2444.48	713.40	524.88	813.09	456.95
Average		87.75	158.28	90.00	112.42	39.07	40.14	335.23	1966.79	3895.35	9687.13	3956.09	2328.99	2207.47	457.00
Number		4	4	3	12	13	20	15	15	56	42	50	45	33	2
Tc-molluscs Bq/kg															
Max	170	130	300	120	130	130	160	740	7100	9450	10300	14200	2240	5390	2090
Min	39	33	26	17	24	0.98	9.3	1.2	44	102	39.8	4.35	20.1	22.7	30.5
Geomean	75.01	60.54	72.08	63.84	53.54	17.91	40.67	46.03	844.05	1166.75	1224.31	570.33	404.25	446.01	490.58
Average	81.50	65.20	98.06	70.24	63.83	35.85	51.41	122.89	1577.34	1972.74	2287.73	1664.72	792.26	948.28	825.36
Number	12	20	16	17	12	15	17	38	39	58	43	50	45	42	9
Tc-Seaweed(FV) Bq/kg															
Max						1800	2300	35600	126000	164000	179000	125000	68500	32600	22500
Min						1.5	190	82.3	30.8	1220	797	943	1450	511	1340
Geomean						142.32	924.58	2138.74	8136.09	18152.60	15937.26	13242.17	7752.46	7948.52	7694.19
Average						561.83	1072.79	6360.84	28252.49	37102.82	33894.03	25321.43	12295.95	12375.50	10763.33
Number						19	29	23	36	39	29	44	37	36	9

Table 9 (cont'd)

Tc-Sediment Bq/kg															
Max												249000			
Min												16800			
Geomean												78904			
Average												118760			

**Table 10      Concentrations of <sup>99</sup>Tc in shellfish (Bq kg<sup>-1</sup> ww)**

		1988	1989	1991	1992	1993	1994	1995	1996	1997	1998	1999
Com-28	Max					4.3	17.0	42.0	150.0	190.0	190.0	180.0
	Min					4.0	12.0	22.0	26.0	43.0	61.0	23.0
	Geomean					4.1	14.3	30.4	58.3	74.8	96.6	56.2
	Average					4.2	14.5	32.0	71.3	91.0	106.3	77.0
	Number					2	2	2	4	4	4	4
Com-32												
	Max				22.0	31.0	390.0	850.0	1700.0	1800.0	1400.0	640.0
	Min				22.0	31.0	5.6	2.5	3.5	28.0	5.0	5.5
	Geomean				22.0	31.0	41.4	47.0	57.7	228.9	130.2	68.6
	Average				22.0	31.0	150.6	262.9	381.5	590.4	407.4	221.6
	Number				1	1	4	5	5	5	7	6
Com-33												
	Max								118.0	465.0	433.0	175.0
	Min								5.0	2.7	0.6	2.2
	Geomean								30.7	64.5	34.6	26.8
	Average								44.2	119.5	84.1	47.9
	Number								12	27	25	18
Com-34												
	Value								250.0	180.0	220.0	260.0
Com-35												
	Max	380	97	220	170	390	2400	8300	13000	16000	7700	4700
	Min	36.0	8.4	3.5	1.8	2.0	9.2	25.0	33.0	44.0	28.0	38.0
	Geomean	103.2	39.2	31.5	24.8	33.6	162.5	661.4	880.4	758.6	478.0	384.3
	Average	141.2	50.9	69.2	48.9	77.4	489.3	1723.3	2285.9	2453.4	1391.3	995.9
	Number	5	5	8	11	11	13	13	14	13	13	15
Com-37												
	Max	1	1	1	4	7	54	250	400	3300	2500	2100
	Min	0.3	0.5	0.5	0.7	0.6	1.7	4.7	4.9	2.3	1.0	1.5
	Geomean	0.6	0.8	0.8	1.7	1.8	9.6	32.9	39.1	82.8	42.8	26.2
	Average	0.7	0.8	0.8	2.5	2.6	27.9	72.3	105.8	663.1	470.5	338.6
	Number	2	2	2	2	4	2	5	5	6	6	7
Com-38												
	Value								63.0	56.0	64.0	27.0
Com-55												
	Value									2.10	3.50	3.00
Com-59												
	Value								14.00	24.00	16.00	9.40
Com-59												
	Value								14.00	24.00	16.00	9.40

**Table 11 Comparison of  $^{99}\text{Tc}$  concentrations in shellfish ( $\text{Bq kg}^{-1}$  ww) from the Irish Sea near Sellafield (compartment 35) measured by BNFL and MAFF**

		1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001
BNFL-Tc-crustaceans(35)																
	Max		150	310	160	320	150	210	2300	9100	36000	5140	5640	1550	4370	168
	Min		35	3.1	45	25	1.3	0.86	0.96	1.1	25.4	231	17.6	54.1	42.3	21.3
	Geomean		70.40	70.15	77.64	76.44	13.53	15.25	34.48	363.99	1128.74	512.5	193.5	171.4	196.8	75.0
	Average		87.75	158.28	90.00	112.42	39.07	40.14	335.23	1966.79	3895.35	739.5	435.3	249.2	377.9	90.4
	Number		4	4	3	12	13	20	15	15	56	30	34	43	45	10
BNFL-Tc-molluscs (35)																
	Max	170	130	300	120	130	130	160	740	7100	9450	505	139	15	27.9	26.4
	Min	39	33	26	17	24	0.98	9.3	1.2	44	102	1.40	1.03	0.77	0.84	1.28
	Geomean	75.01	60.54	72.08	63.84	53.54	17.91	40.67	46.03	844.05	1166.75	21.10	7.54	2.58	3.54	2.55
	Average	81.50	65.20	98.06	70.24	63.83	35.85	51.41	122.89	1577.34	1972.74	54.22	15.01	3.69	5.80	3.97
	Number	12	20	16	17	12	15	17	38	39	58	43	68	57	55	15
MAFF-shellfish (35)																
	Max		380	97		220	170	390	2400	8300	13000	16000	7700	4700		
	Min		36	8.4		3.5	1.8	2	9.20	25.00	33.00	44.00	28.00	38.00		
	Geomean		103.239	39.1513		31.5298	24.7792	33.6094	162.47	661.40	880.44	758.56	478.02	384.33		
	Average		141.2	50.88		69.1625	48.8909	77.4	489.32	1723.31	2285.93	2453.38	1391.31	995.93		
	Number		5	5		8	11	11	13	13	14	13	13	15		

**Table 12      Concentrations of <sup>90</sup>Sr in seawater (Bq m<sup>-3</sup>)**

	1977	1978	1979	1980	1981	1982	1983	1984	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
Com-16-S																							
Max	8.10	3.70	3.70	4.40		5.50	5.80	4.23	4.24		4.30	5.00		3.90				3.88	1.90				
Min	8.10	3.70	3.70	4.40		5.50	5.80	0.75	1.33		2.10	1.37		0.50				2.88	1.10				
Geomean	8.10	3.70	3.70	4.40		5.50	5.80	2.03	2.39		3.21	2.47		2.20				3.19	1.52				
Average	8.10	3.70	3.70	4.40		5.50	5.80	2.24	2.56		3.31	2.60		2.37				3.22	1.53				
Number	1	1	1	1		1	1	23	24		10	39		84				4	18				
Com-16-D																							
Max								0.80	0.93			2.00		1.02					0.90				
Min								0.20	0.70			0.84		0.63					0.90				
Geomean								0.43	0.80			1.30		0.80					0.90				
Average								0.49	0.81			1.36		0.81					0.90				
Number								5	12			4		10					2				
Com-17																							
Max	3.38	3.11	6.30	1.84	2.73	2.82	2.12	2.17	9.74	2.70	3.50	2.30	1.82	2.12	1.27	0.94	1.44	1.40	1.60	1.20	1.44	1.21	1.78
Min	2.66	2.64	0.28	1.84	2.17	2.14	1.96	1.96	0.84	1.79	1.57	1.78	1.67	1.02	1.24	0.55	1.39	1.17	1.17	1.20	1.21	1.21	0.98
Geomean	2.98	2.87	2.04	1.84	2.46	2.46	2.05	2.07	2.64	2.02	2.36	1.97	1.73	1.55	1.25	0.72	1.41	1.28	1.40	1.20	1.32	1.21	1.27
Average	3.00	2.88	2.40	1.84	2.47	2.48	2.05	2.07	2.90	2.04	2.41	1.98	1.73	1.57	1.26	0.75	1.42	1.29	1.40	1.20	1.33	1.21	1.31
Number	4	2	28	1	4	2	3	3	86	7	15	4	3	37	2	2	2	2	18	1	2	1	3
Com-18																							
Max									2.43		0.84			1.14					0.40				
Min									0.36		0.84			0.73					0.40				
Geomean									0.60		0.84			0.90					0.40				
Average									0.69		0.84			0.91					0.40				
Number									28		1			3					2				
Com-19-S																							
Max						5.70		5.07	4.11		5.00			1.89					1.50				
Min						1.40		1.47	1.68		2.50			1.89					1.10				
Geomean						3.43		2.69	2.96		3.60			1.89					1.37				
Average						3.78		2.85	3.04		3.70			1.89					1.38				
Number						6		14	29		6			2					16				
Com-19-D																							
Max						1.10		1.23	1.46										1.20				
Min						0.50		0.43	0.56										0.60				
Geomean						0.72		0.74	0.99										0.85				
Average						0.76		0.80	1.04										0.88				
Number						5		4	16										8				
Com-24																							
Max	11.00	13.00	15.00	18.00	27.00	14.00	12.00	12.00	13.00	17.00	9.30	9.00	9.70	17.00	17.00	8.10	2.50		1.90				
Min	9.30	5.20	4.80	7.00	11.00	0.55	6.50	4.50	2.97	9.40	4.70	5.20	4.40	3.80	3.10	2.70	2.00		1.40				
Geomean	10.11	8.03	7.58	11.97	14.55	4.11	8.91	7.45	4.91	12.17	6.62	6.47	6.67	6.81	5.00	4.73	2.24		1.61				
Average	10.15	8.18	8.00	12.50	15.43	6.52	9.06	7.82	5.55	12.46	6.75	6.58	6.83	7.87	6.04	5.01	2.25		1.61				
Number	2	43	15	6	7	12	7	13	30	5	6	6	7	12	14	12	2		42				
Com-27																							
Max			16.00			7.17			16.00		6.70			3.31					2.30				
Min			3.30			0.48			3.21		3.80			3.31					1.70				
Geomean			7.72			2.61			9.65		5.42			3.31					2.05				
Average			8.36			3.70			10.64		5.57			3.31					2.06				
Number			30			11			20		9			2					10				
Com-28																							
Max								33.70			7.30	19.50	2.80	3.30					3.80		3.80		
Min								15.10			7.10	5.80	2.00	2.10					3.80		1.80		
Geomean								23.18			7.20	8.45	2.37	2.70					3.80		2.66		
Average								24.52			7.20	9.70	2.40	2.75					3.80		2.76		
Number								5			2	4	2	11			0	0	2		9		

Table 12 (cont'd)

	1977	1978	1979	1980	1981	1982	1983	1984	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
Com-29																							
Max								28.70			6.70	6.10	4.30	3.30					4.70		3.50		
Min								11.40			4.40	5.60	3.27	2.80					3.50		1.90		
Geomean								19.02			5.40	5.80	3.76	3.08					4.22		2.55		
Average								19.58			5.48	5.80	3.79	3.09					4.24		2.58		
Number								16			4	3	4	8					10		9		
Com-47																							
Max								7.30				5.50	6.60	3.60						2.40	1.60		
Min								7.30				5.50	2.80	3.60						2.40	1.30		
Geomean						7.30					5.50	4.30	3.60						2.40	1.44			
Average								7.30				5.50	4.30	3.60						2.40	1.45		
Number								1				1	2	2						1	2		
Com-48																							
Max								47.50				4.30	9.10	30.70	4.37				2.40	4.10			
Min								12.90				4.30	3.10	2.50	2.43				2.40	1.90			
Geomean								24.75				4.30	4.75	7.00	3.43				2.40	2.62			
Average								30.20				4.30	5.33	11.95	3.54				2.40	2.77			
Number								2				1	3	8	12				1	3			
Com-50																							
Max								46.20				14.10	24.60	13.60	17.90	12.80			20.10	6.10	3.10		
Min								46.20				14.10	17.60	4.30	12.40	12.80			2.90	3.60	3.10		
Geomean								46.20				14.10	20.50	8.59	14.90	12.80			6.01	4.87	3.10		
Average								46.20				14.10	20.70	9.33	15.15	12.80			8.18	4.98	3.10		
Number								1				1	3	8	8	2			4	4	2		
Com-52																							
Max								52.90			21.90	13.60	23.50	8.80	18.60	12.90			8.00	5.80	3.50		
Min								8.60			16.10	2.90	9.20	8.70	16.30	5.39			2.00	4.00	2.80		
Geomean								20.08			18.27	5.58	16.45	8.75	17.64	8.22			3.51	4.71	3.13		
Average								26.43			18.43	6.97	17.77	8.75	17.66	8.64			4.13	4.75	3.15		
Number								3			3	3	3	4	10	8			4	4	2		
Com-53																							
Max												21.30	4.30	3.50	12.30				2.10	3.40			
Min												21.30	4.30	2.90	12.30				2.00	3.10			
Geomean												21.30	4.30	3.19	12.30				2.05	3.25			
Average												21.30	4.30	3.20	12.30				2.05	3.25			
Number												1	1	4	4				2	2			
Com-55																							
Max								23.90			12.50	15.70	20.50	7.03	18.14	14.38			4.20	5.90	3.90		
Min								22.00			12.50	4.10	5.80	4.20	4.20	3.32			2.10	3.30	2.90		
Geomean								22.93			12.50	8.63	10.54	4.93	9.70	6.08			3.01	4.23	3.60		
Average								22.95			12.50	9.56	11.60	4.98	10.49	6.92			3.13	4.37	3.63		
Number								2			1	5	8	14	12	9			3	3	4		
Com-56																							
Max								39.30			40.70	36.00	25.90	16.00	18.42	16.60			8.50	8.00	4.40		
Min								7.10			12.00	6.00	6.10	4.30	4.20	3.48			3.10	3.20	3.00		
Geomean								23.69			25.50	19.26	13.63	7.83	11.14	10.93			5.94	6.03	3.57		
Average								26.52			27.44	21.50	14.22	8.50	12.03	11.94			6.30	6.24	3.59		
Number								9			18	8	22	51	72	36			13	13	14		
Com-57																							
Max								51.50	37.00		41.40	19.50	18.60	14.82	13.40	10.60	12.90		8.00	8.60	4.40		
Min								19.40	16.50		6.00	5.40	3.20	2.20	3.09	2.62	12.90		2.70	4.10	1.90		
Geomean								26.84	24.54		12.94	7.87	7.68	4.9	5.14	4.44	12.90		4.03	5.58	3.35		
Average								27.76	26.18		14.26	8.79	8.37	5.69	5.48	4.83	12.90		4.26	5.77	3.44		
Number								28	5		21	12	26	55	106	52	2		14	14	18		

**Table 12 (cont'd)**

	1977	1978	1979	1980	1981	1982	1983	1984	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
Com-58																							
Max								57.10	36.90	39.60	39.10	18.90	18.00	18.10	13.00	12.70	13.60		13.80	11.40	13.10		
Min								19.50	20.80	17.70	11.30	8.10	6.06	3.10	3.90	3.01	10.40		3.90	3.80	3.40		
Geomean								32.05	28.76	25.17	18.87	14.15	12.87	9.04	7.35	10.11	12.26		7.29	5.44	5.11		
Average								34.39	29.15	26.04	19.72	14.42	13.09	9.91	7.55	10.40	12.33		7.64	5.79	5.43		
Number								23	21	10	40	15	40	63	170	73	8		32	31	32		
Com-59																							
Max								24.10	24.00	15.40		8.10	8.20	9.19		7.04			5.60		1.90		
Min								2.50	3.99	11.00		7.30	1.58	2.50		2.52			1.40		1.40		
Geomean						7.25		11.02	13.57		7.69	4.28	4.56		4.44			2.61		1.74			
Average								9.18	13.70	13.68		7.70	4.98	5.25		4.86			2.86		1.74		
Number								24	7	4		2	5	14		7			16		9		
Com-60																							
Max								31.10	24.00		13.80	12.90	13.00	10.40	10.20	7.82			4.50	8.70			
Min								11.00	18.40		13.60	3.80	2.60	1.90	2.41	2.13			2.00	2.70			
Geomean								18.81	20.10		13.70	6.53	5.14	4.72	4.61	3.81			2.79	4.62			
Average								19.78	20.27		13.70	7.05	5.93	5.16	5.06	4.30			2.95	4.95			
Number								12	3		2	8	30	26	49	14			8	6			
Com-61																							
Max								25.90	22.70	43.00	14.00	19.20	17.90	19.80	13.80	10.20			12.80	11.20	12.00		
Min								15.20	19.80	17.00	8.90	8.22	3.90	19.80	2.20	3.85			4.40	2.40	3.90		
Geomean								21.29	21.24	20.55	11.50	15.94	10.95	19.80	8.63	7.37			7.47	6.37	6.72		
Average								21.52	21.28	21.13	11.65	16.47	12.10	19.80	9.28	7.87			8.50	7.37	7.75		
Number								13	4	14	6	7	6	2	45	10			4	9	4		

**NBFL data on 90Sr (35)**

								1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001
Com-35																							
Max								1100	1300	530	780		400	360	760	790	505	930	841	498	514	504	309
Min								120	220	180	230		70	190	260	300	471	453	272	259	227	265	295
Geomean								313	4.35	326	508		228	278	435	518	488	578	418	366	403	315	299
Average								313	435	326	508		228	278	435	518	488	578	418	366	403	315	299
Number								29	36	32	16		4	3	5	3	2	4	11	15	21	5	5
	* most of data are lower than LD							* most of data are lower than LD															

**Table 13** Concentrations of  $^{90}\text{Sr}$  ( $\text{Bq kg}^{-1}$  ww) in shellfish in the Irish Sea

		1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001
NBFL date (com-35)																	
Sr-fish																	
	Max	5.30	13.00	4.70	5.50	5.90	4.90	7.70	0.83	0.36	0.72	1.21	1.33	1.18	0.68	0.27	0.18
	Min	1.10	1.80	2.10	3.00	3.20	2.70	0.07	0.09	0.08	0.12	0.14	0.11	0.12	0.10	0.09	0.12
	Geomean	2.00	3.04	3.14	3.91	4.19	3.59	0.81	0.22	0.16	0.24	0.29	0.25	0.26	0.16	0.16	0.14
	Average	2.16	3.25	3.22	3.96	4.25	3.64	2.06	0.25	0.18	0.28	0.36	0.30	0.31	0.17	0.16	0.14
	Number	40	54	61	17	20	19	18	26	34	30	37	46	57	56	48	20
* Before 1993, most of data are lower than detection limit (LD), the data before that date are the estimated upper limit																	
Sr-molluscs																	
	Max	130.00	180.00	230.00	31.00	63.00	27.00	16.00	12.00	20.00	22.00	26.60	40.80	66.00	13.80	9.50	9.99
	Min	1.20	1.10	3.40	5.20	7.80	4.10	1.10	1.50	1.50	0.98	0.27	0.24	0.18	0.20	0.12	0.31
	Geomean	10.40	11.04	12.47	14.26	15.76	11.24	6.72	5.27	4.04	4.50	4.04	3.08	2.80	1.94	1.75	1.57
	Average	14.36	14.65	17.16	16.13	18.62	12.61	8.56	6.12	5.04	5.99	6.22	6.08	6.51	3.11	2.58	2.46
	Number	93	92	75	20	17	13	15	17	37	40	42	49	42	49	43	9
* Before 1992, most of data are lower than LD, the data before that date are the estimated upper limit																	
Sr-Crustean																	
	Max	21.00	11.00	19.00	10.00	21.00	9.70	9.30	4.00	7.90	4.80	3.34	2.76	4.89	1.98	2.25	0.39
	Min	2.30	2.70	3.20	4.00	2.80	2.80	0.36	0.14	0.13	0.23	0.22	0.11	0.16	0.10	0.12	0.16
	Geomean	4.82	5.80	5.87	6.35	6.75	5.86	1.43	0.61	0.69	0.97	0.63	0.51	0.57	0.41	0.45	0.29
	Average	6.08	6.34	6.54	6.61	7.59	6.10	2.36	0.93	1.40	1.20	0.82	0.65	0.90	0.57	0.66	0.31
	Number	12	21	23	19	16	24	23	30	22	26	28	37	26	28	22	4
* Before 1992, most data are lower than the detection limit																	
MAFF shellfish data																	
Com-35																	
	Max			16.00	14.00	13.00	12.00	12.00	16.00	11.00	10.00	6.30	10.00	9.40	8.30		
	Min			0.45	1.50	0.14	0.09	0.15	0.14	0.07	0.14	0.13	0.08	0.25	0.13		
	Geomean			5.93	7.29	1.99	1.01	1.32	0.98	1.13	2.18	1.25	1.07	2.12	1.24		
	Average			9.51	9.26	5.22	3.67	4.06	3.73	2.97	4.22	2.56	2.80	3.75	2.60		
	Number			5	5	8	10	9	9	12	11	12	11	10	13		



Table 13 (cont'd)

		1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001
Com-37	Max								1.40	1.60	1.90	0.70	0.91	1.60	0.99		
	Min								0.07	0.08	0.08	0.07	0.24	0.13	0.48		
	Geomean								0.31	0.36	0.40	0.25	0.46	0.51	0.69		
	Average								0.74	0.70	0.74	0.38	0.54	0.69	0.74		
	Number								2	5	5	5	4	5	2		

**Table 14** Concentrations of  $^{90}\text{Sr}$  in shellfish in the Irish Sea

Com	Location	Material	No. Samples	$^{90}\text{Sr}$	Unit	Year
32	Inner Solway	Shrimps	4	0.07	Bq/kg ww	1993
32	Inner Solway	Shrimps	4	0.082	Bq/kg ww	1994
32	Inner Solway	Shrimps	4	0.16	Bq/kg ww	1995
32	Inner Solway	Shrimps	4	0.092	Bq/kg ww	1996
32	North Solway coast	Cockles	4	1.3	Bq/kg ww	1994
32	North Solway coast	Cockles	4	1.2	Bq/kg ww	1995
32	North Solway coast	Cockles	4	0.7	Bq/kg ww	1996
32	North Solway coast	Cockles	6	0.75	Bq/kg ww	1997
32	North Solway coast	Cockles	8	0.74	Bq/kg ww	1998
32	North Solway coast	Cockles	8	0.99	Bq/kg ww	1999
32	North Solway coast	Crabs	4	0.42	Bq/kg ww	1994
32	North Solway coast	Crabs	4	0.34	Bq/kg ww	1995
32	North Solway coast	Crabs	4	0.38	Bq/kg ww	1996
32	North Solway coast	Crabs	6	0.27	Bq/kg ww	1997
32	North Solway coast	Crabs	7	0.35	Bq/kg ww	1998
32	North Solway coast	Lobsters	4	0.083	Bq/kg ww	1994
32	North Solway coast	Lobsters	4	0.08	Bq/kg ww	1995
32	North Solway coast	Lobsters	4	0.065	Bq/kg ww	1996
32	North Solway coast	Lobsters	8	0.13	Bq/kg ww	1998
32	North Solway coast	Mussels	4	0.64	Bq/kg ww	1996
32	North Solway coast	Mussels	7	0.91	Bq/kg ww	1997
32	North Solway coast	Mussels	8	0.64	Bq/kg ww	1998
32	North Solway coast	Mussels	8	0.48	Bq/kg ww	1999
32	Southernness	Winkles	4	5	Bq/kg ww	1992
32	Southernness	Winkles	4	1.4	Bq/kg ww	1993
32	Southernness	Winkles	4	1.6	Bq/kg ww	1994
32	Southernness	Winkles	4	1.9	Bq/kg ww	1995
32	Southernness	Winkles	4	1.24	Bq/kg ww	1997
32	Southernness	Winkles	4	1.6	Bq/kg ww	1998
35	Drigg	Winkles	3	14	Bq/kg ww	1988
35	Drigg	Winkles	4	14	Bq/kg ww	1989
35	Drigg	Winkles	4	7.5	Bq/kg ww	1991
35	Nethertown	Winkles	12	16	Bq/kg ww	1988
35	Nethertown	Winkles	12	12	Bq/kg ww	1989
35	Nethertown	Winkles	12	12	Bq/kg ww	1991
35	Nethertown	Mussels	4	6.9	Bq/kg ww	1992
35	Nethertown	Winkles	12	16	Bq/kg ww	1993
35	Nethertown	Winkles	12	7.1	Bq/kg ww	1994
35	Nethertown	Winkles	12	10	Bq/kg ww	1995
35	Nethertown	Winkles	12	6	Bq/kg ww	1996
35	Nethertown	Winkles	12	10	Bq/kg ww	1997
35	Nethertown	Winkles	12	7.3	Bq/kg ww	1998
35	Nethertown	Winkles	12	5.9	Bq/kg ww	1999
35	Ravenglass	Cockles	4	0.53	Bq/kg ww	1992
35	Ravenglass	Cockles	4	1.8	Bq/kg ww	1994
35	Ravenglass	Cockles	5	2	Bq/kg ww	1995
35	Ravenglass	Cockles	4	2.6	Bq/kg ww	1996
35	Ravenglass	Cockles	4	2.3	Bq/kg ww	1998

**Table 14 (cont'd)**

Com	Location	Material	No. Samples	<sup>90</sup> Sr	Unit	Year
35	Ravenglass	Cockles	4	2.5	Bq/kg ww	1999
35	Ravenglass	Crabs	2	0.89	Bq/kg ww	1991
35	Ravenglass	Crabs	4	0.19	Bq/kg ww	1992
35	Ravenglass	Crabs	4	0.62	Bq/kg ww	1993
35	Ravenglass	Crabs	4	0.68	Bq/kg ww	1994
35	Ravenglass	Crabs	4	1	Bq/kg ww	1995
35	Ravenglass	Crabs	4	0.64	Bq/kg ww	1996
35	Ravenglass	Crabs	4	0.89	Bq/kg ww	1997
35	Ravenglass	Crabs	4	0.96	Bq/kg ww	1998
35	Ravenglass	Crabs	4	1.1	Bq/kg ww	1999
35	Ravenglass	Lobsters	2	0.14	Bq/kg ww	1991
35	Ravenglass	Lobsters	4	0.14	Bq/kg ww	1993
35	Ravenglass	Lobsters	4	0.3	Bq/kg ww	1994
35	Ravenglass	Lobsters	4	0.42	Bq/kg ww	1995
35	Ravenglass	Lobsters	4	0.24	Bq/kg ww	1996
35	Ravenglass	Lobsters	4	0.3	Bq/kg ww	1997
35	Ravenglass	Lobsters	4	0.25	Bq/kg ww	1998
35	Ravenglass	Lobsters	4	0.31	Bq/kg ww	1999
35	Sellfield Coastal area	Crabs	1	1.5	Bq/kg ww	1989
35	Sellfield Coastal area	Crabs	8	0.97	Bq/kg ww	1991
35	Sellfield Coastal area	Crabs	9	0.52	Bq/kg ww	1992
35	Sellfield Coastal area	Crabs	8	1.1	Bq/kg ww	1993
35	Sellfield Coastal area	Crabs	9	1.3	Bq/kg ww	1994
35	Sellfield Coastal area	Crabs	8	7.4	Bq/kg ww	1995
35	Sellfield Coastal area	Crabs	8	1.2	Bq/kg ww	1996
35	Sellfield Coastal area	Crabs	8	1.5	Bq/kg ww	1997
35	Sellfield Coastal area	Crabs	8	1.8	Bq/kg ww	1998
35	Sellfield Coastal area	Crabs	8	1.6	Bq/kg ww	1999
35	Sellfield Coastal area	Limpets	4	9.2	Bq/kg ww	1994
35	Sellfield Coastal area	Limpets	4	10	Bq/kg ww	1995
35	Sellfield Coastal area	Limpets	4	3.9	Bq/kg ww	1996
35	Sellfield Coastal area	Limpets	4	8.6	Bq/kg ww	1997
35	Sellfield Coastal area	Limpets	4	9.4	Bq/kg ww	1998
35	Sellfield Coastal area	Limpets	4	6.1	Bq/kg ww	1999
35	Sellfield Coastal area	Lobsters	2	0.45	Bq/kg ww	1988
35	Sellfield Coastal area	Lobsters	10	0.25	Bq/kg ww	1991
35	Sellfield Coastal area	Lobsters	9	0.29	Bq/kg ww	1992
35	Sellfield Coastal area	Lobsters	8	0.42	Bq/kg ww	1993
35	Sellfield Coastal area	Lobsters	8	0.53	Bq/kg ww	1994
35	Sellfield Coastal area	Lobsters	8	0.75	Bq/kg ww	1995
35	Sellfield Coastal area	Lobsters	9	0.47	Bq/kg ww	1996
35	Sellfield Coastal area	Lobsters	8	0.64	Bq/kg ww	1997
35	Sellfield Coastal area	Lobsters	8	0.42	Bq/kg ww	1998
35	Sellfield Coastal area	Lobsters	8	0.7	Bq/kg ww	1999
35	Sellfield Coastal area	Mussels	4	1.6	Bq/kg ww	1994
35	Sellfield Coastal area	Mussels	4	2.6	Bq/kg ww	1995
35	Sellfield Coastal area	Mussels	4	4.7	Bq/kg ww	1996
35	Sellfield Coastal area	Mussels	4	2.1	Bq/kg ww	1997
35	Sellfield Coastal area	Mussels	4	1.5	Bq/kg ww	1998
35	Sellfield Coastal area	Mussels	4	2	Bq/kg ww	1999

**Table 14 (cont'd)**

Com	Location	Material	No. Samples	<sup>90</sup> Sr	Unit	Year
35	Sellfield Coastal area	Whelks	2	0.17	Bq/kg ww	1996
35	Sellfield Coastal area	Whelks	1	0.076	Bq/kg ww	1997
35	Sellfield Coastal area	Whelks	2	0.15	Bq/kg ww	1999
35	Sellfield Coastal area	Winkles	12	7.8	Bq/kg ww	1988
35	Sellfield Coastal area	Winkles	12	6.8	Bq/kg ww	1989
35	Sellfield Coastal area	Winkles	12	7	Bq/kg ww	1991
35	Sellfield Coastal area	Winkles	12	8	Bq/kg ww	1992
35	Sellfield Coastal area	Winkles	12	6.7	Bq/kg ww	1993
35	Sellfield Coastal area	Winkles	4	2	Bq/kg ww	1994
35	Sellfield Coastal area	Winkles	4	3.6	Bq/kg ww	1995
35	Sellfield Coastal area	Winkles	4	4.4	Bq/kg ww	1996
35	Sellfield Coastal area	Winkles	4	1.6	Bq/kg ww	1997
35	Sellfield Coastal area	Winkles	4	5.2	Bq/kg ww	1998
35	Sellfield Coastal area	Winkles	4	4.7	Bq/kg ww	1999
35	St.Bees	Winkles	4	9.3	Bq/kg ww	1988
35	St.Bees	Winkles	4	12	Bq/kg ww	1989
35	St.Bees	Winkles	4	13	Bq/kg ww	1991
35	St.Bees	Winkles	4	8	Bq/kg ww	1992
35	St.Bees	Limpets	4	12	Bq/kg ww	1992
35	St.Bees	Winkles	4	8.3	Bq/kg ww	1993
35	St.Bees	Winkles	4	11	Bq/kg ww	1994
35	St.Bees	Winkles	4	8.5	Bq/kg ww	1995
35	St.Bees	Winkles	4	6.3	Bq/kg ww	1996
35	St.Bees	Winkles	4	5	Bq/kg ww	1997
35	St.Bees	Winkles	4	8.4	Bq/kg ww	1998
35	St.Bees	Winkles	4	8.3	Bq/kg ww	1999
35	White Haven	Nephrops	4	0.094	Bq/kg ww	1992
35	White Haven	Whelks	3	0.15	Bq/kg ww	1992
35	White Haven	Nephrops	4	0.15	Bq/kg ww	1993
35	White Haven	Whelks	3	0.16	Bq/kg ww	1993
35	White Haven	Whelks	3	0.066	Bq/kg ww	1994
35	White Haven	Nephrops	4	0.11	Bq/kg ww	1994
35	White Haven	Nephrops	4	0.14	Bq/kg ww	1995
35	White Haven	Nephrops	4	0.13	Bq/kg ww	1996
35	White Haven	Nephrops	3	0.078	Bq/kg ww	1997
35	White Haven	Nephrops	4	0.13	Bq/kg ww	1999
35	White Haven	Whelks	3	0.32	Bq/kg ww	1999
36	Ravenglass	Cockles	4	2.5	Bq/kg ww	1997
37	Morecambe Bay	Cockles	4	1.3	Bq/kg ww	1988
37	Morecambe Bay	Cockles	4	1	Bq/kg ww	1989
37	Morecambe Bay	Cockles	4	0.81	Bq/kg ww	1991
37	Morecambe Bay	Cockles	4	0.72	Bq/kg ww	1992
37	Morecambe Bay	Cockles	4	0.84	Bq/kg ww	1993
37	Morecambe Bay	Cockles	2	0.71	Bq/kg ww	1995
37	Morecambe Bay	Cockles	4	0.55	Bq/kg ww	1996
37	Morecambe Bay	Cockles	4	0.43	Bq/kg ww	1997
37	Morecambe Bay	Cockles	4	0.44	Bq/kg ww	1998
37	Morecambe Bay	Shrimps	4	0.11	Bq/kg ww	1988
37	Morecambe Bay	Shrimps	4	0.098	Bq/kg ww	1989

**Table 14 (cont'd)**

Com	Location	Material	No. Samples	<sup>90</sup> Sr	Unit	Year
37	Morecambe Bay	Shrimps	4	0.07	Bq/kg ww	1991
37	Morecambe Bay	Shrimps	4	0.051	Bq/kg ww	1992
37	Morecambe Bay	Shrimps	4	0.08	Bq/kg ww	1993
37	Morecambe Bay	Shrimps	4	0.1	Bq/kg ww	1994
37	Morecambe Bay	Shrimps	4	0.12	Bq/kg ww	1995
37	Morecambe Bay	Shrimps	4	0.13	Bq/kg ww	1996
37	Morecambe Bay	Cockles	4	0.49	Bq/kg ww	1999
38	Fleetwood	Whelks	4	0.14	Bq/kg ww	1996
38	Fleetwood	Whelks	4	0.13	Bq/kg ww	1997
38	Fleetwood	Whelks	4	0.036	Bq/kg ww	1998
38	Fleetwood	Whelks	4	0.022	Bq/kg ww	1999

**Table 15** Concentrations of  $^{239,240}\text{Pu}$  in seawater,  $\text{Bq m}^{-3}$ 

		1972	1979	1980	1984	1985	1987	1988	1989	1990	1991	1992	1995	1997
Com-16														
	Max	4.30E-02		2.30E-02		1.37E-02	1.71E-02			2.07E-02			2.25E-02	
	Min	3.00E-03		2.20E-03		9.74E-03	1.13E-02			5.40E-03			4.58E-03	
	Geomean	1.78E-02		1.29E-02		1.21E-02	1.38E-02			1.07E-02			1.02E-02	
	Average	2.12E-02		1.39E-02		1.22E-02	1.40E-02			1.13E-02			1.07E-02	
	Number	23		27		6	5			15			58	
Com-17														
	Max	5.60E-02			3.20E-02	2.31E-02				5.94E-03			8.37E-02	
	Min	2.00E-03			3.40E-08	1.06E-02				5.94E-03			3.41E-03	
	Geomean	2.14E-02			5.18E-03	1.43E-02				5.94E-03			1.20E-02	
	Average	2.62E-02			1.72E-02	1.50E-02				5.94E-03			1.50E-02	
	Number	21			11	8				2			96	
Com-19														
	Max			2.80E-02		1.43E-02	1.53E-02			1.09E-02			1.78E-02	
	Min			2.70E-03		9.10E-03	1.25E-02			1.09E-02			3.92E-03	
	Geomean			1.26E-02		1.17E-02	1.42E-02			1.09E-02			9.84E-03	
	Average			1.42E-02		1.19E-02	1.43E-02			1.09E-02			1.04E-02	
	Number			18		9	3			2			60	
Com-23,24														
	Max		2.33E-02	1.20E-02		1.32E-02				2.16E-02		1.15E-02	2.16E-02	
	Min		5.18E-03	9.00E-03		1.06E-02				6.41E-03		7.50E-03	4.05E-03	
	Geomean		1.28E-02	9.91E-03		1.23E-02				1.21E-02		8.65E-03	9.37E-03	
	Average		1.38E-02	1.00E-02		1.23E-02				1.35E-02		8.83E-03	1.03E-02	
	Number		12	3		3				6		3	60	
Com-27														
	Max		1.07E-02	1.60E-02		1.77E-02				5.62E-02			3.84E-02	
	Min		7.03E-03	6.00E-03		9.38E-03				6.76E-03			5.37E-03	
	Geomean		8.67E-03	1.03E-02		1.49E-02				1.30E-02			1.45E-02	
	Average		8.87E-03	1.07E-02		1.52E-02				1.83E-02			1.60E-02	
	Number		2	7		7				11			66	

**Table 15 (cont'd)**

		1972	1979	1980	1984	1985	1987	1988	1989	1990	1991	1992	1995	1997
Com-28														
	Max							2.18E-01	1.01E-01	8.77E-02			4.90E-02	5.18E-02
	Min							2.18E-01	1.01E-01	3.05E-02			4.90E-02	2.25E-02
	Geomean							2.18E-01	1.01E-01	5.81E-02			4.90E-02	3.27E-02
	Average							2.18E-01	1.01E-01	6.28E-02			4.90E-02	3.46E-02
	Number							1	1	10			2	8
Com-29														
	Max							1.14E-01	6.19E-02	6.06E-02			6.57E-02	5.49E-02
	Min							6.43E-02	4.17E-02	2.22E-02			3.82E-02	2.19E-02
	Geomean							8.54E-02	5.22E-02	4.18E-02			5.38E-02	3.67E-02
	Average							8.73E-02	5.27E-02	4.33E-02			5.47E-02	3.81E-02
	Number							5	4	18			12	10
Com-30														
	Value												7.00E-02	
Com-31														
	Value												4.79E-02	
Cokm-33														
								1988	1989	1990	1993	1994	1995	
	Min							4.45E-01	4.22E-01	2.31E-01	3.80E-01	2.55E-01	4.49E-01	
	Geomean							8.70E-02	2.48E-01	1.80E-01	1.34E-01	9.50E-02	2.80E-02	
	Average							2.55E-01	3.38E-01	1.98E-01	2.17E-01	1.77E-01	1.73E-01	
	Number							2.85E-01	3.44E-01	1.99E-01	2.38E-01	1.93E-01	2.44E-01	
								8	9	6	6	6	8	
Com-34														
	Value								4.27E-01				4.51E-02	
Com-35														
	Value							3.17E+00	8.60E+00				5.88E-01	
Com-36														
	Value							1.46E-01						

Table 15 (cont'd)

		1972	1979	1980	1984	1985	1987	1988	1989	1990	1991	1992	1995	1997
Com-48														
	Max								5.00E-02	4.79E-02	1.56E-02		2.40E-02	4.05E-03
	Min								1.39E-02	1.21E-02	7.10E-03		1.21E-02	3.05E-03
	Geomean								2.64E-02	3.03E-02	1.05E-02		1.70E-02	3.51E-03
	Average								3.20E-02	3.60E-02	1.14E-02		1.81E-02	3.55E-03
	Number								2	6	4		2	2
Com-50														
	Max							8.44E-02	4.76E-02	5.58E-02	2.58E-02		3.33E-02	1.04E-02
	Min							5.14E-02	2.15E-02	3.44E-02	1.25E-02		1.14E-02	8.43E-03
	Geomean							6.59E-02	3.36E-02	4.36E-02	1.80E-02		1.89E-02	9.36E-03
	Average							6.79E-02	3.49E-02	4.43E-02	1.92E-02		2.04E-02	9.42E-03
	Number							2	4	8	4		4	2
Com-52														
	Max							5.51E-02	3.10E-02	4.17E-02	3.24E-02		2.56E-02	1.12E-02
	Min							5.51E-02	2.43E-02	1.65E-02	1.11E-02		1.35E-02	8.31E-03
	Geomean							5.51E-02	2.72E-02	2.39E-02	1.72E-02		1.75E-02	9.65E-03
	Average							5.51E-02	2.73E-02	2.56E-02	1.85E-02		1.81E-02	9.76E-03
	Number							1	3	14	10		6	2
Com-55														
	Max							3.80E-02	2.93E-02	3.67E-02	6.20E-02		5.84E-02	4.06E-02
	Min							1.44E-02	1.44E-02	2.02E-02	1.13E-02		7.27E-03	8.50E-03
	Geomean							2.44E-02	2.09E-02	2.61E-02	2.43E-02		2.16E-02	1.59E-02
	Average							2.63E-02	2.17E-02	2.66E-02	2.74E-02		2.70E-02	1.82E-02
	Number							3	6	16	20		6	6
Com-56														
	Max						4.80E-02	3.37E-02	2.59E-02	7.63E-02	6.20E-02		5.42E-02	1.77E-02
	Min						4.80E-02	1.44E-02	6.70E-03	6.59E-03	9.34E-03		1.27E-02	5.22E-03
	Geomean						4.80E-02	2.04E-02	1.26E-02	2.81E-02	2.29E-02		1.98E-02	1.05E-02
	Average						4.80E-02	2.19E-02	1.39E-02	3.24E-02	2.68E-02		2.13E-02	1.14E-02
	Number						1	3	11	40	46		13	14



**Table 15 (cont'd)**

		1972	1979	1980	1984	1985	1987	1988	1989	1990	1991	1992	1995	1997
Com-57														
	Max						1.92E-01	6.80E-02	3.86E-02	9.20E-02	4.79E-02		5.16E-02	3.38E-02
	Min						1.92E-01	7.30E-03	5.00E-03	9.32E-03	3.42E-03		6.96E-03	3.35E-03
	Geomean						1.92E-01	3.72E-02	1.63E-02	2.06E-02	1.41E-02		2.44E-02	1.42E-02
	Average						1.92E-01	4.99E-02	2.17E-02	2.61E-02	1.75E-02		2.85E-02	1.69E-02
	Number						1	4	9	38	44		28	18
Com-58														
	Max						2.58E-02	1.50E-02	7.70E-03	1.81E-02	1.67E-02	3.43E-03	2.99E-02	1.77E-02
	Min						1.64E-03	8.10E-03	3.20E-03	5.39E-03	5.81E-03	1.95E-03	1.78E-03	3.38E-03
	Geomean						1.82E-02	1.15E-02	5.57E-03	8.59E-03	1.03E-02	2.35E-03	6.83E-03	6.13E-03
	Average						1.85E-02	1.19E-02	5.83E-03	9.14E-03	1.10E-02	2.44E-03	9.04E-03	7.21E-03
	Number						4	3	9	28	38	3	20	9
Com-59														
	Max					2.02E-02		2.27E-02	6.60E-03	1.47E-02	1.18E-02		5.25E-02	2.06E-02
	Min					1.80E-02		2.27E-02	6.60E-03	7.02E-03	1.18E-02		3.85E-03	9.02E-03
	Geomean					1.94E-02		2.27E-02	6.60E-03	1.04E-02	1.18E-02		1.19E-02	1.40E-02
	Average					1.95E-02		2.27E-02	6.60E-03	1.08E-02	1.18E-02		1.56E-02	1.44E-02
	Number					3		1	1	8	2		30	9
Com-60														
	Max							1.55E-02	5.30E-03	1.55E-02	2.70E-02		9.79E-03	
	Min							1.05E-02	2.80E-03	7.10E-03	5.58E-03		2.29E-03	
	Geomean							1.28E-02	3.62E-03	8.72E-03	1.19E-02		5.08E-03	
	Average							1.30E-02	3.77E-03	9.10E-03	1.37E-02		5.63E-03	
	Number							2	3	12	14		10	

**Table 16** Concentrations of <sup>239,240</sup>Pu in fish (F) and shellfish (SF), Bq kg<sup>-1</sup> ww

		1988	1989	1991	1992	1993	1994	1995	1996	1997	1998	1999
Com-17-SF												
	Value	0.00008	0.00005		0.00012	0.00026	0.0013	0.00029	0.00023	7.5E-05	0.0005	0.00063
Com-28-F												
	Max	0.0026	0.0035	0.0045	0.0033	0.0047	0.0038	0.0055	0.002	0.0022	0.0082	0.0049
	Min	0.00029	0.00032	0.00019	0.00021	0.00032	0.00018	0.00086	0.00024	0.00011	0.00038	0.00033
	Geomean	0.00088	0.00102	0.00096	0.00065	0.00098	0.00069	0.00178	0.00052	0.00038	0.00159	0.00134
	Average	0.00117	0.0014	0.00168	0.0013	0.00188	0.00149	0.00252	0.00085	0.00084	0.00329	0.00224
	Number	4	5	5	3	3	3	3	3	3	3	3
Com-28-SF												
	Max	0.26	0.32	0.28	0.21	0.21	0.18	0.25	0.15	0.2	0.16	0.16
	Min	0.00075	0.00017	0.00017	0.00014	0.00019	0.00011	9.1E-05	0.00016	0.00044	0.001	0.00058
	Geomean	0.01141	0.00933	0.01343	0.00948	0.00899	0.00695	0.00813	0.00869	0.01103	0.01391	0.0093
	Average	0.07319	0.08634	0.08379	0.06446	0.06335	0.05308	0.07307	0.04779	0.06116	0.0515	0.04742
	Number	4	4	4	4	4	4	4	4	4	4	4
Com-31-F												
		0.007	0.0035	0.0064	0.0015	0.0024	0.0018	0.0032	0.0021	0.0021	0.0028	0.016
Com-31-SF												
	Max	0.17	0.099	0.15	0.14	0.14	0.16	0.1	0.11	0.12	0.18	0.11
	Min	0.12	0.064	0.089	0.097	0.13	0.086	0.085	0.028	0.07	0.024	0.081
	Geomean	0.14283	0.0796	0.11554	0.11653	0.13491	0.1173	0.0922	0.0555	0.08378	0.06573	0.09439
	Average	0.145	0.0815	0.1195	0.1185	0.135	0.123	0.0925	0.069	0.08667	0.102	0.0955
	Number	2	2	2	2	2	2	2	2	3	2	2
Com-32-F												
	Max	0.001	0.0014	0.0023		0.019	0.0064	0.02	0.01	0.022	0.014	0.028
	Min	0.001	0.0014	0.0023		0.0055	0.0027	0.0021	0.0026	0.00039	0.00019	0.003
	Geomean	0.001	0.0014	0.0023		0.01022	0.00416	0.00648	0.0051	0.00293	0.00139	0.00917
	Average	0.001	0.0014	0.0023		0.01225	0.00455	0.01105	0.0063	0.0112	0.00506	0.0155
	Number	1	1	1		2	2	2	2	2	3	2
Com-32-SF												
	Max	7.7	7.2	4.7	9.2	5.1	4.1	4.3	3.7	3.8	4.1	2.7
	Min	3.6	5.4	2	4.5	0.026	0.02	0.019	0.016	0.016	0.017	0.013
	Geomean	5.30952	6.18992	3.45828	6.43428	1.07842	0.62064	0.65153	0.5908	0.54453	0.49034	0.31839
	Average	5.525	6.23333	3.7	6.85	2.8815	1.83143	2.08414	1.78514	1.54657	1.33489	0.98771
	Number	4	3	3	2	4	7	7	7	7	9	7
Com-33-F												
	Value	0.00019	0.00018		0.0018	0.00091	0.0012	0.00047	0.00026	0.0025	0.0011	0.00021
Com-33-SF												
	Value	0.47	0.36								0.0082	
Com-34-F												
	Max	0.0023	0.0022	0.0028	0.14	0.0019	0.005	0.008	0.00098	0.0018	0.001	0.00087
	Min	0.00028	0.00065	0.0012	0.0012	0.00096	0.00093	0.0014	0.00026	0.00065	0.0006	0.00051
	Geomean	0.00078	0.00126	0.00183	0.01296	0.00143	0.00172	0.00304	0.00054	0.00108	0.00077	0.00067
	Average	0.0011	0.00142	0.002	0.0706	0.00149	0.00234	0.00397	0.00062	0.00123	0.0008	0.00069
	Number	3	3	2	2	3	3	3	3	2	2	2

**Table 16 (cont'd)**

		1988	1989	1991	1992	1993	1994	1995	1996	1997	1998	1999
Com-34-SF												
	Max	0.26	0.29	0.24	0.2	0.2	0.18	0.14	0.18	0.15	0.12	0.12
	Min	0.15	0.21	0.19	0.003	0.099	0.13	0.12	0.099	0.072	0.11	0.1
	Geomean	0.19748	0.24678	0.21354	0.02449	0.14071	0.15297	0.12961	0.13349	0.10392	0.11489	0.10954
	Average	0.205	0.25	0.215	0.1015	0.1495	0.155	0.13	0.1395	0.111	0.115	0.11
	Number	2	2	2	2	2	2	2	2	2	2	2
		1988	1989	1991	1992	1993	1994	1995	1996	1997	1998	1999
Com-35-F												
	Max	0.1	0.087	0.034	0.51	0.014	0.02	0.072	0.33	0.02	0.017	0.08
	Min	0.00042	0.0055	0.0022	0.0028	0.0017	0.0012	0.0018	0.0018	0.0015	0.0024	0.0028
	Geomean	0.01232	0.01507	0.01011	0.02066	0.00518	0.00645	0.0065	0.00969	0.00536	0.00621	0.01131
	Average	0.0271	0.02346	0.01423	0.10473	0.00644	0.00954	0.01343	0.04287	0.00707	0.00733	0.02171
	Number	10	9	10	9	9	10	9	10	9	9	9
Com-35-SF												
	Max	33	50	25	22	23	32	25	19	17	37	17
	Min	0.3	0.24	0.096	0.0028	0.24	0.24	0.16	0.15	0.083	0.12	0.12
	Geomean	7.09426	6.77192	4.41898	2.58274	5.19605	5.61101	4.16652	4.57198	3.41244	3.91153	3.1542
	Average	14.4774	14.9196	10.5786	8.78807	10.4212	10.6743	8.83762	8.60571	6.91729	8.9348	6.4792
	Number	23	23	25	26	25	21	21	21	21	25	25
Com-37-F												
	Max	0.092	0.086	0.045	0.57	0.025	0.21	0.13	0.35	0.01	0.26	0.35
	Min	0.00078	0.0016	0.002	0.0017	0.0018	0.002	0.0011	0.0001	0.0012	0.0015	0.00032
	Geomean	0.00505	0.00753	0.00583	0.01931	0.00365	0.00891	0.00842	0.00368	0.00297	0.00708	0.00447
	Average	0.03153	0.03023	0.0164	0.12956	0.0077	0.0478	0.03312	0.06285	0.00418	0.0667	0.07198
	Number	3	3	3	5	4	5	5	6	4	4	5
Com-37-SF												
	Max	3.7	3.4	3	3	3.3	2.4	3.4	2.5	2.1	2.4	2.5
	Min	0.044	0.035	0.039	0.00081	0.025	0.028	0.025	0.023	0.029	0.017	0.012
	Geomean	0.79311	0.94463	0.72171	0.03661	0.66891	0.55957	0.75438	0.7537	0.74413	0.47218	0.3903
	Average	1.45067	1.67786	1.36483	0.96792	1.44083	1.03967	1.27813	1.29757	1.22843	1.15225	1.14557
	Number	6	7	6	4	6	6	8	7	7	8	7
Com-46-SF												
	Max										0.0068	0.0034
	Min										0.00048	0.0015
	Geomean										0.00214	0.00254
	Average										0.00343	0.0027
	Number										3	3
Com-55-F												
	Max	0.0069	0.0033	0.0014	0.006	0.0052	0.0041	0.0087	0.0034	0.00098	0.0025	0.001
	Min	0.00026	0.00033	0.00025	0.00019	0.00023	0.00034	0.00015	0.00022	0.00047	0.00033	0.00055
	Geomean	0.00134	0.00073	0.00059	0.00079	0.00088	0.00084	0.00068	0.0012	0.00065	0.00072	0.00074
	Average	0.00358	0.00133	0.00083	0.00221	0.002	0.00162	0.00303	0.00197	0.00068	0.0011	0.00076
	Number	2	3	2	3	3	3	3	3	3	3	3
Com-55-SF												
	Max	0.03	0.016	0.32	0.019	0.025	0.015	0.019	0.013	0.031	0.011	0.022
	Min	0.0068	0.011	0.0089	0.0098	0.0058	0.0067	0.0059	0.0085	0.008	0.0074	0.011
	Geomean	0.01268	0.01283	0.03496	0.01307	0.01168	0.01147	0.01014	0.0103	0.01331	0.00881	0.01427
	Average	0.0156	0.013	0.11463	0.0136	0.01393	0.01223	0.0114	0.01047	0.01617	0.00893	0.015
	Number	3	3	3	3	3	3	3	3	3	3	3

**Table 16 (cont'd)**

		1988	1989	1991	1992	1993	1994	1995	1996	1997	1998	1999
Com-57-SF												
	Value	0.0039	0.0048	0.0027	0.0018	0.0038	0.0025	0.0023	0.0031	0.0035	0.0021	0.0028
Com-59-F												
	Max	0.0069	0.0033	0.0014	0.006	0.0052	0.0041	0.0087	0.0034	0.00098	0.0025	0.001
	Min	0.00026	0.00033	0.00025	0.00019	0.00023	0.00034	0.00015	0.00022	0.00047	0.00033	0.00055
	Geomean	0.00134	0.00073	0.00059	0.00079	0.00088	0.00084	0.00068	0.0012	0.00065	0.00072	0.00074
	Average	0.00358	0.00133	0.00083	0.00221	0.002	0.00162	0.00303	0.00197	0.00068	0.0011	0.00076
	Number	2	3	2	3	3	3	3	3	3	3	3
Com-59-SF												
	Value	0.0015	0.0036	0.0045	0.0043	0.0036	0.0014	0.0014	0.0012	0.003	0.0027	0.0045

**Table 17** Concentrations of <sup>239,240</sup>Pu in shellfish from the Irish sea near Sellafield by BNFL, Bq kg<sup>-1</sup> ww

		1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001
Pu-molluscs-BNFL-35												
	Max					22.439	32.926	30.818	91.100	20.268	25.626	7.594
	Min					8.420	0.837	0.932	0.787	0.737	0.409	0.319
	Geomean					12.247	8.391	7.919	6.613	6.841	6.413	4.312
	Average					12.892	10.706	10.449	10.136	8.486	8.316	5.469
	Number					10	43	44	47	39	47	9
Pu-Crustacean-BNFL-35												
	Max					0.432	1.070	2.690	3.483	3.200	2.781	0.797
	Min					0.247	0.112	0.089	0.066	0.086	0.121	0.244
	Geomean					0.325	0.278	0.268	0.281	0.373	0.373	0.382
	Average					0.332	0.346	0.448	0.429	0.524	0.506	0.442
	Number					4	19	26	26	23	20	3
238Pu												
Pu-molluscs-BNFL-34												
	Max					4.6612	7.7737	6.5824	4.5843	4.4576	5.2052	1.5664
	Min					1.58	0.1404	0.168	0.1779	0.1573	0.1666	0.0625
	Geomean					2.514	1.6947	1.7144	1.3132	1.4213	1.4759	0.8433
	Average					2.6524	2.2097	2.2133	1.7506	1.7933	1.7896	1.0709
	Number					11	43	44	47	40	45	9
Pu-crustaceans-BNFL-34												
	Max					0.0968	0.2397	0.616	0.162	0.6004	0.5361	0.1643
	Min					0.0594	0.0198	0.013	0.0164	0.022	0.0216	0.0464
	Geomean					0.0782	0.0594	0.0592	0.0514	0.0831	0.097	0.0814
	Average					0.0794	0.0736	0.0974	0.0619	0.1179	0.1393	0.0938
	Number					4	20	27	27	24	19	3

**Table 18** Concentrations of  $^{239,240}\text{Pu}$  in sediment,  $\text{Bq kg}^{-1}$  dw

		1988	1989	1991	1992	1993	1994	1995	1996	1997	1998	1999
Com-28												
	Max	15	15	17	16	14	13	15	19	18	17	19
	Min	1.6	1.1	0.45	1.7	2.3	0.28	3.9	3.1	6.7	2	0.56
	Geomean	5.27	4.92	5.80	9.52	7.57	4.84	8.16	9.56	12.86	9.13	6.97
	Average	7.57	7.77	10.96	11.62	8.98	8.18	9.30	11.18	13.54	11.34	11.31
	Number	3	3	6	6	5	5	5	5	5	5	5
Com-32												
	Max	590	500	380	420	320	320	260	340	290	280	310
	Min	240	170	120	130	100	130	83	160	87	30	34
	Geomean	355	273	202	229	166	191	138	200	138	136	121
	Average	383	298	218	248	180	200	149	208	151	170	151
	Number	4	4	5	5	5	5	5	5	5	5	5
Com-33												
	Value			39	34	39	30	23	22	23	22	
Com-35												
	Max	1300	1100	970	940	890	630	510	690	630	710	520
	Min	810	740	610	590	630	480	460	510	480	440	520
	Geomean	1007	870	756	732	737	564	485	609	530	559	520
	Average	1027	883	770	743	745	568	485	613	533	575	520
	Number	3	3	3	4	4	4	4	4	4	2	1
Com-24 (Barent Sea)												
	Max					3.2						
	Min					0.5						
	Geomean					1.1909						
	Average					1.3433						
	Number					21						

**Table 19** Concentrations of <sup>238</sup>Pu in fish (F) and shellfish (SF), Bq kg<sup>-1</sup> ww

		1988	1989	1991	1992	1993	1994	1995	1996	1997	1998	1999
Com-17-SF												
	Value	0.00002	0.00001		2.1E-05	4.9E-05	0.00028	5.6E-05	5.8E-05	1.4E-05	9.7E-05	0.00014
Com-28-F												
	Max	0.00049	0.00069	0.00089	0.00067	0.00088	0.00043	0.00099	0.00034	0.00041	0.0016	0.00081
	Min	0.00007	0.00006	0.00004	4.9E-05	0.00006	3.6E-05	0.00016	5.1E-05	1.4E-05	7.3E-05	3.9E-05
	Geomean	0.00019	0.0002	0.00021	0.00014	0.00019	0.00011	0.00033	0.0001	6.4E-05	0.00031	0.0002
	Average	0.00024	0.00028	0.00034	0.00027	0.00035	0.00018	0.00046	0.00015	0.00016	0.00064	0.00037
	Number	4	5	5	3	3	3	3	3	3	3	3
Com-28-SF												
	Max	0.051	0.067	0.055	0.04	0.039	0.037	0.048	0.027	0.035	0.029	0.03
	Min	0.00017	0.00003	0.00002	4.6E-05	2.5E-05	1.9E-05	1.7E-05	2.5E-05	7.2E-05	0.00021	9.3E-05
	Geomean	0.00211	0.00179	0.00224	0.00201	0.00148	0.00125	0.00149	0.00151	0.00189	0.00256	0.00168
	Average	0.0143	0.01796	0.01626	0.01216	0.01175	0.01066	0.01391	0.00858	0.01074	0.00937	0.00892
	Number	4	4	4	4	4	4	4	4	4	4	4
Com-31-F												
		0.0016	0.00071	0.0012	0.00032	0.00047	0.00037	0.0006	0.00041	0.00042	0.0013	0.0034
Com-31-SF												
	Max	0.035	0.02	0.031	0.024	0.027	0.03	0.02	0.02	0.015	0.034	0.022
	Min	0.023	0.014	0.017	0.019	0.026	0.016	0.016	0.0055	0.014	0.0048	0.015
	Geomean	0.02837	0.01673	0.02296	0.02135	0.0265	0.02191	0.01789	0.01049	0.01433	0.01277	0.01817
	Average	0.029	0.017	0.024	0.0215	0.0265	0.023	0.018	0.01275	0.01433	0.0194	0.0185
	Number	2	2	2	2	2	2	2	2	3	2	2
Com-32-F												
	Max	0.00028	0.00033	0.00055	0.00062	0.0037	0.0012	0.0038	0.0021	0.00027	0.00058	0.007
	Min	0.00028	0.00033	0.00055	0.00062	0.0011	0.00059	0.00044	0.00045	0.00027	0.00024	0.0025
	Geomean	0.00028	0.00033	0.00055	0.00062	0.00202	0.00084	0.00129	0.00097	0.00027	0.00037	0.00418
	Average	0.00028	0.00033	0.00055	0.00062	0.0024	0.0009	0.00212	0.00128	0.00027	0.00041	0.00475
	Number	1	1	1	1	2	2	2	2	1	2	2
Com-32-SF												
	Max	1.6	1.4	0.96	1.8	1.1	0.82	0.88	0.69	0.65	0.79	0.52
	Min	0.77	1.1	0.41	0.93	0.0048	0.0037	0.0036	0.003	0.0033	0.0042	0.0036
	Geomean	1.10268	1.26034	0.72295	1.29383	0.21633	0.12559	0.12665	0.11343	0.10471	0.09762	0.0471
	Average	1.1425	1.26667	0.77667	1.365	0.5987	0.37224	0.40266	0.34386	0.28633	0.25991	0.16709
	Number	4	3	3	2	4	7	7	7	7	9	7
Com-33-F												
	Value	0.00004	0.00005		0.00038	0.00023	0.00025	0.0001	7.6E-05	4.4E-05	0.00023	3.1E-05
Com-33-SF												
	Value	0.09	0.075								0.0015	
Com-34-F												
	Max	0.00043	0.0005	0.00052	0.00027	0.00038	0.00097	0.00051	0.00017	0.00034	0.00018	0.00014
	Min	0.00006	0.00013	0.00026	0.00018	0.00019	0.00018	0.00015	5.3E-05	0.00011	0.00011	9.3E-05
	Geomean	0.00016	0.00027	0.00037	0.00022	0.00028	0.00033	0.00028	0.00011	0.00019	0.00014	0.00011
	Average	0.00022	0.00031	0.00039	0.00023	0.0003	0.00045	0.00031	0.00012	0.00023	0.00015	0.00012
	Number	3	3	2	2	3	3	3	3	2	2	2

**Table 19 (cont'd)**

		1988	1989	1991	1992	1993	1994	1995	1996	1997	1998	1999
Com-34-SF												
	Max	0.055	0.059	0.049	0.042	0.037	0.035	0.026	0.036	0.027	0.022	0.022
	Min	0.033	0.043	0.037	0.029	0.021	0.024	0.024	0.019	0.013	0.021	0.018
	Geomean	0.0426	0.05037	0.04258	0.0349	0.02787	0.02898	0.02498	0.02615	0.01873	0.02149	0.0199
	Average	0.044	0.051	0.043	0.0355	0.029	0.0295	0.025	0.0275	0.02	0.0215	0.02
	Number	2	2	2	2	2	2	2	2	2	2	2
Com-35-F												
	Max	0.022	0.02	0.0061	0.0077	0.028	1.8	0.015	0.068	0.0043	0.0035	0.017
	Min	0.0001	0.0011	0.00045	0.00059	0.00039	0.00022	0.00035	0.00037	0.00029	0.00045	0.0006
	Geomean	0.00275	0.00329	0.00218	0.0016	0.00187	0.00278	0.00132	0.00201	0.0011	0.00129	0.00221
	Average	0.00609	0.00523	0.00307	0.00219	0.0065	0.182	0.00278	0.00888	0.00149	0.00153	0.00425
	Number	10	9	10	9	9	10	9	10	9	9	9
Com-35-SF												
	Max	7.5	12	5.8	4.9	5.2	4.1	5.3	3.9	3.7	7.6	3.4
	Min	0.065	0.05	0.016	0.012	0.012	0.049	0.03	0.029	0.014	0.023	0.007
	Geomean	1.62309	1.54136	0.89065	0.92325	1.04797	0.78131	0.86809	0.94683	0.69545	0.79947	0.58389
	Average	3.31874	3.44304	2.37896	2.10112	2.32704	1.68229	1.85205	1.78071	1.43171	1.84024	1.29748
	Number	23	23	25	26	25	21	21	21	21	25	25
Com-37-F												
	Max	0.018	0.018	0.0085	0.0066	0.0047	0.041	0.024	0.066	0.07	0.048	0.066
	Min	0.00014	0.00033	0.00042	0.00016	0.00037	0.00026	0.00021	0.00016	0.00024	0.00027	6.6E-05
	Geomean	0.00099	0.00162	0.0012	0.00064	0.00073	0.00158	0.00162	0.00142	0.0014	0.00127	0.00085
	Average	0.00618	0.00635	0.00314	0.00189	0.00147	0.00919	0.00616	0.0142	0.01781	0.01231	0.01357
	Number	3	3	3	4	4	5	5	5	4	4	5
Com-37-SF												
	Max	0.82	0.71	0.62	0.59	0.64	0.48	0.65	0.46	0.39	0.43	0.48
	Min	0.0093	0.0065	0.0078	0.0085	0.0048	0.0056	0.0048	0.0042	0.0056	0.0034	0.0025
	Geomean	0.16643	0.1913	0.14513	0.11036	0.12893	0.10919	0.14635	0.1415	0.13813	0.08751	0.07206
	Average	0.30822	0.34379	0.2763	0.2097	0.27897	0.20427	0.24623	0.24346	0.22509	0.21164	0.21001
	Number	6	7	6	5	6	6	8	7	7	8	7
Com-46-SF												
	Max										0.0019	0.0009
	Min										6.8E-05	0.00018
	Geomean										0.00032	0.00032
	Average										0.00074	0.00043
	Number										3	3
Com-55-F												
	Max	0.00098	0.00021	0.00025	0.00035	0.0006	0.00046	0.00066	0.0006	0.00012	0.0003	0.00014
	Min	0.00006	0.00004	0.00004	4.9E-05	0.00011	5.7E-05	2.6E-05	4.7E-05	9.4E-05	0.00007	0.0001
	Geomean	0.00024	8.8E-05	1E-04	0.00012	0.00032	0.00014	8.7E-05	0.00019	0.0001	0.00012	0.00012
	Average	0.00052	0.00011	0.00015	0.00016	0.0004	0.00021	0.00024	0.0003	0.0001	0.00015	0.00012
	Number	2	3	2	3	3	3	3	3	3	3	3
Com-55-SF												
	Max	0.0058	0.0028	0.061	0.0034	0.0042	0.0049	0.0031	0.0031	0.0034	0.0035	0.0034
	Min	0.0017	0.0022	0.0023	0.0026	0.0019	0.0016	0.002	0.0018	0.002	0.0013	0.002
	Geomean	0.00261	0.00258	0.00715	0.00291	0.00271	0.00266	0.00262	0.00227	0.0025	0.00234	0.00261
	Average	0.0031	0.0026	0.02197	0.00293	0.00287	0.00297	0.00267	0.00233	0.00257	0.00253	0.00267
	Number	3	3	3	3	3	3	3	3	3	3	3



**Table 19 (cont'd)**

		1988	1989	1991	1992	1993	1994	1995	1996	1997	1998	1999
Com-57-SF												
	Value	0.00038	0.00033	0.00024	0.00015	0.00037	0.00015	0.00019	0.00025	0.00023	0.00053	0.00031
Com-59-F												
	Max	0.00098	0.00021	0.00025	0.00035	0.0006	0.00046	0.00066	0.0006	0.00012	0.0003	0.00014
	Min	0.00006	0.00004	0.00004	4.9E-05	0.00011	5.7E-05	2.6E-05	4.7E-05	9.4E-05	0.00007	0.0001
	Geomean	0.00024	8.8E-05	1E-04	0.00012	0.00032	0.00014	8.7E-05	0.00019	0.0001	0.00012	0.00012
	Average	0.00052	0.00011	0.00015	0.00016	0.0004	0.00021	0.00024	0.0003	0.0001	0.00015	0.00012
	Number	2	3	2	3	3	3	3	3	3	3	3
Com-59-SF												
	Value	0.00023	0.0007	0.00078	0.00083	0.00054	0.00018	0.00015	0.00014	0.00045	0.00042	0.001

**Table 20** Concentrations of  $^{238}\text{Pu}$  in the sediment,  $\text{Bq kg}^{-1}$  dw

		1988	1989	1991	1992	1993	1994	1995	1996	1997	1998	1999
Com-28												
	Max	2.5	2.5	3.3	2.7	2.5	2.2	2.8	3.4	3.4	3.1	3.5
	Min	0.27	0.18	0.062	0.21	0.41	0.05	0.66	0.52	1.2	0.36	0.11
	Geomean	0.91	0.86	1.04	1.10	1.41	0.87	1.46	1.74	2.40	1.63	1.25
	Average	1.29	1.36	2.07	1.65	1.68	1.47	1.67	2.04	2.54	2.01	2.00
	Number	3	3	6	6	5	5	5	5	5	5	5
Com-32												
	Max	120	110	77	86	68	63	54	67	57	54	60
	Min	50	36	26	26	20	26	16	33	17	5.9	6.1
	Geomean	75	59	42	46	34	38	28	39	27	28	23
	Average	80	65	45	50	37	39	30	41	29	35	29
	Number	4	4	5	5	5	5	5	5	5	5	5
Com-33												
	Value			7.1	6.4	7.6	5.3	4.2	4	4.1	3.9	
Com-35												
	Max	300	240	230	200	200	140	100	140	130	130	110
	Min	190	160	130	130	130	100	96	110	100	86	110
	Geomean	221	190	168	158	160	119	99	124	109	106	110
	Average	227	193	173	160	163	120	99	125	110	108	110
	Number	3	3	3	4	4	4	4	4	4	2	1
Com-24(Barents Sea)												
	Max					0.14						
	Min					0.021						
	Geomean					0.0565						
	Average					0.063						
	Number					18						

**Table 21** Inventories of transuranium elements in North Sea sediment, Bq/m<sup>2</sup>

	Com.	55	56	57	58	59	60
<sup>238</sup> Pu							
	Max	7.2	8.7	75	50	100	135
	Min	7.2	1.1	0.78	16	3.1	21
	Geomean	7.2	3.0935	18.369	28.284	11.178	40.645
	Average	7.2	4.9	28.241	33	20.055	49.889
	Number	1	2	14	2	11	9
<sup>239,240</sup> Pu							
	Max	55	42	678	429	821	1409
	Min	55	7.3	3.3	300	46	241
	Geomean	55	17.51	149.44	358.75	124.67	484.84
	Average	55	24.65	235.09	364.5	190.69	571.22
	Number	1	2	14	2	13	9
<sup>241</sup> Am							
	Max	26		640	160	370	730
	Min	26		2	40	22	190
	Geomean	26		83.228	80	124.45	382.62
	Average	26		186.5	100	183.58	418.89
	Number	1		12	2	12	9

**Table 22 Concentrations of <sup>241</sup>Am in fish (F) and shellfish (SF), Bq kg<sup>-1</sup> ww**

		1988	1989	1991	1992	1993	1994	1995	1996	1997	1998	1999
		241Am	241Am	241Am	241Am	241Am	241Am	241Am	241Am	241Am	241Am	241Am
Com-17-SF												
	Value	0.00007	0.00005		0.00016	0.00042	0.0022	0.00043	0.00026	9.7E-05	0.00078	0.0011
Com-28-F												
	Max	0.0037	0.0054	0.0069	0.005	0.0081	0.0044	0.0088	0.0035	0.0034	0.0064	0.0076
	Min	0.0004	0.00032	0.00025	0.00027	0.00054	0.0003	0.0012	0.00037	0.00016	0.00058	0.00045
	Geomean	0.00111	0.00128	0.00133	0.00081	0.00164	0.00093	0.00251	0.0009	0.00055	0.00146	0.00187
	Average	0.00154	0.00203	0.00259	0.00189	0.00321	0.00177	0.00383	0.00148	0.00129	0.00261	0.00332
	Number	4	5	5	3	3	3	3	3	3	3	3
Com-28-SF												
	Max	0.14	0.18	0.19	0.11	0.11	0.13	0.19	0.14	0.19	0.32	0.13
	Min	0.0008	0.00018	0.0001	0.00015	0.00039	0.00011	4.4E-05	0.00014	0.00044	0.0014	0.00073
	Geomean	0.01479	0.01113	0.01754	0.01162	0.01207	0.00959	0.00899	0.01268	0.02695	0.02796	0.01613
	Average	0.0596	0.06095	0.08428	0.05354	0.05622	0.0576	0.07286	0.06779	0.09771	0.12013	0.05381
	Number	4	4	4	4	4	4	4	4	5	4	4
Com-31-F												
		0.012	0.0067	0.012	0.0028	0.0043	0.0038	0.006	0.0043	0.004	0.0035	0.084
Com-31-SF												
	Max	0.2	0.11	0.1	0.15	0.16	0.1	0.093	0.15	0.8	0.026	0.13
	Min	0.035	0.024	0.074	0.038	0.051	0.055	0.06	0.01	0.03	0.012	0.066
	Geomean	0.08367	0.05138	0.08602	0.0755	0.09033	0.07416	0.0747	0.03873	0.12879	0.01766	0.09263
	Average	0.1175	0.067	0.087	0.094	0.1055	0.0775	0.0765	0.08	0.30633	0.019	0.098
	Number	2	2	2	2	2	2	2	2	3	2	2
Com-32-F												
	Max	0.0015	0.0022	0.0032	0.0044	0.029	0.011	0.031	0.018	0.05	0.034	0.079
	Min	0.0015	0.0022	0.0032	0.0044	0.008	0.0034	0.0032	0.0037	0.05	0.0013	0.079
	Geomean	0.0015	0.0022	0.0032	0.0044	0.01523	0.00612	0.00996	0.00816	0.05	0.00438	0.079
	Average	0.0015	0.0022	0.0032	0.0044	0.0185	0.0072	0.0171	0.01085	0.05	0.0124	0.079
	Number	1	1	1	1	2	2	2	2	1	3	1
Com-32-SF												
	Max	15	12	9.9	19	13	10	11	7.5	9.5	10	6.8
	Min	5.2	7.5	3	7.2	0.044	0.036	0.032	0.028	0.023	0.0023	0.036
	Geomean	8.70744	9.65489	5.44977	11.6962	2.05799	1.55007	1.67202	1.62738	1.00377	1.00811	0.92694
	Average	9.475	9.83333	6.45	13.1	6.061	3.70229	4.19457	3.75829	3.06043	3.19137	2.22371
	Number	4	3	2	2	4	7	7	7	7	9	7
Com-33-F												
	Value	0.00033	0.00033	0.72	0.003	0.0015	0.0021	0.00081	0.00055	0.00039	0.0018	0.00029
Com-33-SF												
	Value	0.68	0.43								0.079	
Com-34-F												
	Max	0.0028	0.0031	0.0042	0.0019	0.029	0.0072	0.0046	0.0017	0.0026	0.0022	0.0016
	Min	0.00047	0.0012	0.0014	0.0013	0.0015	0.0013	0.0016	0.00059	0.00089	0.0013	0.00072
	Geomean	0.0012	0.00173	0.00242	0.00157	0.00464	0.00246	0.00245	0.00096	0.00152	0.00169	0.00107
	Average	0.00152	0.0019	0.0028	0.0016	0.01093	0.00337	0.00273	0.00106	0.00175	0.00175	0.00116
	Number	3	3	2	2	3	3	3	3	2	2	2

**Table 22 (cont'd)**

	1988	1989	1991	1992	1993	1994	1995	1996	1997	1998	1999
Com-34-SF											
Max	0.46	0.49	0.4	0.34	0.33	0.31	0.23	0.31	0.24	0.19	0.22
Min	0.042	0.065	0.062	0.072	0.035	0.048	0.027	0.031	0.02	0.036	0.038
Geomean	0.139	0.17847	0.15748	0.15646	0.10747	0.12198	0.0788	0.09803	0.06928	0.0827	0.09143
Average	0.251	0.2775	0.231	0.206	0.1825	0.179	0.1285	0.1705	0.13	0.113	0.129
Number	2	2	2	2	2	2	2	2	2	2	2
Com-35-F											
Max	0.16	0.12	0.046	0.056	0.021	0.033	0.11	4.2	0.031	0.035	0.14
Min	0.00076	0.0067	0.0035	0.0047	0.003	0.002	0.0031	0.003	0.0029	0.0049	0.0039
Geomean	0.021	0.02124	0.01718	0.01297	0.00924	0.01127	0.01107	0.02209	0.00998	0.01204	0.01805
Average	0.04344	0.0314	0.02454	0.01761	0.01129	0.01634	0.02177	0.43687	0.01278	0.0147	0.03568
Number	10	9	10	9	9	10	9	10	9	9	9
Com-35-SF											
Max	57	84	42	40	40	34	47	39	31	110	33
Min	1.6	0.86	0.42	0.93	0.74	1.1	0.39	0.51	0.19	0.41	0.43
Geomean	16.8235	14.5831	10.5024	10.5607	10.3224	8.59868	7.93118	8.44859	8.13325	8.8171	7.88158
Average	24.8652	25.8735	19.2075	17.0043	18.0672	14.5125	15.4812	14.7204	13.4589	17.2437	12.9343
Number	23	23	32	30	29	24	25	25	27	30	30
Com-37-F											
Max	0.13	0.13	0.46	0.72	0.039	0.32	0.18	0.47	0.1	0.41	0.59
Min	0.0012	0.0023	0.0042	0.0014	0.0032	0.0022	0.0022	9.9E-05	0.0032	0.0032	0.00076
Geomean	0.00793	0.0118	0.02747	0.01482	0.00645	0.01271	0.01526	0.00582	0.0087	0.01325	0.01378
Average	0.0448	0.04593	0.1339	0.15542	0.01243	0.07258	0.0545	0.08582	0.028	0.10588	0.14343
Number	3	3	4	5	4	5	5	6	4	4	5
Com-37-SF											
Max	8.4	7.6	7.6	7.9	7.9	6.5	8.4	6	5.4	6	6.9
Min	0.063	0.054	0.058	0.066	0.04	0.043	0.039	0.033	0.047	0.028	0.023
Geomean	1.15897	1.52403	1.35215	1.18066	1.07466	0.90258	1.389	1.34621	1.36921	0.9681	0.78105
Average	2.66217	3.29914	2.924	2.6932	3.11833	2.07217	2.74988	2.68471	2.51671	2.68	2.62843
Number	6	7	7	5	6	6	8	7	7	8	7
Com-46-SF											
Max										0.0027	0.0021
Min										0.0011	0.00077
Geomean										0.00148	0.00145
Average										0.00163	0.00159
Number										3	3
Com-55-F											
Max	0.00098	0.00083	0.0016	0.005	0.0015	0.001	0.0013	0.0048	0.0011	0.00092	0.00058
Min	0.00036	0.00026	0.00019	0.00025	0.00041	0.00038	0.00024	0.00055	0.0004	0.00045	0.00051
Geomean	0.00059	0.0004	0.00055	0.00086	0.00079	0.00066	0.00043	0.00125	0.0007	0.00058	0.00055
Average	0.00067	0.00046	0.0009	0.00192	0.00091	0.00071	0.0006	0.00203	0.00076	0.00061	0.00055
Number	2	3	2	3	3	3	3	3	3	3	3
Com-55-SF											
Max	0.026	0.012	0.63	0.013	0.015	0.017	0.016	0.013	0.012	0.015	0.013
Min	0.0075	0.0042	0.0053	0.006	0.0082	0.0057	0.0072	0.0046	0.0064	0.0034	0.0064
Geomean	0.01155	0.00707	0.03109	0.0095	0.01106	0.00915	0.01082	0.00842	0.00945	0.00794	0.00918
Average	0.0138	0.00773	0.21477	0.01	0.0114	0.0102	0.0114	0.0092	0.0098	0.0094	0.00957
Number	3	3	3	3	3	3	3	3	3	3	3

**Table 22 (cont'd)**

	1988	1989	1991	1992	1993	1994	1995	1996	1997	1998	1999
Com-57-SF											
Value	0.0022	0.021	0.0021	0.0013	0.0027	0.0016	0.0011	0.0012	0.0012	0.0011	0.0028
Com-59-F											
Max	0.00098	0.00083	0.0016	0.005	0.0015	0.001	0.0013	0.0048	0.0011	0.00092	0.00058
Min	0.00036	0.00026	0.00019	0.00025	0.00041	0.00038	0.00024	0.00055	0.0004	0.00045	0.00051
Geomean	0.00059	0.0004	0.00055	0.00086	0.00079	0.00066	0.00043	0.00125	0.0007	0.00058	0.00055
Average	0.00067	0.00046	0.0009	0.00192	0.00091	0.00071	0.0006	0.00203	0.00076	0.00061	0.00055
Number	2	3	2	3	3	3	3	3	3	3	3
Com-59-SF											
Value	0.002	0.0046	0.0038	0.0049	0.0044	0.0026	0.0016	0.0018	0.0042	0.0036	0.0023

**Table 23 Concentrations of <sup>241</sup>Am in samples from the Irish Sea near Sellafield (by BNFL)**

		1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001
Am-Sed-BNFL(35)																	
	Max	4900.0	4500.0	2400.0	5500.0	4400.0	2900.0	1900.0	1520.0	2300.0	1270.0	1170.0	943.0	3020.0	967.0	1070.0	1340.0
	Min	170.0	20.0	36.0	37.0	46.0	32.0	58.0	50.5	56.0	49.1	55.1	76.6	26.1	102.0	117.0	126.0
	Geomean	782.3	653.8	751.7	750.2	730.9	615.3	545.9	419.7	411.6	361.6	426.1	398.8	402.9	349.8	253.4	342.2
	Average	1055.5	923.7	981.7	1025.5	976.8	821.4	706.6	573.1	546.6	438.3	530.5	503.3	585.9	433.9	331.7	471.3
	Number	60	63	67	68	67	83	76	72	78	65	63	58	77	49	29	11
Am-W-BNFL(35)																	
	Max		12.00	51.00	65.00	3.20	7.80			8.98	5.39						
	Min		1.50	1.50	2.30	3.20	3.00			2.07	5.39						
	Geomean		4.39	6.27	5.80	3.20	4.84			4.29	5.39						
	Average		5.19	8.27	7.48	3.20	5.40			5.38	5.39						
	Number		8	36	37	1	2			4	1						
Am-Fish-BNFL(35)																	
	Max									0.0720	0.0570						
	Min									0.0130	0.0110						
	Geomean									0.0272	0.0226						
	Average									0.0358	0.0251						
	Number									4	9						
Am-Crustaceans-BNFL(35)																	
	Max		3.10	1.80	5.50	1.40				0.55	8.00	1.49	0.25	0.86	1.47		
	Min		0.77	0.68	0.73	0.71				0.05	0.20	1.49	0.25	0.86	1.47		
	Geomean		1.54	1.11	2.20	1.00				0.22	0.96	1.49	0.25	0.86	1.47		
	Average		1.94	1.24	2.79	1.06				0.33	2.15	1.49	0.25	0.86	1.47		
	Number		2	2	7	2				3	7	1	1	1	1		
Am-Molluscs-BNFL(35)																	
	Max	200.00	170.00	88.00	55.00	68.00	93.00	53.00	50.20	55.40	40.60	57.70	47.40	56.90	48.70	60.60	63.90
	Min	1.10	1.80	2.10	0.61	0.94	0.96	1.28	1.30	0.01	0.02	1.52	2.65	2.45	1.47	1.26	3.95
	Geomean	27.70	24.76	24.12	15.79	16.47	15.40	14.11	12.19	9.99	7.31	13.73	17.40	14.51	14.78	17.34	15.00
	Average	37.64	32.64	30.25	21.45	22.55	22.48	19.38	17.25	16.20	15.50	19.38	21.32	18.96	18.18	22.66	18.92
	Number	92	92	84	55	53	55	50	58	74	55	50	53	50	50	51	10

**Table 23 (cont'd)**

		1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001
Am-Seaweed(FV)-BNFL(35)																	
	Max					32.00	19.00	17.10	18.00	9.10	14.70	15.20	10.90	16.30	19.00	11.00	
	Min					16.00	5.60	2.25	4.50	2.25	2.16	2.13	2.34	1.87	1.78	2.51	
	Geomean					22.63	10.26	9.75	9.41	3.81	4.97	4.68	4.54	4.54	4.22	5.02	
	Average					24.00	11.74	11.78	10.57	4.69	5.99	5.51	5.12	5.32	5.24	5.76	
	Number					2	5	6	5	3	25	40	27	44	36	8	



**Table 24      Concentrations of  $^3\text{H}$  in seawater, Bq m $^{-3}$** 

		1984	1988	1989	1990	1991	1992	1995	1996	1997
Com-57										
	Max	1620	1890	1540	850	1069	930	3130	1800	3960
	Min	570	480	1000	620	640	457	630	1180	1140
	geomean	842	723	1162	765	773	709	1684	1420	2628
	average	886	808	1187	773	794	742	2060	1438	2880
	Number	20	17	3	3	3	3	3	4	5
Com-58										
	Max	4570	3230	3460	1940	2188	2259	3560	1790	4310
	Min	1100	580	1130	1140	363	1371	2660	1280	2340
	geomean	1669	1501	1692	1546	1290	1916	3129	1431	3377
	average	1881	1661	1823	1564	1427	1944	3142	1442	3420
	Number	11	15	8	7	8	6	6	5	8

**Table 25      Concentration of <sup>129</sup>I (Bq/kg ww) in Biota in the east Irish Sea**

Sample	Seaweed				Fish			
Species	Fucus ves.		Porphyra		Cod		Plaice	
Year	2000	2001	2000	2001	2000	2001	2000	2001
Max	32.600	26.200	2.650	1.690	0.975	0.120	0.033	1.600
Min	0.012	0.005	0.693	0.309	0.036	0.050	0.007	0.005
Geomean	5.357	3.434	1.417	0.660	0.155	0.085	0.017	0.013
Average	8.922	6.740	1.498	0.778	0.219	0.088	0.018	0.142
Number	33	15	10	7	14	14	19	12

**Table 26 Concentrations of naturally occurring radionuclides in fish and shellfish from the West Irish Sea (com.35), Bq kg<sup>-1</sup> ww**

		1991	1992	1993	1994	1995	1996	1997	1998	1999
Po-210										
	Max	750	280	67	75	50	37	51	69	53
	Min	23	1.6	13	10	10	4.3	1	1.1	0.77
	Geomean	96.53	41.47	25.69	20.08	15.94	13.57	16.65	17.94	15.72
	Average	181.00	70.50	30.00	23.79	18.00	15.49	21.50	23.59	21.19
	Number	11	13	13	14	14	14	14	15	16
Pb-210										
	Max		19.000	9.100	4.400	4.600	3.100	3.800	5.700	3.700
	Min		1.200	0.330	0.120	0.002	0.017	0.017	0.014	0.018
	Geomean		4.090	2.199	1.227	0.383	0.258	0.449	0.339	0.475
	Average		7.733	4.038	2.041	1.422	0.966	1.437	2.081	1.452
	Number		3	8	9	9	11	8	10	11
Ra226										
	Max	2.900	0.190	0.200	0.590	1.800	1.100	9.600	0.082	0.050
	Min	0.260	0.030	0.200	0.590	1.800	1.100	0.480	0.069	0.050
	Geomean	0.868	0.086	0.200	0.590	1.800	1.100	2.147	0.075	0.050
	Average	1.580	0.110	0.200	0.590	1.800	1.100	5.040	0.076	0.050
	Number	2	3	1	1	1	1	2	2	1
Th-228										
	Max	0.920	0.780	1.500	0.570	1.100	0.640	0.990	1.000	1.100
	Min	0.130	0.017	0.035	0.025	0.022	0.019	0.029	0.025	0.066
	Geomean	0.346	0.115	0.234	0.179	0.146	0.150	0.169	0.172	0.223
	Average	0.525	0.313	0.551	0.311	0.384	0.334	0.364	0.404	0.427
	Number	2	5	4	4	5	4	4	6	4
Th-230										
	Max	28.000	14.000	4.700	1.600	1.800	2.100	2.900	2.000	1.800
	Min	0.068	0.066	0.041	0.013	0.016	0.010	0.019	0.010	0.013
	Geomean	1.380	0.604	0.310	0.165	0.181	0.152	0.178	0.212	0.150
	Average	14.034	3.916	1.426	0.573	0.835	0.779	0.904	0.754	0.627
	Number	2	4	4	4	4	4	4	5	4
U-234										
	Max	15.000	5.600	4.500	1.600	1.400	1.800	2.000	2.000	1.800
	Min	0.100	0.051	0.033	0.019	0.024	0.021	0.029	0.022	0.045
	Geomean	0.766	0.316	0.281	0.144	0.122	0.156	0.166	0.129	0.181
	Average	5.133	1.920	1.561	0.573	0.493	0.640	0.703	0.690	0.639
	Number	3	3	3	3	3	3	3	3	3
U-235										
	Max	0.5700	0.2200	0.2200	0.0640	0.0450	0.0660	0.0700	0.0790	0.0800
	Min	0.0052	0.0015	0.0014	0.0007	0.0008	0.0009	0.0003	0.0011	0.0003
	Geomean	0.0329	0.0103	0.0125	0.0056	0.0042	0.0061	0.0027	0.0052	0.0034
	Average	0.1957	0.0749	0.0759	0.0229	0.0160	0.0236	0.0185	0.0272	0.0213
	Number	3	3	3	3	3	3	4	3	4

**Table 26 (cont'd)**

U-238										
	Max	14.550	5.500	4.400	1.500	1.300	1.700	1.800	1.900	1.700
	Min	0.096	0.049	0.031	0.016	0.023	0.020	0.027	0.020	0.009
	Geomean	0.722	0.300	0.267	0.131	0.115	0.146	0.149	0.119	0.080
	Average	4.972	1.883	1.524	0.536	0.458	0.604	0.632	0.655	0.455
	Number	3	3	3	3	3	3	3	3	4

**Table 27** Concentrations of naturally occurring radionuclides (Bq kg<sup>-1</sup> ww) in biota (from UK)

Material	Location	No. Sample	Year	<sup>210</sup> Po	<sup>210</sup> Pb	<sup>226</sup> Ra	<sup>228</sup> Th	<sup>230</sup> Th	<sup>232</sup> Th	<sup>234</sup> U	<sup>235</sup> U	<sup>238</sup> U
Bass	Ribble Estuary	1	1991			0.0027	0.00096	0.001				
Cockles	Southern North Sea	2	1996			0.31	0.19	0.25				
Cockles	Southern North Sea	2	1997			0.4	0.26	0.34				
Cockles	Southern North Sea	2	1998			0.44	0.23	0.33				
Cod	Parton	3	1991	1.6			0.017	0.0032	0.00044	0.0053	0.0003	0.005
Cod	Parton	3	1992	2.5	0.05							
Cod	Parton	4	1993	0.8	0.025	0.024	0.024	0.013	0.0051	0.0049	0.00026	0.0044
Cod	Parton	5	1994	0.63	0.026		0.014	0.0014	0.0006	0.0036	0.000031	0.0035
Cod	Parton	4	1995	0.064	0.011		0.022	0.0036	0.0015	0.0048	0.00015	0.0046
Cod	Parton	1	1996	0.68	0.017		0.016	0.002	0.0007	0.005	0.00021	0.0043
Cod	Parton	2	1997	0.91	0.017		0.014	0.0076	0.0017	0.0052	0.0003	0.0046
Cod	Parton	2	1998	0.89	0.014		0.025	0.0019	0.0008	0.0041		0.0035
Crabs	Parton	3	1991	44		0.26	0.13	0.068	0.011	0.1	0.0052	0.096
Crabs	Parton	3	1992	56	3	0.11	0.078	0.096	0.012	0.11	0.0033	0.1
Crabs	Parton	4	1993	32	6.4		0.1	0.053	0.019	0.15	0.0064	0.14
Crabs	Parton	5	1994	21	1.4		0.14	0.058	0.023	0.099	0.004	0.093
Crabs	Parton	4	1995	22	0.17		0.095	0.025	0.01	0.054	0.0021	0.051
Crabs	Parton	4	1996	17	0.12		0.068	0.026	0.011	0.1	0.0039	0.091
Crabs	Parton	4	1997	23			0.078	0.027	0.012	0.079	0.0028	0.068
Crabs	Parton	4	1998	26	0.017		0.087	0.031	0.013	0.049	0.0016	0.044
Crabs	Sellafield coast area	4	1999	14	0.21		0.066	0.033	0.011	0.045	0.0019	0.039
Lobster	Parton	4	1992	59	1.2	0.03	0.03	0.066	0.0068	0.051	0.0015	0.049
Lobster	Parton	4	1993	17	0.33		0.035	0.041	0.0063	0.033	0.0014	0.031
Lobster	Parton	5	1994	11	0.12		0.025	0.013	0.0038	0.019	0.0007	0.016
Lobster	Parton	4	1995	13	0.07		0.044	0.016	0.0062	0.024	0.0008	0.023
Lobster	Parton	3	1996	12	0.026		0.019	0.01	0.0039	0.021	0.0009	0.02
Lobster	Parton	4	1997	13			0.029	0.019	0.0076	0.029	0.0009	0.027
Lobster	Parton	4	1998	8.8	0.017		0.032	0.01	0.0055	0.022	0.0011	0.02

**Table 27 (cont'd)**

Material	Location	No. Sample	Year	<sup>210</sup> Po	<sup>210</sup> Pb	<sup>226</sup> Ra	<sup>228</sup> Th	<sup>230</sup> Th	<sup>232</sup> Th	<sup>234</sup> U	<sup>235</sup> U	<sup>238</sup> U
Lobster	Sellafield coast area	2	1999	0.77	0.018		0.025	0.0049	0.003	0.01	0.0003	0.009
Mussels	Portishead	1	1999	19	1.4							
Plaice	Ribble Estuary	1	1992				0.027	0.0021	0.0012			
Salmon	Ribble Estuary	1	1992				0.0007	0.0008	0.0003			
Shrimp	Ribble Estuary	2	1999			0.067		0.012	0.022	0.0052		
Shrimps	Ribble Estuary	1	1992				0.0069	0.016	0.0039			
Shrimps	Ribble Estuary	1	1996			0.15	0.004	0.025	0.005			
Shrimps	Ribble Estuary	2	1997				0.011	0.013	0.0047			
Shrimps	Ribble Estuary	2	1998		0.033	0.0078	0.01	0.0041				
Whelks	Sellafield offshore area	1	1999	26	0.17		0.073	0.013	0.0074	0.073	0.0029	0.07
Winkles	Parton	4	1991	120		2.9	0.92	28	0.72	15	0.57	14.55
Winkles	Parton	6	1992	110	19	0.19	0.66	14	0.015	5.6	0.22	5.5
Winkles	Parton	4	1993	50	6.8		1.5	4.7	1.1	4.5	0.22	4.4
Winkles	Parton	4	1994	32	4.4	0.59	0.57	1.6	0.41	1.6	0.064	1.5
Winkles	Parton	4	1995	21	4.6	1.8	0.66	1.8	0.45	1.4	0.045	1.3
Winkles	Parton	4	1996	17	3.1	1.1	0.64	2.1	0.5	1.8	0.066	1.7
Winkles	Parton	4	1997	28	3.8	9.6	0.99	2.9	0.7	2	0.07	1.8
Winkles	Parton	4	1998	28	4.6	0.082	1	2	0.76	2	0.079	1.9
Winkles	Parton	4	1999	26	3.7	0.05	1.1	1.8	0.72	1.8	0.08	1.7
Shellfish	USA		1987					0.029	0.03	0.018		1.9
Fish	USA		1987					0.0012	0.0012	0.018		
Winkles	Parton	8	1989-1990		4.8		1	9.7	0.68	5.6	0.19	5.5
Winkles	Parton	2	1989-1990				0.66	1.4	1.2	1.4	0.027	1.2
Lobster	Parton	2	1989-1990		0.0055	0.0045	0.035			0.037	0.0025	0.032
Crabs	Parton	1	1989-1990		0.013	0.1	0.075			0.2	0.007	0.18
Cockles	Wash	1	1989-1990							0.62	0.018	0.53
Cockles	Dee East	1	1989-1990				0.57					
Cockles	Ribble East	1	1989-1990		0.37	0.76	0.58					
Shrimp	Ribble East	1	1989-1990				0.0084					

\* Except USA, Other locations are in UK

**Table 27 (cont'd)**

Material	Location	No. Sample	Year	<sup>210</sup> Po	<sup>210</sup> Pb	<sup>226</sup> Ra	<sup>228</sup> Th	<sup>230</sup> Th	<sup>232</sup> Th	<sup>234</sup> U	<sup>235</sup> U	<sup>238</sup> U
Bass	Ribble East	1	1989-1990				0.004					
Cod	Parton	1	1989-1990		0.0024	0.007	0.012			0.0095	0.0005	0.008
Winkles	Torness	1	1989-1990							0.95	0.02	0.84
Winkles	Nethertown	1	1989-1990							2	0.071	1.9
Winkles	Saltom Bay	1	1989-1990				2.4		2			50
Salmon	Ribble Estuary	1	1992				0.0007	0.0008	0.0003			
Shrimp	Ribble Estuary	2	1999			0.067		0.012	0.022	0.0052		
Shrimps	Ribble Estuary	1	1992				0.0069	0.016	0.0039			
Shrimps	Ribble Estuary	1	1996			0.15	0.004	0.025	0.005			
Shrimps	Ribble Estuary	2	1997				0.011	0.013	0.0047			
Shrimps	Ribble Estuary	2	1998		0.033	0.0078	0.01	0.0041				
Whelks	Sellafield offshore area	1	1999	26	0.17		0.073	0.013	0.0074	0.073	0.0029	0.07
Winkles	Parton	4	1991	120		2.9	0.92	28	0.72	15	0.57	14.55
Winkles	Parton	6	1992	110	19	0.19	0.66	14	0.015	5.6	0.22	5.5
Winkles	Parton	4	1993	50	6.8		1.5	4.7	1.1	4.5	0.22	4.4
Winkles	Parton	4	1994	32	4.4	0.59	0.57	1.6	0.41	1.6	0.064	1.5
Winkles	Parton	4	1995	21	4.6	1.8	0.66	1.8	0.45	1.4	0.045	1.3
Winkles	Parton	4	1996	17	3.1	1.1	0.64	2.1	0.5	1.8	0.066	1.7
Winkles	Parton	4	1997	28	3.8	9.6	0.99	2.9	0.7	2	0.07	1.8
Winkles	Parton	4	1998	28	4.6	0.082	1	2	0.76	2	0.079	1.9
Winkles	Parton	4	1999	26	3.7	0.05	1.1	1.8	0.72	1.8	0.08	1.7
Shellfish	USA		1987					0.029	0.03	0.018		1.9
Fish	USA		1987					0.0012	0.0012	0.018		
Winkles	Parton	8	1989-1990		4.8		1	9.7	0.68	5.6	0.19	5.5
Winkles	Parton	2	1989-1990				0.66	1.4	1.2	1.4	0.027	1.2
Lobster	Parton	2	1989-1990		0.0055	0.0045	0.035			0.037	0.0025	0.032
Crabs	Parton	1	1989-1990		0.013	0.1	0.075			0.2	0.007	0.18
Cockles	Wash	1	1989-1990							0.62	0.018	0.53

\* Except USA, Other locations are in UK

**Table 27 (cont'd)**

Material	Location	Year	<sup>210</sup> Po	<sup>210</sup> Pb	<sup>226</sup> Ra	<sup>228</sup> Th	<sup>230</sup> Th	<sup>232</sup> Th	<sup>234</sup> U	<sup>235</sup> U	<sup>238</sup> U	<sup>40</sup> K
Cockles	Dee East	1989-1990	1				0.57					
Cockles	Ribble East	1989-1990	1		0.37	0.76	0.58					
Shrimp	Ribble East	1989-1990	1				0.0084					
Bass	Ribble East	1989-1990	1				0.004					
Cod	Parton	1989-1990	1		0.0024	0.007	0.012			0.0095	0.0005	0.008
Winkles	Torness	1989-1990	1							0.95	0.02	0.84
Winkles	Nethertown	1989-1990	1							2	0.071	1.9
Winkles	Saltom Bay	1989-1990	1				2.4		2			50
Mussels	Aberdeen	1989	625	75.5				0.69			29	606
Mussels	Poolewe	1989	168	13.6				0.07			2.92	281
Mussels	Sandyhills	1989	143	5				0.23			1.11	416
Mussels	Stonehaven	1989	158	17.4				0.19			3.5	268
Mussels	Tongue	1989	197	8				0.161			2.08	211
Mussels	Colwyn Bay	1989	105	2.8				0.16			1.01	315
Mussels	Hartlepool	1989	104	7.5				0.35			1.62	372
Mussels	Lowestoft	1989	186	4.7				0.95			3.45	511
Mussels	Ravenglass	1989	160	6.7				0.3			2.72	396
Mussels	Whitehaven	1989	3124	284				0.72			37.1	445
Winkle	Anstruther	1989	13.1	1.11				0.007			1.36	136
Winkle	Aberdeen	1989	67	4				2.5			2.59	526
Winkle	Sandyhills	1989	28.1	4.32				0.59			2.72	525
Winkle	Stonehaven	1989	53.4	3.31				0.22			2.46	493
Winkle	Tongue	1989	79.6	16.1				0.54			3.49	440
Winkle	Colwyn Bay	1989	58.7	2.7				0.23			2.07	324
Winkle	Ravenglass	1989	60	5.3				0.81			2.18	481
Winkle	Whitehaven	1989	399	68				0.61			18.9	456
Winkle	Plymouth	1989	51	2.1				0.11			2.69	379
Sediment	Tongue	1991	20.1	13.9				15.6			13.4	597
Sediment	Poolewe	1991	10.9	7.81				9.3			13.1	495
Sediment	Aberdeen	1991	6.44	5.65				10.6			8.9	760



**Table 27 (cont'd)**

Material	Location	Year	<sup>210</sup> Po	<sup>210</sup> Pb	<sup>226</sup> Ra	<sup>228</sup> Th	<sup>230</sup> Th	<sup>232</sup> Th	<sup>234</sup> U	<sup>235</sup> U	<sup>238</sup> U	<sup>40</sup> K
Sediment	Stonehaven	1991	12.5	13.1				14.3			14	668
Sediment	Anstruther	1991	9.31	8.58				9.9			12.1	193
Sediment	Sandyhills	1991	23.1	10.4				15.5			14	476
Sediment	Whitehaven	1991	56.6	12.8				5.3			22.1	103
Sediment	Ravenglass	1991	30.2	19.5				15.5			16.2	413
Sediment	Blackhall	1991	16.7	33.7				24.4			32.3	489
Sediment	Hartlepool	1991	5.63	5.34				3.4			5.2	175
Sediment	Colwyn Bay	1991	7.23	5.36				6.9			6.6	185
Sediment	Gt Yarmouth	1991	2.03	186				2.6			3.6	77
Sediment	Eastbourne	1991	1.37	3.56				3			4.6	25
Sediment	Plymouth	1991	8.71	5.96				10.9			9.8	201

**Table 28** Concentrations of naturally occurring radionuclides in seawater (Bq m<sup>-3</sup>), sediment and seaweed (Bq kg<sup>-1</sup> dw)

Material	Location	No. Sample	Year	<sup>210</sup> Po	<sup>210</sup> Pb	<sup>226</sup> Ra	<sup>228</sup> Th	<sup>230</sup> Th	<sup>232</sup> Th	<sup>234</sup> U	<sup>235</sup> U	<sup>238</sup> U	<sup>40</sup> K
Sediment	North Sea		1993				5~19	4~16	4~16				
Sediment	UK (coast)	9	1991	8.73	7.3				9.23			8.47	
Sediment	Irish Sea estuaries		1991				8~38	9~78	7~40			12~31	
Seawater	North Sea		1989-1990	1	1	1.5	1	0.05	0.1	4.7	1.9	41	
Seawater	Worldwide		1989-1990	2	3	1	0.2			46		40	
Seawater	UK	9	1991	2.6	3.4	0						39	
Seawater	Antarctic	8	1987	1.9	1	0							
Seawater	Oosterschelde (NL)		1987-1990	0.8	0.9	4.3							
Seawater	North Sea		1987-1990	0.7	0.8	5.3							
Seawater	North Sea coast (NL)		1987-1990	0.5	0	5							
Seawater	Waddensea		1987-1990	1.3	0.7	5.7							
Seawater	Nieuwe waterweg (NL)		1987-1990	3.3	10	19							
Seawater	Westerschelde (NL)		1987-1990	2.1	3	18							
Fucus Ves.	Torness (UK)	2	1989-1990				0.2	0.031	0.18	3.5	0.069	3	
Fucus Ves.	Hartlepool (UK)	21	1989-1990				0.33	0.046	0.044	1.2	0.038	1.1	
Fucus Ves.	Alderney (UK)	2	1989-1990							4.5	0.151	4	
Fucus Ves.	Devoport (UK)	1	1989-1990				0.26	0.08	0.08				
Fucus Ves.	Cemlyn (UK)	2	1989-1990				0.41	0.12	0.071				
Fucus Ves.	Heysham (UK)	1	1989-1990				0.4	0.14	0.048	2.5	0.066	2.2	
Fucus Ves.	Port William (UK)	2	1989-1990							2.1	0.89	1.9	
Fucus Ves.	Tongue (UK)		1991	6.21	1.33				0.12			12.2	955
Fucus Ves.	Poolewe (UK)		1991	10.9	2.18				0.03			9.5	700
Fucus Ves.	Aberdeen (UK)		1991	9.54	4.16				0.18			4.5	1285
Fucus Ves.	Stonehaven (UK)		1991	8.82	1.75				0.16			9.9	1198
Fucus Ves.	Anstruther (UK)		1991	2.72	1.31				0.04			12	1003
Fucus Ves.	Sandyhills (UK)		1991	8.98	0.71				0.15			3.8	1175
Fucus Ves.	Whitehaven (UK)		1991	51	27.4				0.38			18.6	1363
Fucus Ves.	Ravenglass (UK)		1991	16	3.79				0.82			4.8	1317
Fucus Ves.	Hartlepool (UK)		1991	2.92	0.85				0.38			7.1	1439

Table 28 (cont'd)

Material	Location	No. Sample	Year	<sup>210</sup> Po	<sup>210</sup> Pb	<sup>226</sup> Ra	<sup>228</sup> Th	<sup>230</sup> Th	<sup>232</sup> Th	<sup>234</sup> U	<sup>235</sup> U	<sup>238</sup> U	<sup>40</sup> K
Fucus Ves.	Colwyn Bay (UK)		1991	6.53	1.57				0.22			12.4	1127
Fucus Ves.	Gt Yarmouth (UK)		1991	6.5	1.68				0.29			5.1	374
Fucus Ves.	Eastbourne UK)		1991	8.66	1.25				0.08			8.9	886
Fucus Ves.	Plymouth (UK)		1991	4.56	1.52				0.19			14.7	985

**Table 29**      **Concentrations of  $^{210}\text{Po}$  in seaweed (*Fucus ves.*) from France, Bq kg<sup>-1</sup> dw**

Date	Phare de la Falaise (Seine estuary)	Tour de Controle Honfleur (Seine estuary)	Pointe de Moulard	Pirou	Wimereux
29/03/90	13.56	14.41	6.60		11.03
11/05/90	12.72	17.88			7.03
22/05/90	17.57	21.38	4.26		
12/06/90	19.01	14.49	6.70		9.32
21/06/90	12.48	13.45			
10/07/90	11.16	11.00	4.31		12.28
26/07/90	14.26	13.46	4.07		
07/08/90	15.65	11.31	3.81		9.94
22/09/90	12.38	13.47	3.93		10.73
11/10/90	17.50	19.49	3.02		7.85
15/11/90				15.90	
13/12/90	13.73	21.72	3.94		7.09
18/01/91	13.51	10.09	5.55		9.06
28/02/91	11.33	10.40	6.31	16.96	9.72
28/03/91	12.49	12.00	3.89	20.01	8.72
Max	19.01	21.72	6.70	20.01	12.28
Min	11.16	10.09	3.02	15.90	7.03
Geomean	13.91	14.15	4.56	17.54	9.21
Average	14.10	14.61	4.70	17.62	9.34
Number	14	14	12	3	11

**Table 30** Concentrations of 210Po in mussels (*Mytilus edulis*) from France and Ireland, Bq kg<sup>-1</sup> dw

Date	Digue Nord	Phare de la Falaise	Outfall	Pointe de Moulard	Pirou	Agon Coutain ville	Wimereux	Date	Sutton
	(Seine estuary)	(Seine estuary)	(Seine estuary)						Ireland
29/03/90	346	225	702	138		293	193	09/93	160
11/05/90	219	290		106			217	10/93	190
22/05/90	137	189		109				11/93	250
12/06/90	197	205	176	96.4			219	12/93	220
21/06/90	191	200						02/94	150
10/07/90	162	177		111			277	03/94	180
26/07/90	157	164		102		332		04/94	220
07/08/90	134	121		91				05/94	160
22/09/90	139	157		94.9		255	201	06/94	360
11/10/90	141	156	138	136			215	06/94	160
15/11/90					315		244	07/94	170
13/12/90	129	177		145			214	09/94	210
18/01/91	147	189		208	351		287	10/94	180
28/02/91	194	153		273	309		262	11/94	220
28/03/91	236	213		219	282		151	12/94	160
01/05/91	220								
01/07/91	170								
01/10/91	260								
01/11/91	150								
01/06/92	210								
01/10/92	210								
01/03/93	300								
01/05/93	160								
Max	346	290	702	273	351	332	287		360
Min	129	121	138	91	282	255	151		150
Geomean	184	183	257	132	313	292	222		194
Average	191	187	339	141	314	293	225		199
Number	22	14	3	13	4	3	11		15

**Table 31** Concentrations of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in biota from European waters

Material	Specie	No.sample	Location	Year	$^{210}\text{Po}$	$^{210}\text{Pb}$	Unit
Mussels	Mytilus edulis		Oosterschelde (NL)	1989-1990	110	4	Bq/kg dw
Mussels	Mytilus edulis		Westerschelde (NL)	1989-1991	300	4	Bq/kg dw
Mussels	Mytilus edulis		North Sea coast (NL)	1989-1992	300	4	Bq/kg dw
Mussels	Mytilus edulis		Waddensea	1989-1993	300	4	Bq/kg dw
Mussels	Mytilus edulis		Nieuwe waterweg (NL)	1989-1994	350	5	Bq/kg dw
Shrimp	Crangon crangon		Oosterschelde (NL)	1989-1995	90	1	Bq/kg dw
Shrimp	Crangon Crangon		Westerschelde (NL)	1989-1996	235	3	Bq/kg dw
Shrimp	Crangon Crangon		North Sea coast (NL)	1989-1997	270	1	Bq/kg dw
Shrimp	Crangon Crangon		Waddensea	1989-1998	180	1	Bq/kg dw
Shrimp	Crangon Crangon		Nieuwe waterweg (NL)	1989-1999	290	1	Bq/kg dw
Mussels	Mytilus edulis	41	Waddensea (DK)	1993-1994	220		Bq/kg dw
Mussels	Mytilus edulis	41	Fjords (DK)	1993-1994	100		Bq/kg dw
Mussels	Mytilus edulis	41	Risø (DK)	1993-1994	180		Bq/kg dw
Mussels	Mytilus edulis	41	Baltic	1993-1994	190		Bq/kg dw
Cod		13	Denmark	1993-1994	0.35		Bq/kg ww
Herring		14	Denmark	1993-1994	0.65		Bq/kg ww
Plaice		14	Denmark	1993-1994	0.96		Bq/kg ww
Herring		4	Irish coast	1993-1995	1.74	0.7	Bq/kg ww
Mackerel		3	Irish coast	1993-1995	1.91	0.7	Bq/kg ww
Salmon		4	Irish coast	1993-1995	0.16		Bq/kg ww
Prawn		8	Irish coast	1993-1995	5.3	0.2	Bq/kg ww
Lobster		2	Irish coast	1993-1995	2.5		Bq/kg ww
Oysters		2	Irish coast	1993-1995	29.5		Bq/kg ww
Whiting		4	Irish coast	1993-1995	0.9	0.3	Bq/kg ww
Cod		3	Irish coast	1993-1995	1.3	0.8	Bq/kg ww
Plaice		3	Irish coast	1993-1995	3	0.6	Bq/kg ww
Ray		4	Irish coast	1993-1995	0.8		Bq/kg ww
Mussels		68	Irish coast	1993-1995	25.5	0.6	Bq/kg ww
Mussels	Mytilus edulis		Irish coast	1994	196		Bq/kg dw
Mussels	Mytilus edulis		Seine estuary (Fr)	1990-1993	224		Bq/kg dw
Mussels	Mytilus edulis		Wimereux (Fr)	1990-1991	194		Bq/kg dw
Mussels	Mytilus edulis		Revenglass (England)	1984	279		Bq/kg dw
Mussels	Mytilus edulis		Hunterston (Scotland)	1984	111		Bq/kg dw
Mussels	Mytilus edulis		Firth of Forth(Scotland)	1985	246	24.6	Bq/kg dw
Mussels	Mytilus edulis		Mon St. Michel (Fr)	1985	258	2.6	Bq/kg dw
Mussels	M. galloprovincialis		Sete (Fr)	1985	201	7	Bq/kg dw
Mussels	M. galloprovincialis		Monaco	1984	442	10.8	Bq/kg dw
Mussels	M. galloprovincialis		Monaco	1984	459	13.9	Bq/kg dw
Mussels	M. galloprovincialis		Monaco	1984	428	10.8	Bq/kg dw

**Table 32 Concentrations of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in biota from coastal waters of UK**

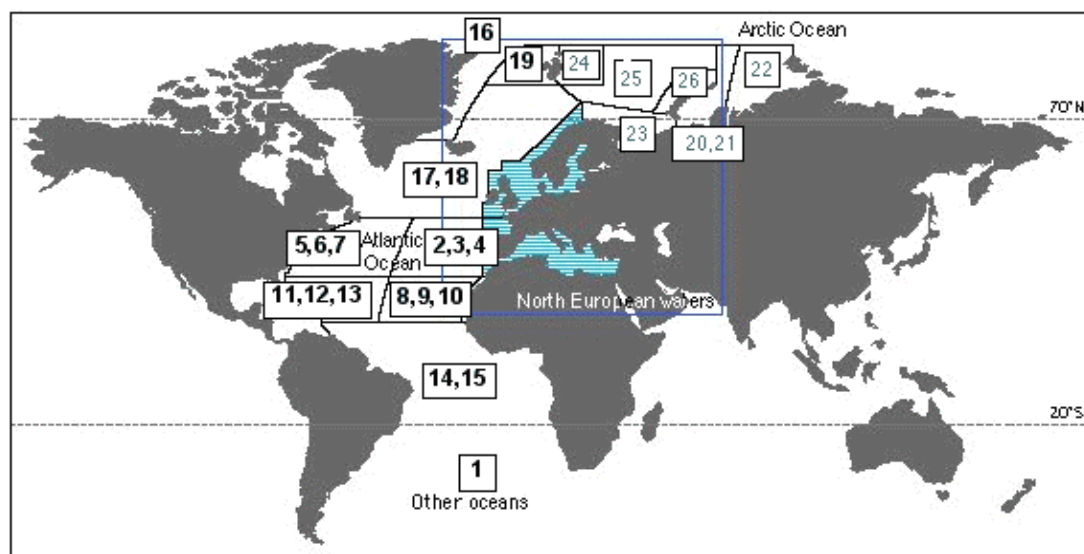
Material	No.sample	Location	Year	$^{210}\text{Po}$
Winkles	3	Sandside Bay	1989-1990	20.4
Winkles	4	Torness	1989-1990	11
Winkles	1	Lynemouth	1989-1990	7
Winkles	1	Tynemouth	1989-1990	9.4
Winkles	3	Hartlepool	1989-1990	7.4
Winkles	3	Cromer	1989-1990	12.7
Winkles	2	Newlyn	1989-1990	8.5
Winkles	3	Tarne Bay	1989-1990	29
Winkles	1	Drigg	1989-1990	24.6
Winkles	9	Nethertown	1989-1990	38.3
Winkles	5	Fleswick Bay	1989-1990	63.8
Winkles	6	Saltom Bay	1989-1990	25.8
Winkles	24	Parton	1989-1990	168
Winkles	4	North Harrington	1989-1990	96.4
Winkles	3	Southernness	1989-1990	23.5
Winkles	1	Kinlochleven	1989-1990	37.9
Mussels	1	Cleethorpes Pier	1989-1990	25.6
Mussels	4	Wash	1989-1990	48.2
Mussels	6	Lowestoft	1989-1990	25.2
Mussels	2	Newlyn	1989-1990	22.8
Mussels	1	Carbis Bay	1989-1990	30.8
Mussels	1	Holyhead Penhos	1989-1990	43
Mussels	4	Ravenglass	1989-1990	73.7
Mussels	12	Nethertown	1989-1990	115
Mussels	1	Portling Bay	1989-1990	53.7
Mussels	1	Kinlochleven	1989-1990	54.8
Mussels	1	Fort William Pier	1989-1990	40.6
Cod	3	Parton	1989-1990	4.5
Lobster	1	Saltom Bay	1989-1990	62.6
Lobster	2	Parton	1989-1990	31.4
Crabs	2	Newlyn	1989-1990	59.7
Crabs	3	Parton	1989-1990	129
Cockles	2	Ribble Estuary	1989-1990	29.2
<i>Fucus ves.</i>		Sandside Bay	1989-1990	3.5
<i>Fucus ves.</i>		Torness	1989-1990	2.5
<i>Fucus ves.</i>		Hartlepool	1989-1990	0.7
<i>Fucus ves.</i>		Immingham	1989-1990	1.3
<i>Fucus ves.</i>		Bradwell	1989-1990	1.3
<i>Fucus ves.</i>		Alderney	1989-1990	2
<i>Fucus ves.</i>		Devonport	1989-1990	0.9
<i>Fucus ves.</i>		Hinkley	1989-1990	0.8
<i>Fucus ves.</i>		Avonmouth	1989-1990	1.2
<i>Fucus ves.</i>		Swansea	1989-1990	0.9
<i>Fucus ves.</i>		Holyhead Penhos	1989-1990	1.8
<i>Fucus ves.</i>		Cemlyn bay	1989-1990	1.3
<i>Fucus ves.</i>		Heysham	1989-1990	1.2

**Table 32 (cont'd)**

<i>Fucus ves.</i>		Sellafield	1989-1990	3.2
<i>Fucus ves.</i>		St. Bees	1989-1990	3.2
<i>Fucus ves.</i>		Parton	1989-1990	6
<i>Fucus ves.</i>		North Harrington	1989-1990	8.5
<i>Fucus ves.</i>		Port william	1989-1990	2.3



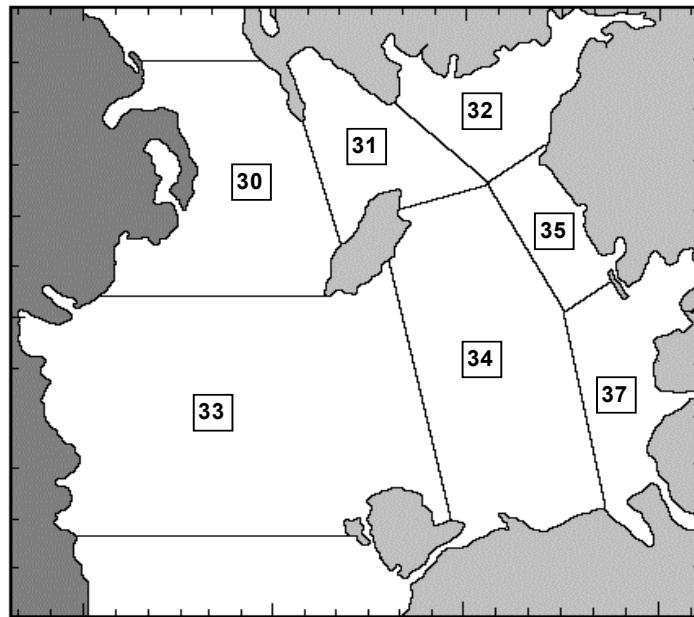
**Figure 1a** Word Marine compartments in MARINA – II model



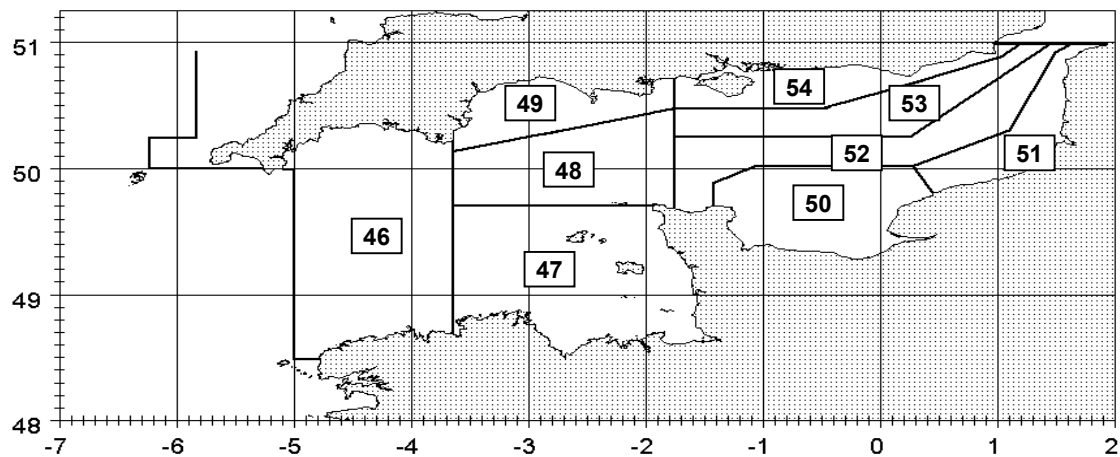
**Figure 1b** North European water compartments in MARINA - II model.



**Figure 1c      Irish Sea compartments in MARINA - II model**



**Figure 1d – English Channel compartments in MARINA - II model**



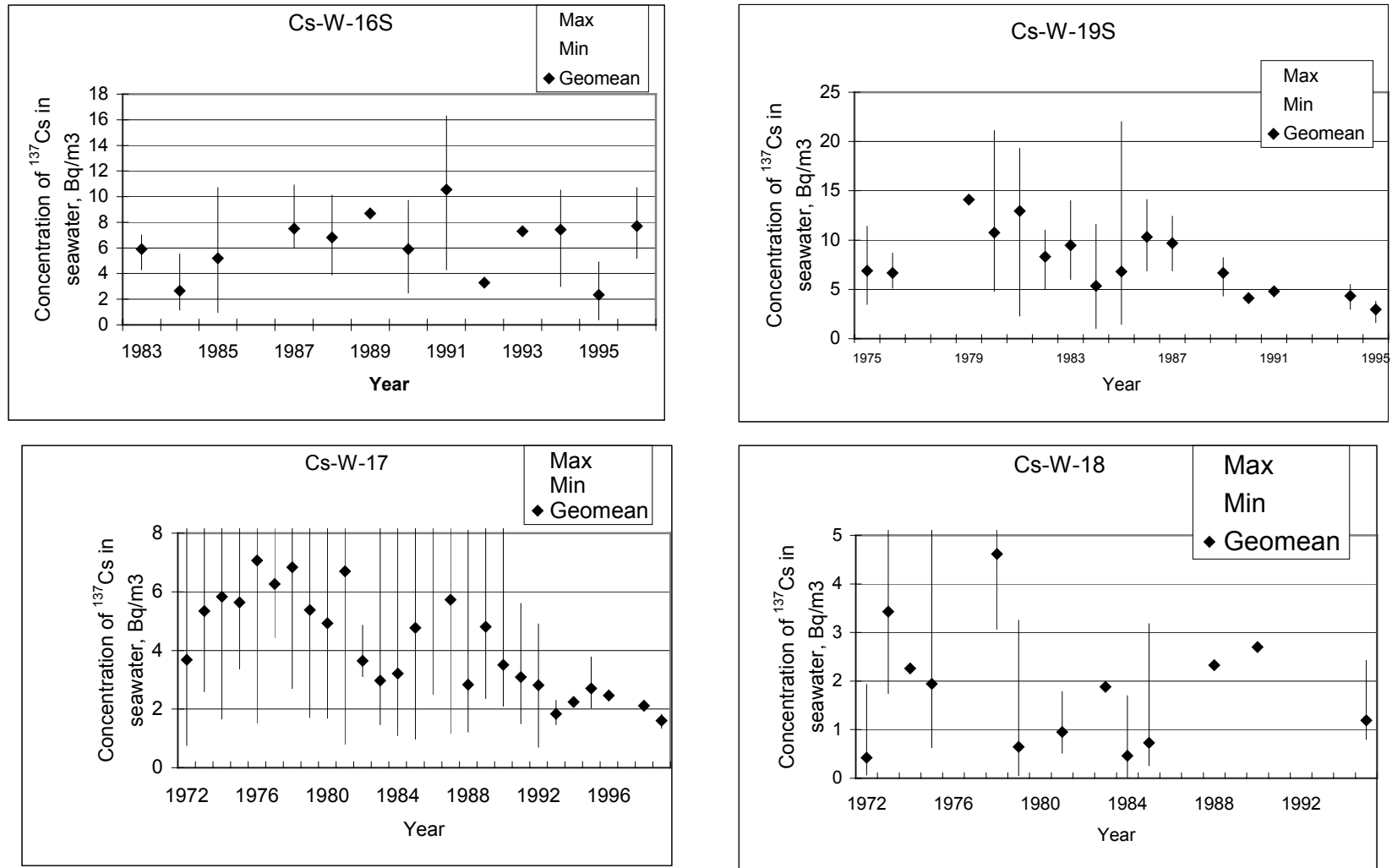
**Figure 2** Temporal variation of  $^{137}\text{Cs}$  concentrations ( $\text{Bq m}^{-3}$ ) in seawater

Figure 2 (cont'd)

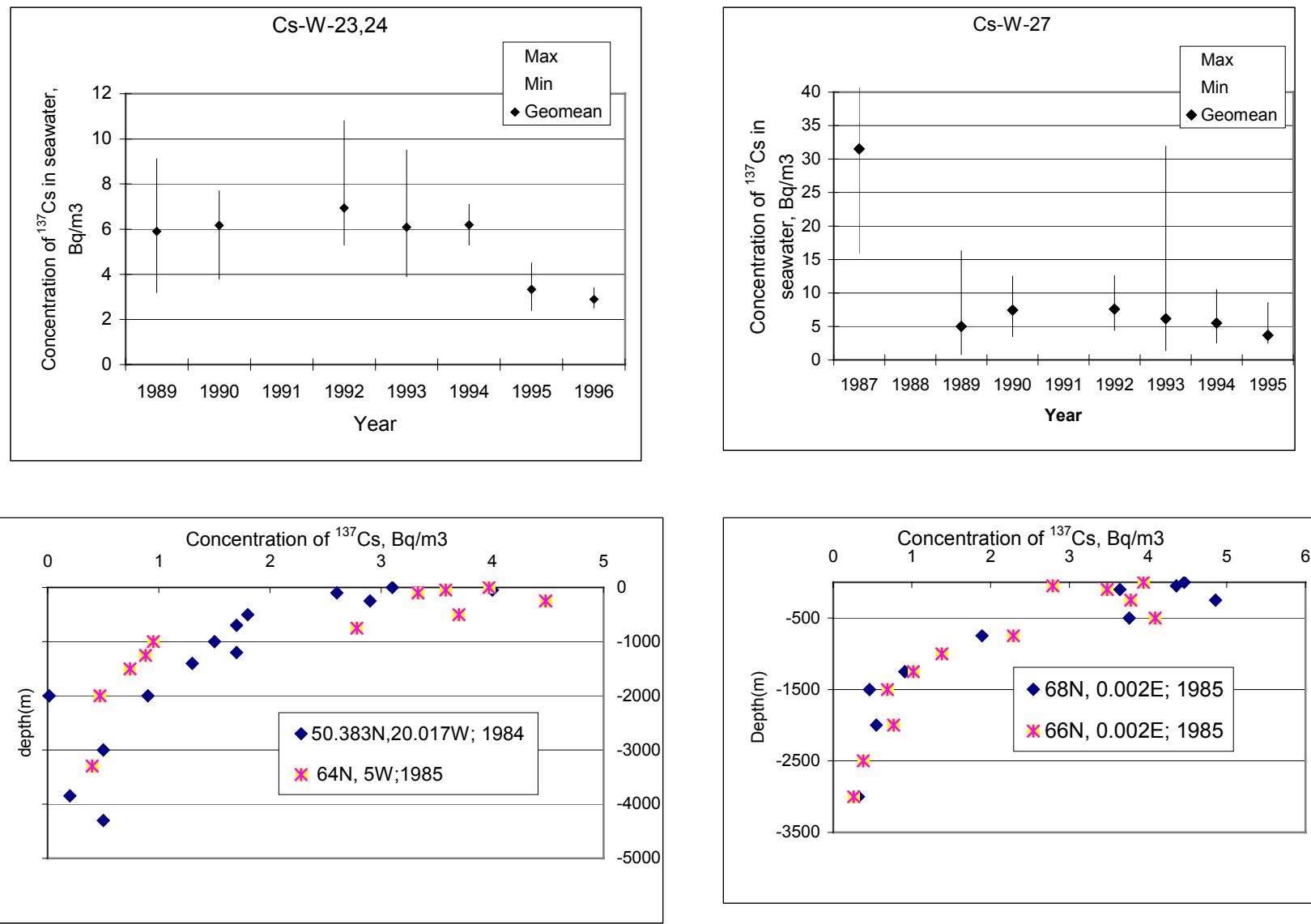


Figure 2 (cont'd)

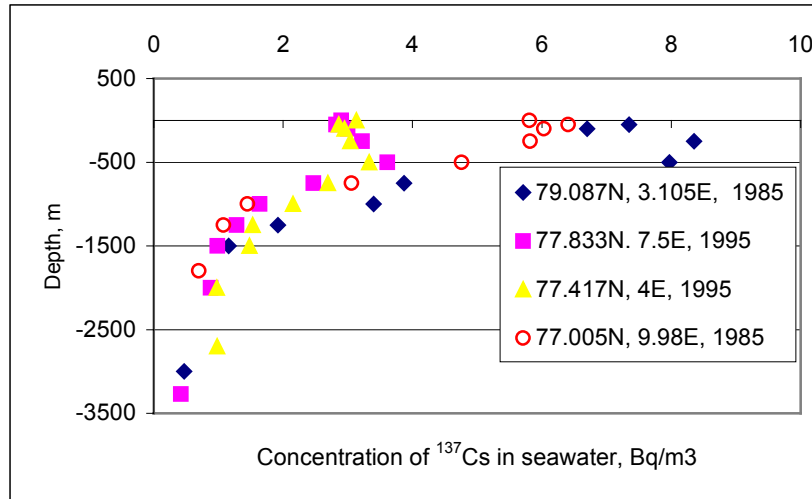
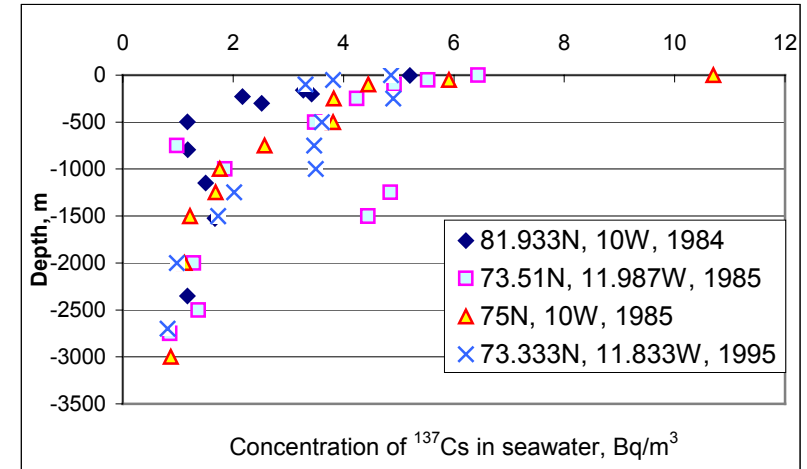
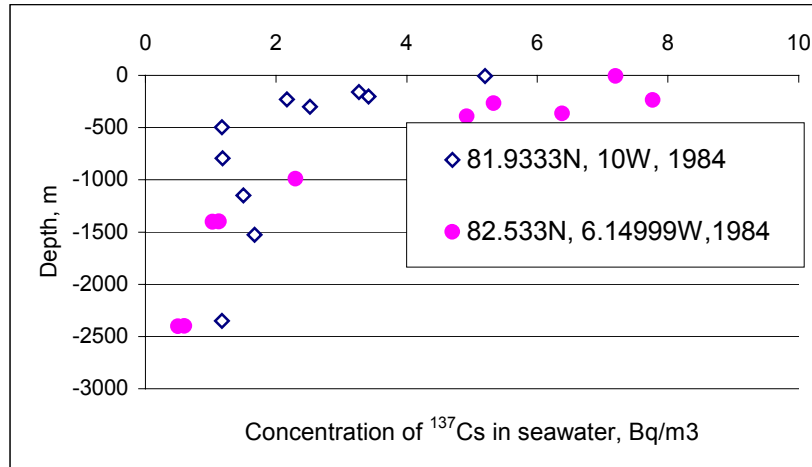


Figure 2 (cont'd)

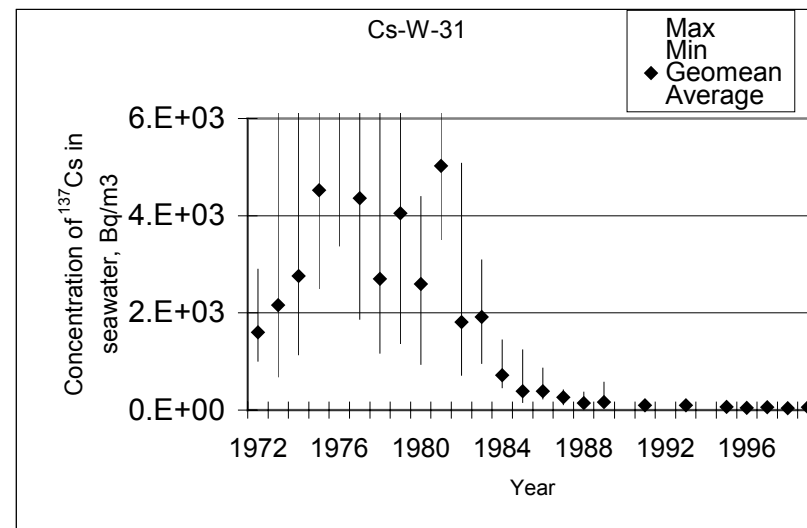
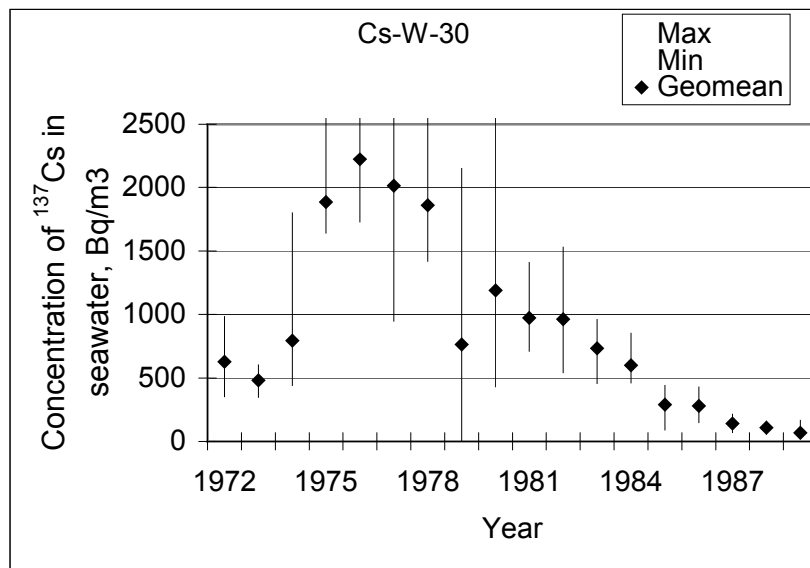
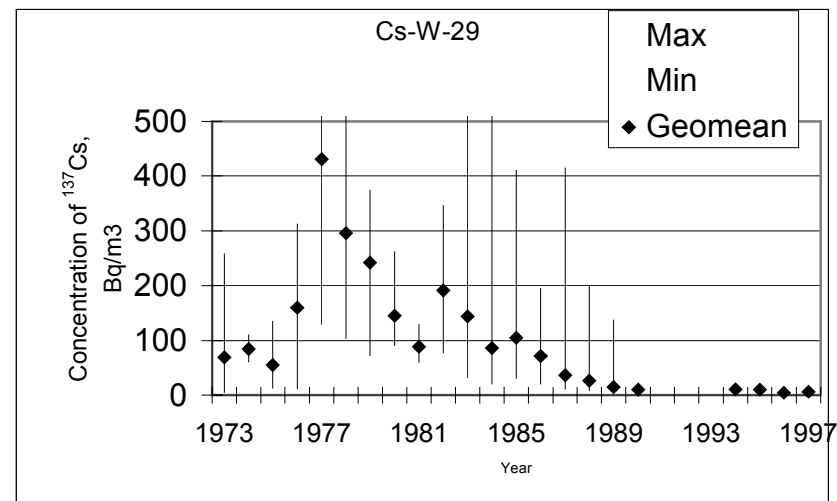
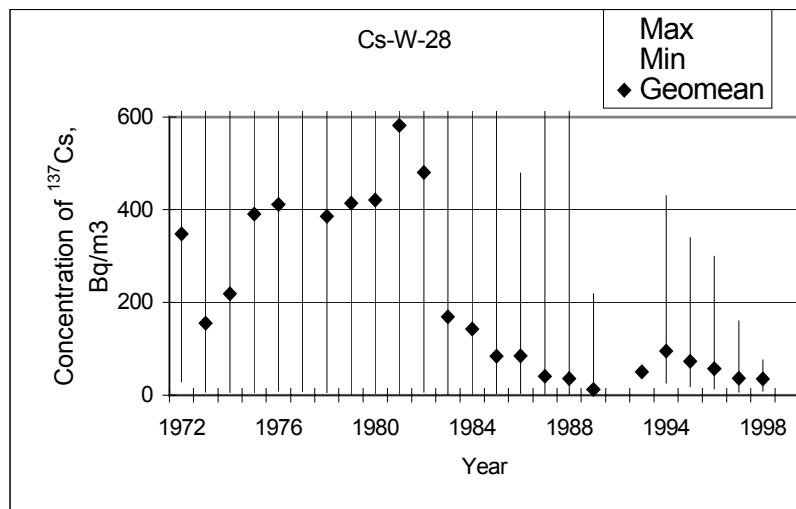


Figure 2 (cont'd)

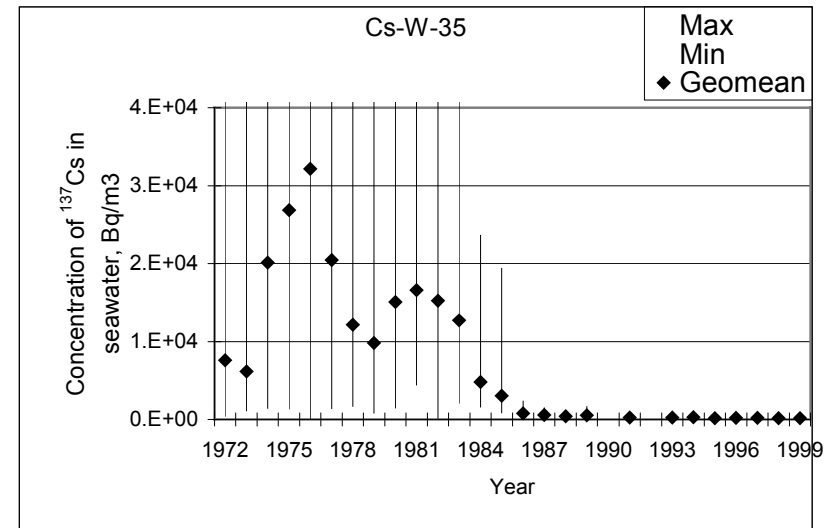
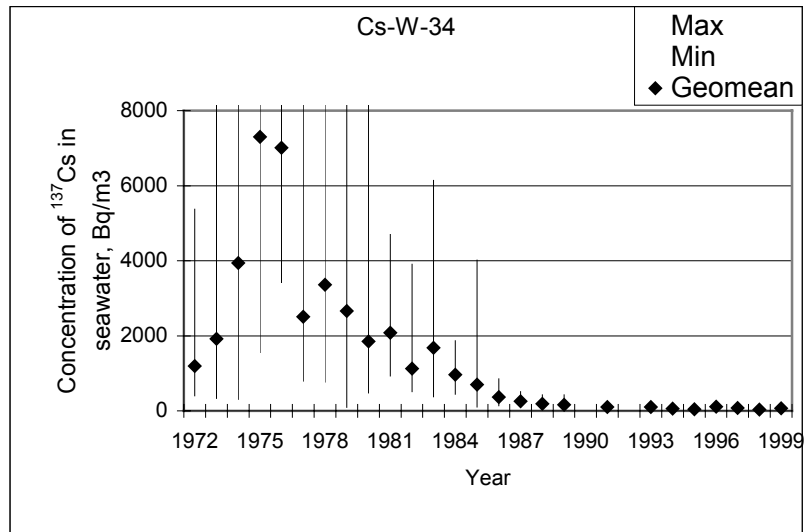
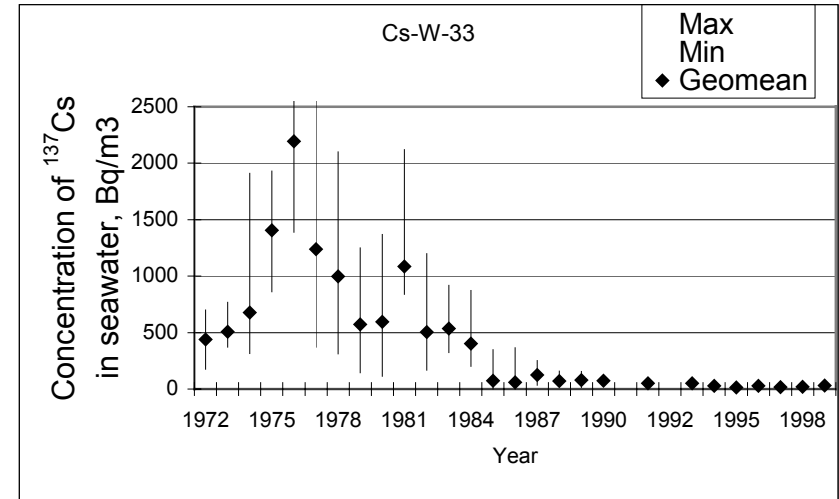
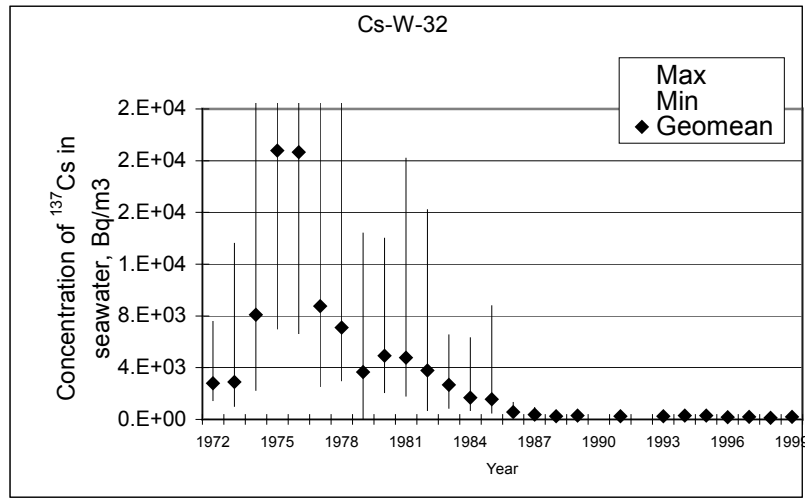




Figure 2 (cont'd)

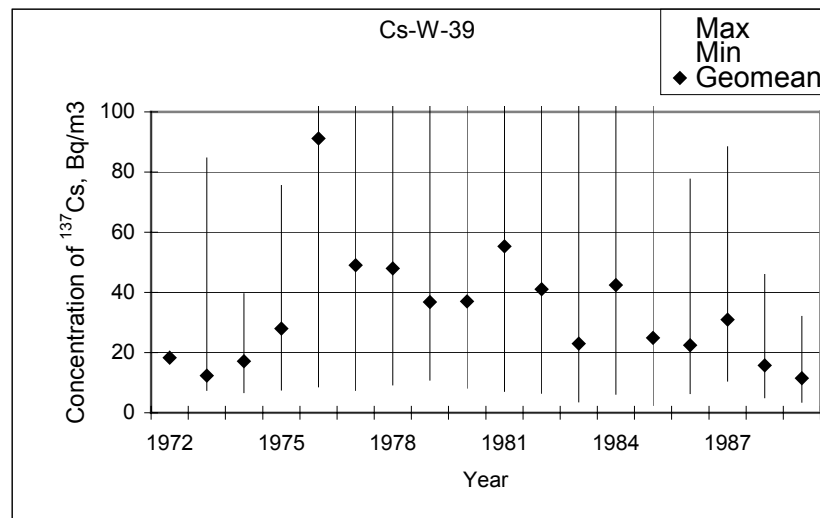
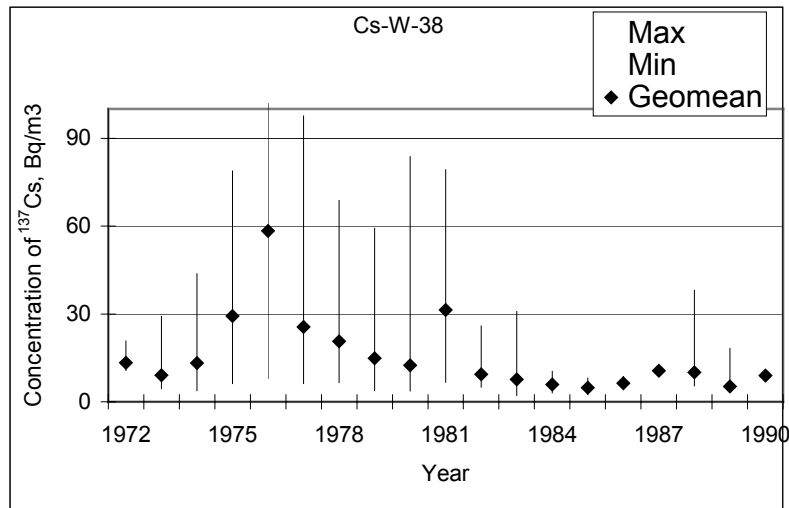
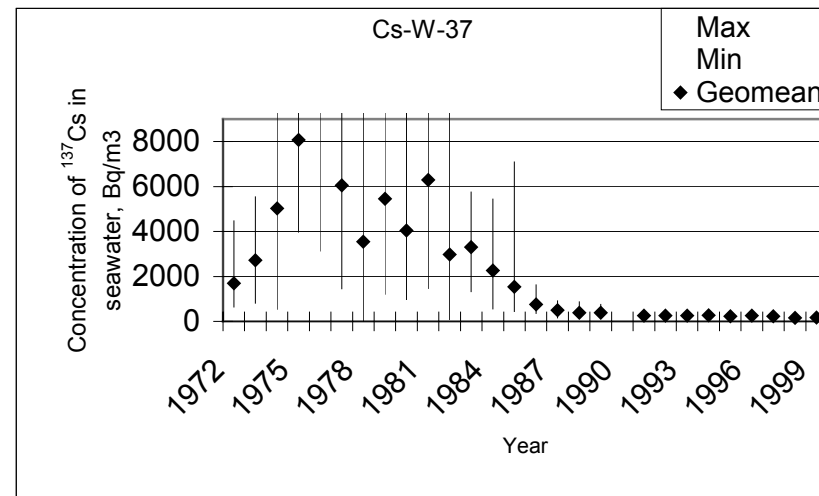
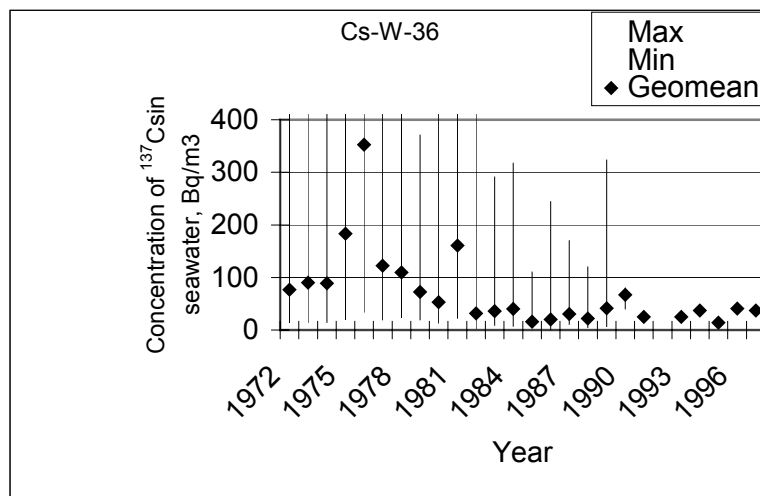


Figure 2 (cont'd)

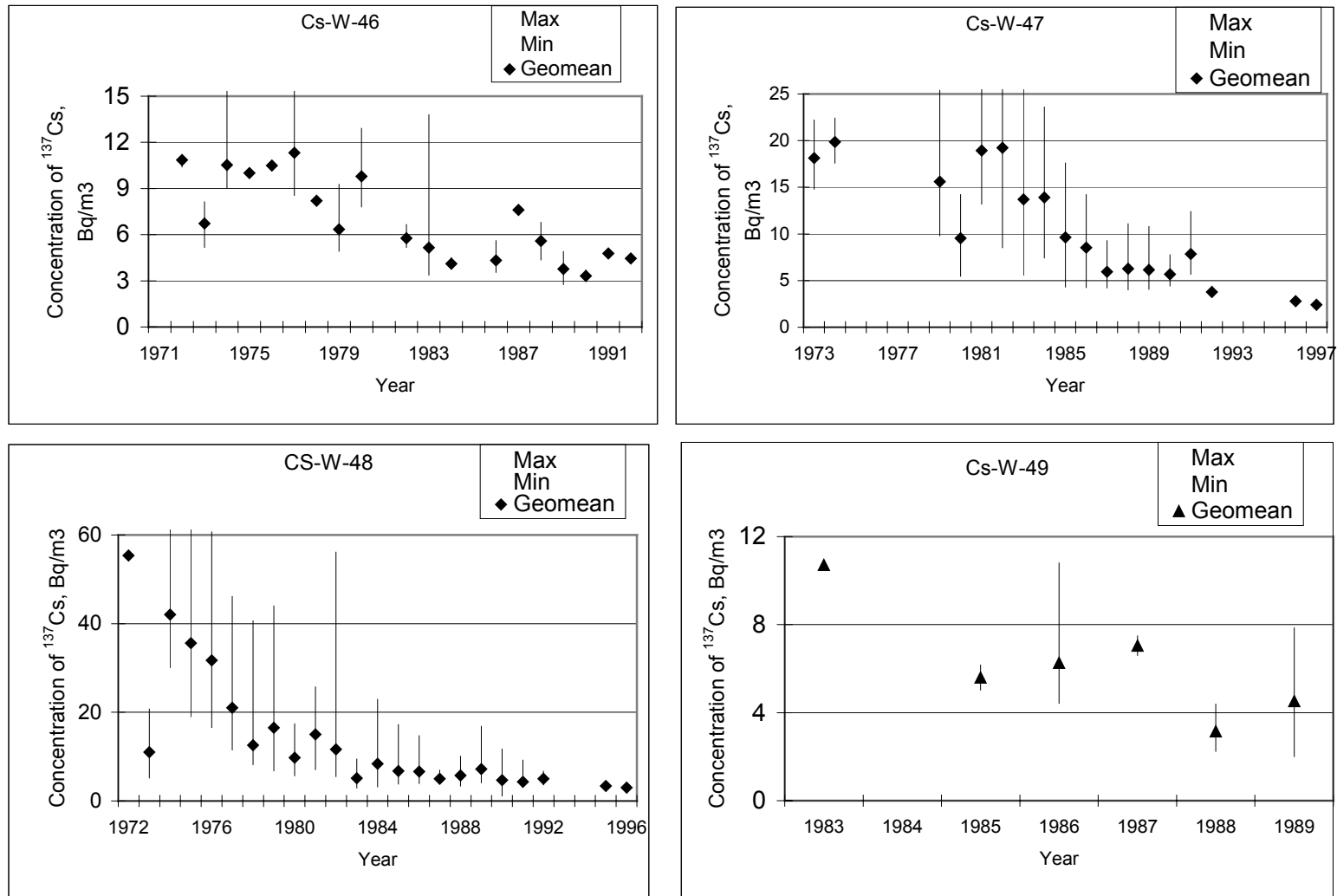


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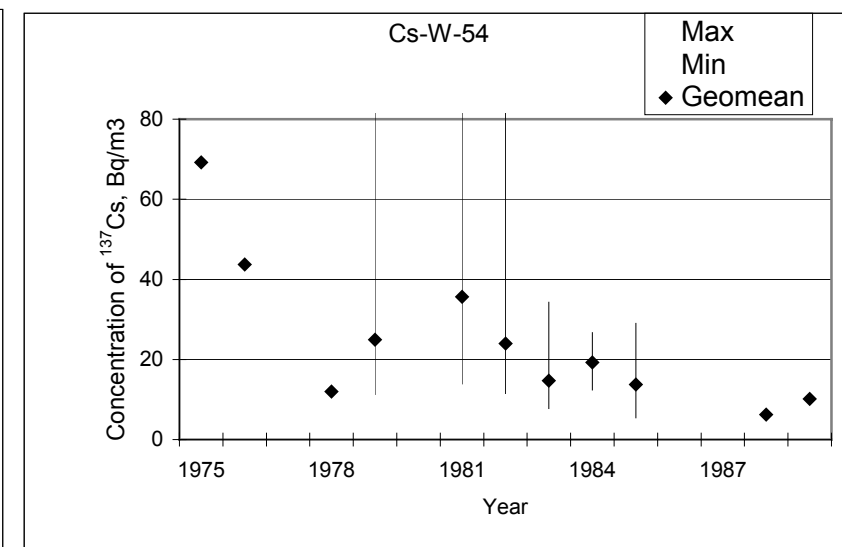
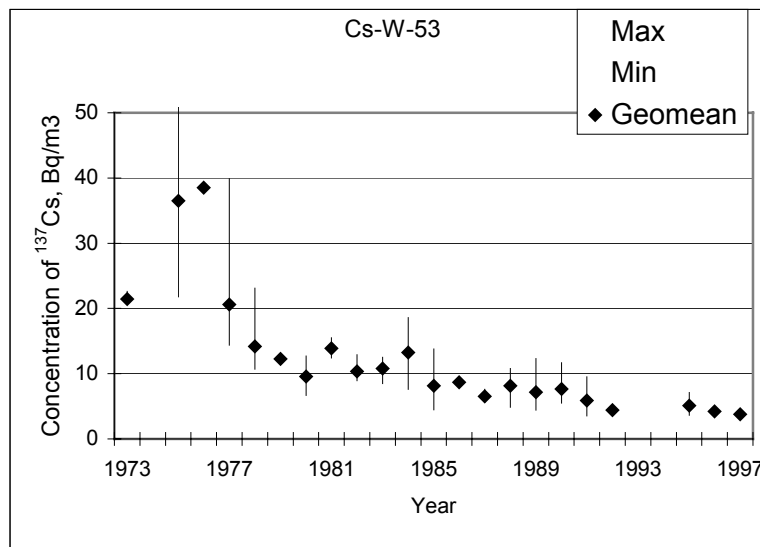
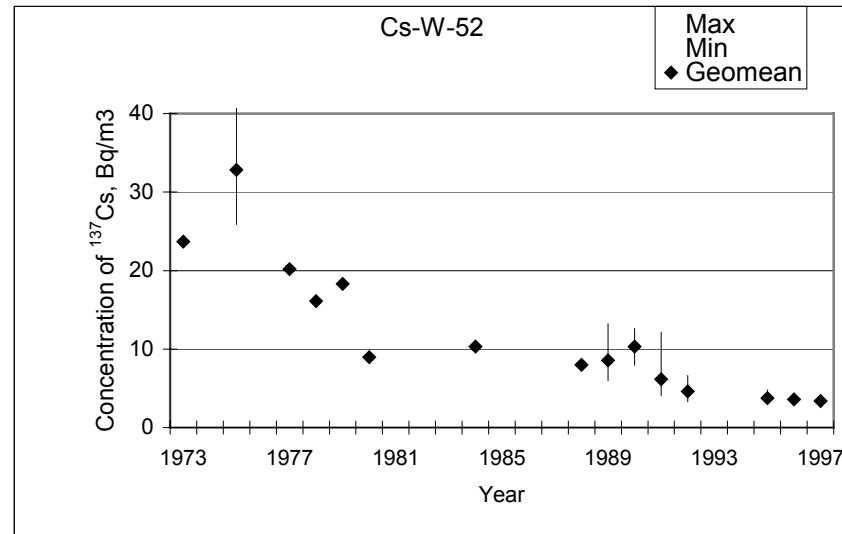
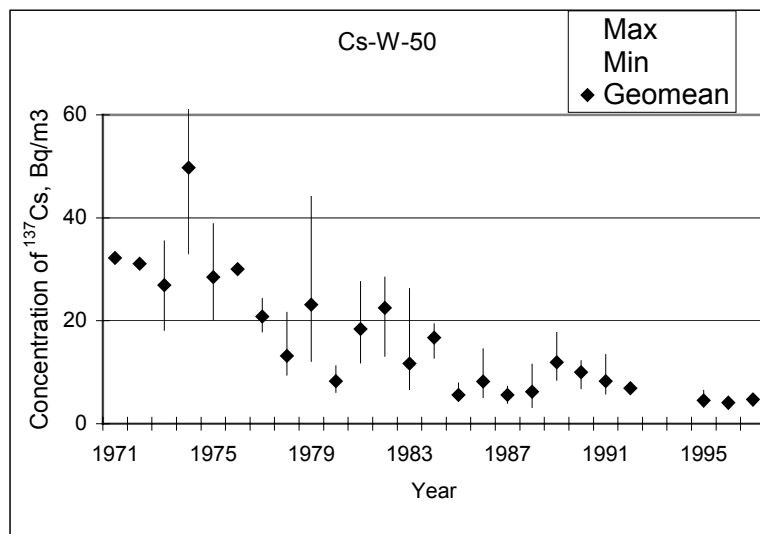


Figure 2 (cont'd)

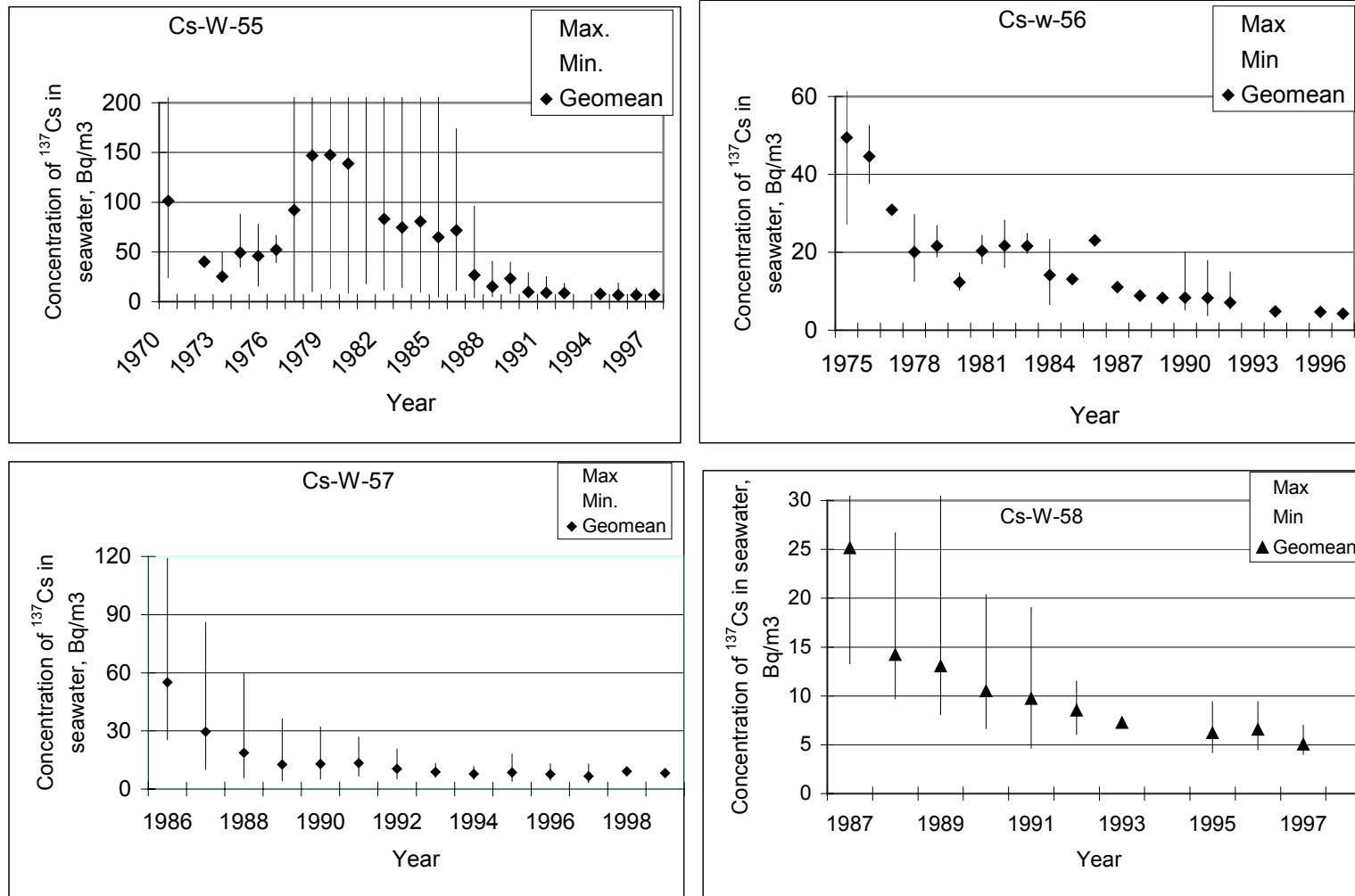
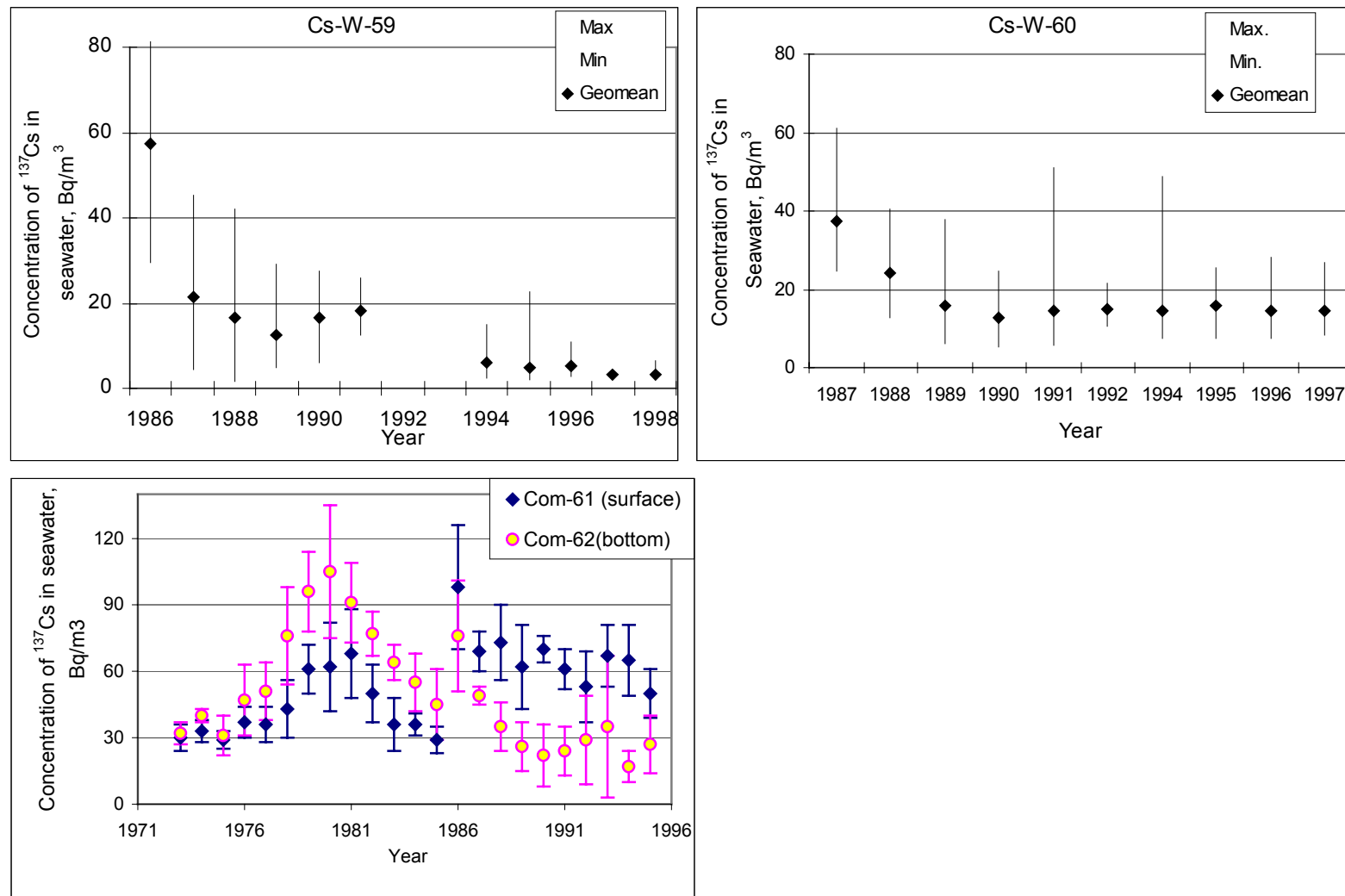
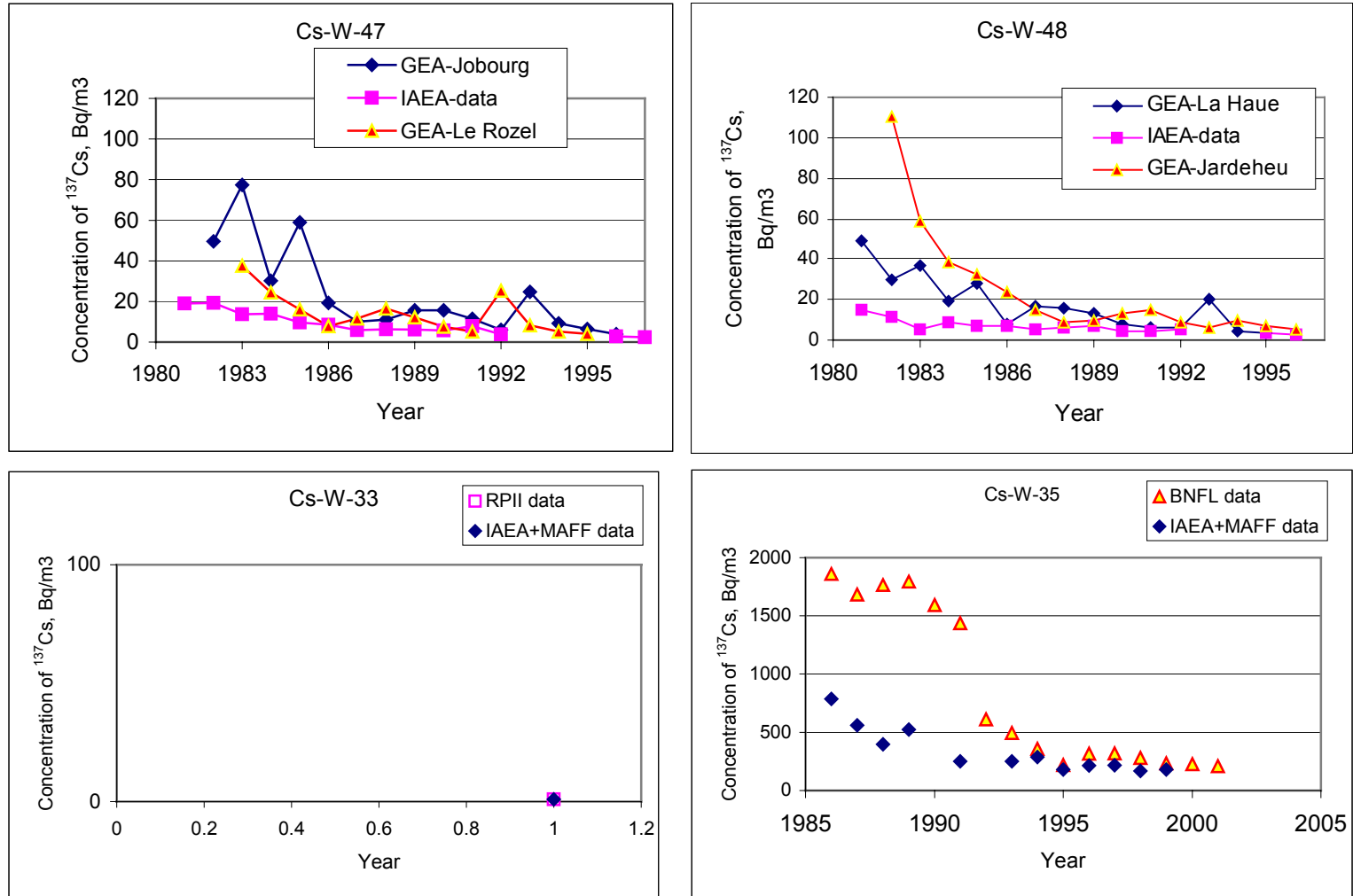


Figure 2 (cont'd)



**Figure 3** Comparison of  $^{137}\text{Cs}$  concentrations in seawater from different sources



**Figure 4** Temporal variation of  $^{137}\text{Cs}$  concentrations in fish and shellfish,  $\text{Bq kg}^{-1}$  wet weight

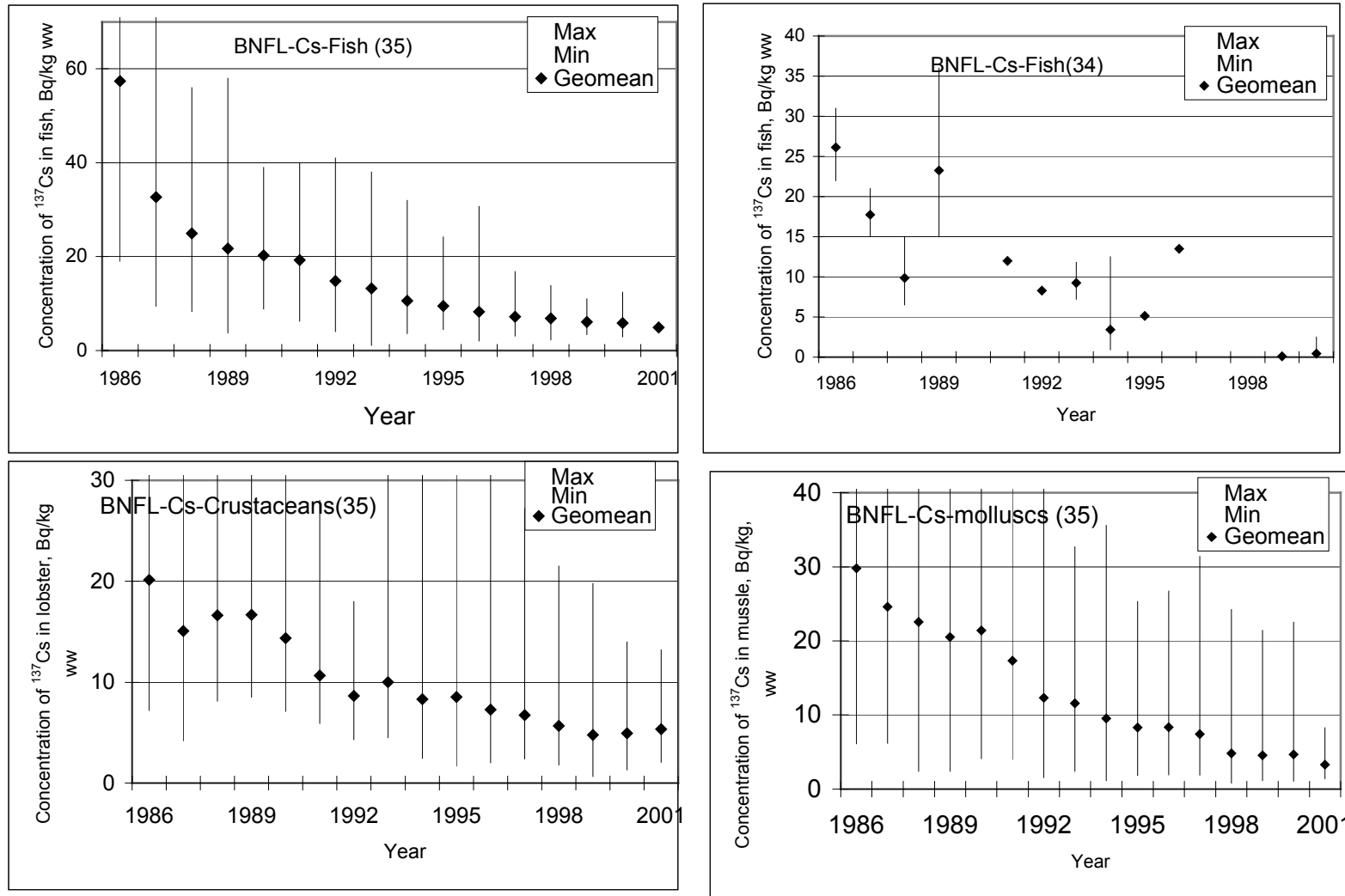


Figure 4 (cont'd)

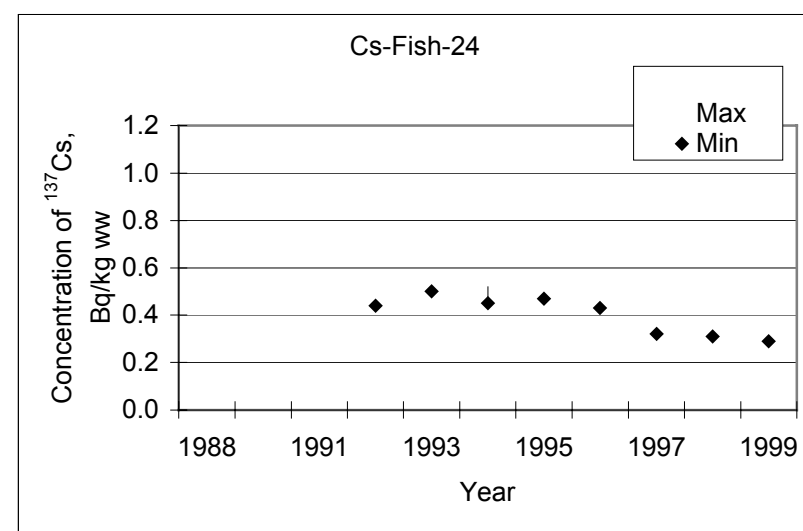
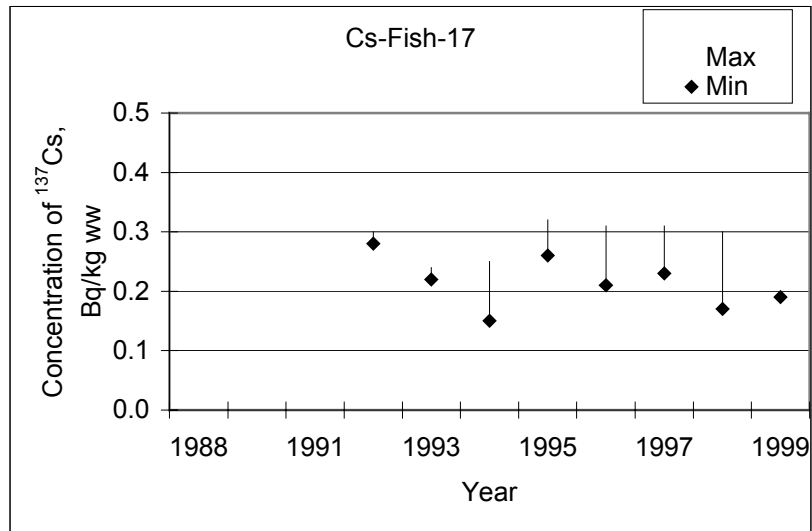
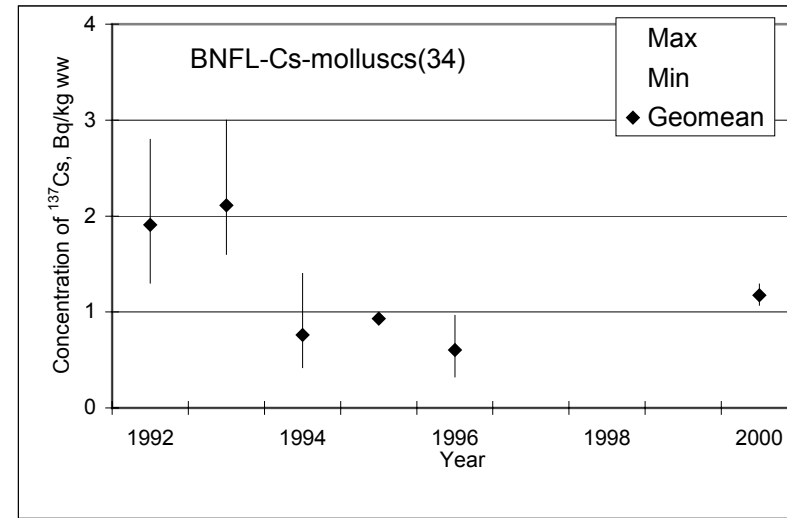
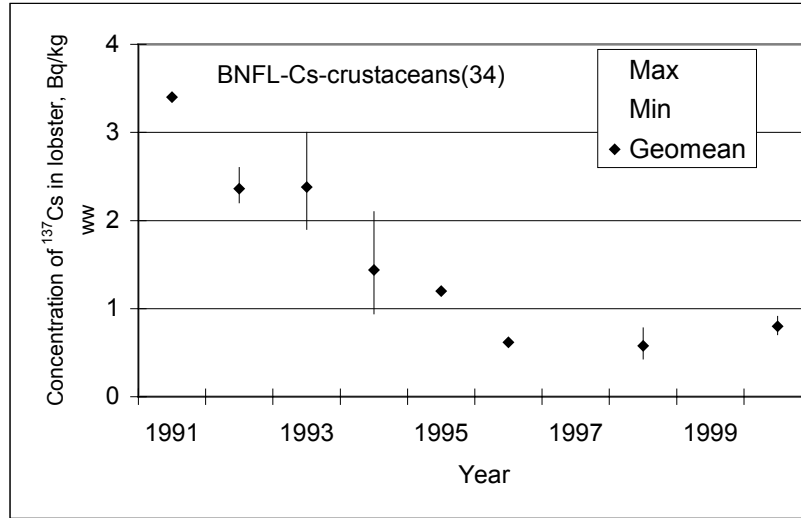




Figure 4 (cont'd)

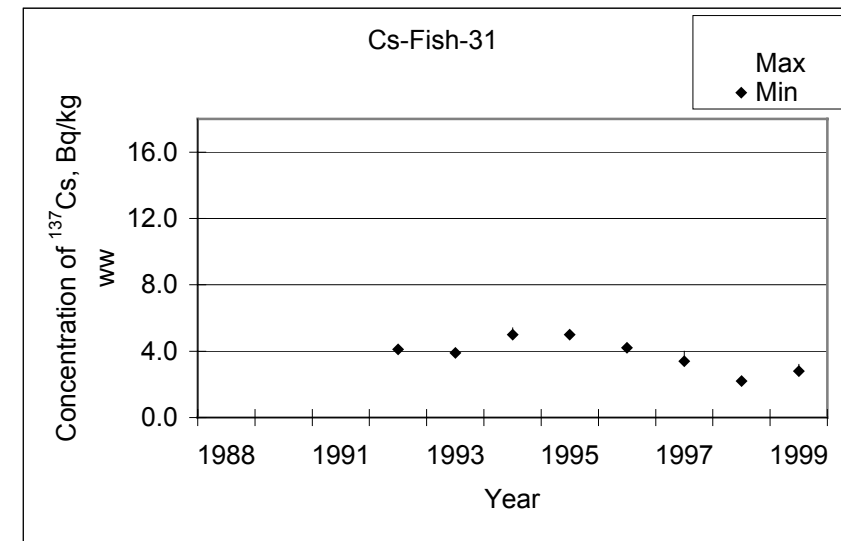
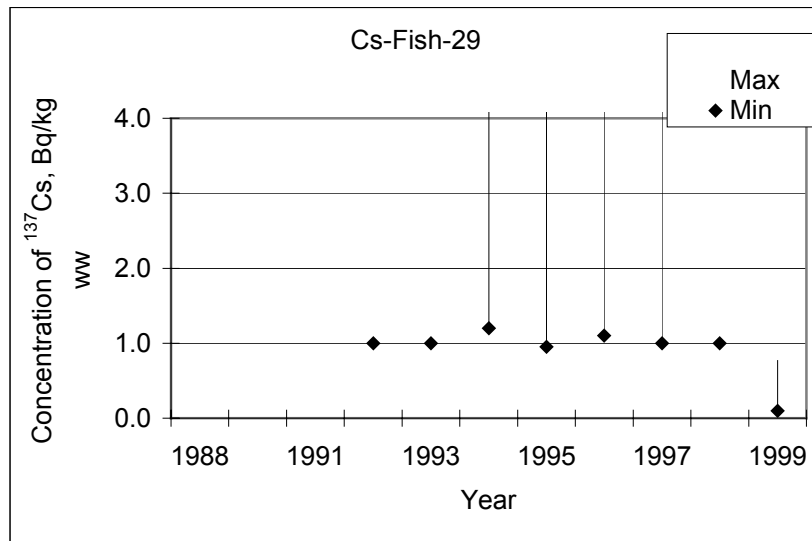
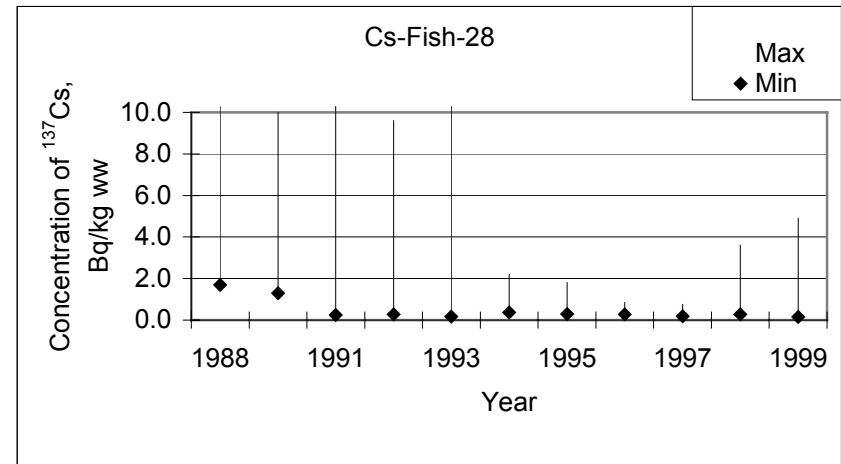
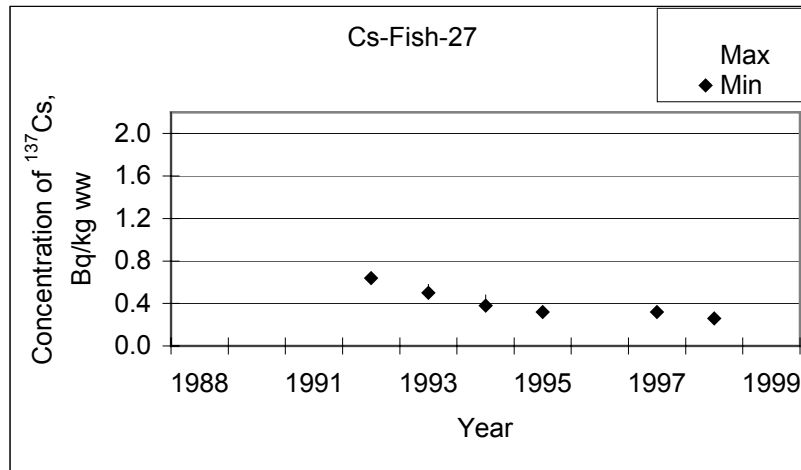


Figure 4 (cont'd)

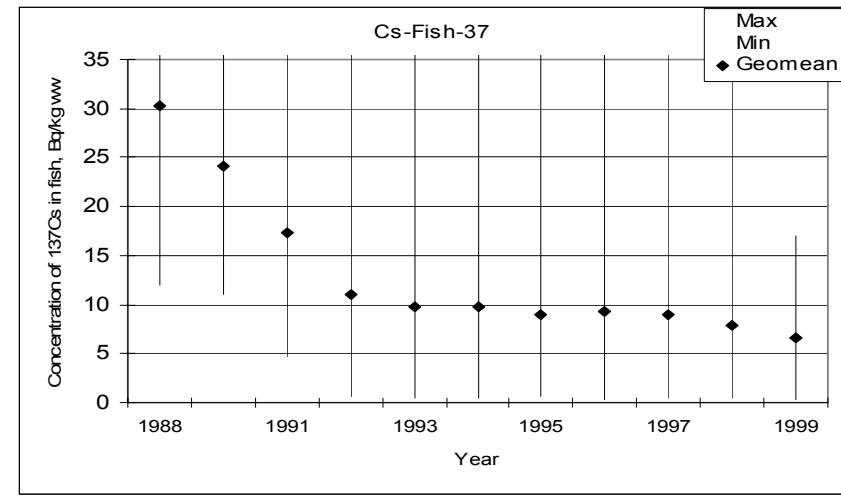
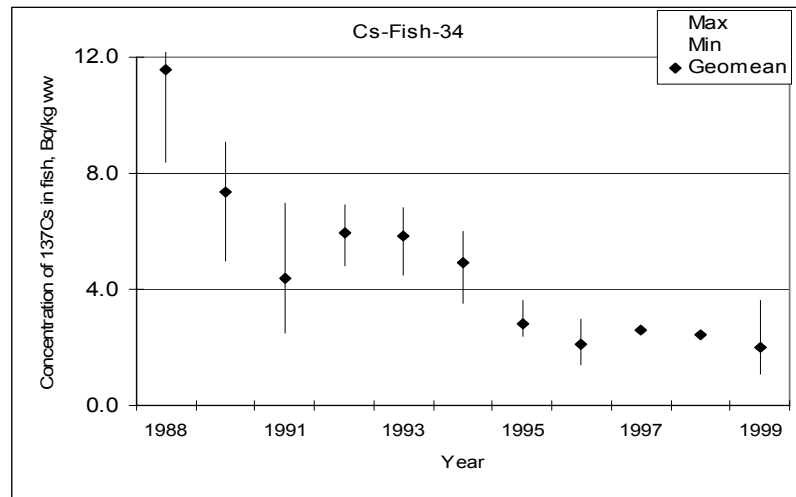
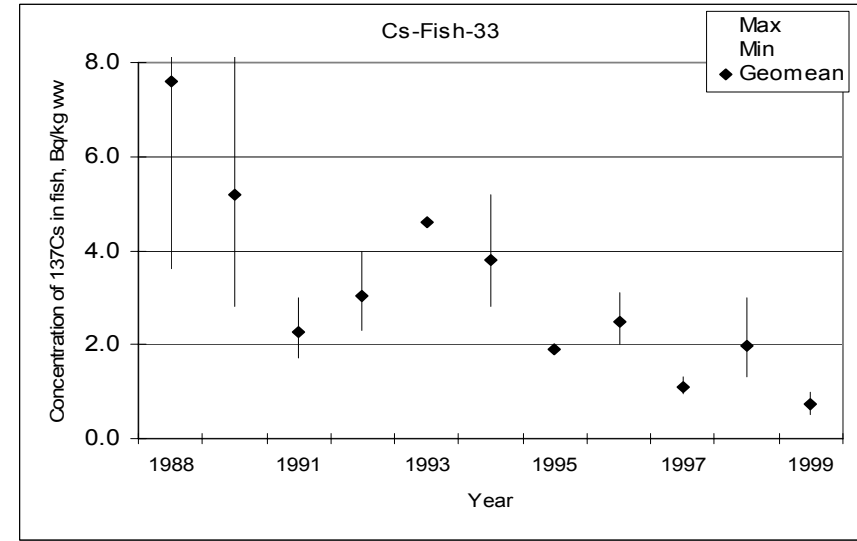
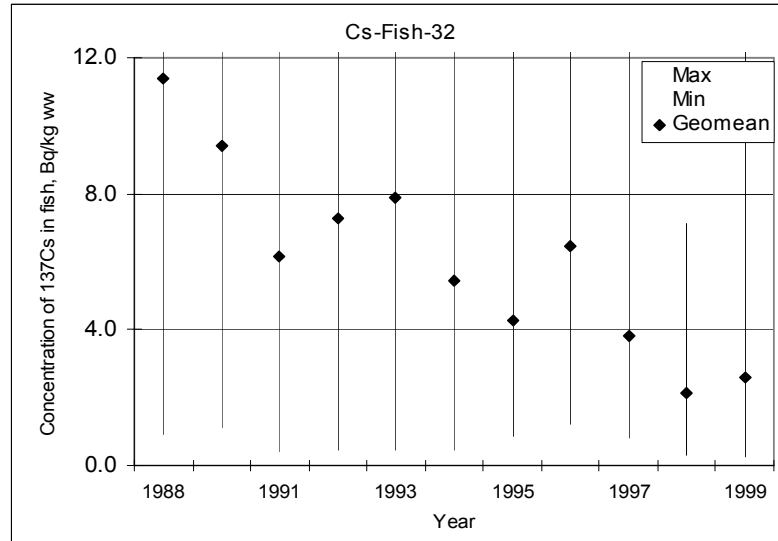
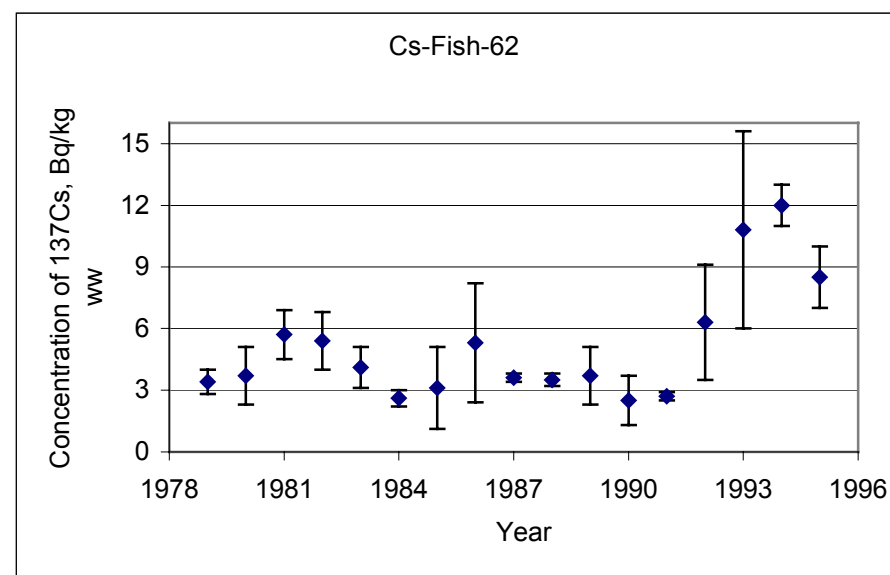
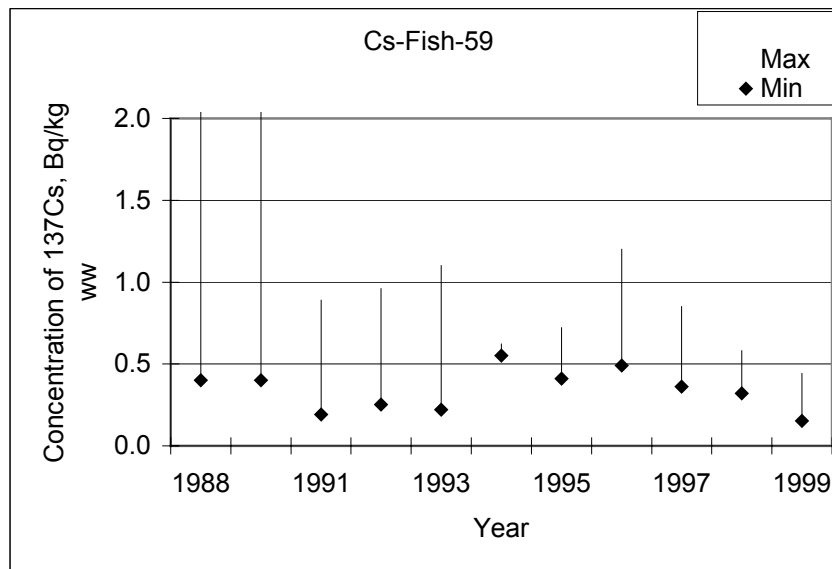
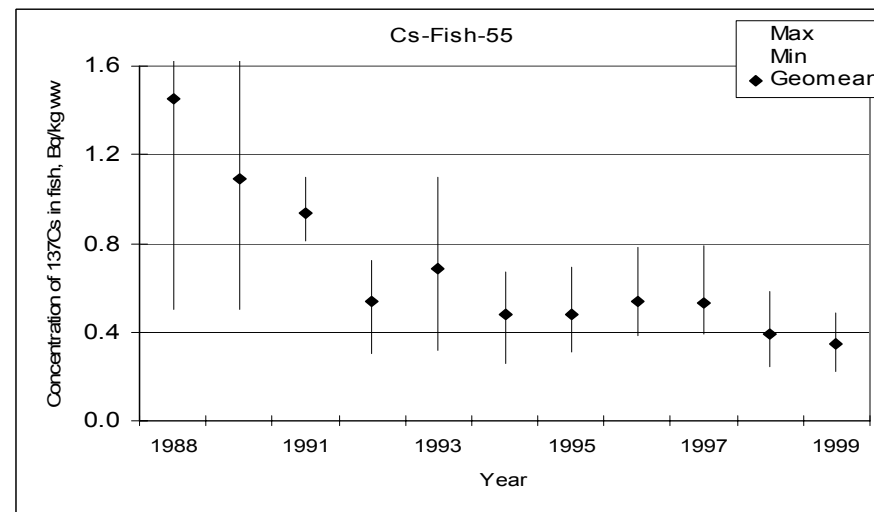
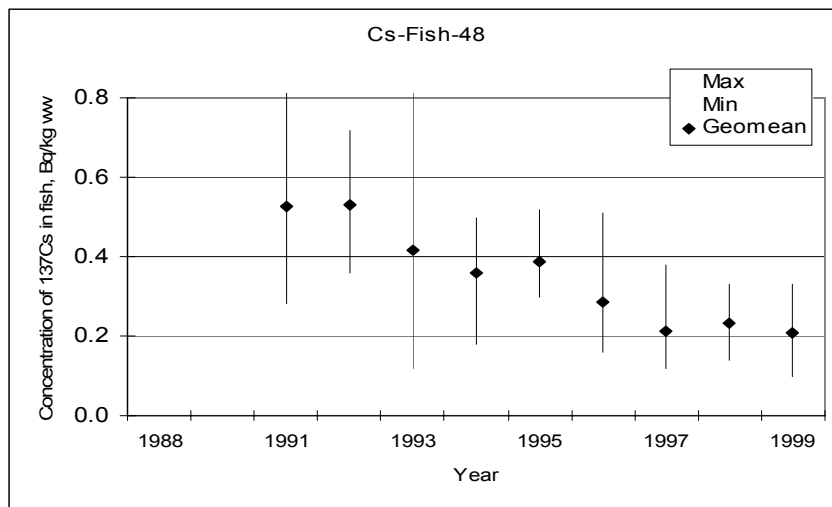


Figure 4 (cont'd)



**Figure 5** Temporal variation of  $^{137}\text{Cs}$  concentrations in seaweed (*Fucus ves.*), Bq kg<sup>-1</sup> wet weight

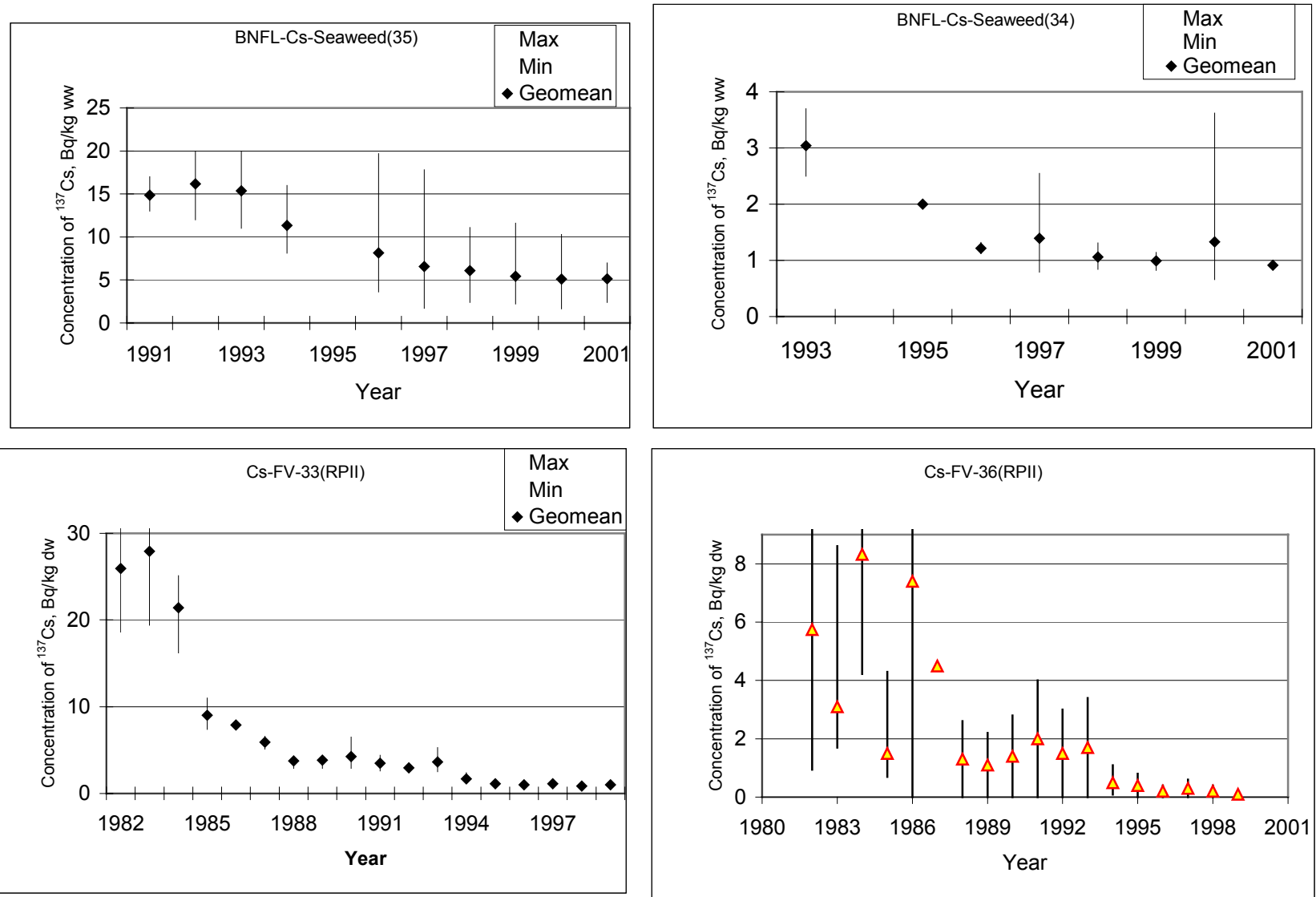


Figure 5 (cont'd)

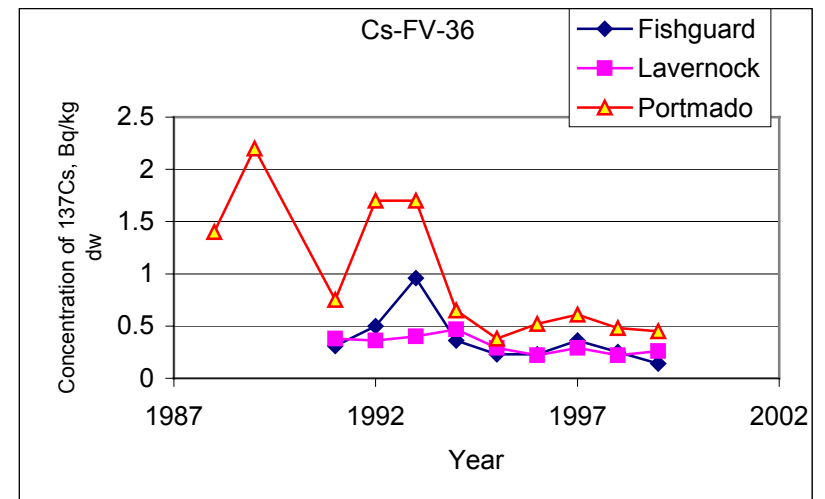
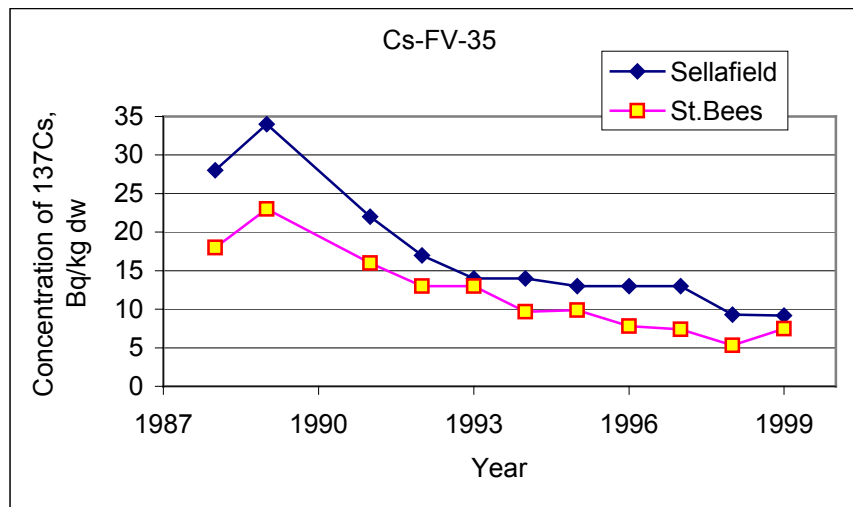
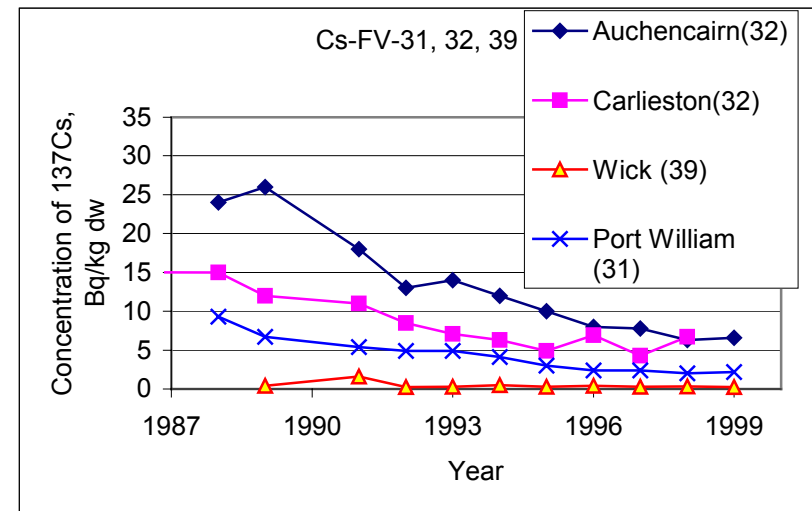
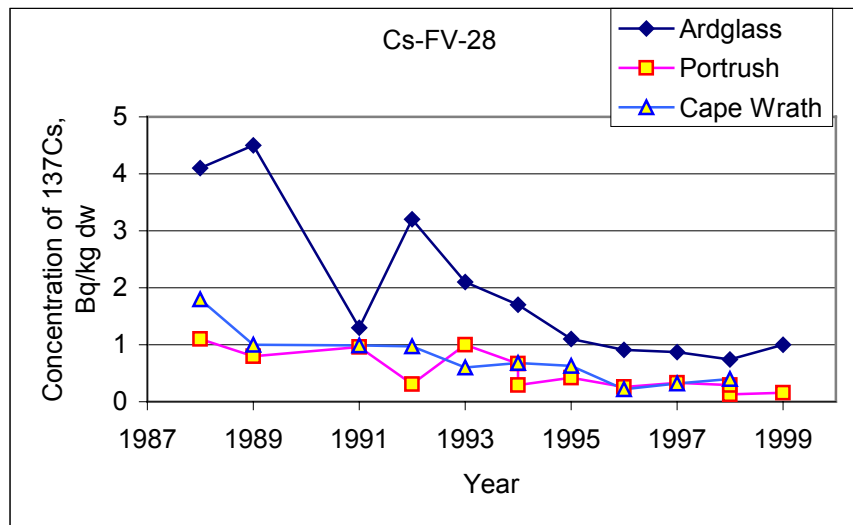
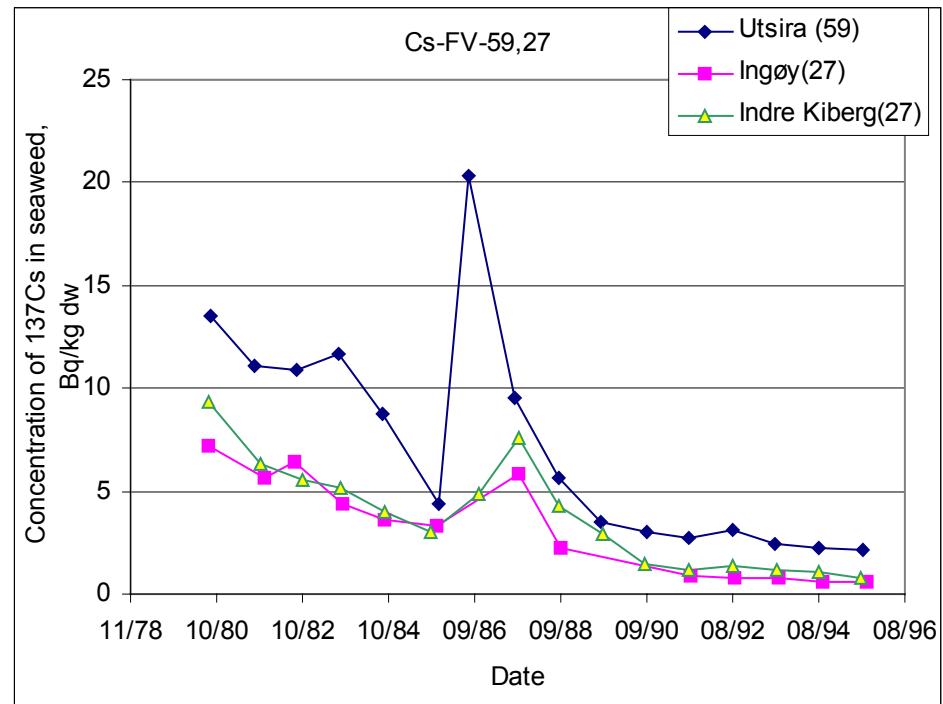
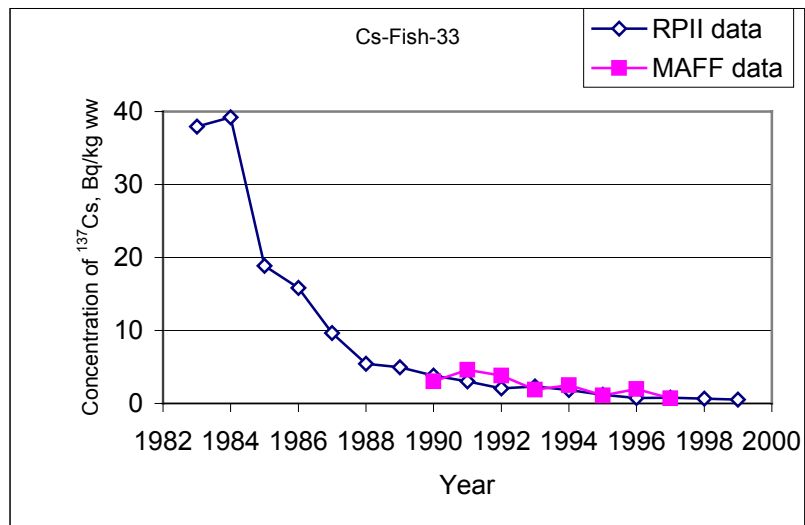
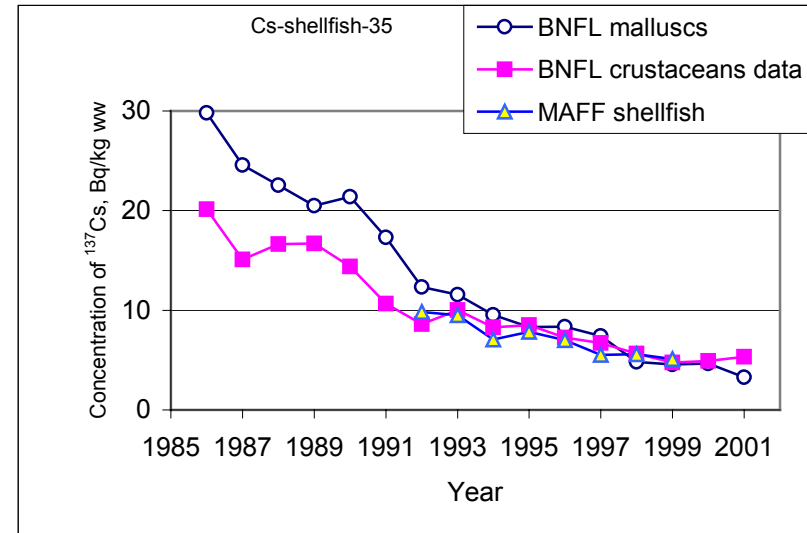
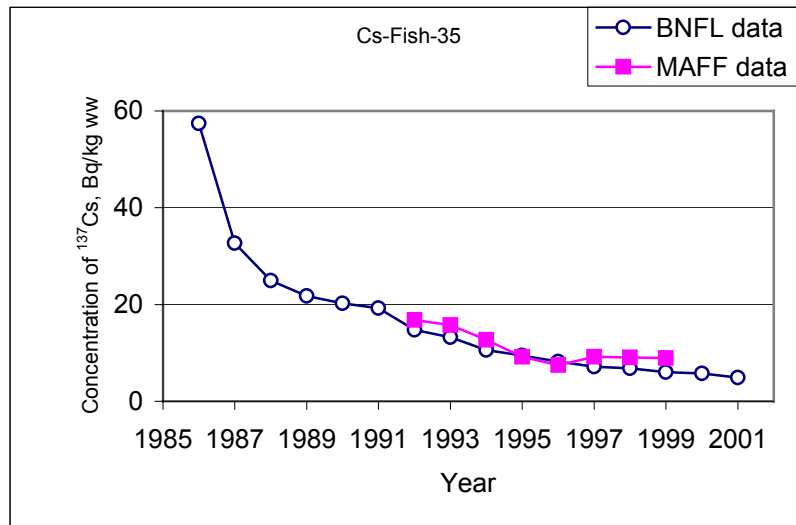


Figure 5 (cont'd)



**Figure 6 Comparison of  $^{137}\text{Cs}$  concentrations in biota from different sources**



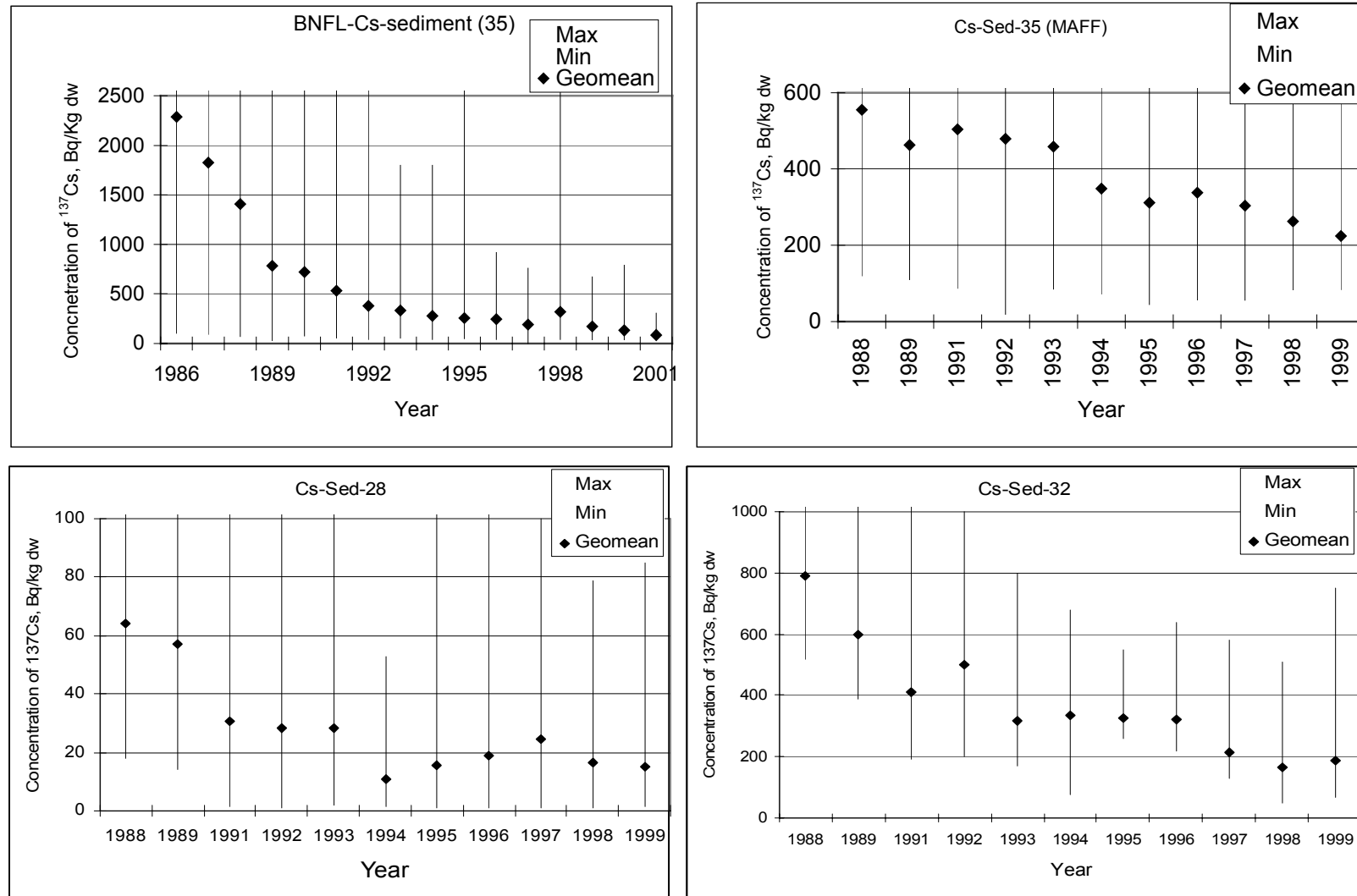
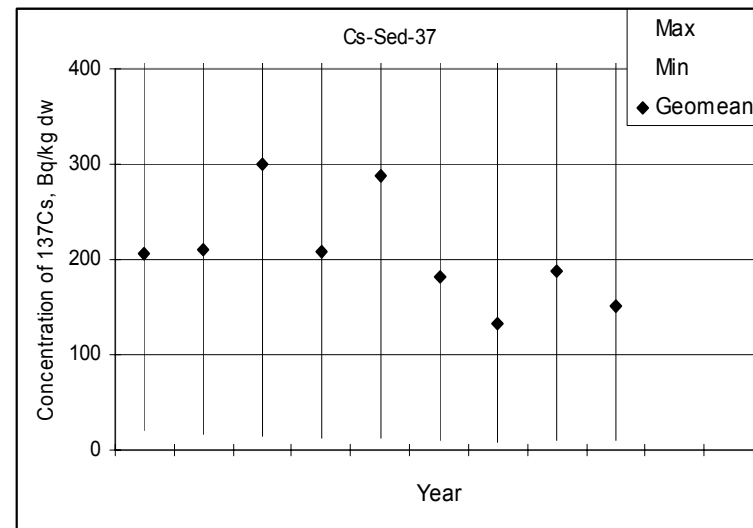
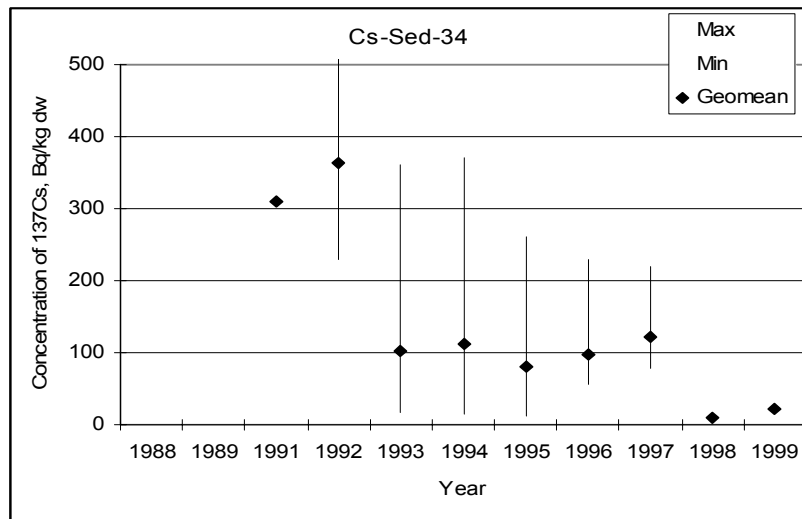
**Figure 7** Temporal variation of  $^{137}\text{Cs}$  concentration in sediments from the Irish Sea



Figure 7 (cont'd)



**Figure 8** Temporal variation of  $^{99}\text{Tc}$  concentrations ( $\text{Bq m}^{-3}$ ) in seawater from different locations

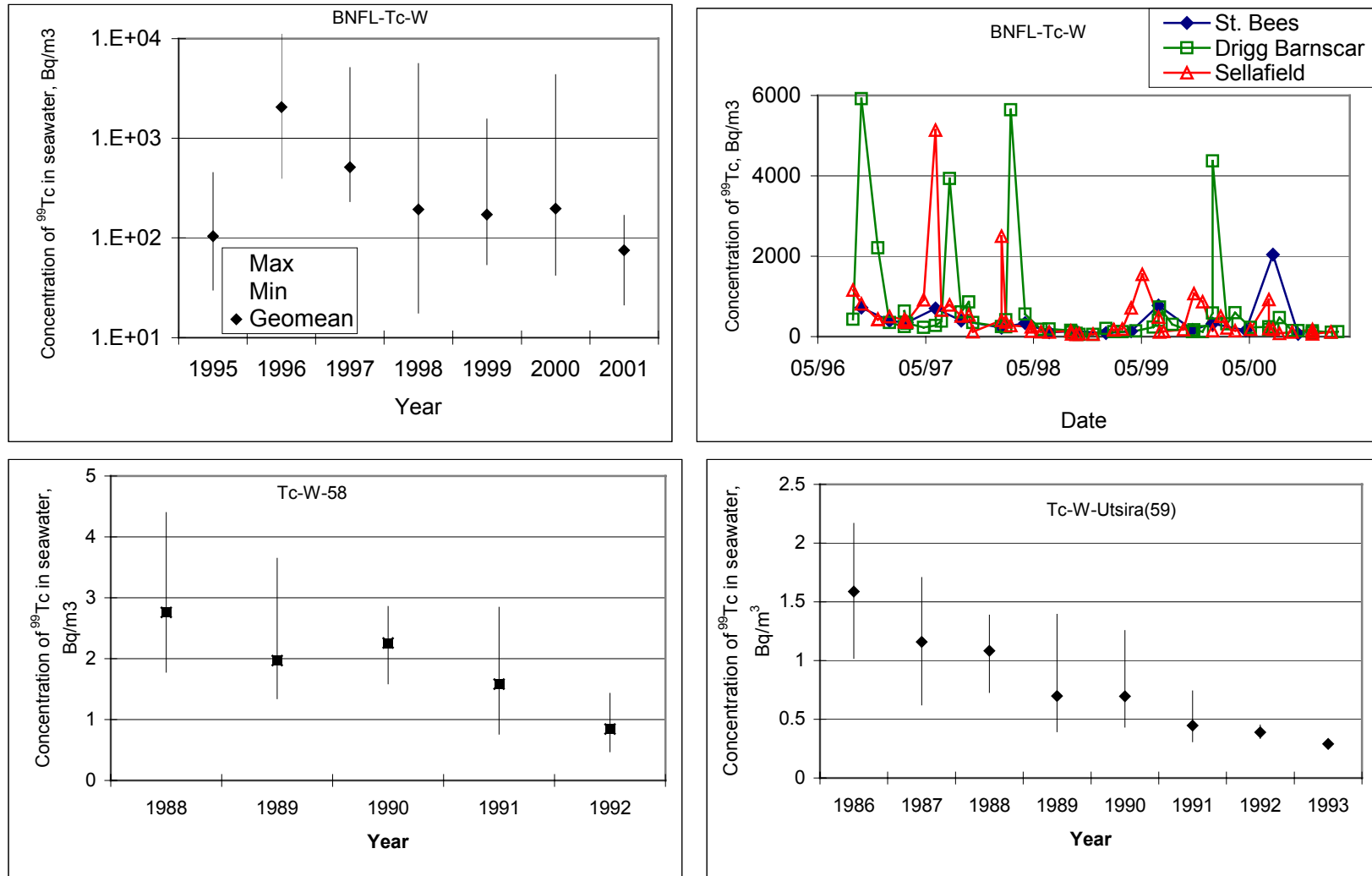


Figure 8 (cont'd)

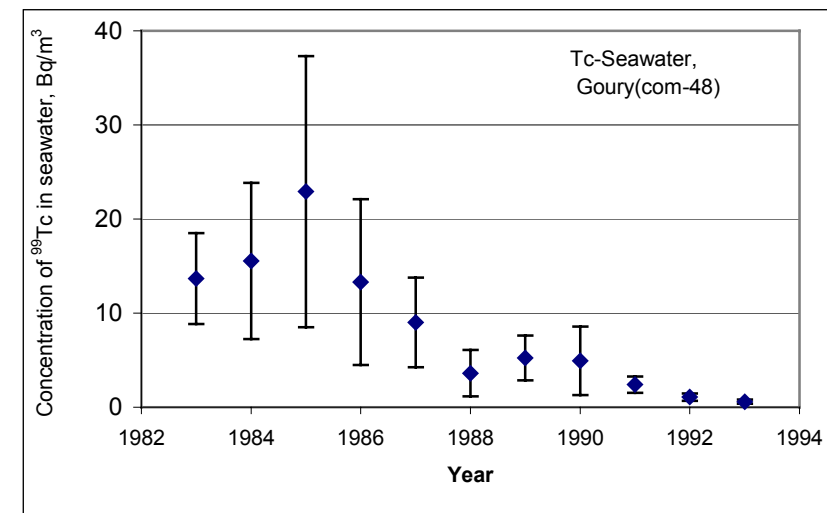
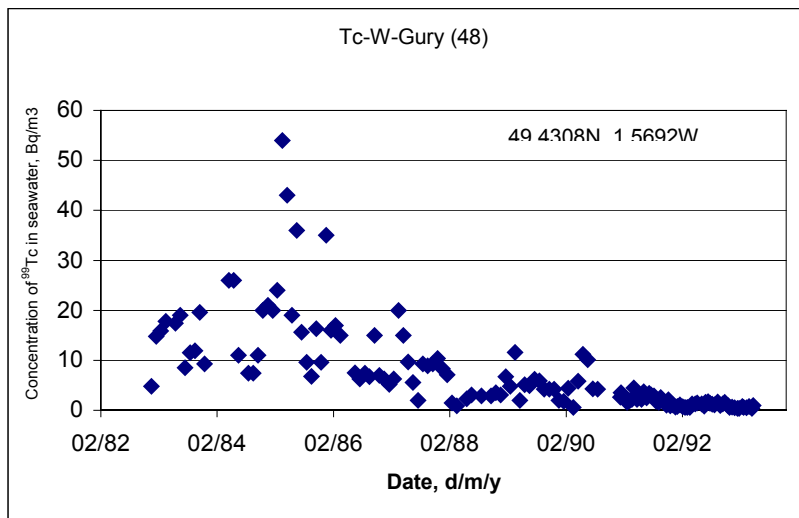
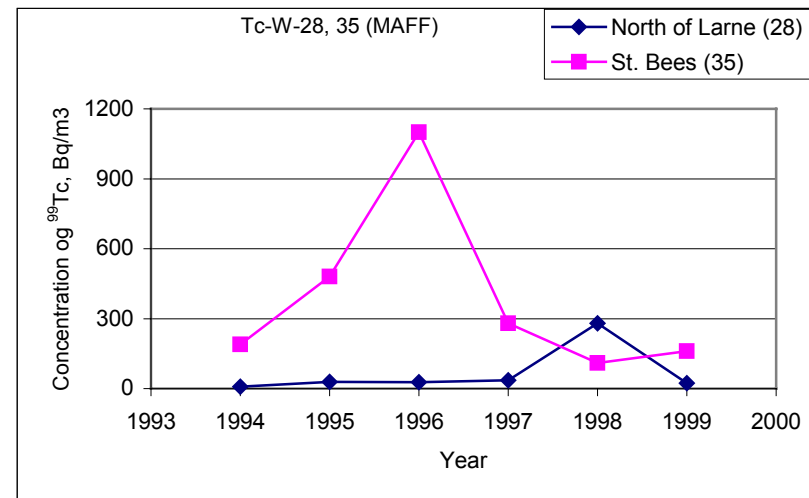
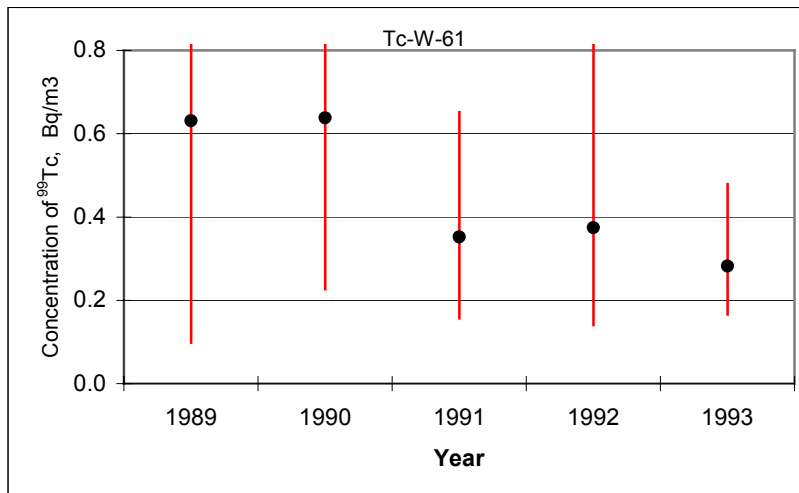
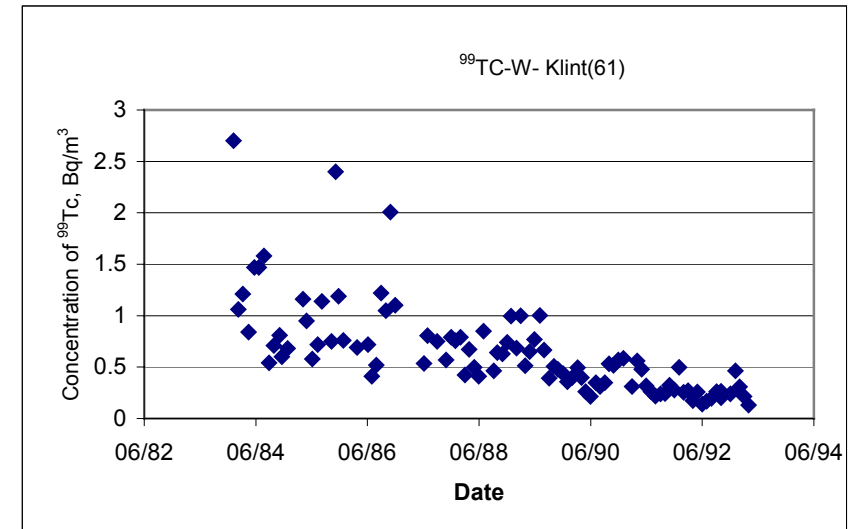
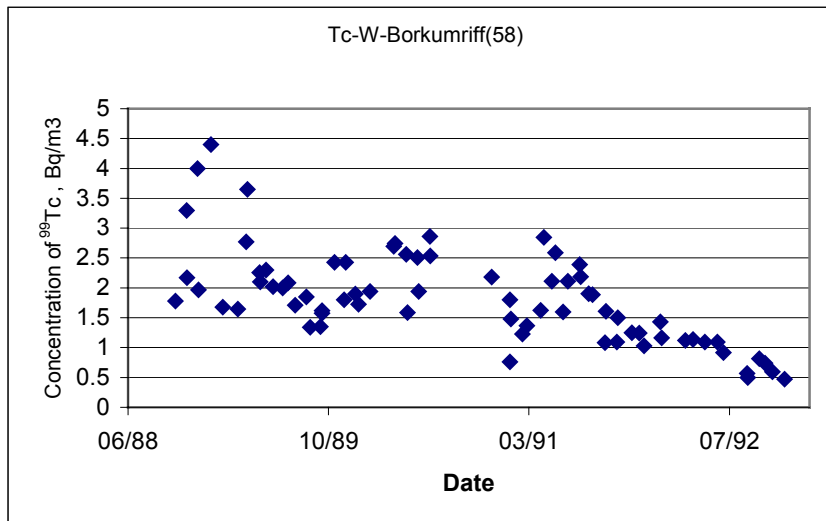
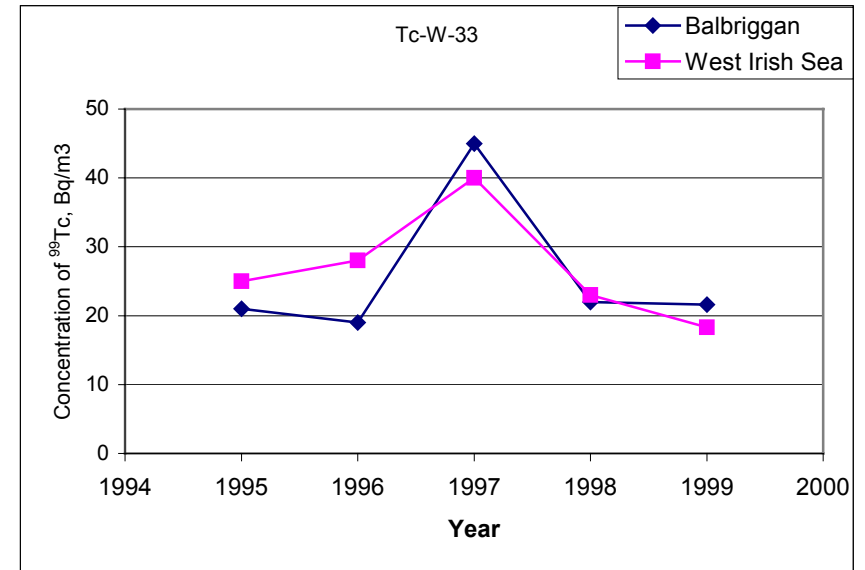
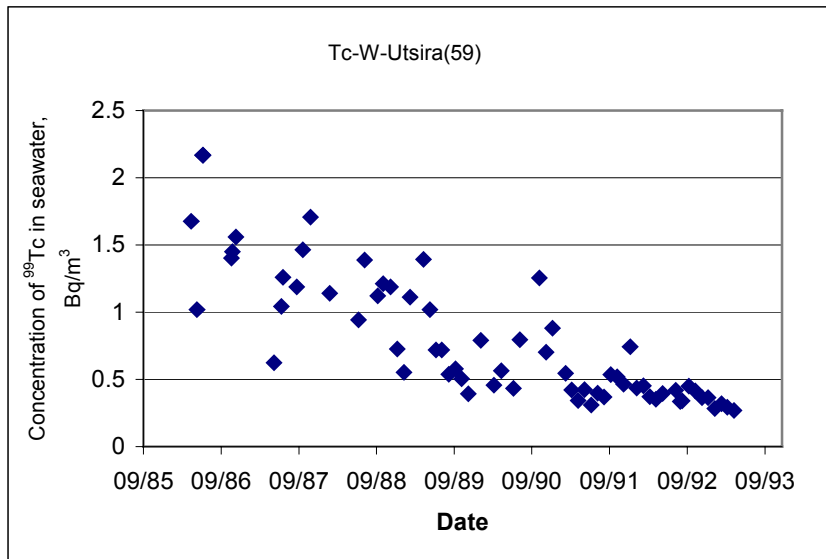
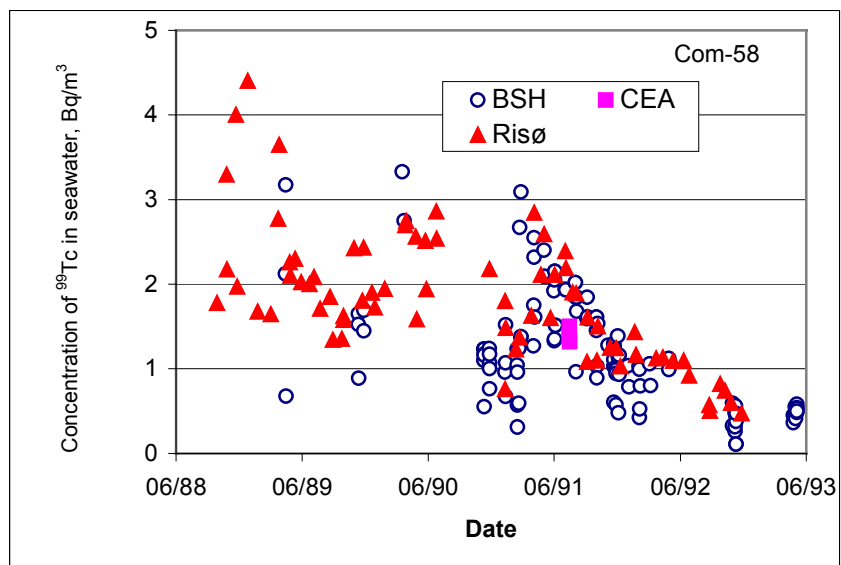


Figure 8 (cont'd)



**Figure 9**      **Comparison of  $^{99}\text{Tc}$  concentrations in seawater measured by different laboratories**



**Figure 10** Temporal variation of  $^{99}\text{Tc}$  concentrations in fish and shellfish (Bq kg<sup>-1</sup> ww) from different locations

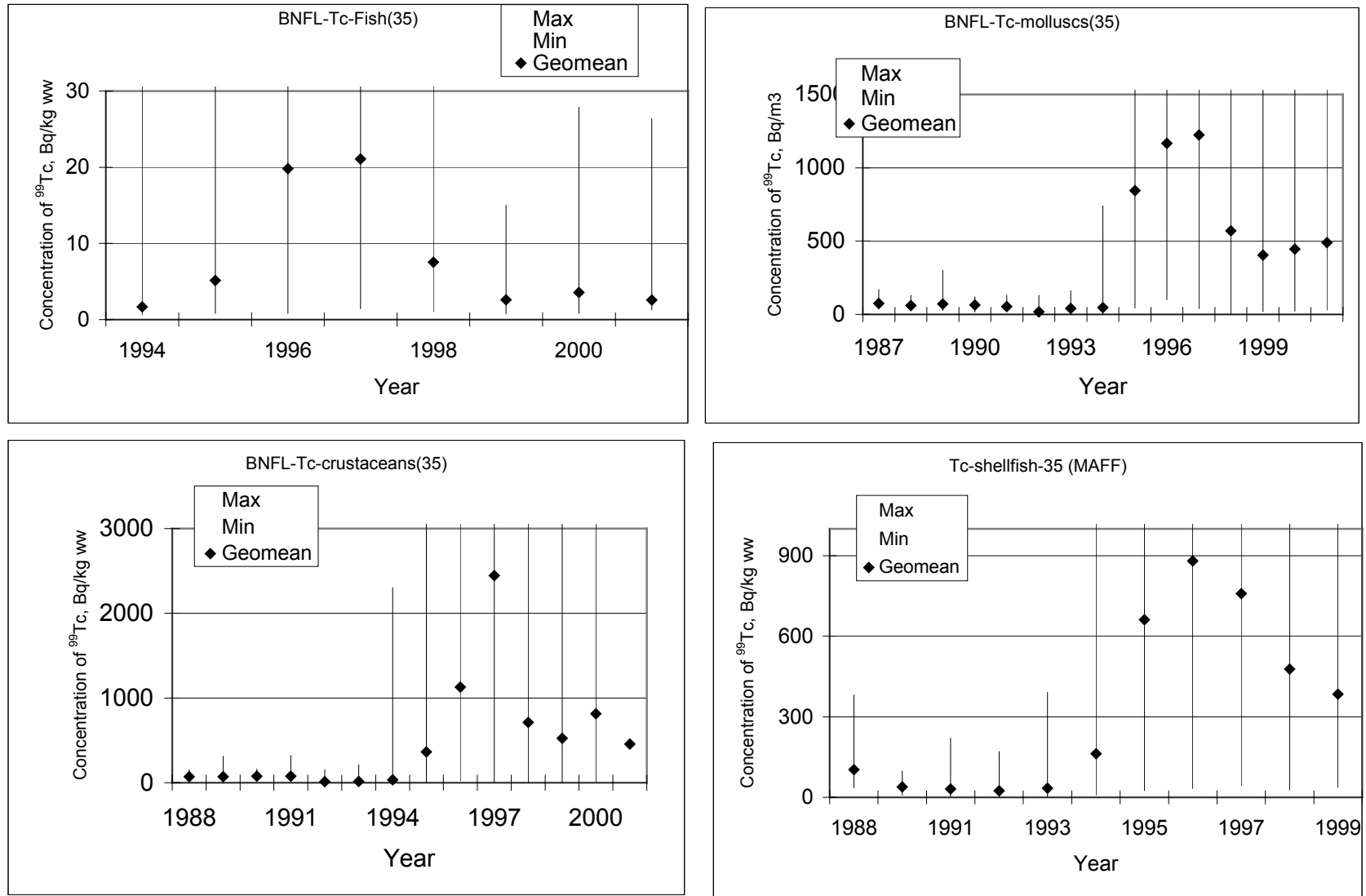


Figure 10 (cont'd)

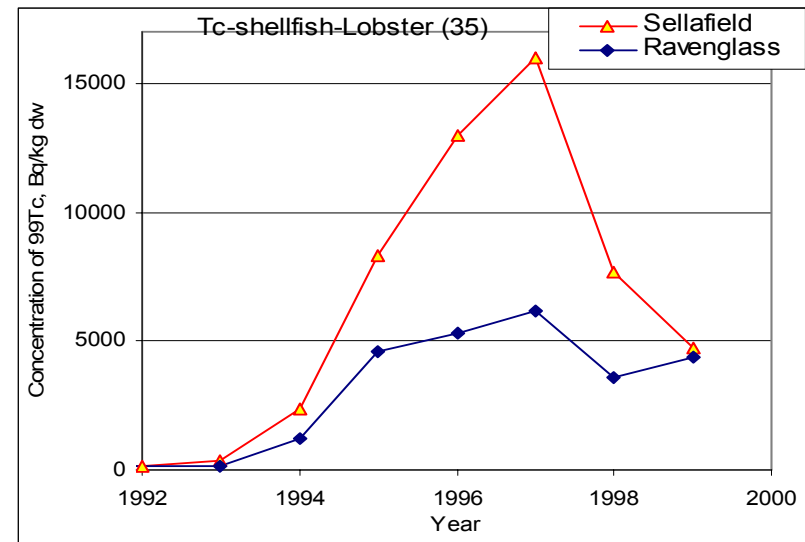
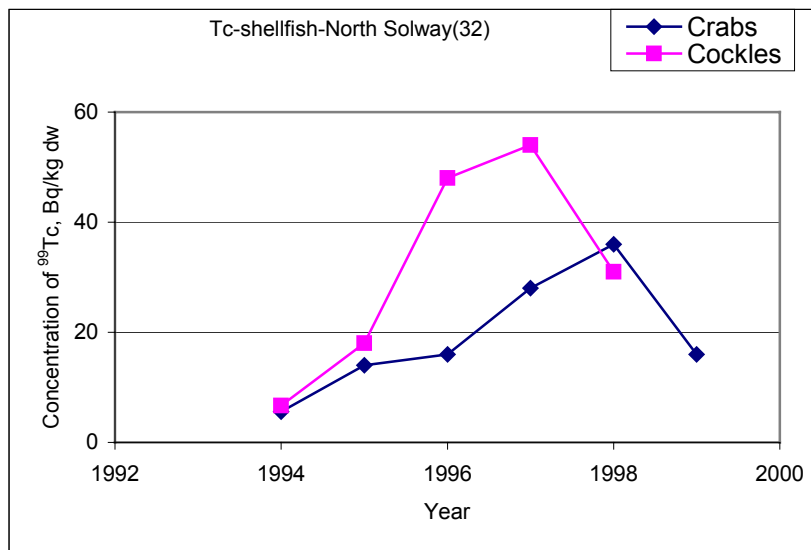
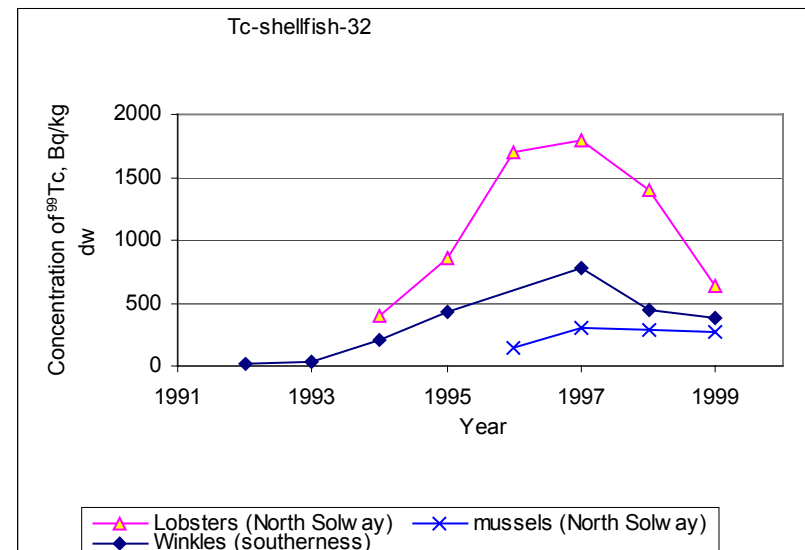
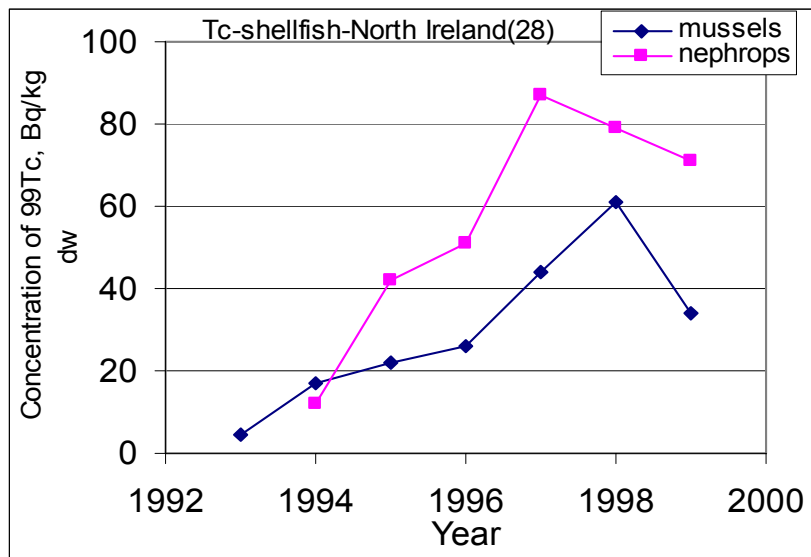
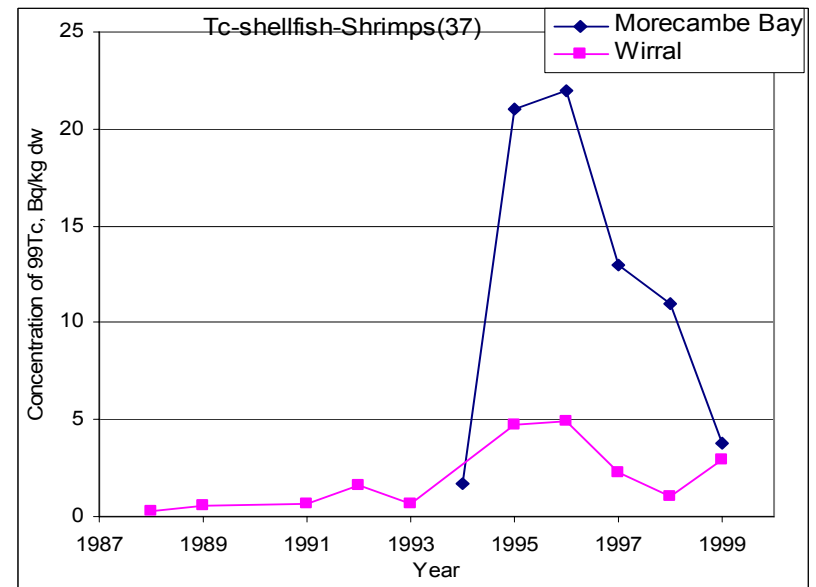
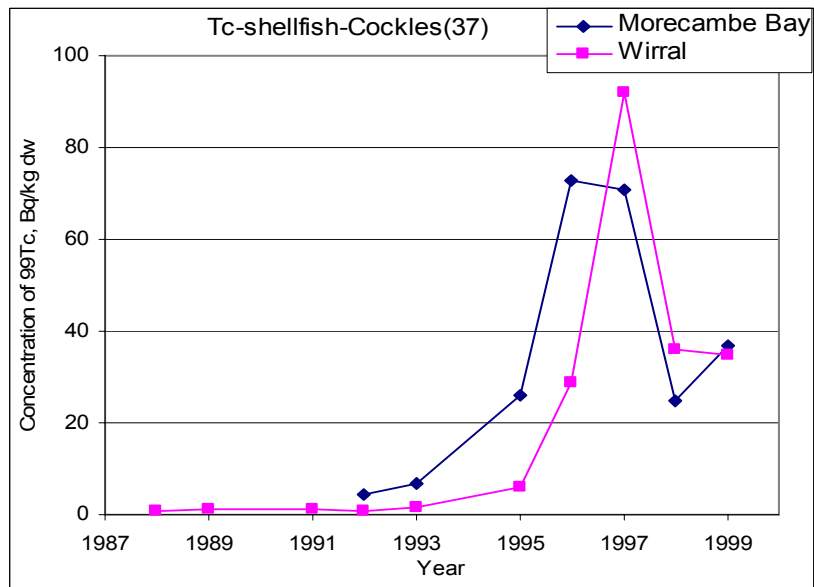
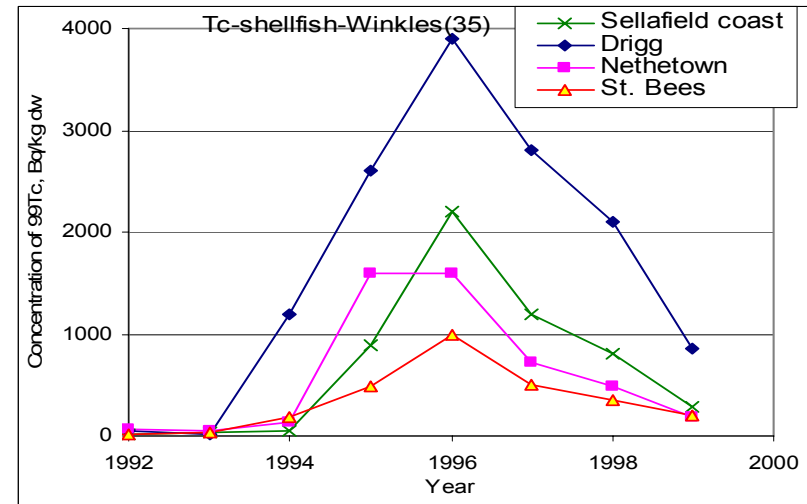
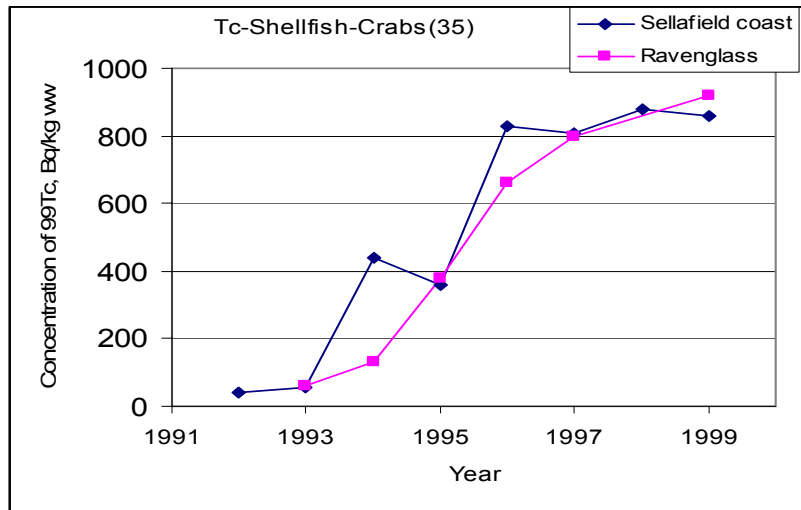


Figure 10 (cont'd)





**Figure 11** Temporal variation of  $^{99}\text{Tc}$  concentrations in seaweed (*Fucus ves.*) from different locations

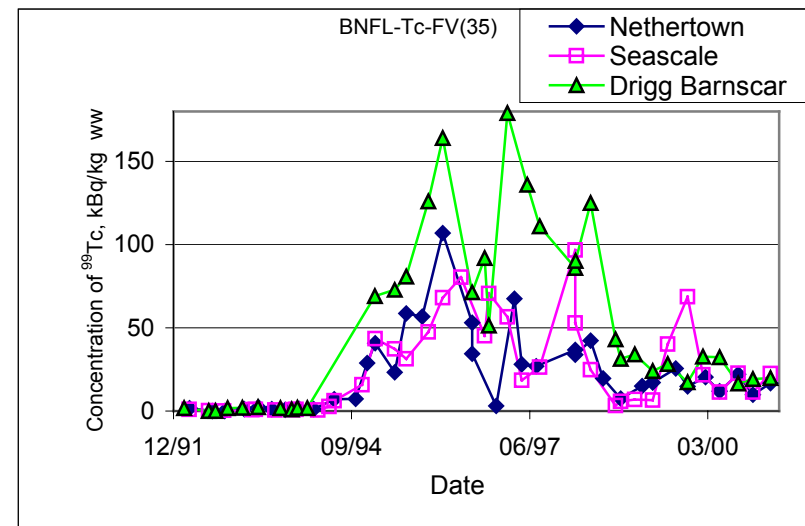
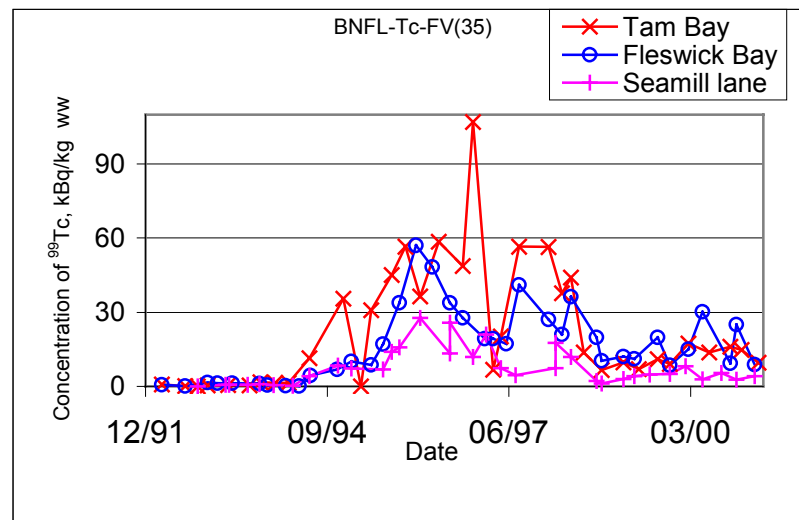
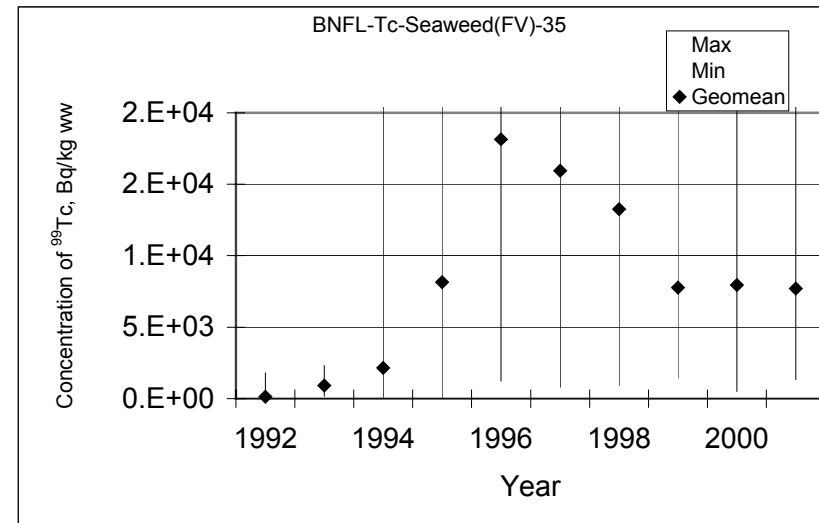
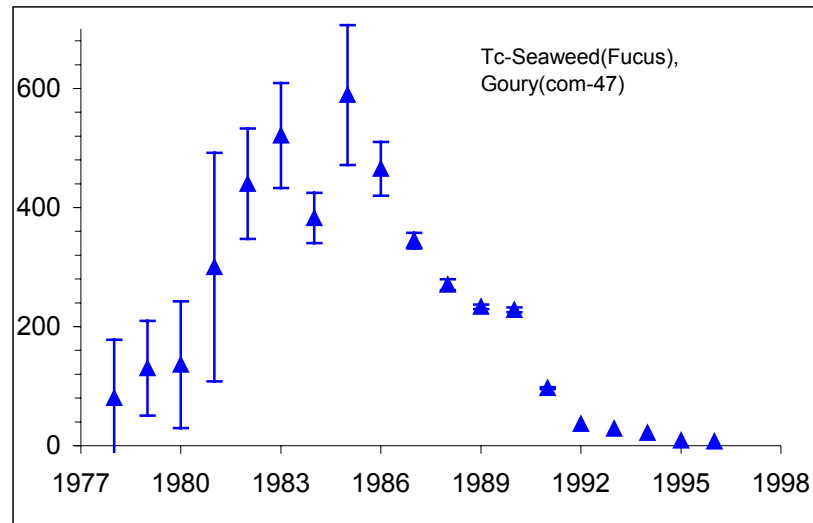


Figure 11 (cont'd)

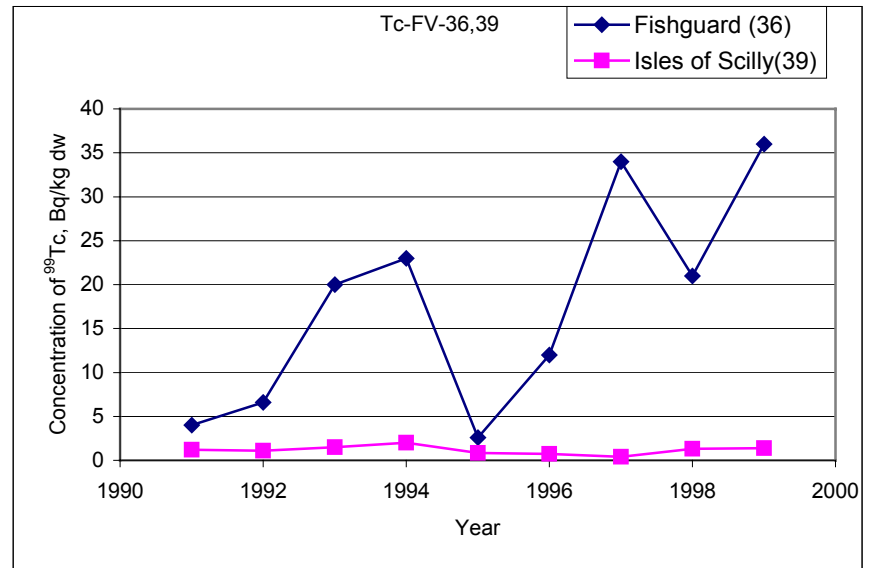
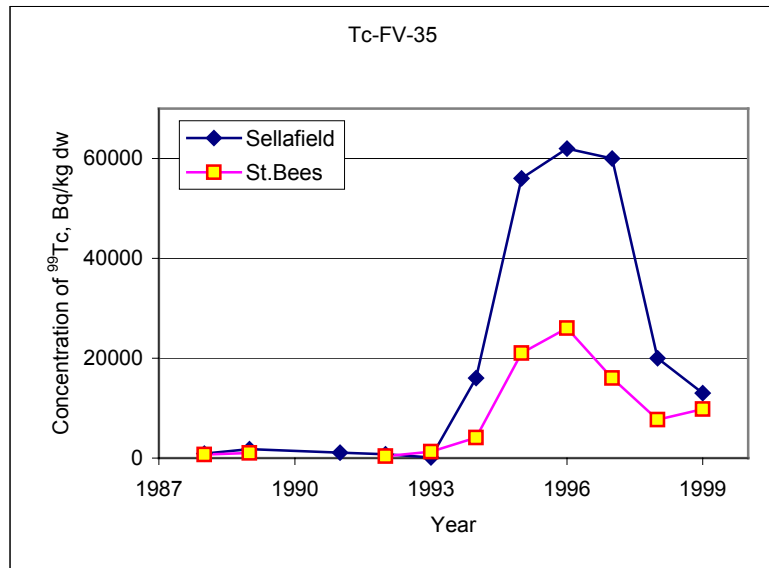
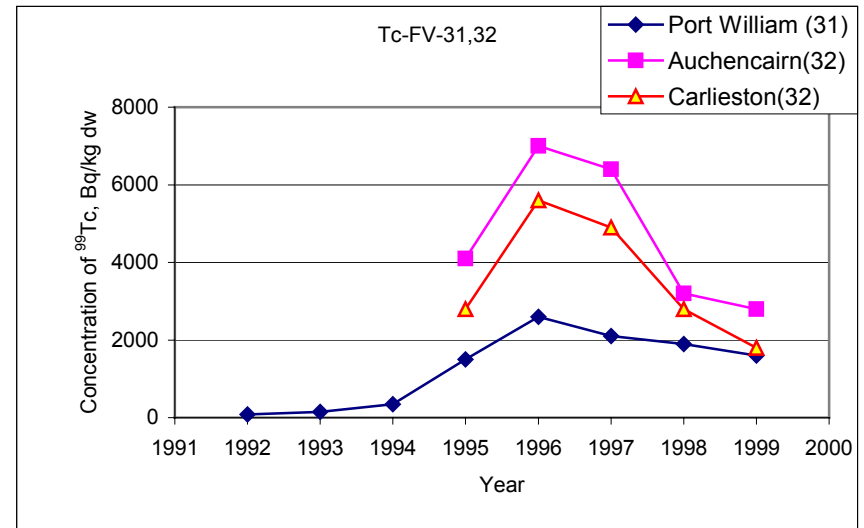
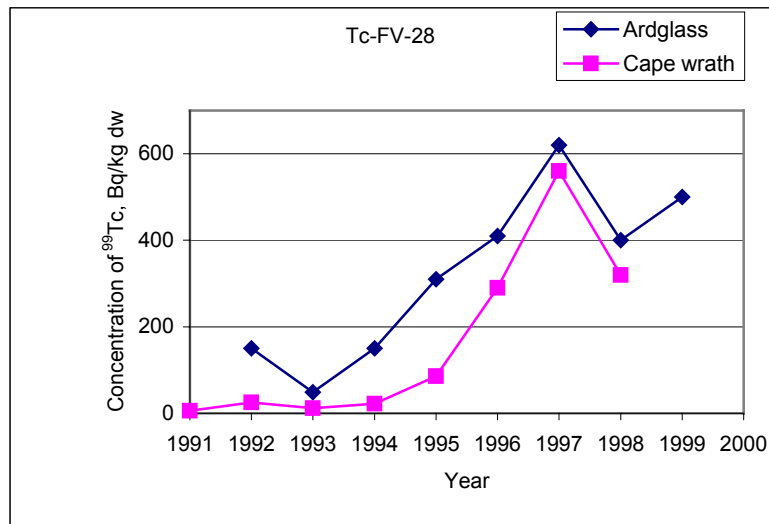


Figure 11 (cont'd)

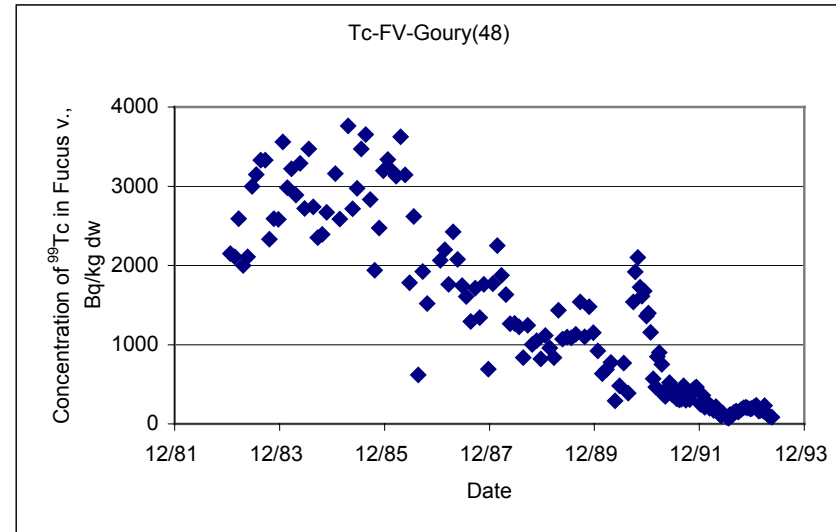
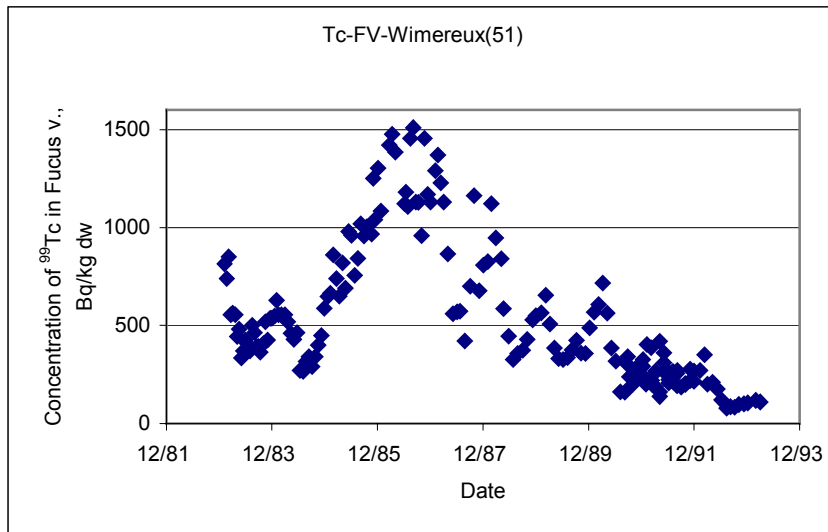
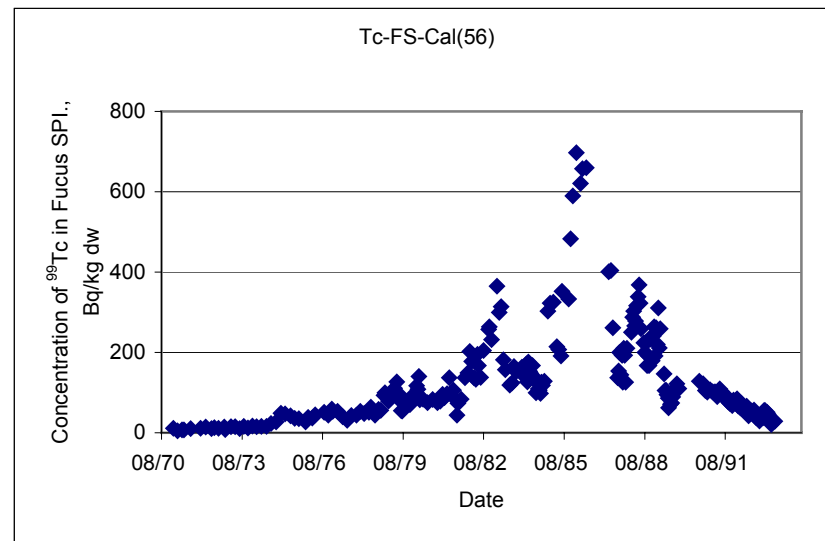
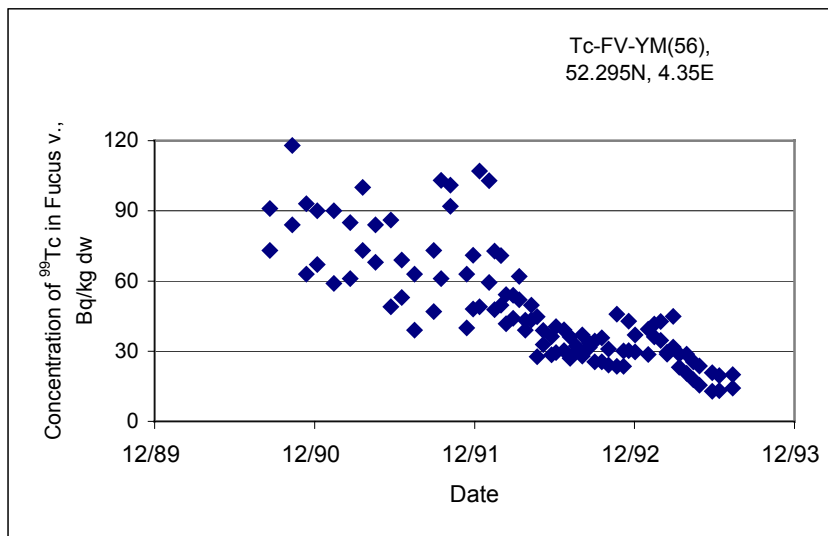


Figure 11 (cont'd)

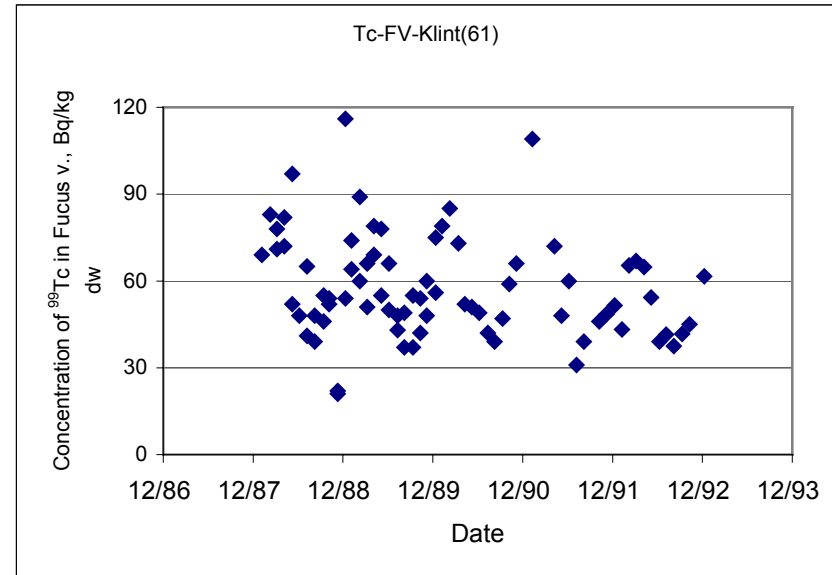
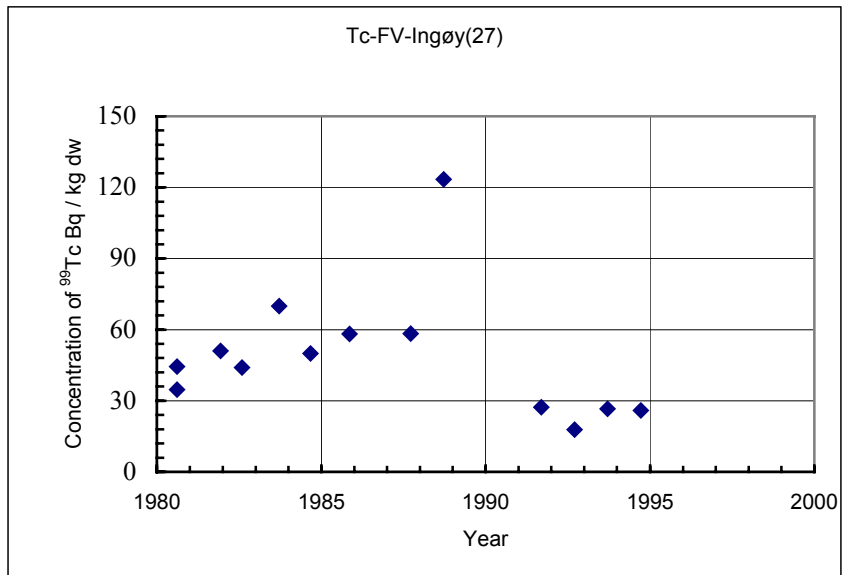
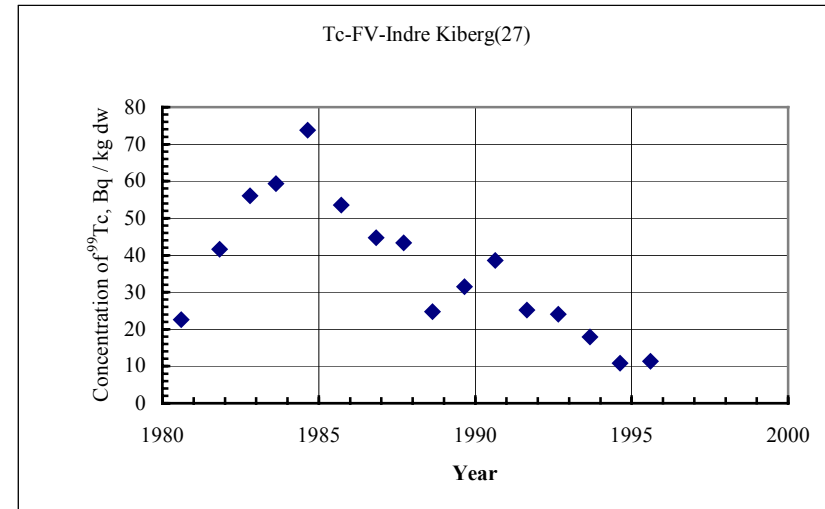
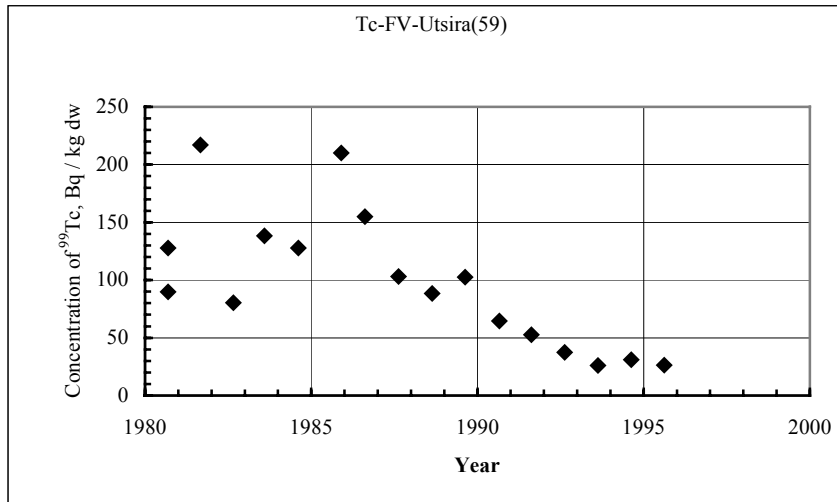
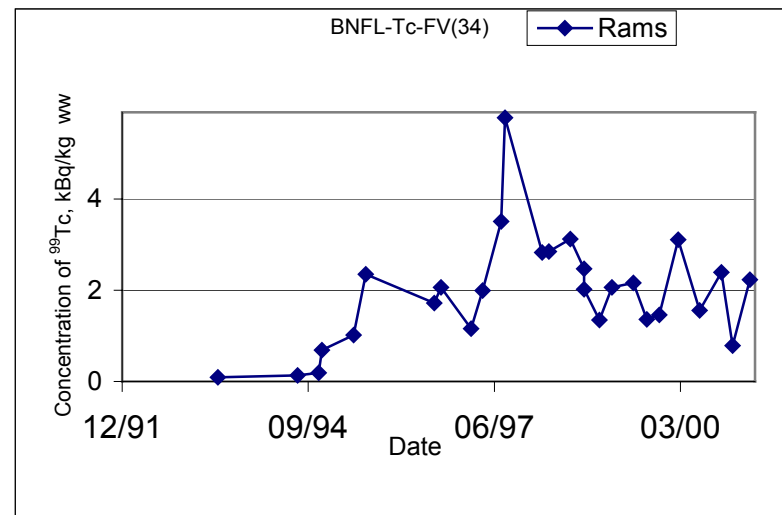
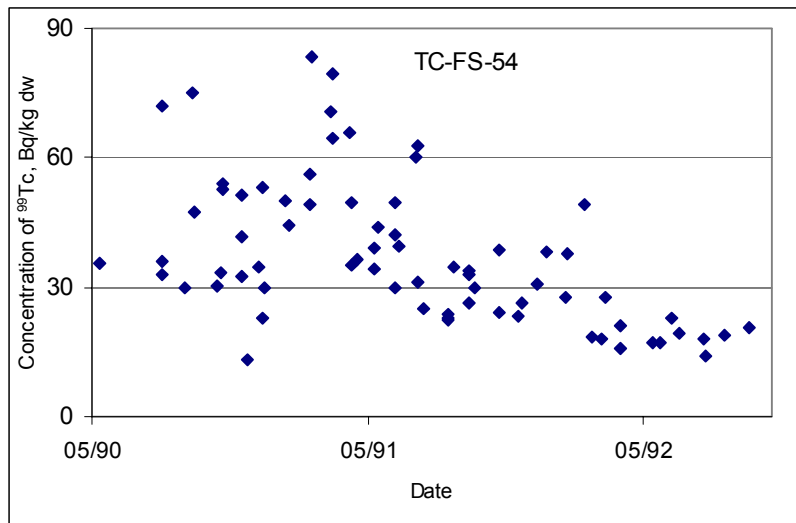
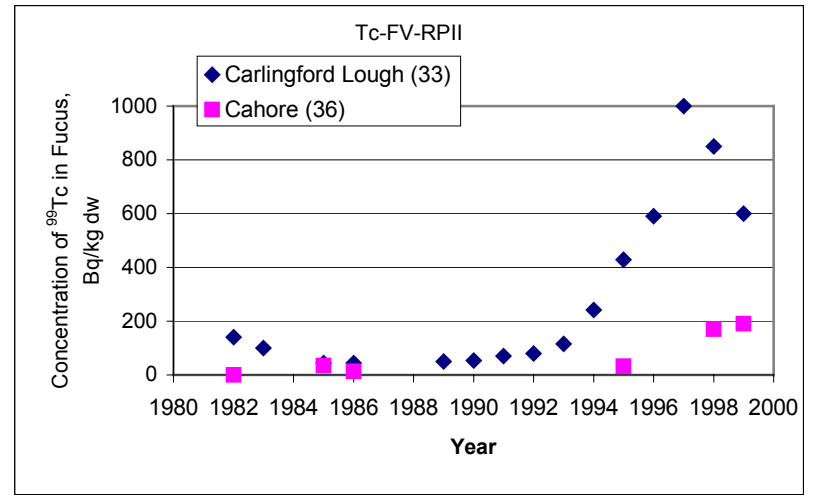
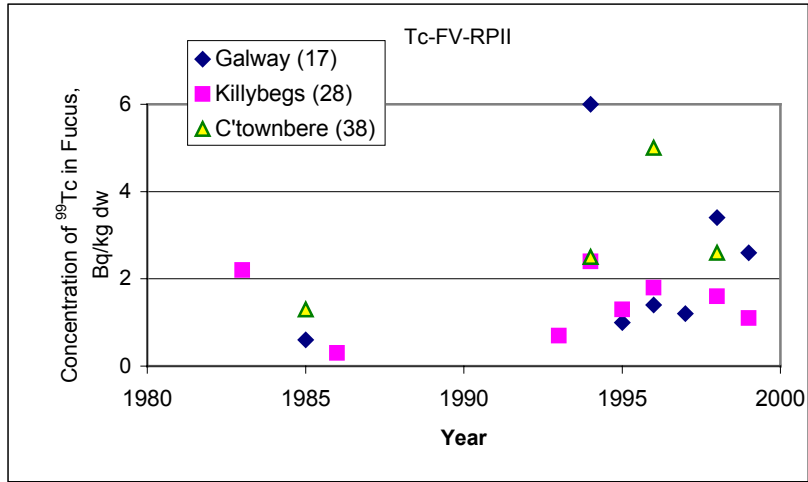


Figure 11 (cont'd)





**Figure 12** Temporal variation of  $^{90}\text{Sr}$  concentrations ( $\text{Bq m}^{-3}$ ) in seawater from different locations and the water column depth profiles

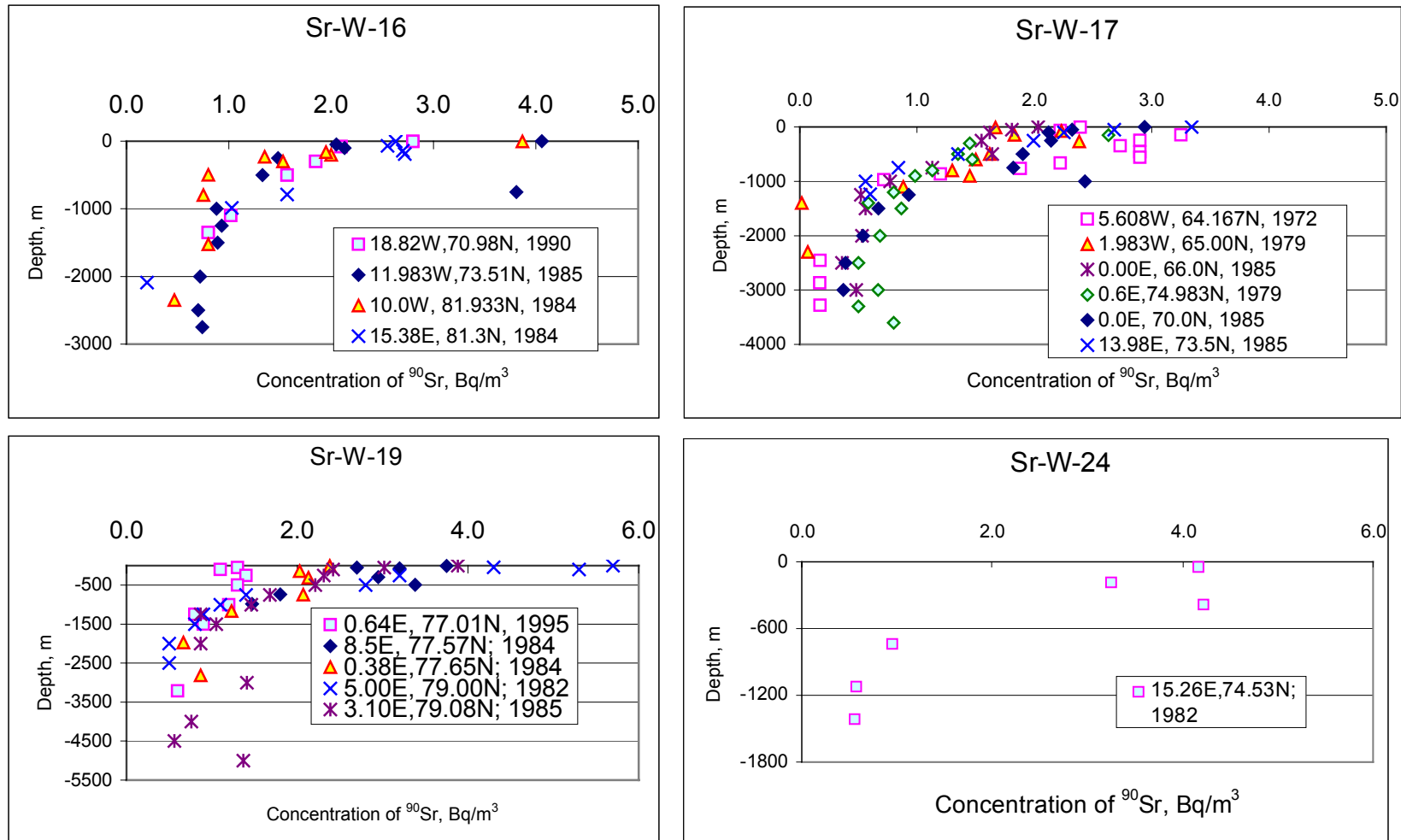


Figure 12 (cont'd)

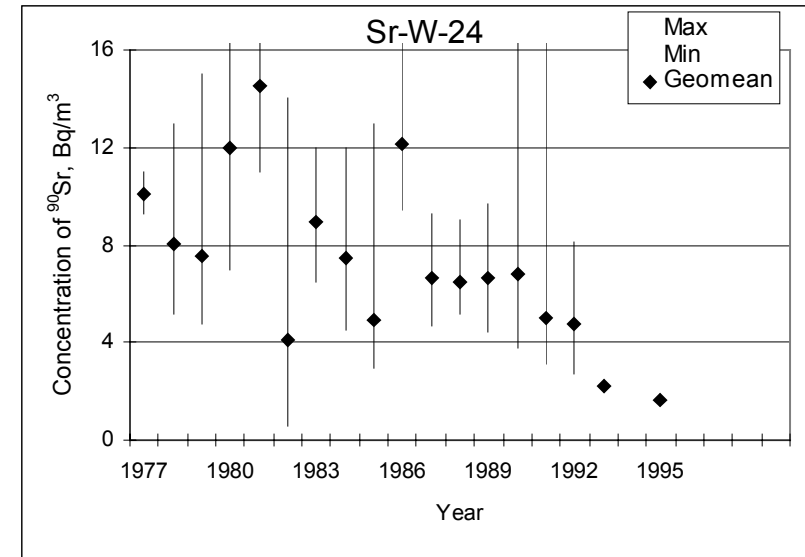
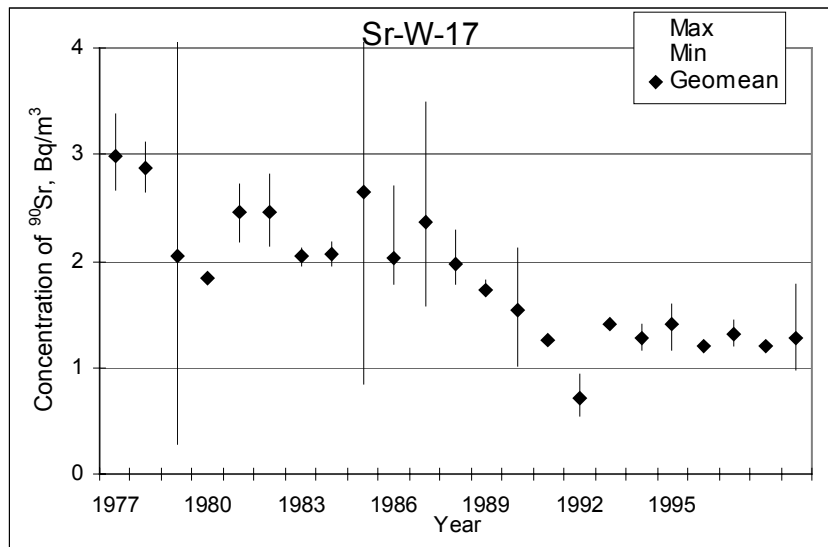
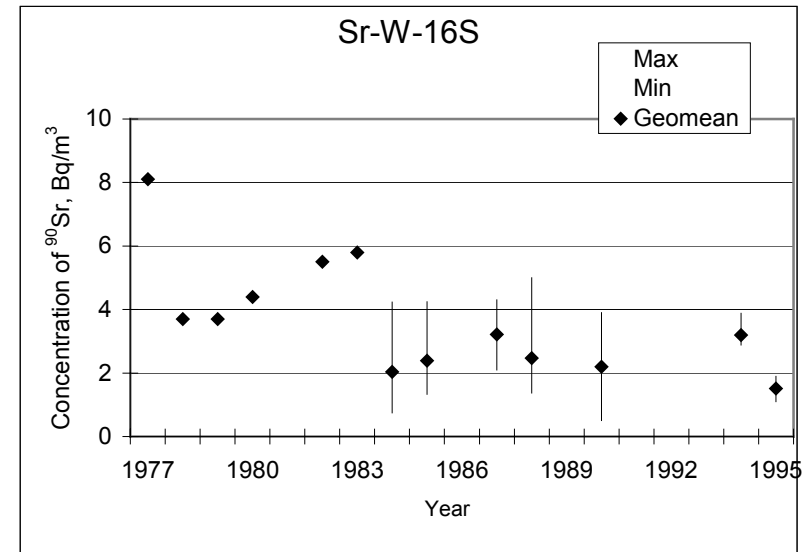
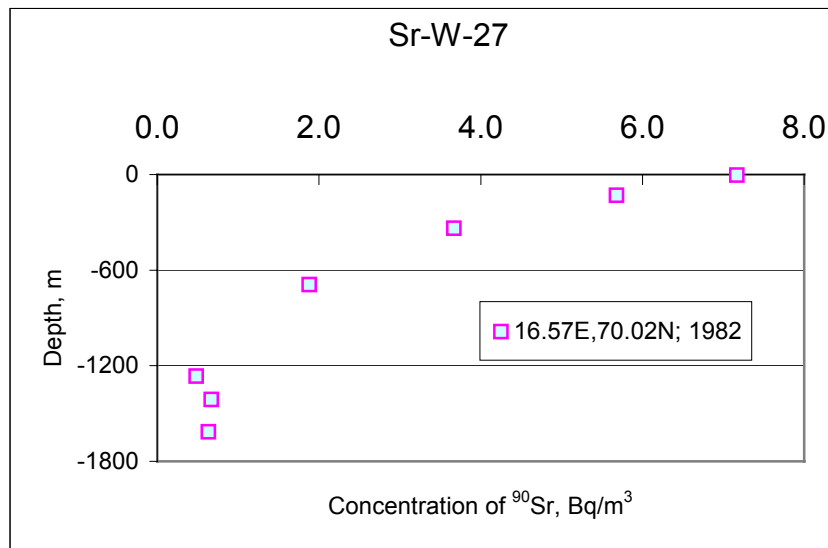




Figure 12 (cont'd)

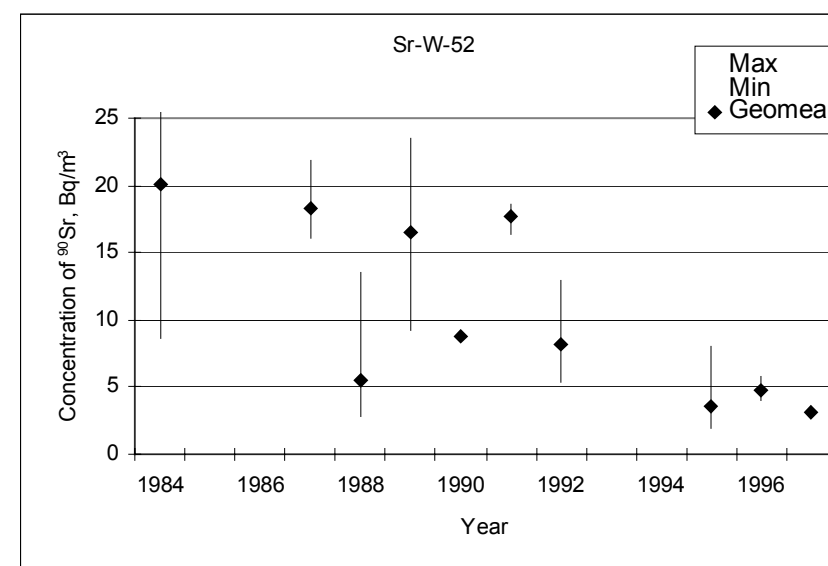
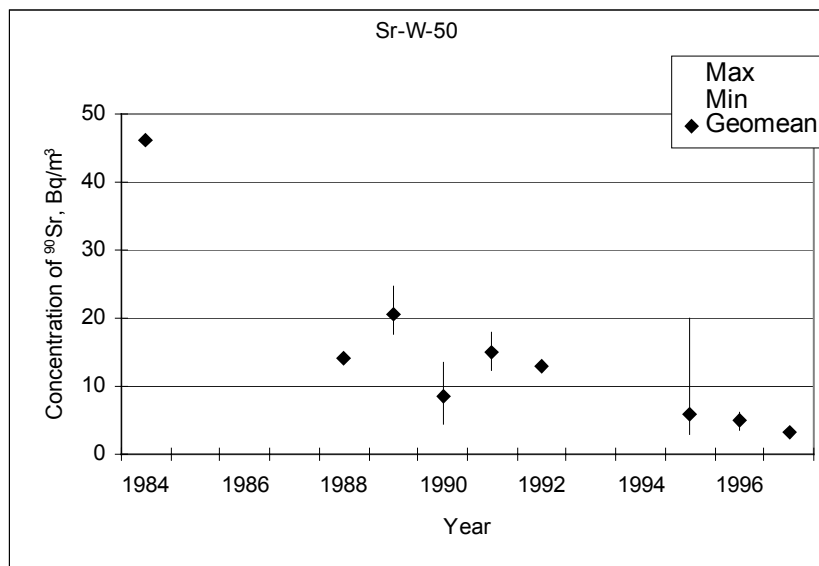
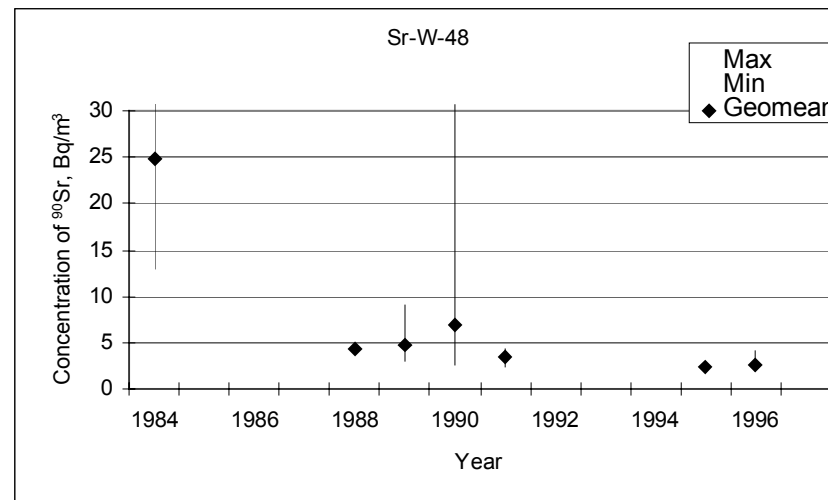
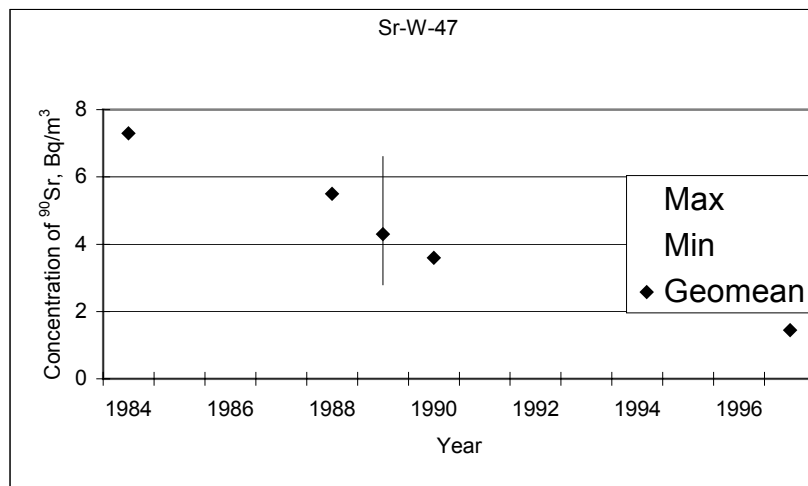


Figure 12 (cont'd)

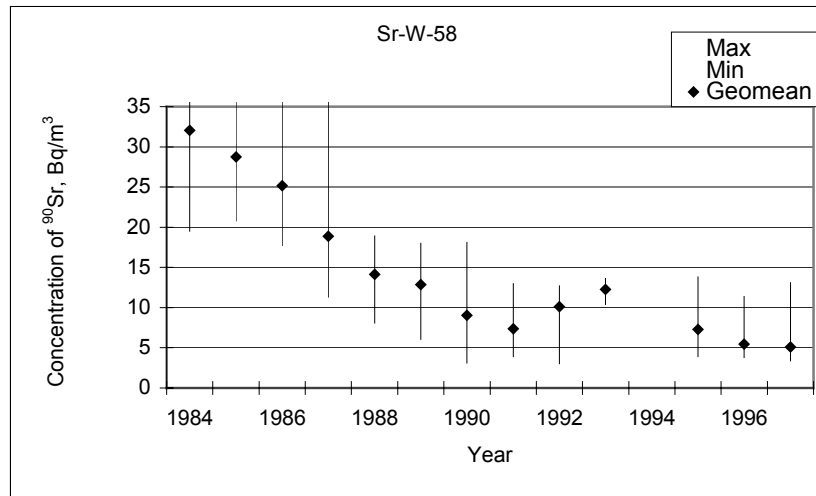
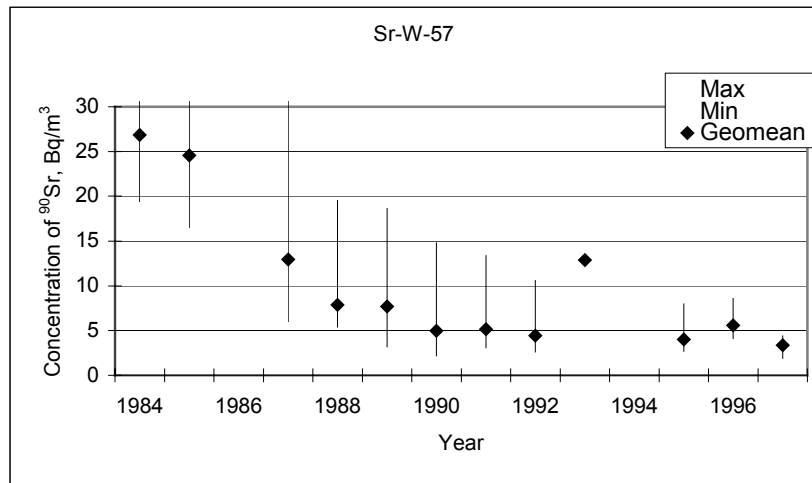
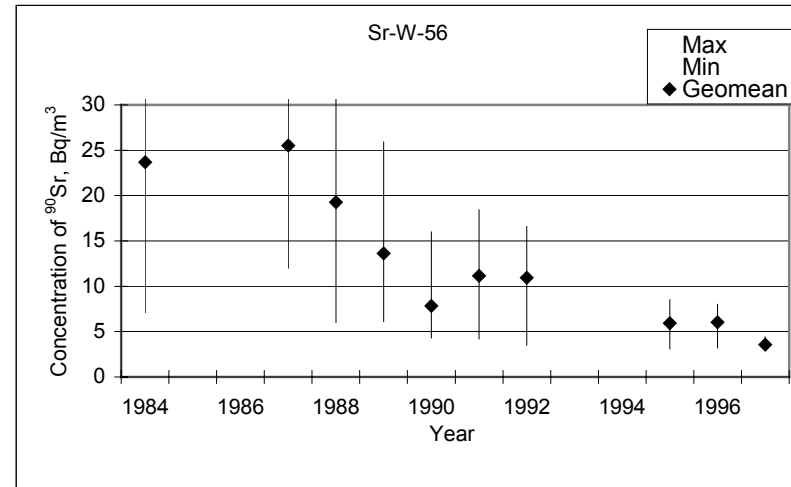
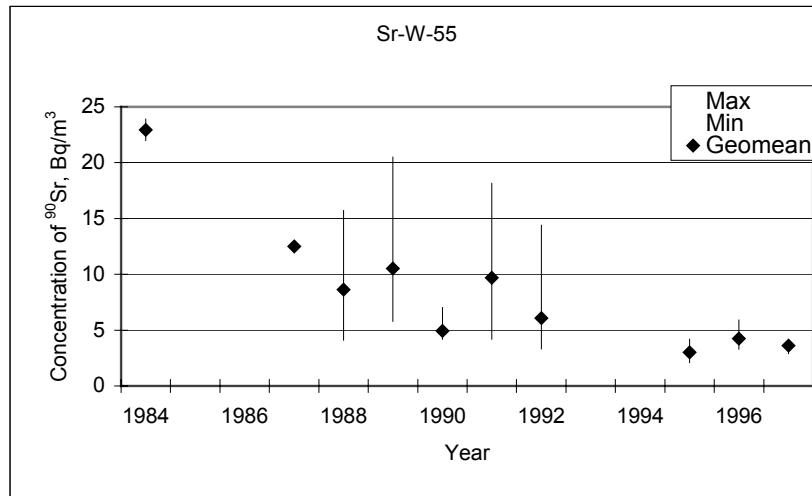
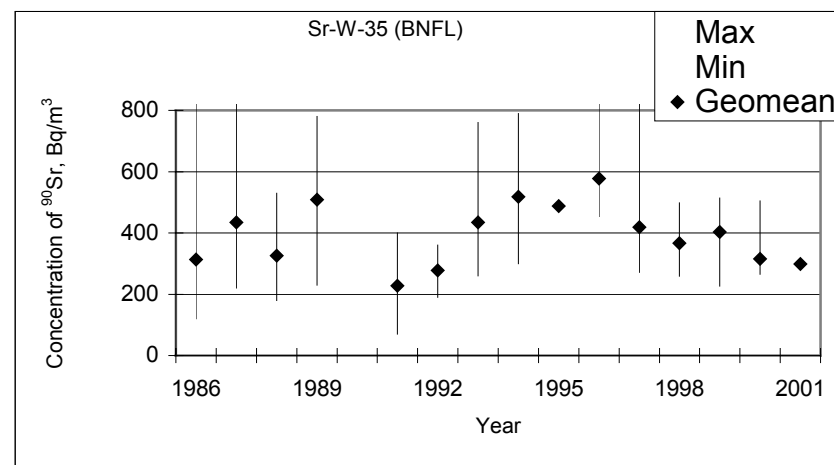
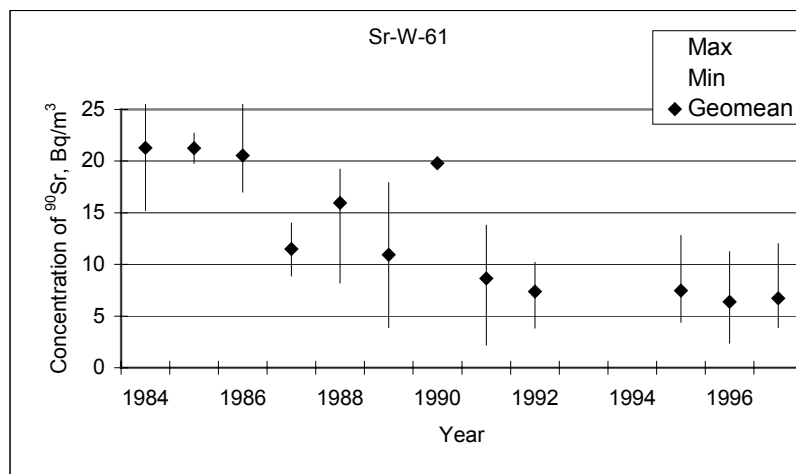
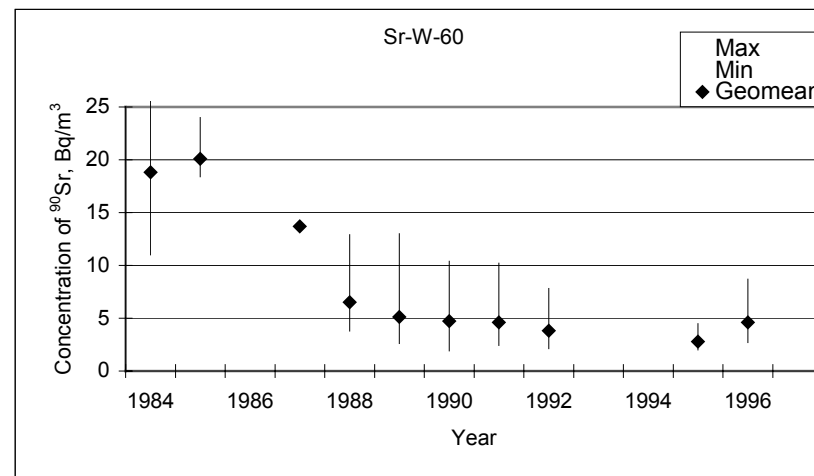
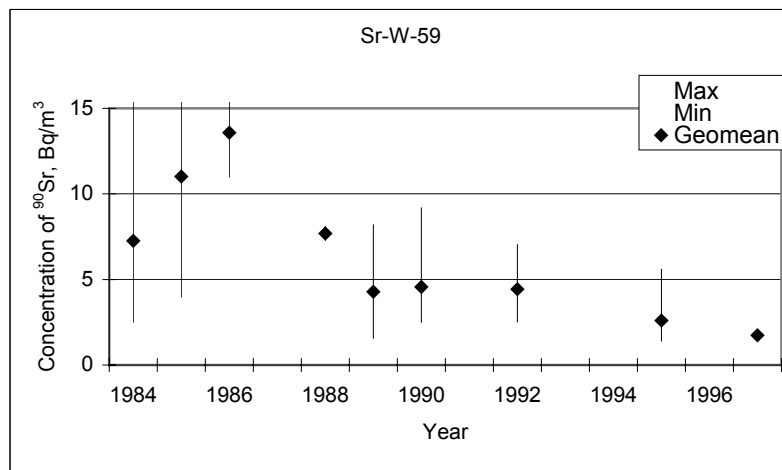


Figure 12 (cont'd)



**Figure 13** Temporal variation of  $^{90}\text{Sr}$  concentrations in fish and shellfish ( $\text{Bq kg}^{-1}$  ww) from different location

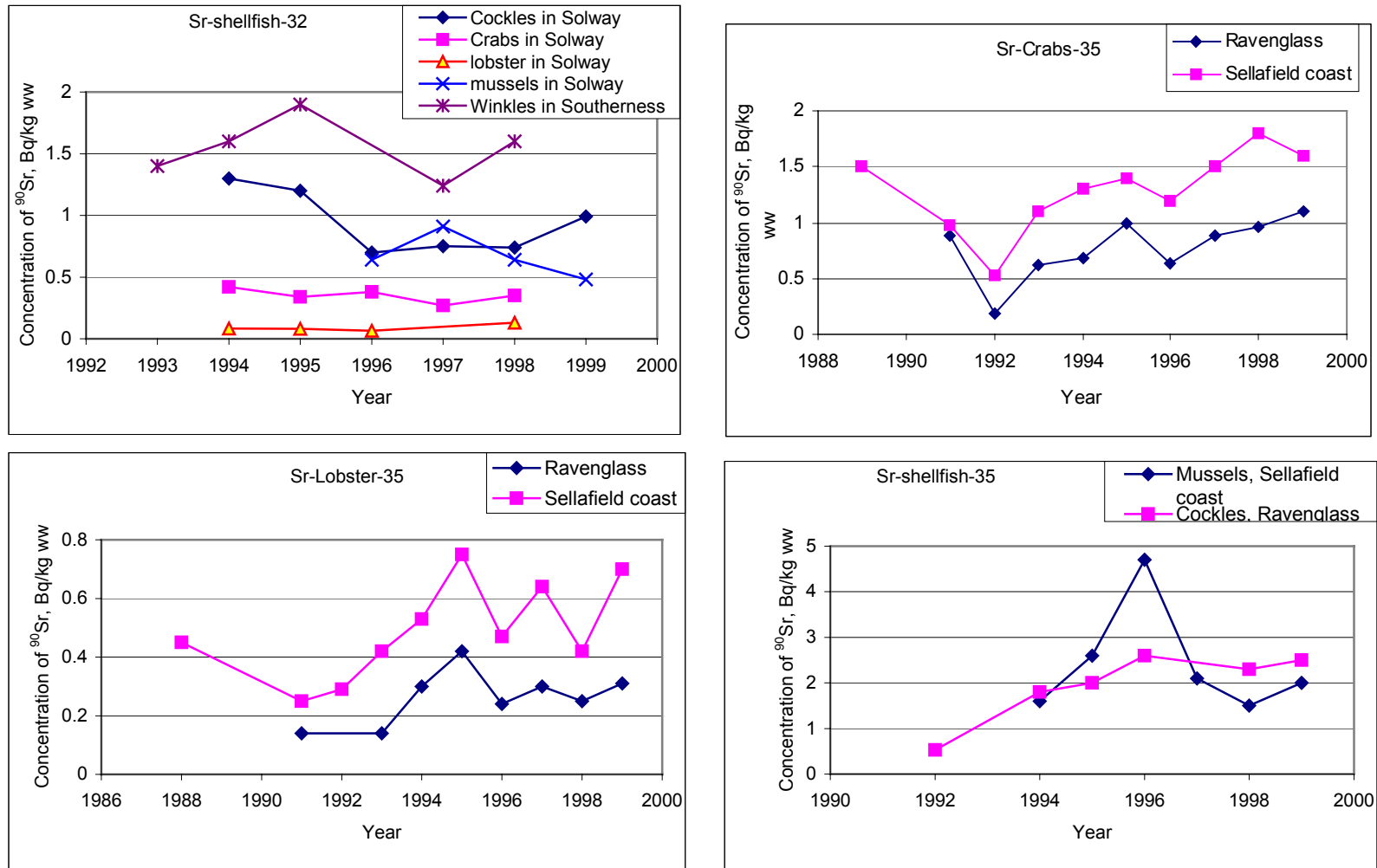


Figure 13 (cont'd)

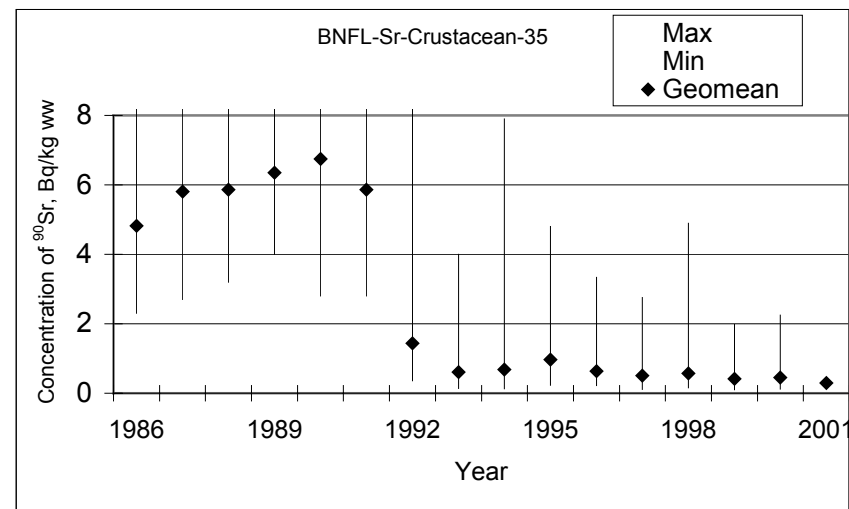
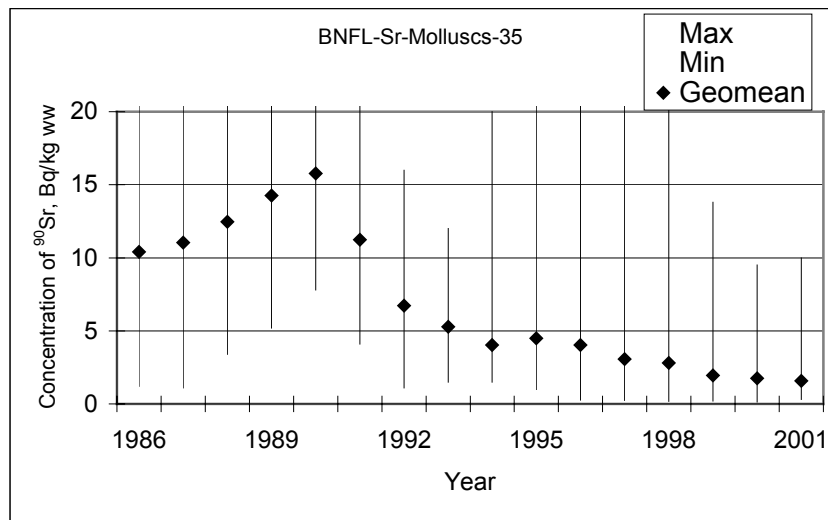
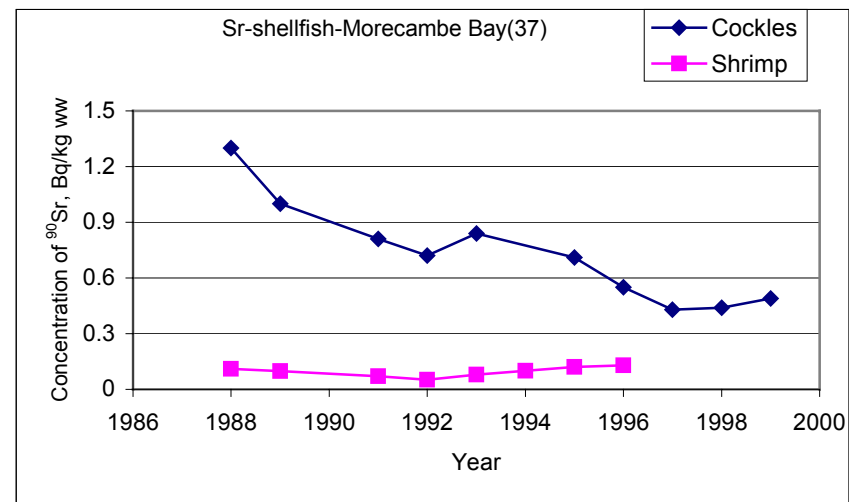
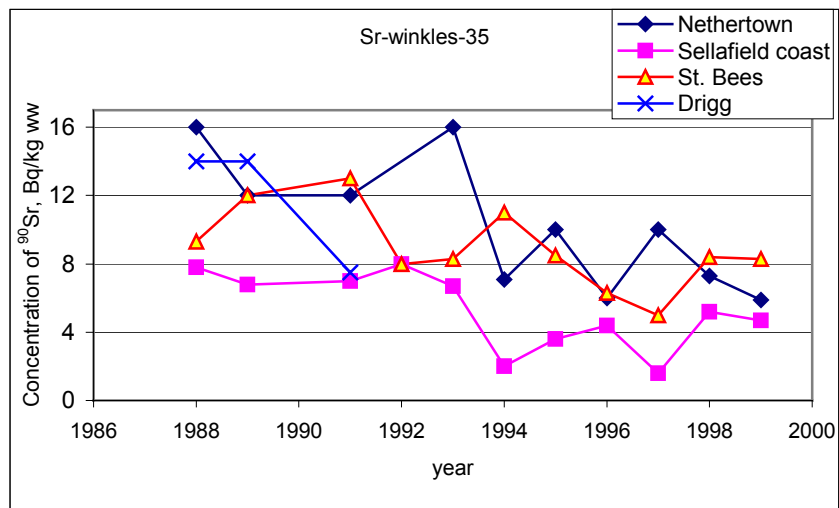


Figure 13 (cont'd)

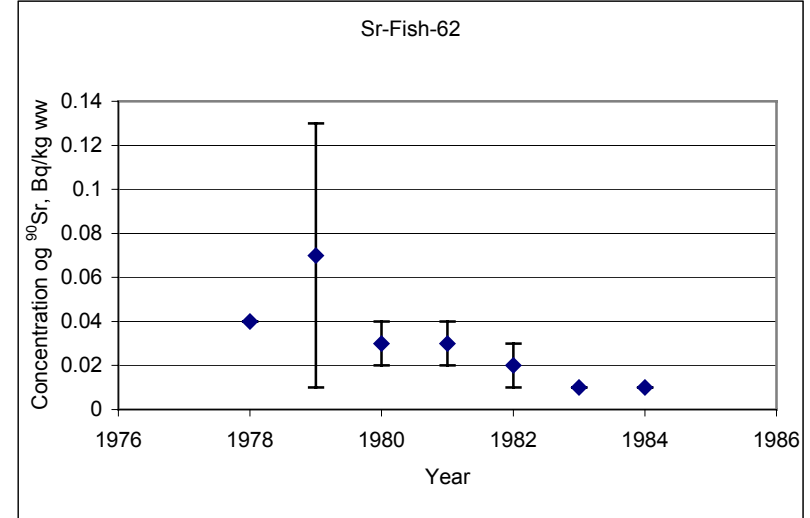
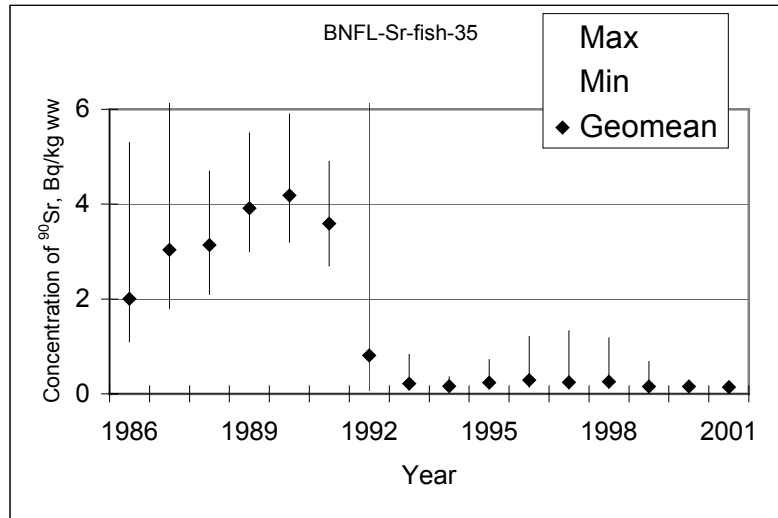


Figure 14 Distribution of dissolved  $^{239+240}\text{Pu}$  in the surface water of the Irish Sea ( $\text{mBq m}^{-3}$ )

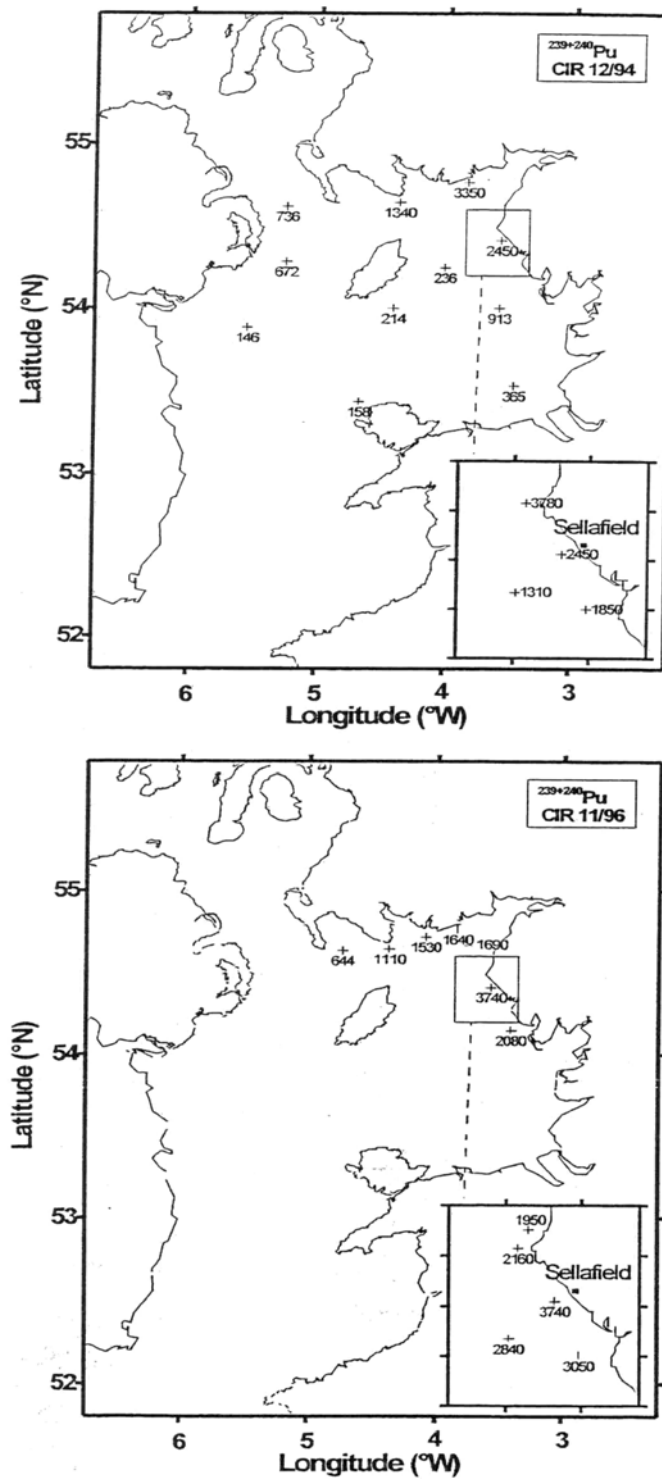
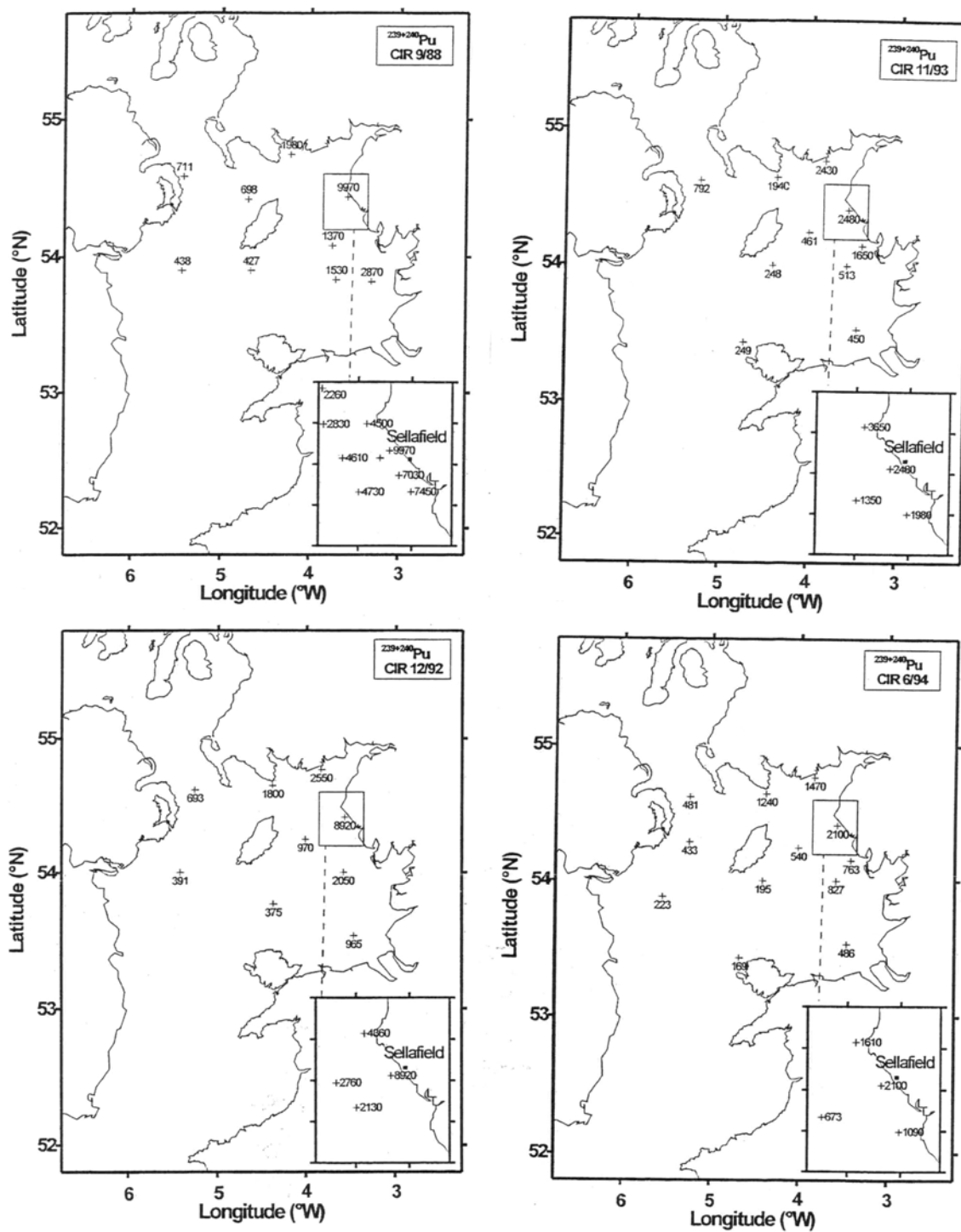


Figure 14 (cont'd)





**Figure 15** Temporal variation of  $^{239+240}\text{Pu}$  concentrations in biota, Bq kg<sup>-1</sup> wet weight

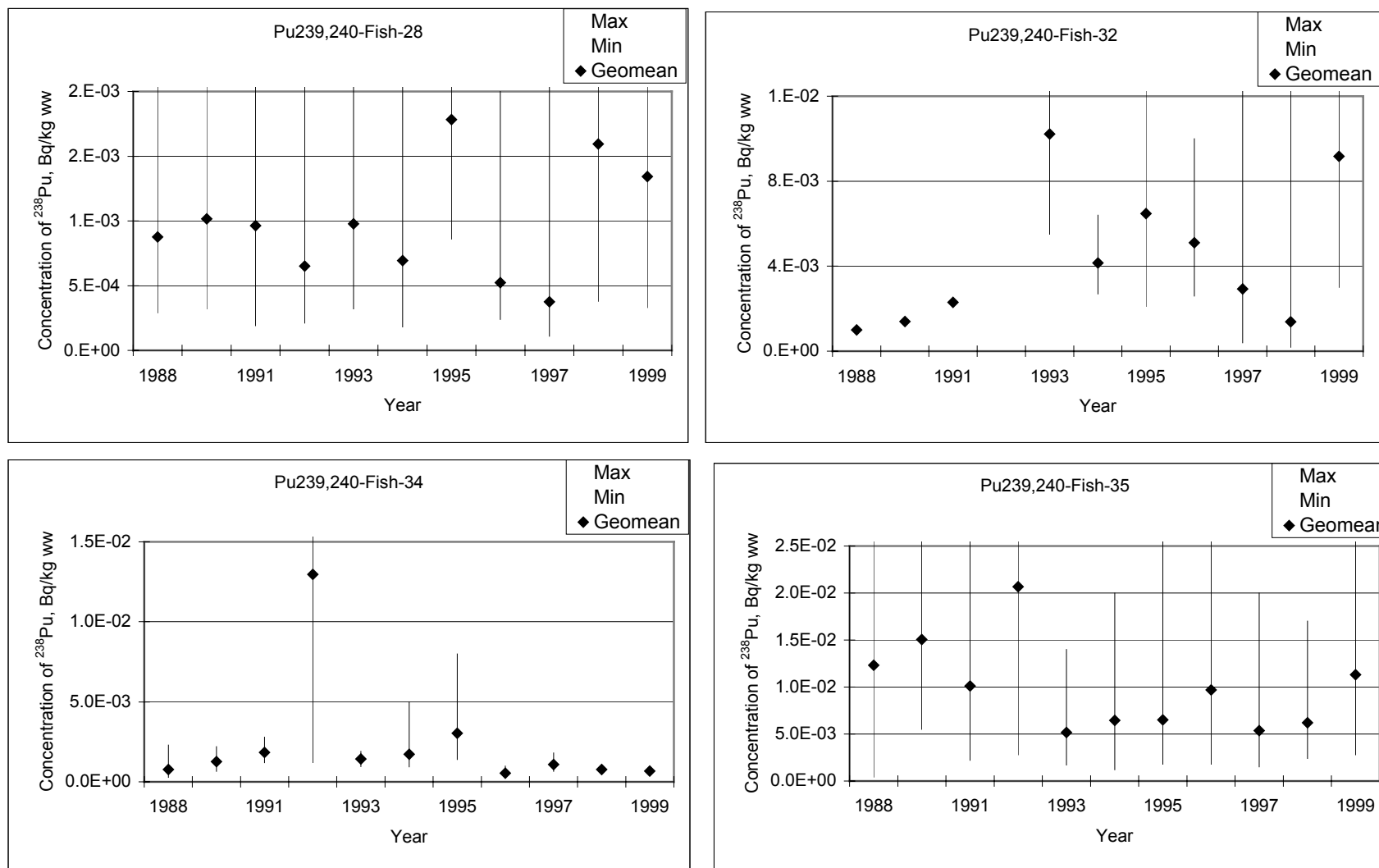


Figure 15 (cont'd)

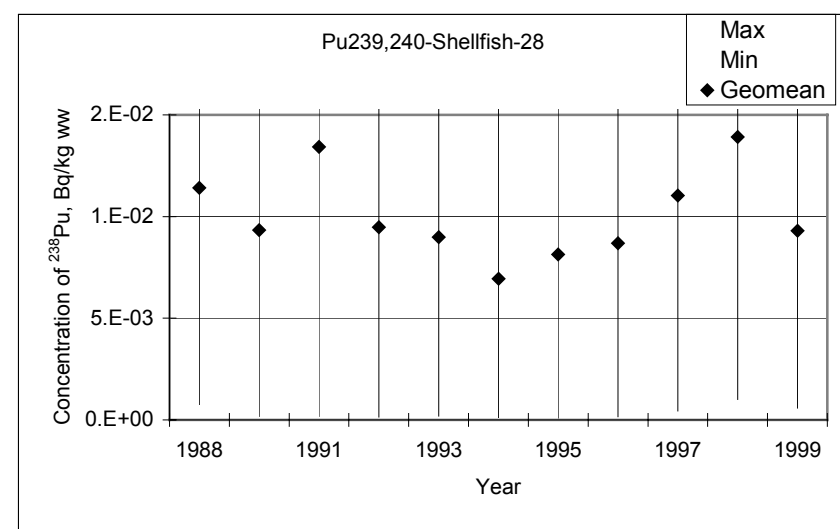
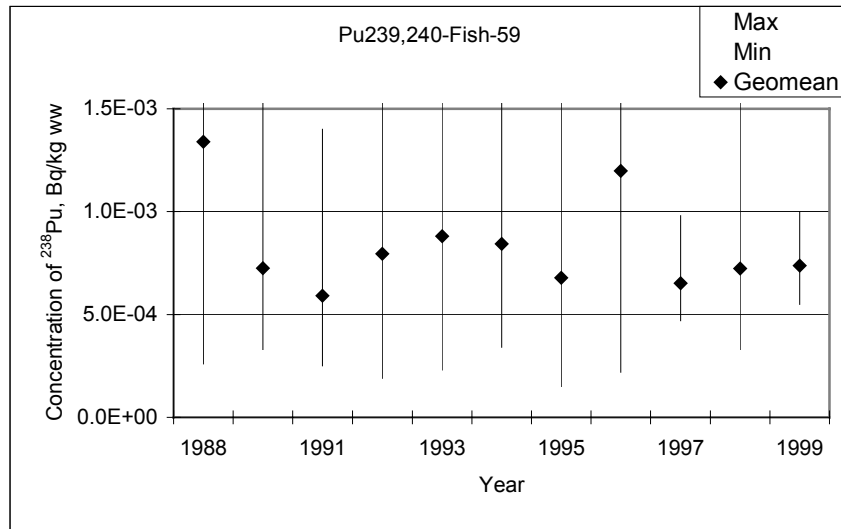
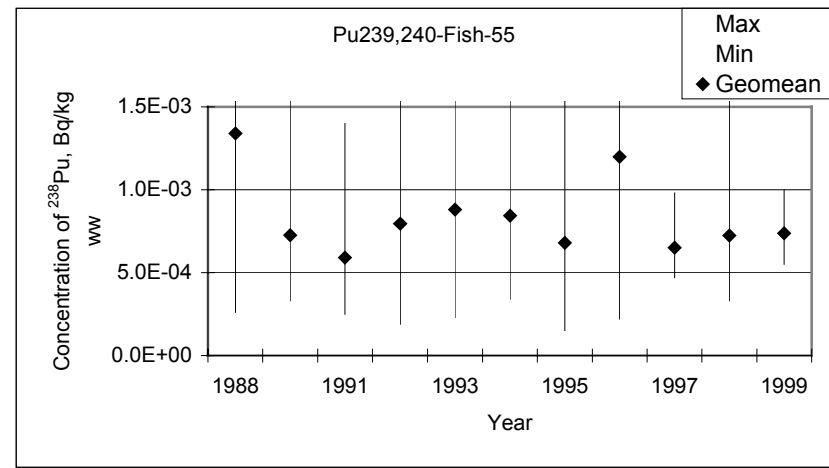
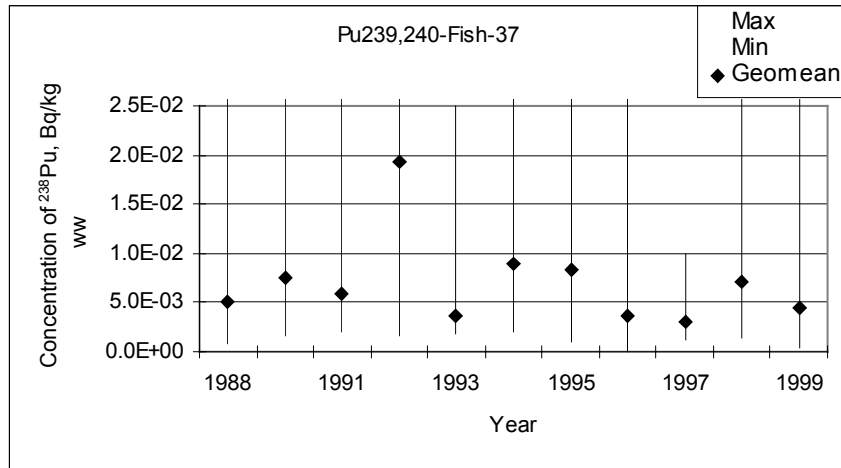


Figure 15 (cont'd)

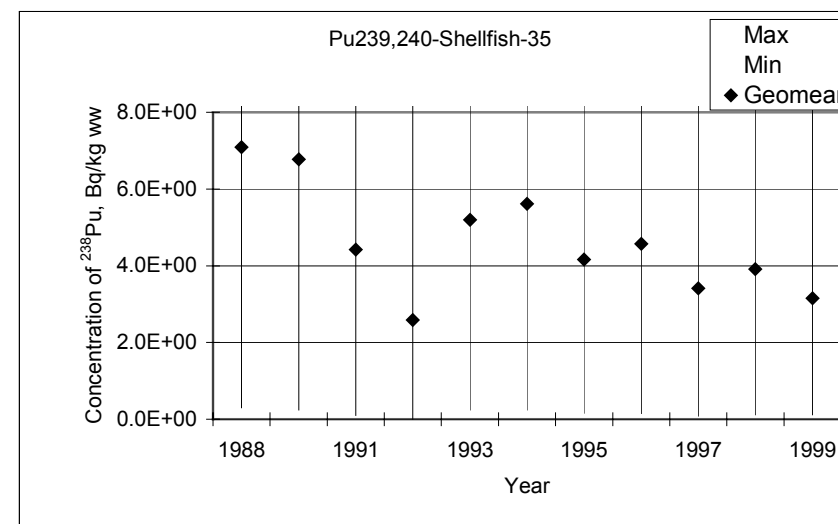
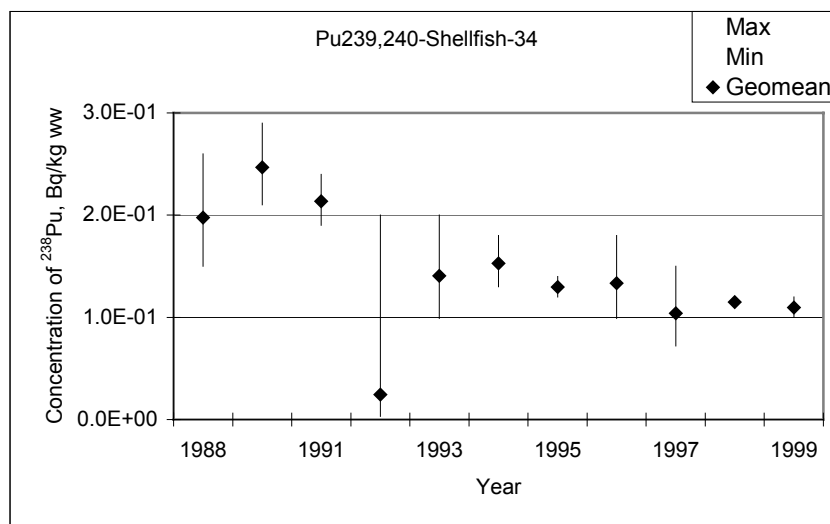
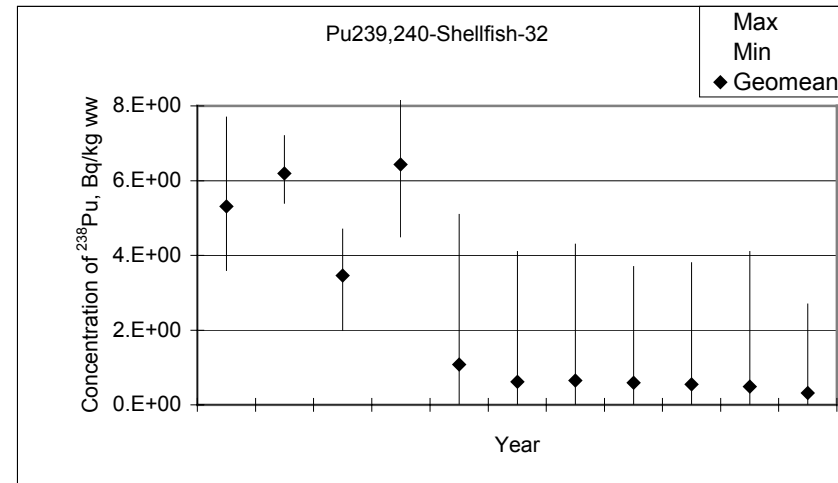
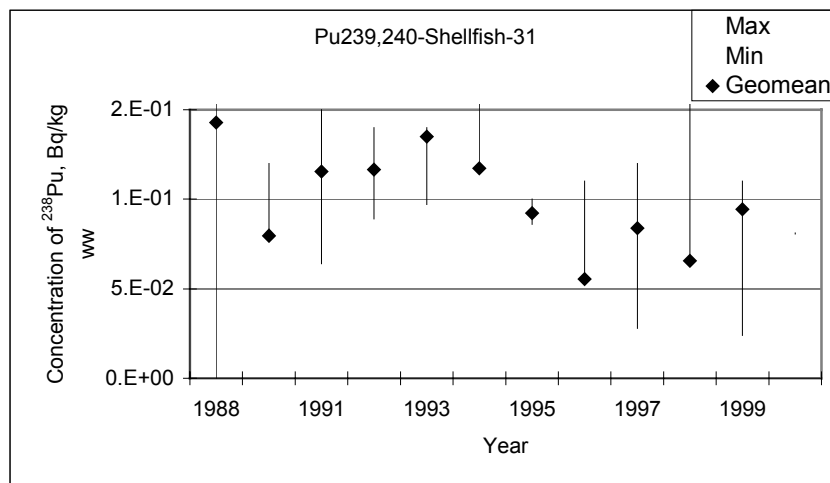


Figure 15 (cont'd)

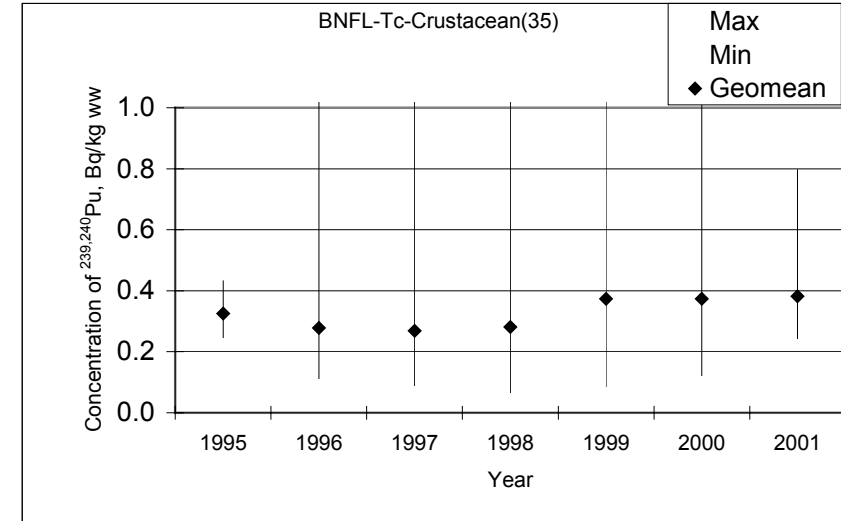
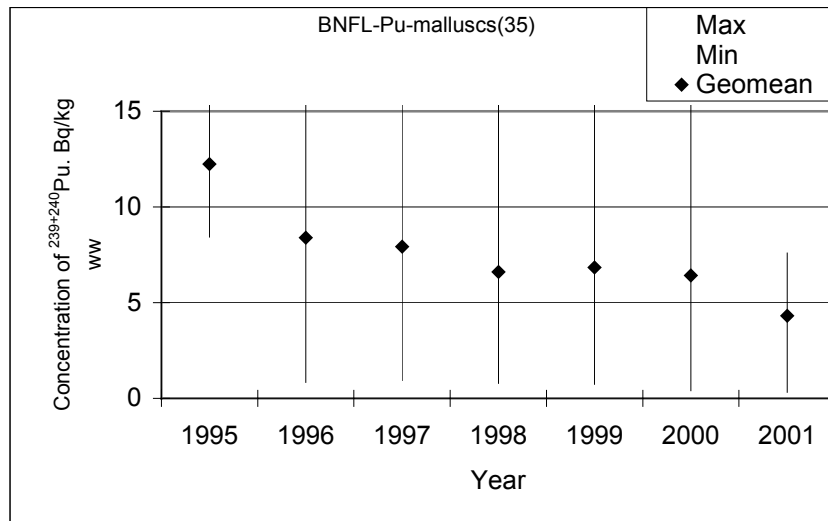
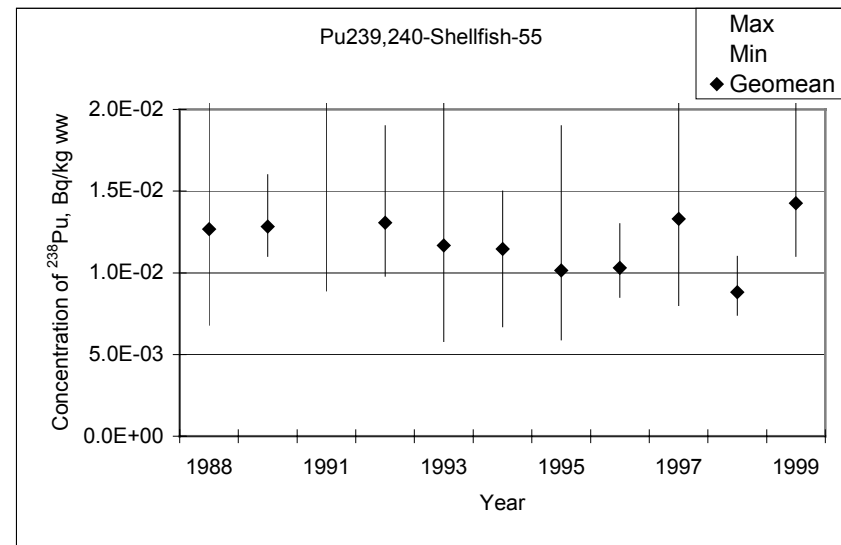
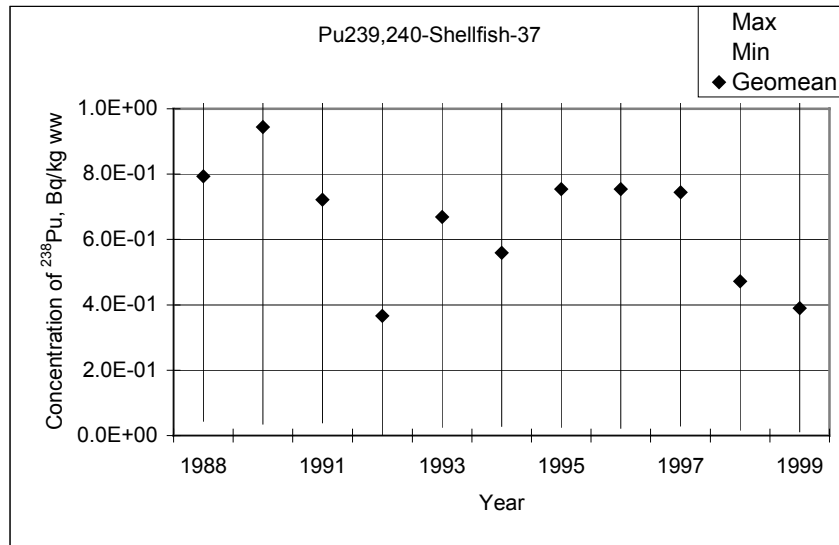
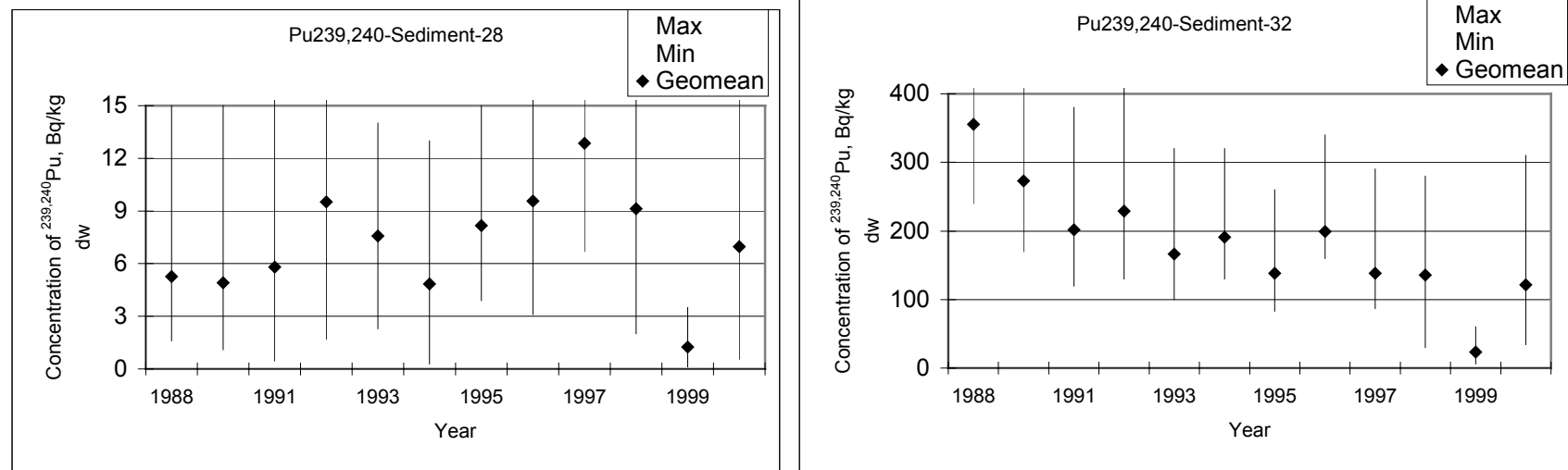


Figure 15 (cont'd)



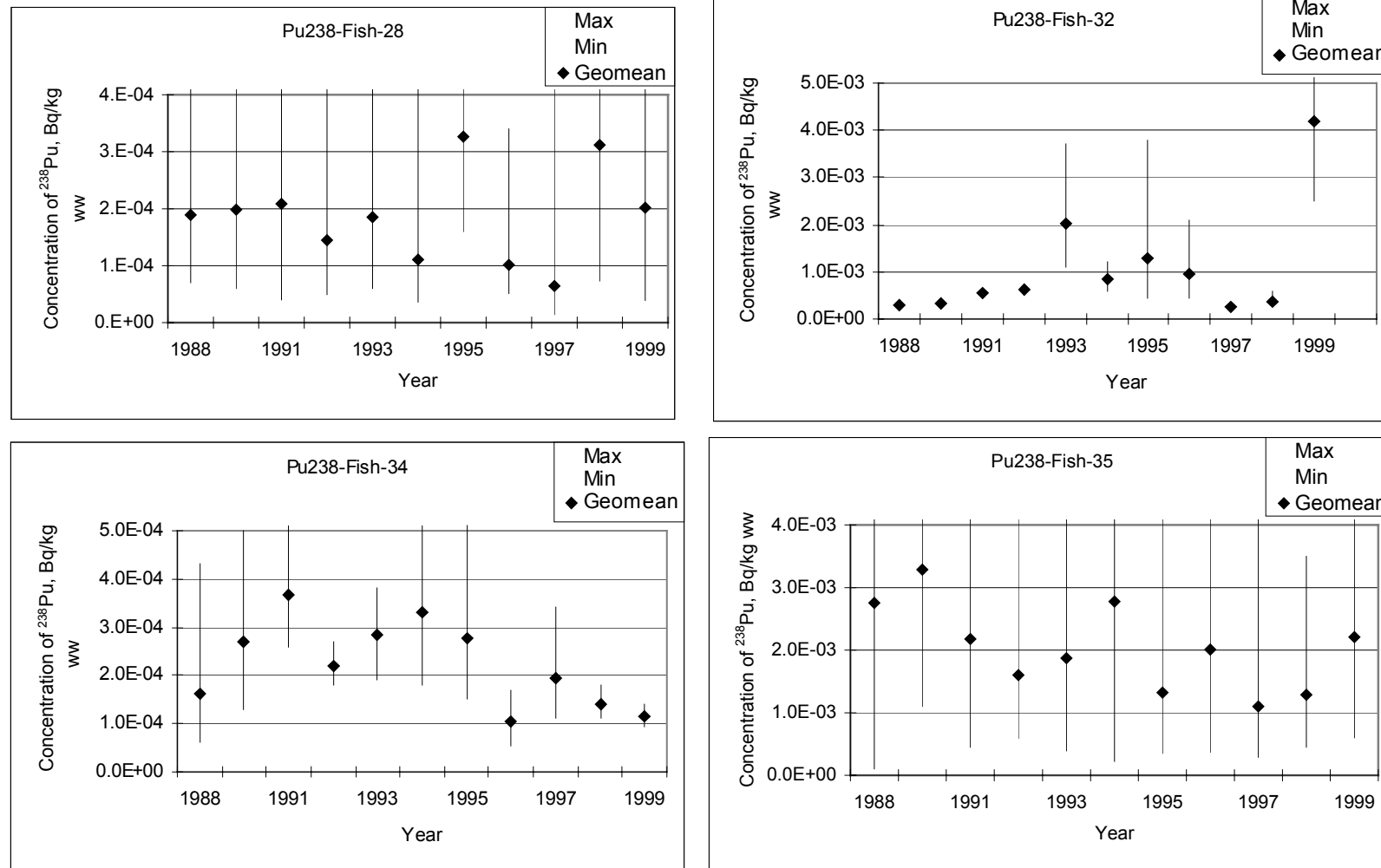
**Figure 16** Temporal variation of  $^{238}\text{Pu}$  concentrations in biota,  $\text{Bq kg}^{-1}$  ww

Figure 16 (cont'd)

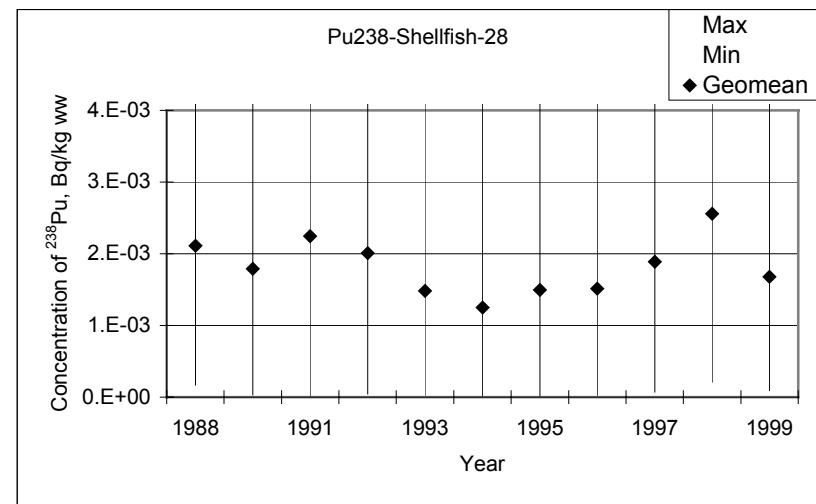
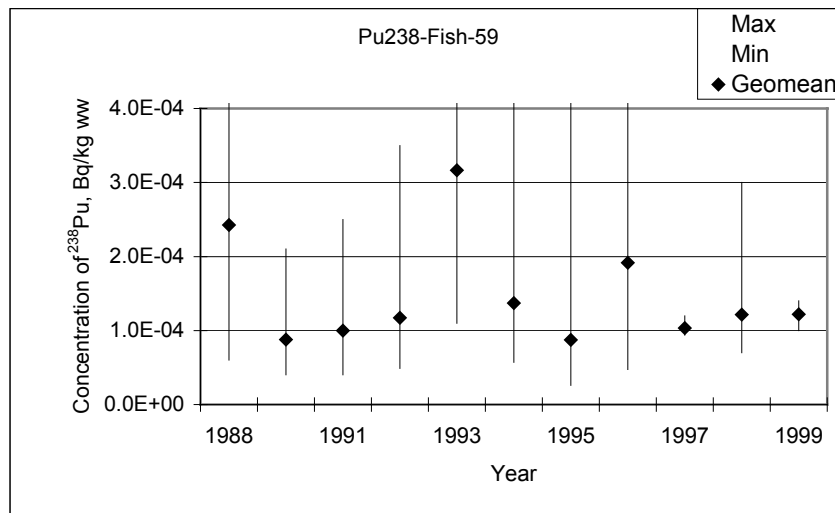
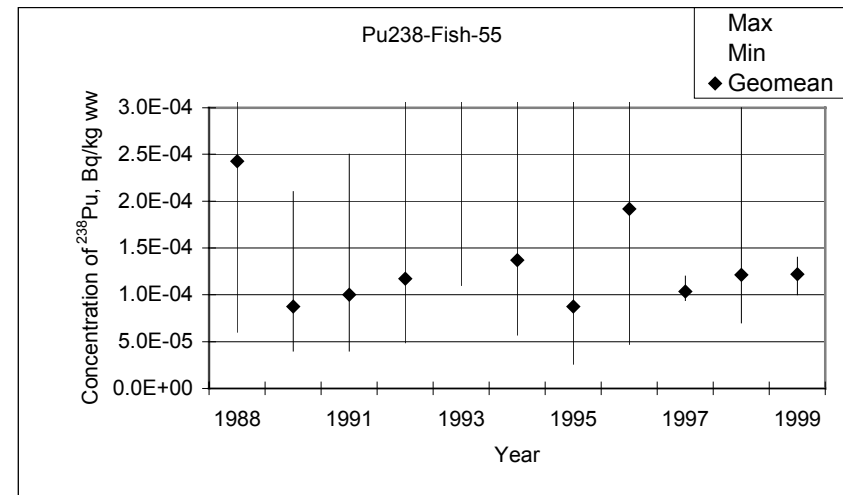
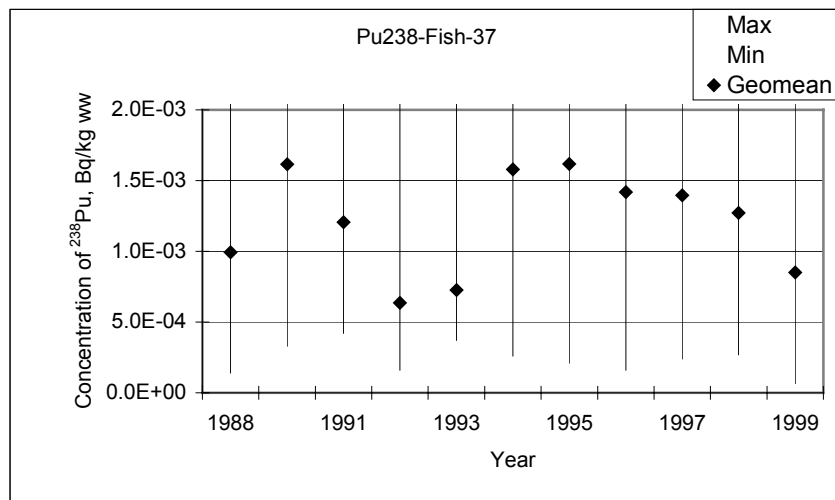


Figure 16 (cont'd)

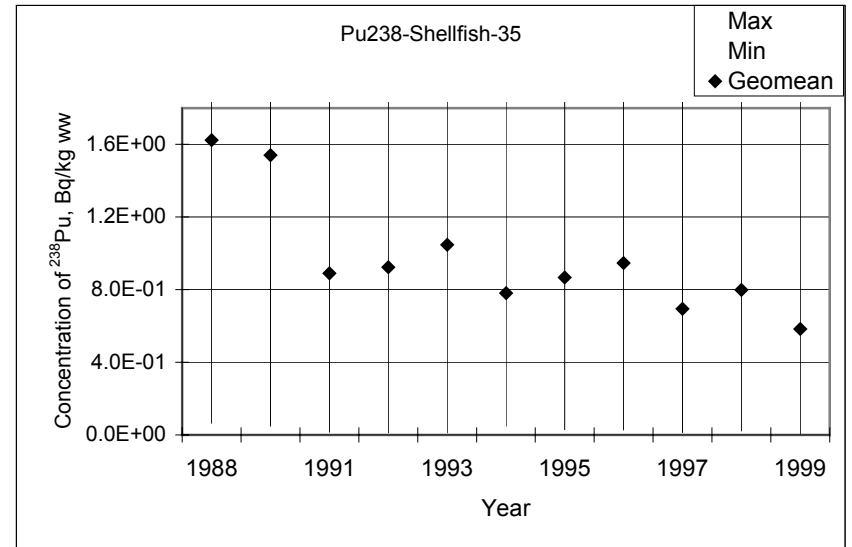
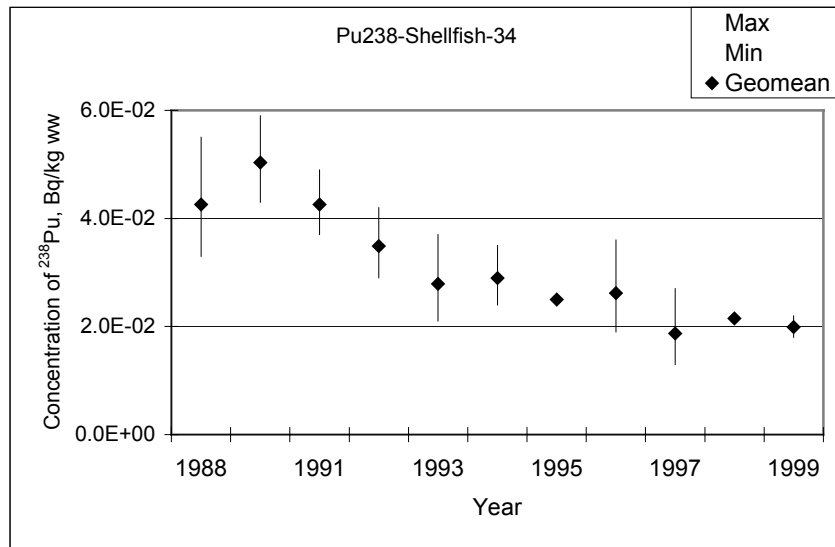
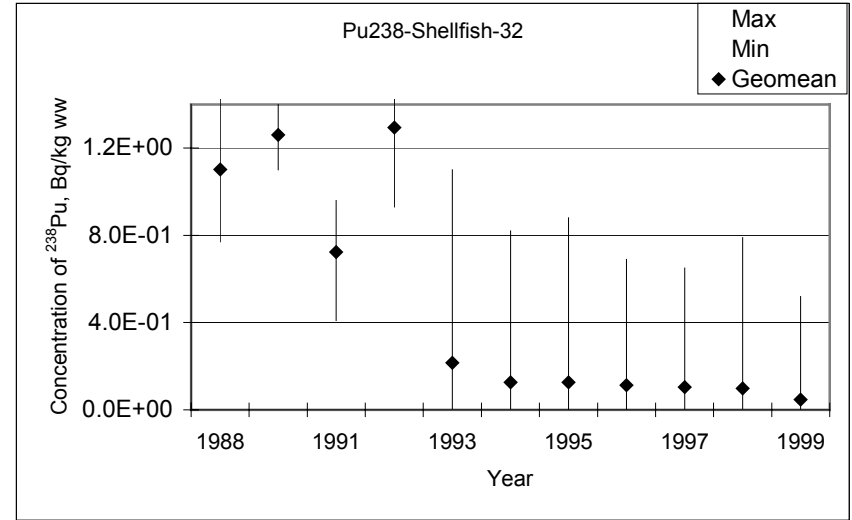
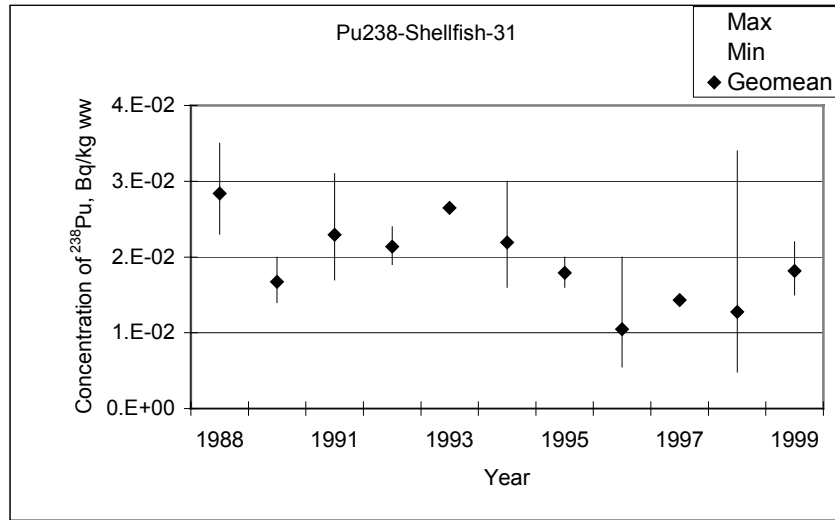




Figure 16 (cont'd)

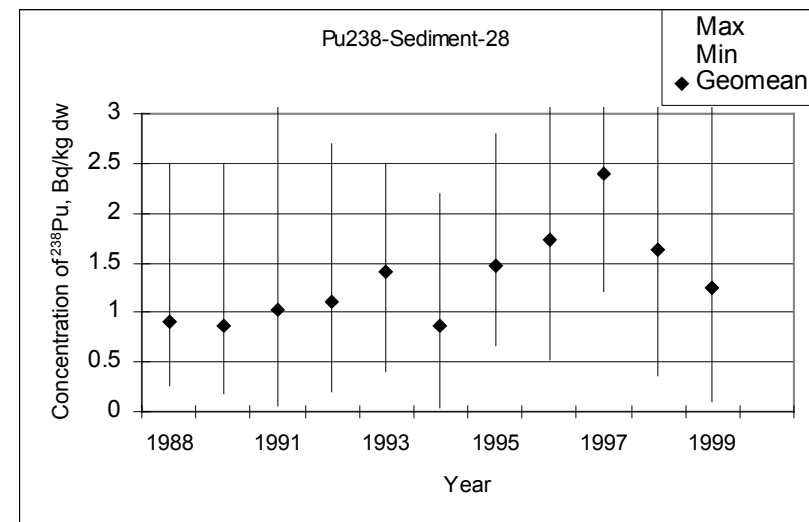
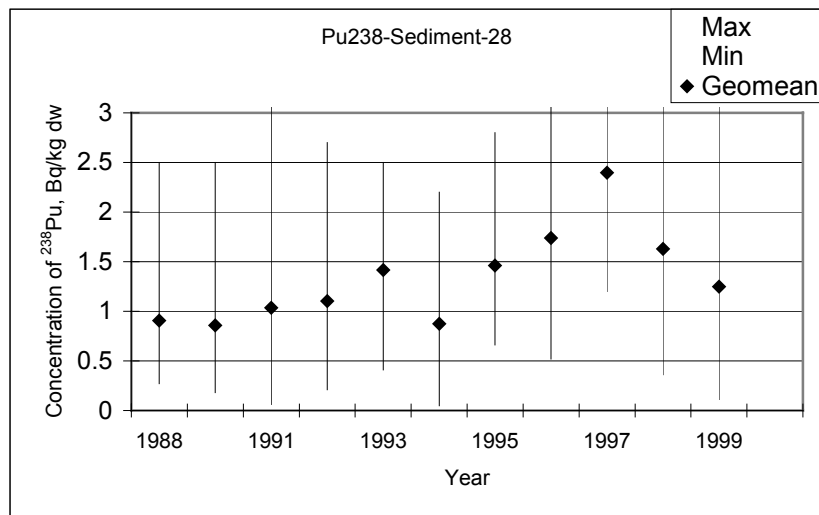
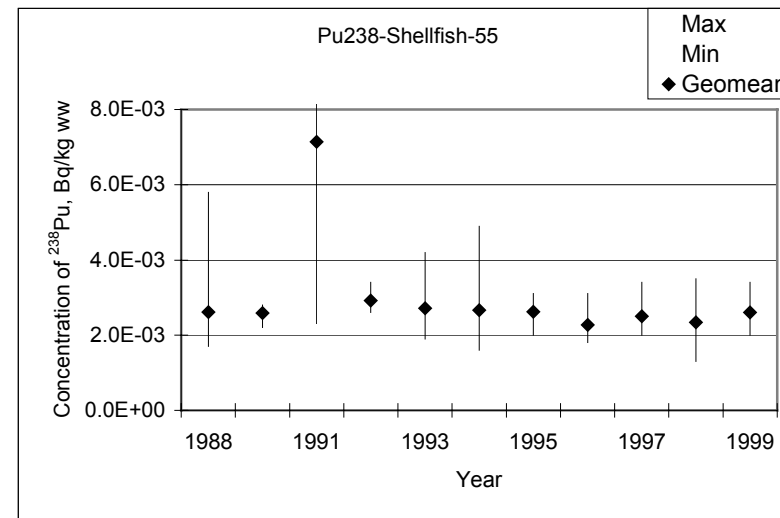
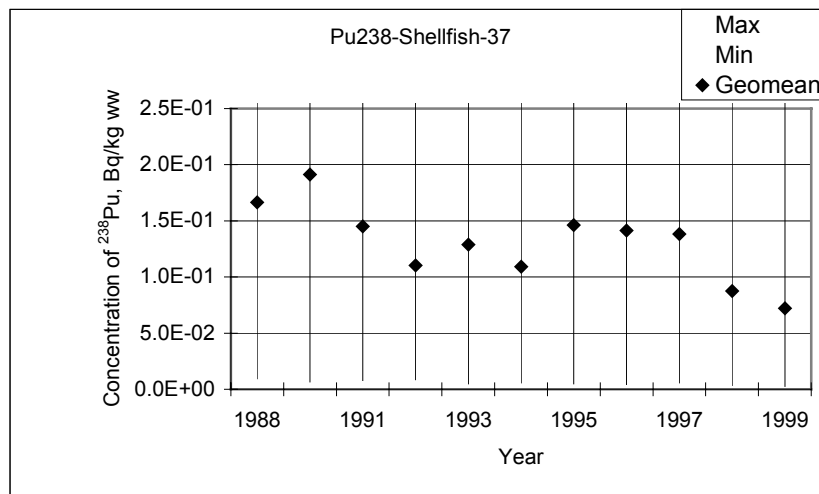
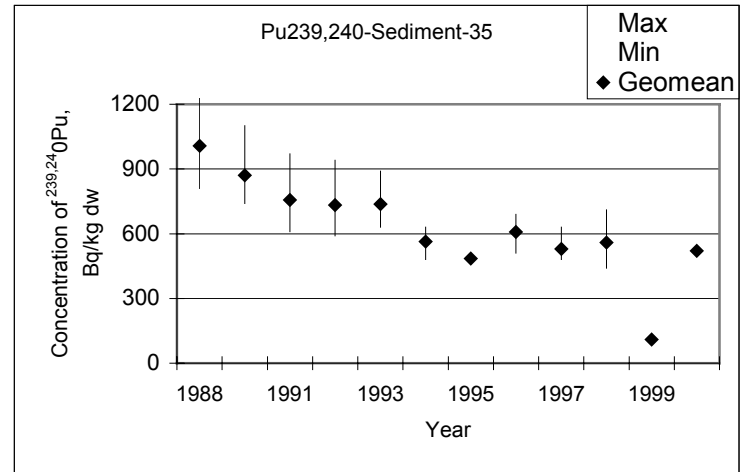
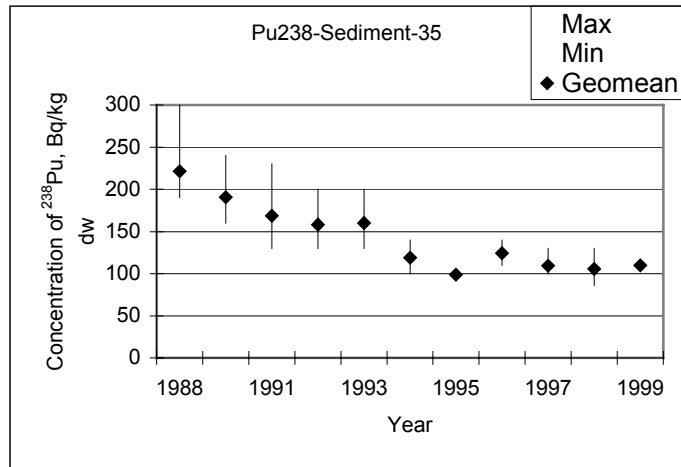
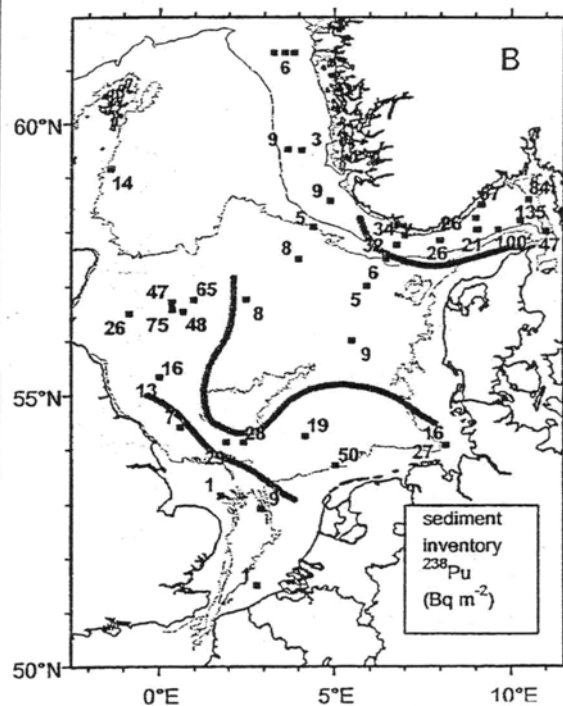
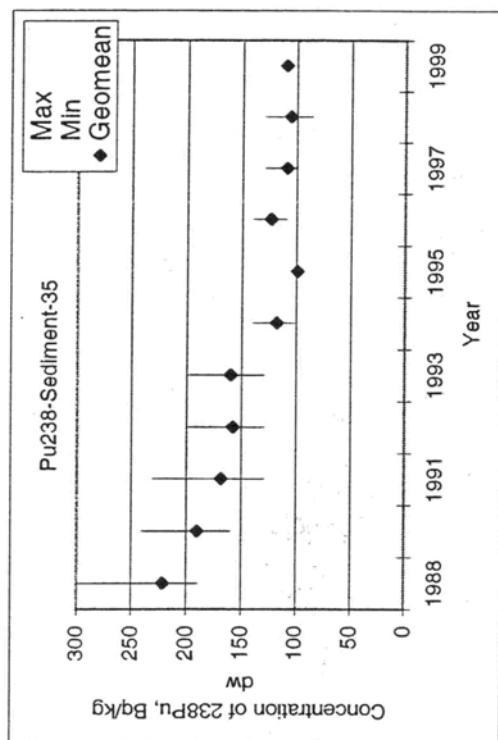
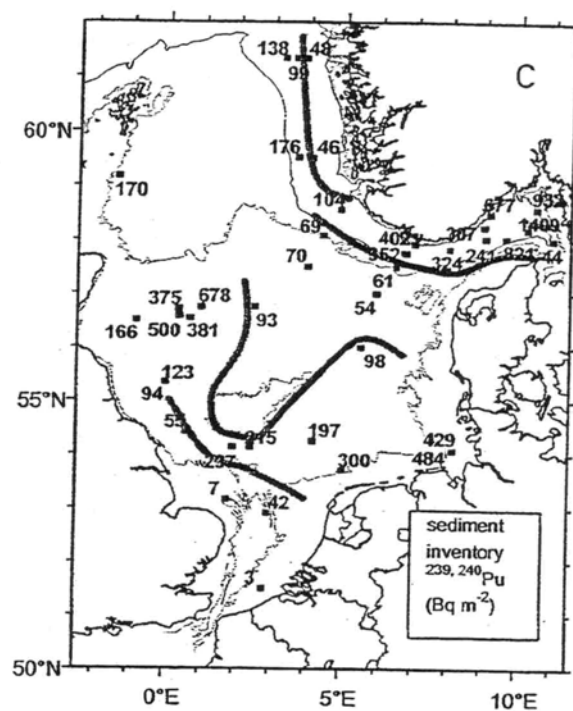
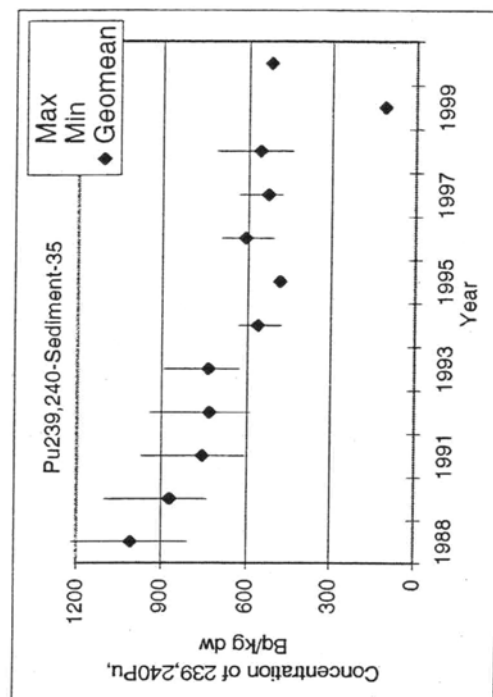


Figure 16 (cont'd)



**Figure 17**      **Temporal variation of plutonium isotopes in sediments, Bq kg<sup>-1</sup> dry weight**



**Figure 18** Concentrations of  $^{239,240}\text{Pu}$  ( $\text{Bq kg}^{-1}$  dw) in surface sediments (0-5cm) in (a)1983, (b) 1988 and (c)1995

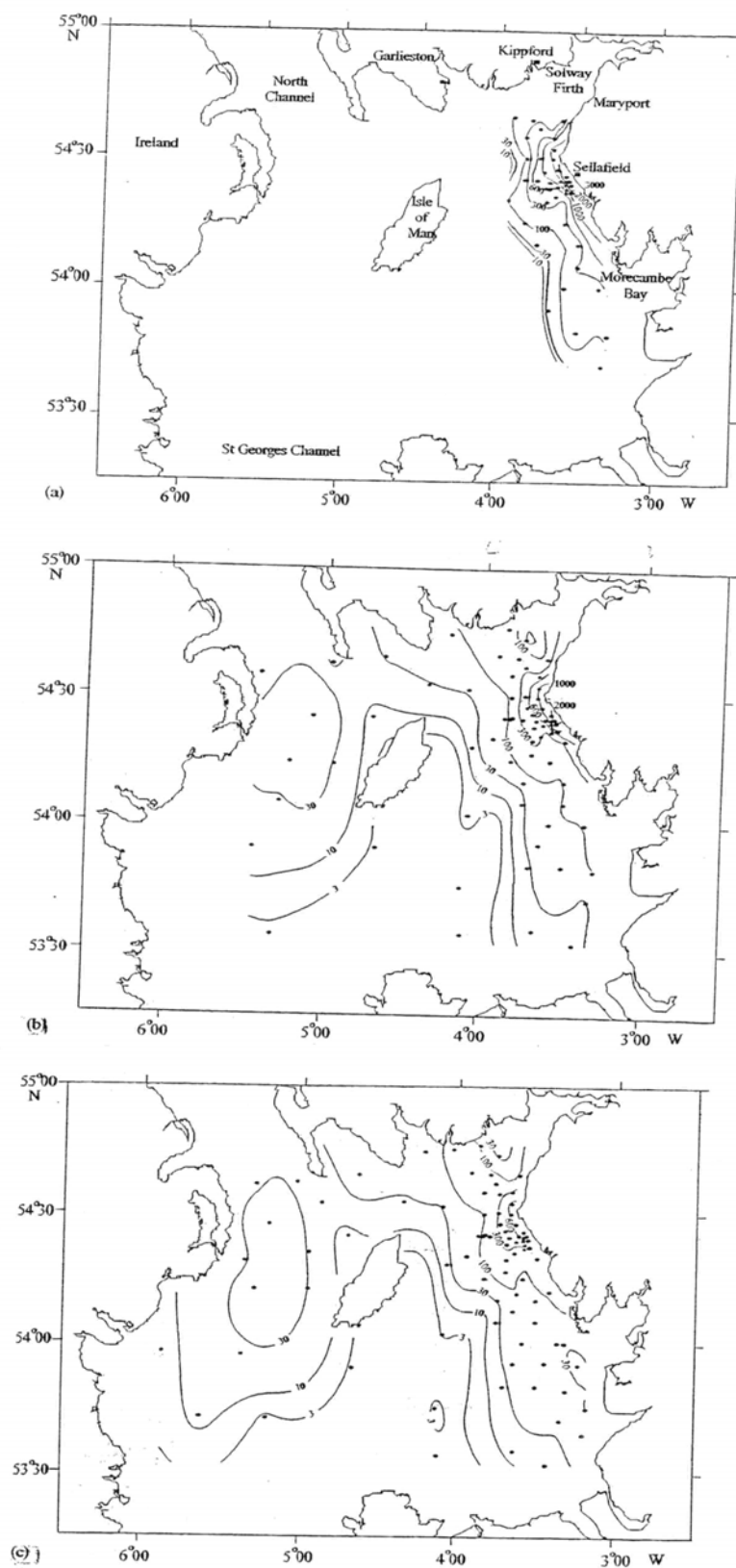
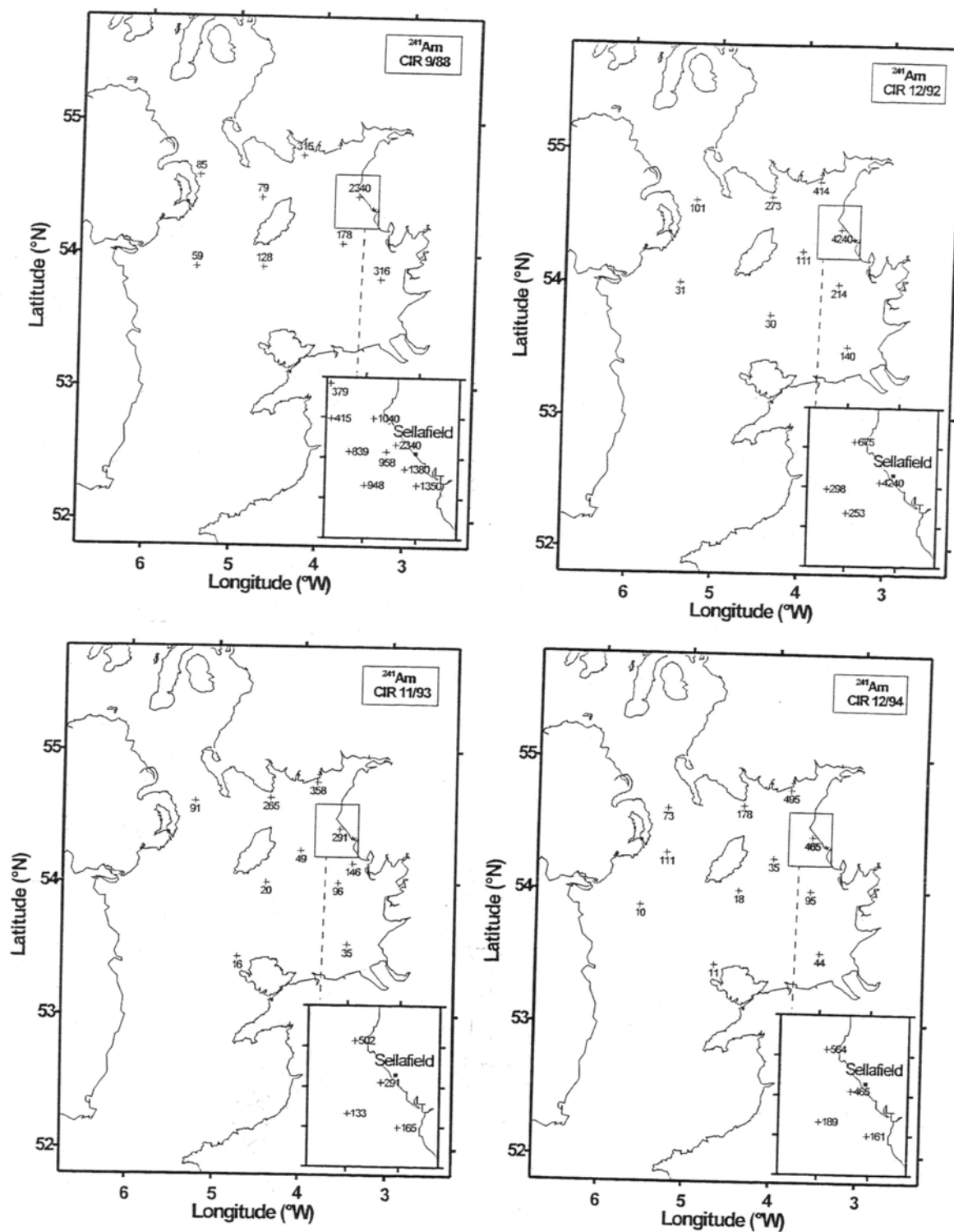


Figure 19 Distribution of dissolved  $^{241}\text{Am}$  in the surface water of the Irish Sea ( $\text{mBq m}^{-3}$ )



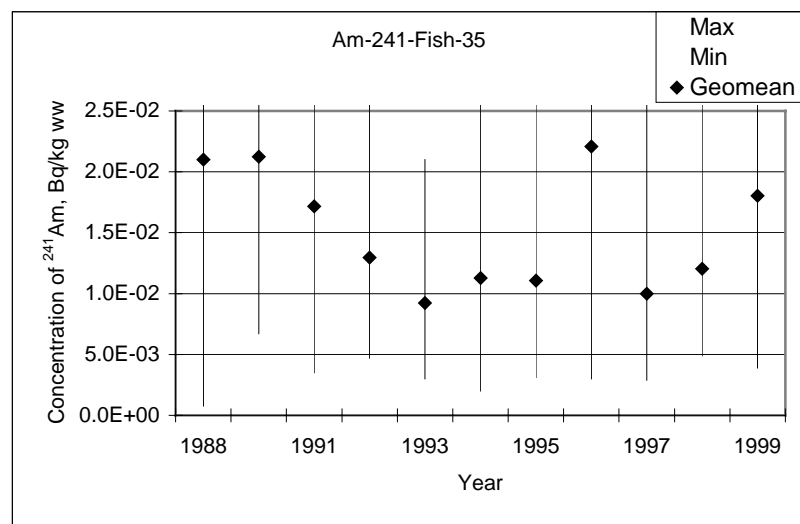
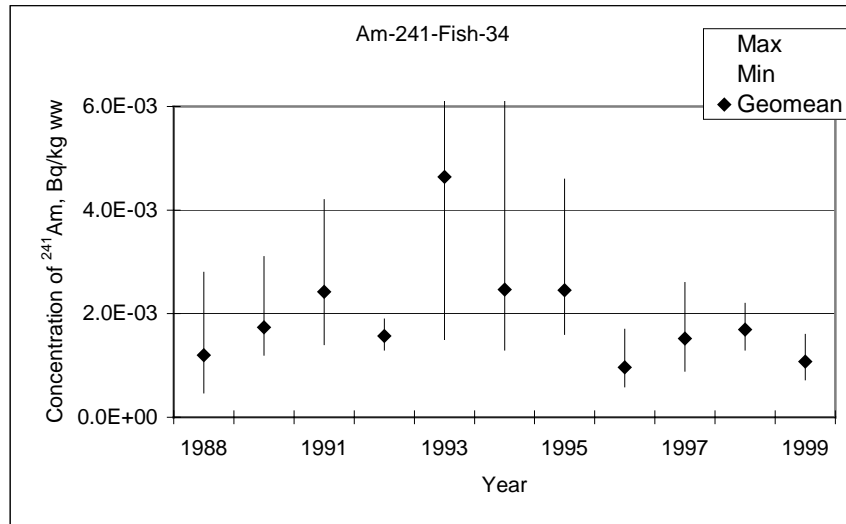
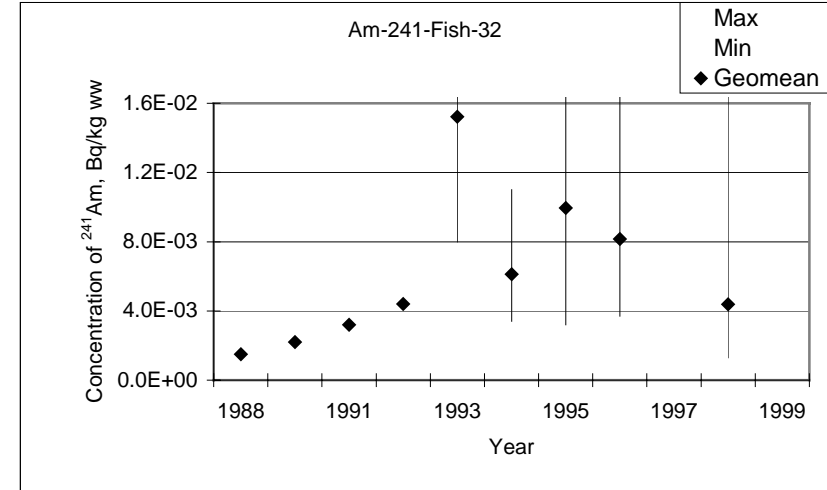
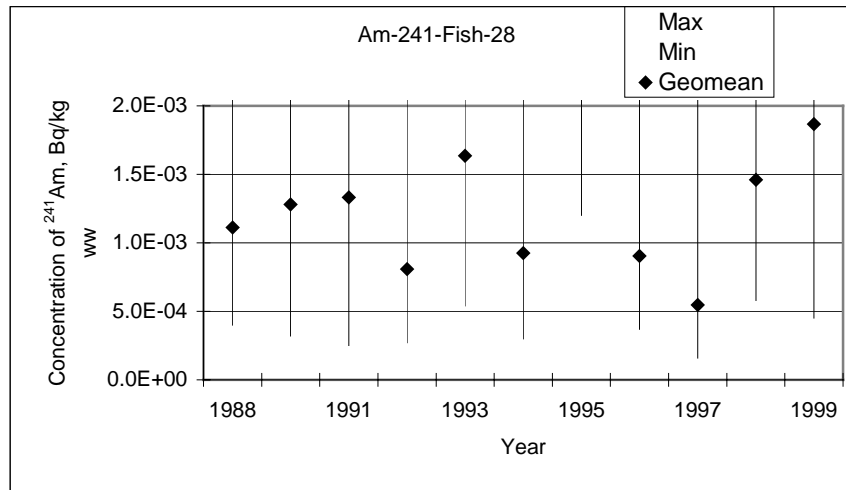
**Figure 20** Temporal variation of  $^{241}\text{Am}$  concentrations in biota and sediment

Figure 20 (cont'd)

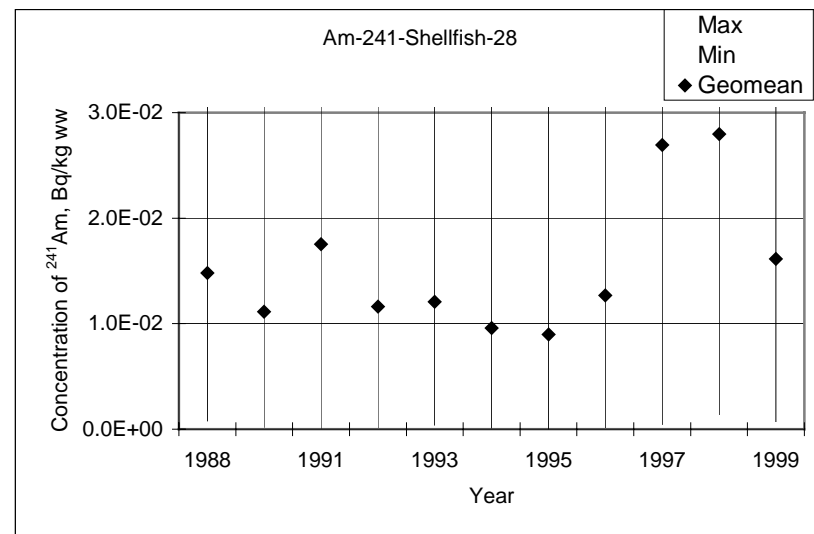
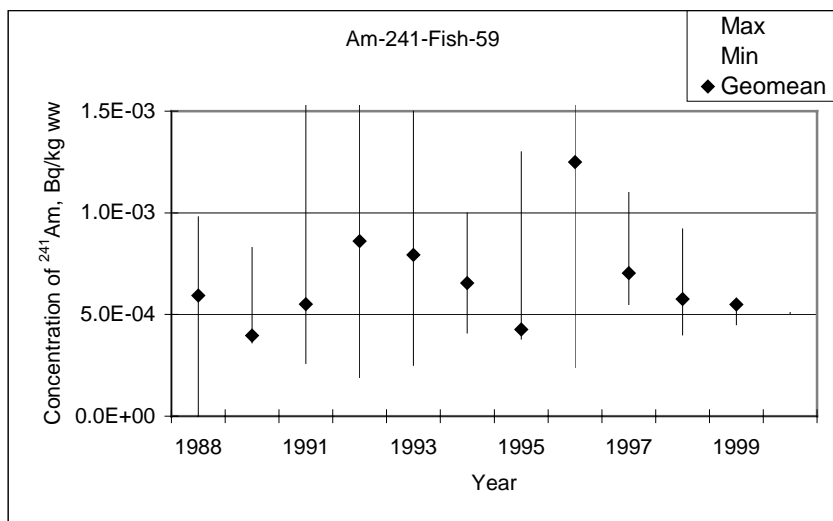
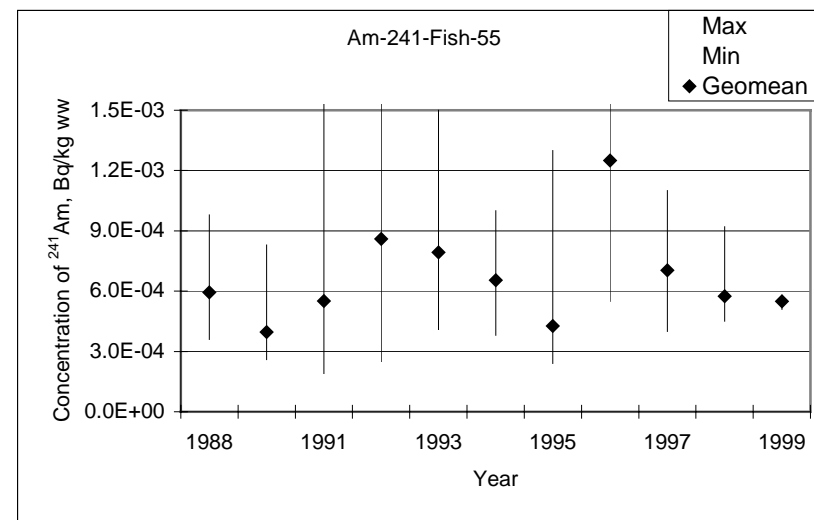
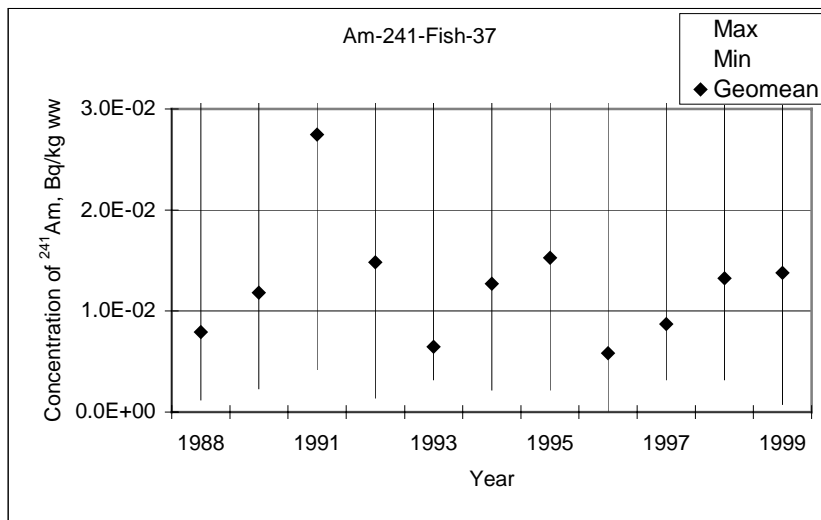


Figure 20 (cont'd)

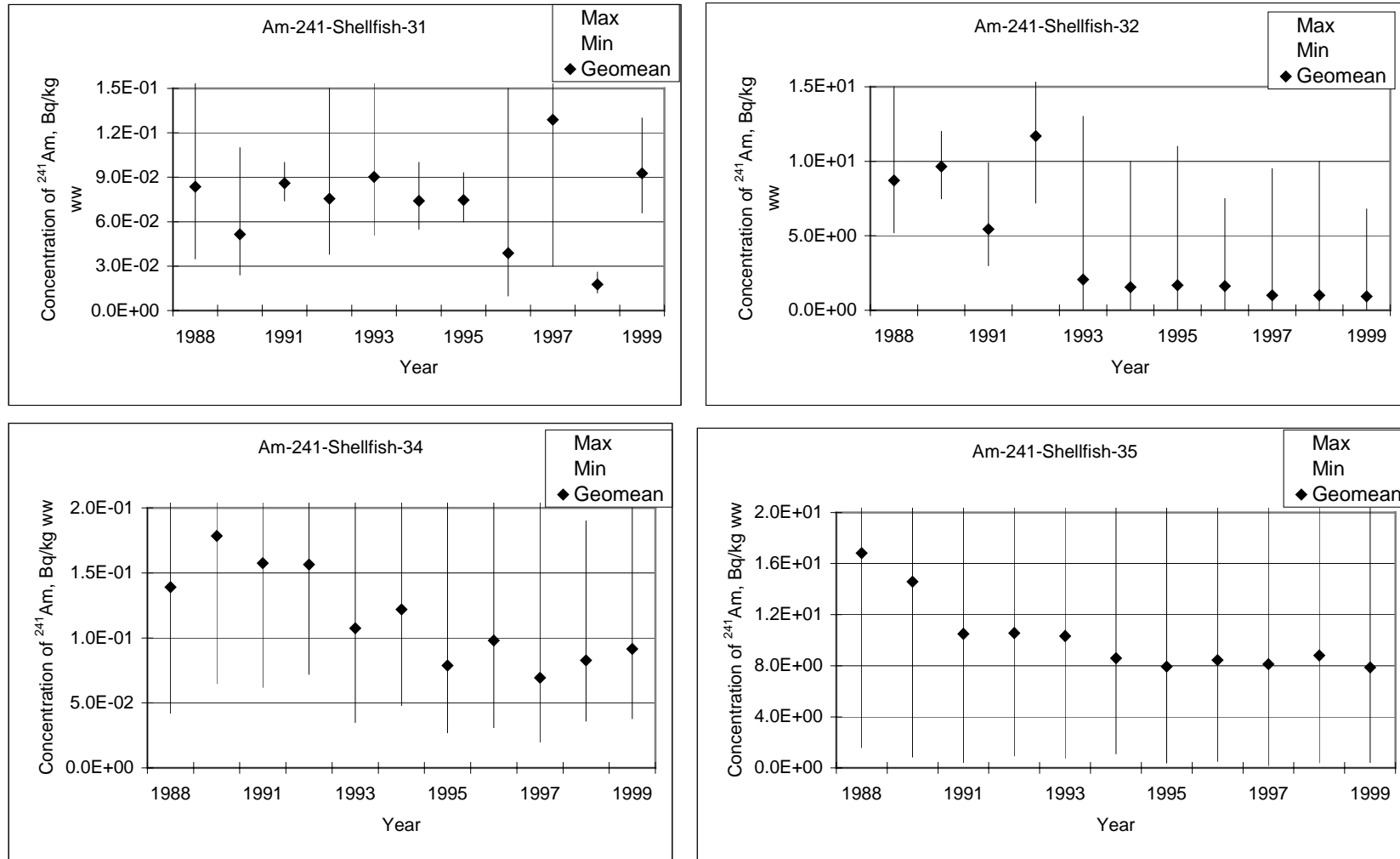




Figure 20 (cont'd)

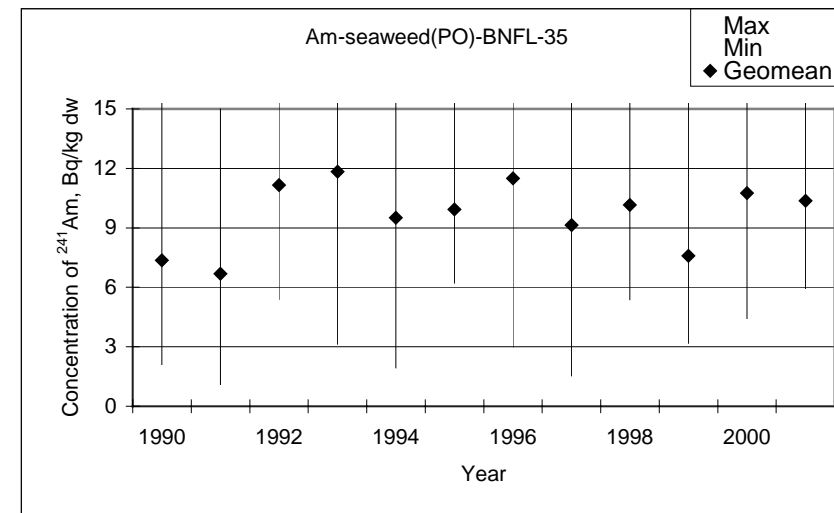
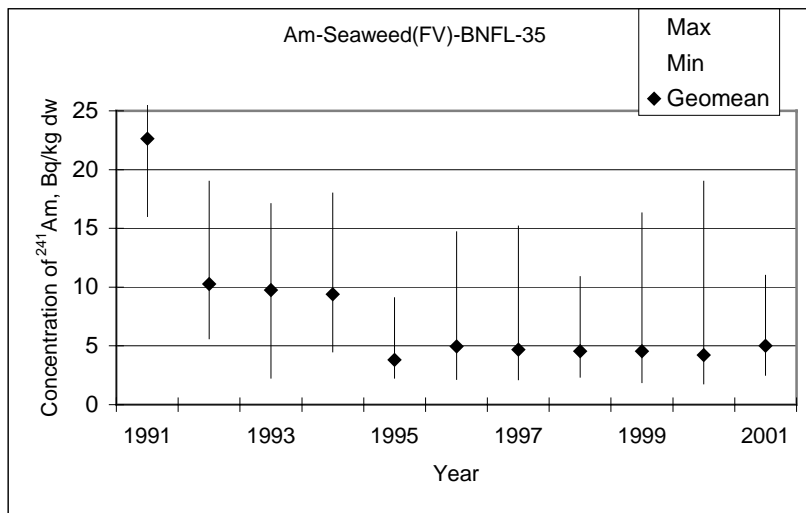
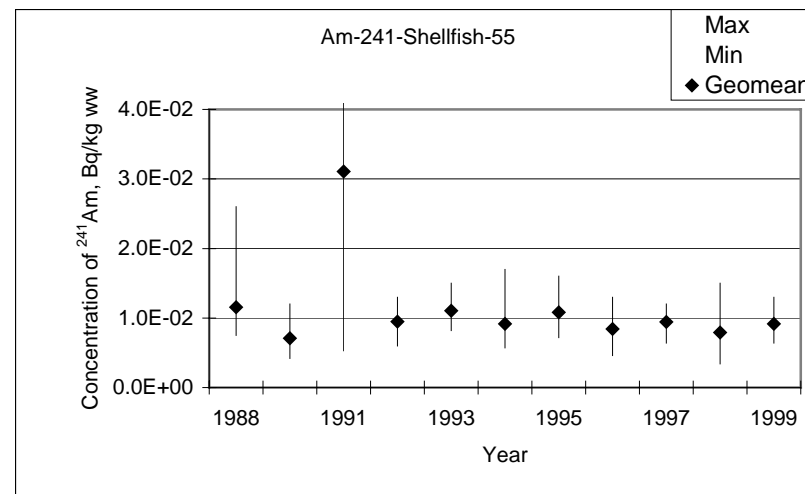
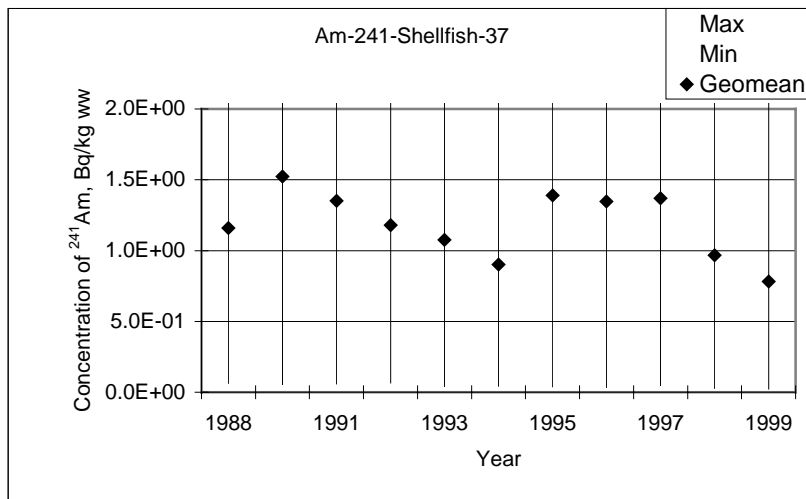
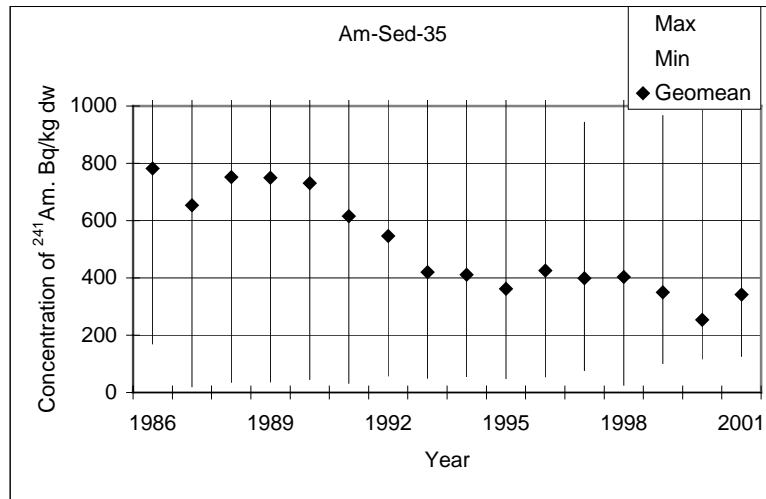
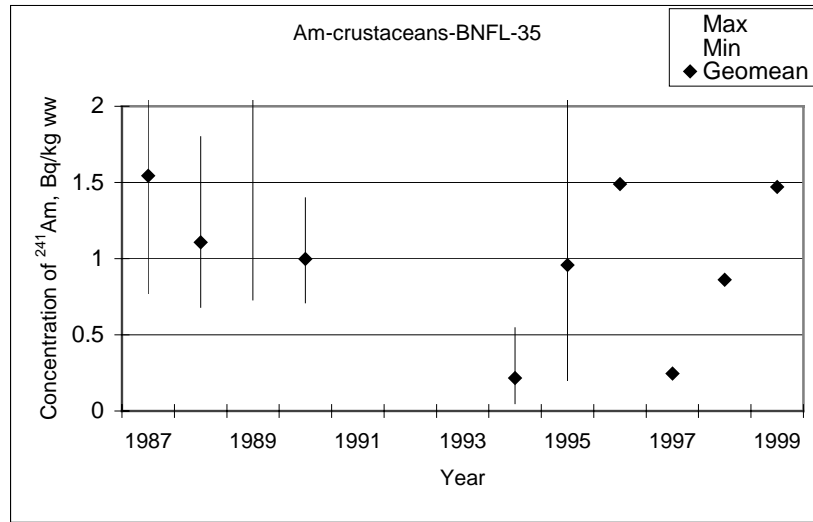
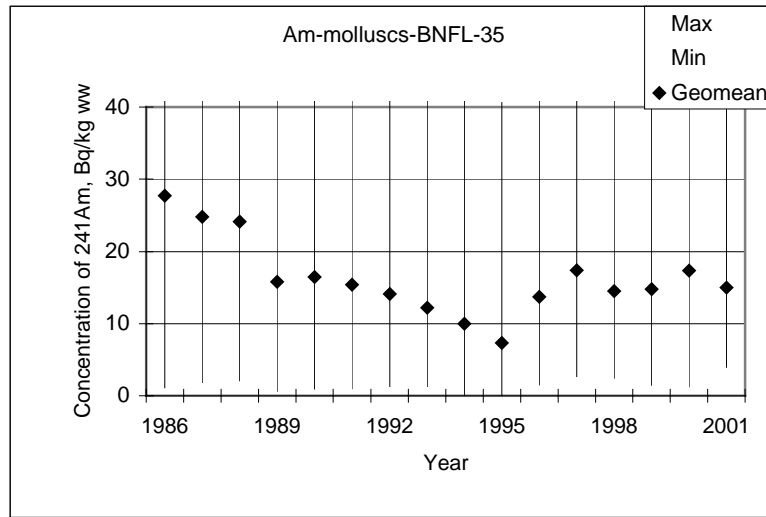


Figure 20 (cont'd)



**Figure 21**      Temporal variation of  $^{60}\text{Co}$  concentrations in shellfish in the Irish Sea,  $\text{Bq kg}^{-1}$  wet weight

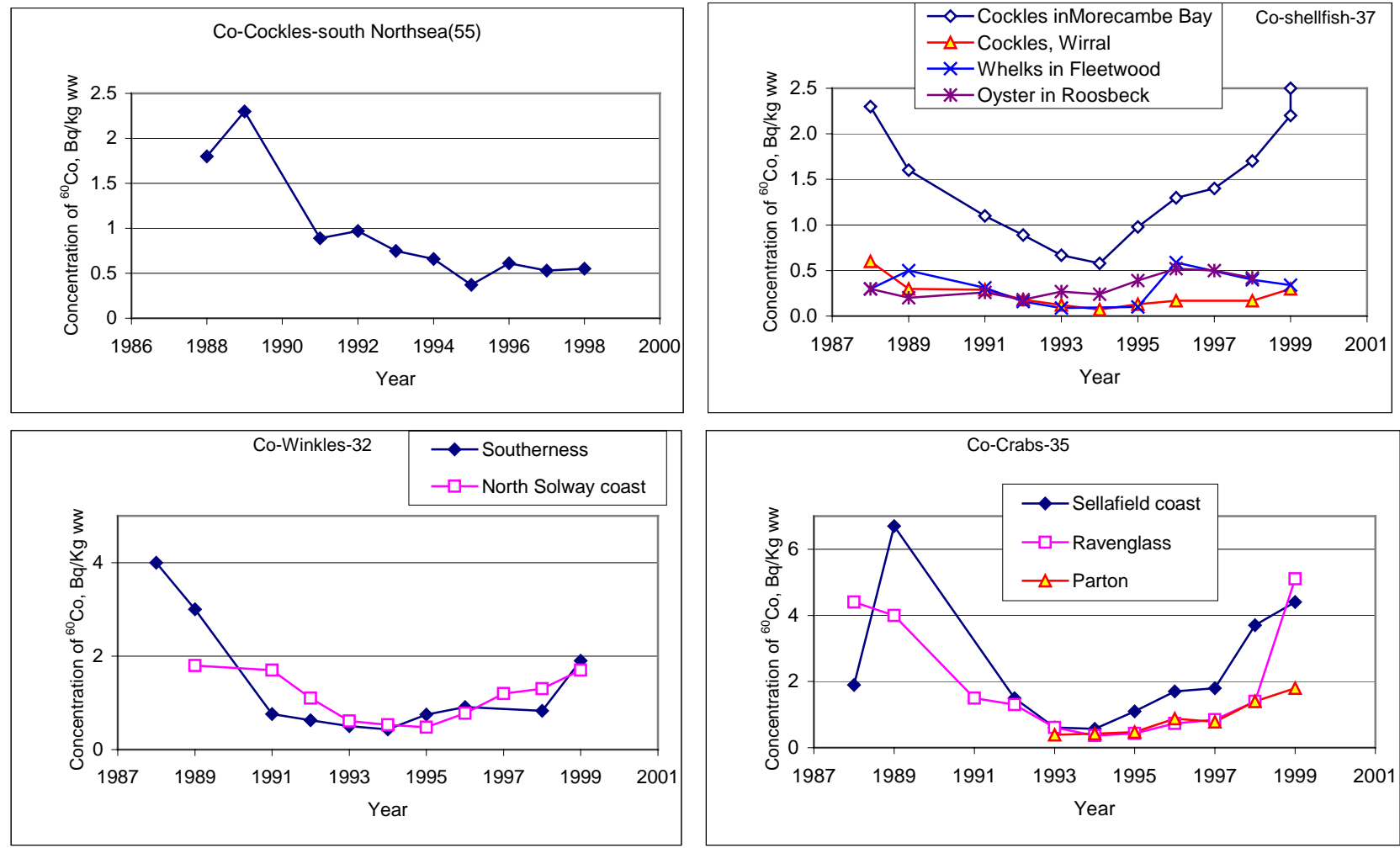
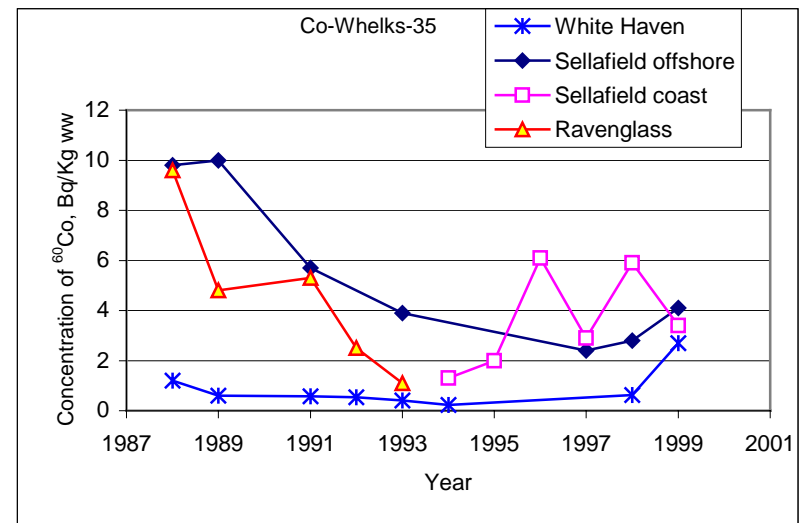
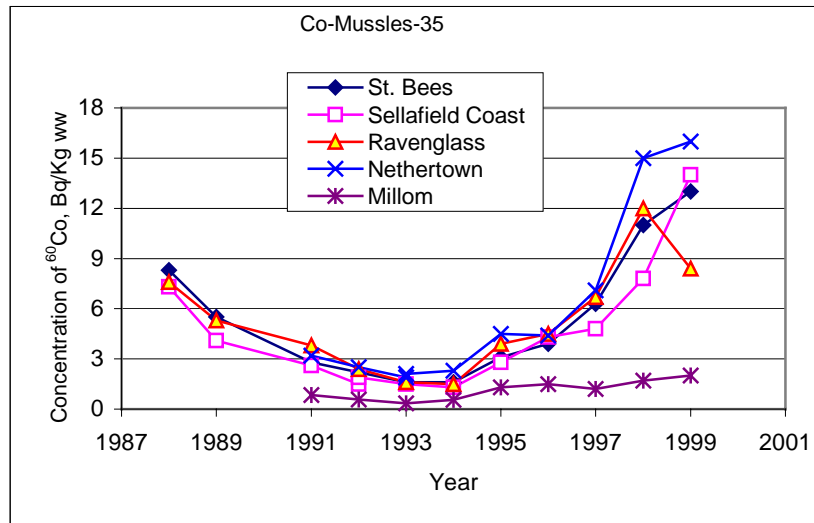
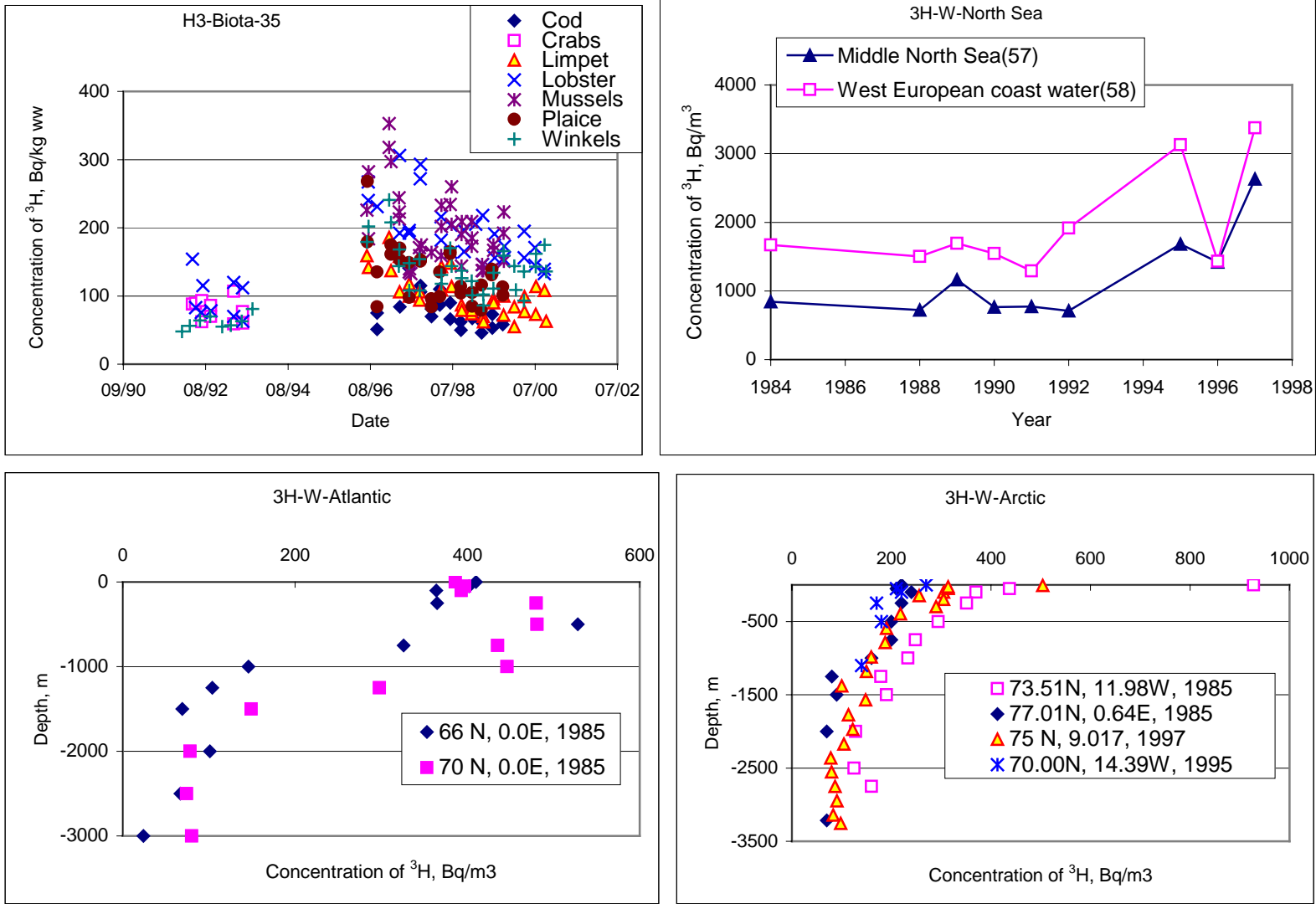


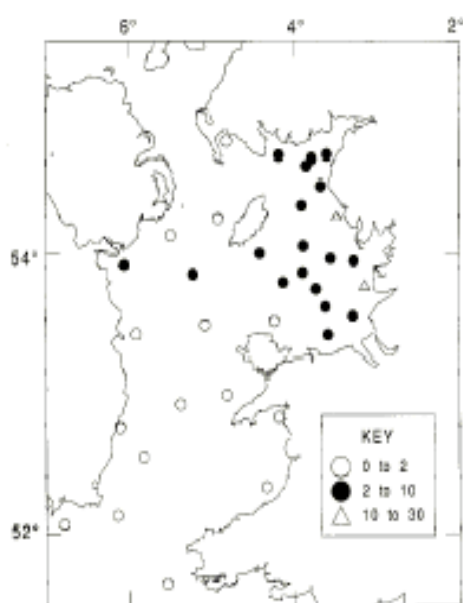
Figure 21 (cont'd)



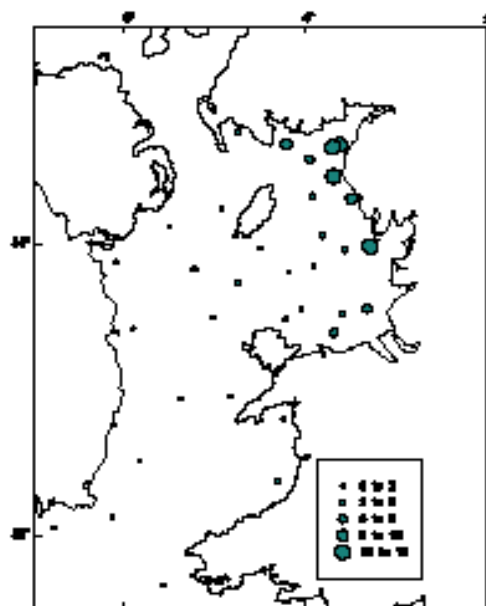
**Figure 22**      Temporal variation of  $^3\text{H}$  concentrations in biota and seawater



**Figure 23**      **Distribution of tritium in surface seawater in the Irish Sea in 1997 and 1999**

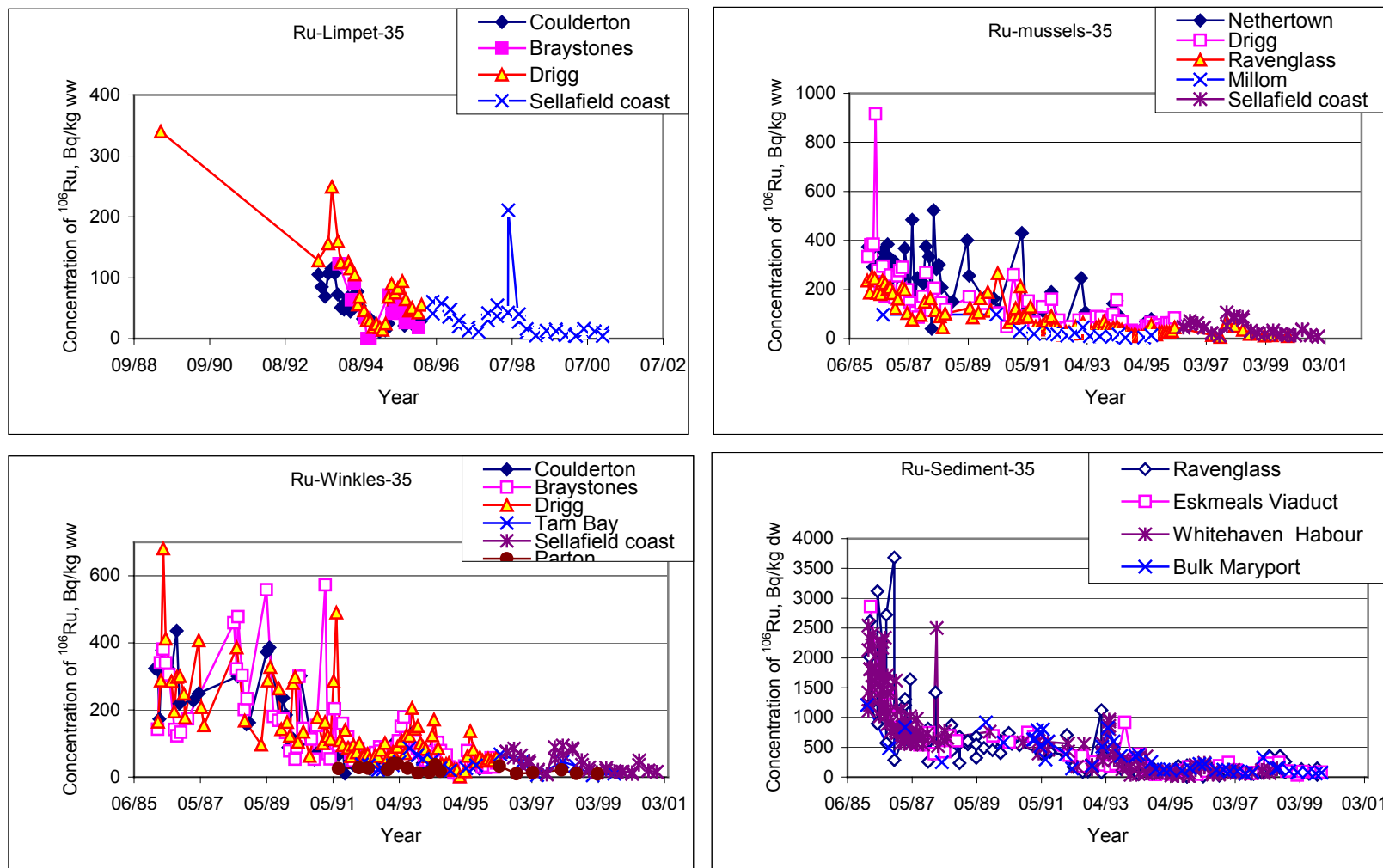


Concentrations ( $\text{Bq kg}^{-1}$ ) of tritium in water from the Irish Sea, September-October, 1997

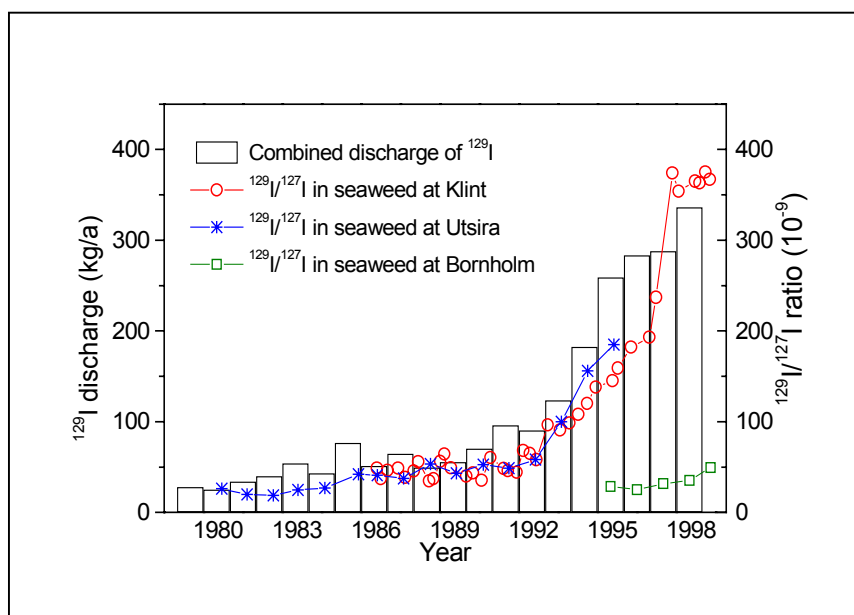


Concentrations ( $\text{Bq kg}^{-1}$ ) of tritium in surface seawater from the Irish Sea, September, 1999

**Figure 24** Temporal variation of  $^{106}\text{Ru}$  concentrations in shellfish and sediment in the east Irish Sea



**Figure 25** Temporal variation of  $^{129}\text{I}/^{127}\text{I}$  ratios in seaweed (*Fucus*) from Klint, Utsira and Bornholm



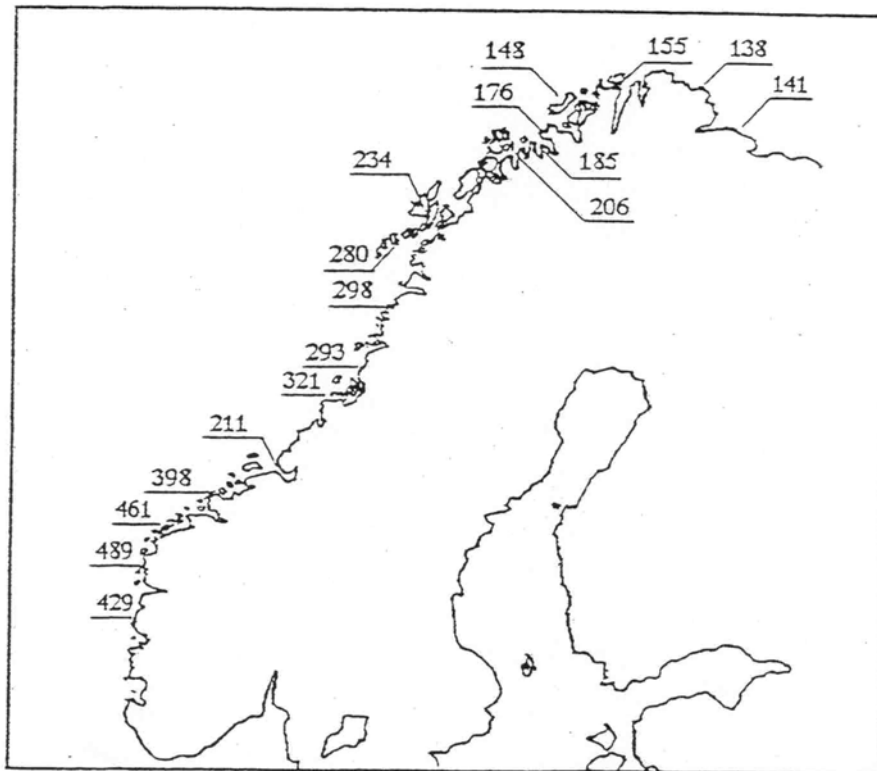
The  $^{129}\text{I}/^{127}\text{I}$  ratios in seaweed from Klint, Utsira and Bornholm and the combined discharges of  $^{129}\text{I}$  from Cap de la Hague and Sellafield (year represents sampling and discharge times,) respectively. The combined discharge was calculated by assuming the discharge from Sellafield reached these areas 2 years after that from Cap de la Hague (from Hou et al. 2000)



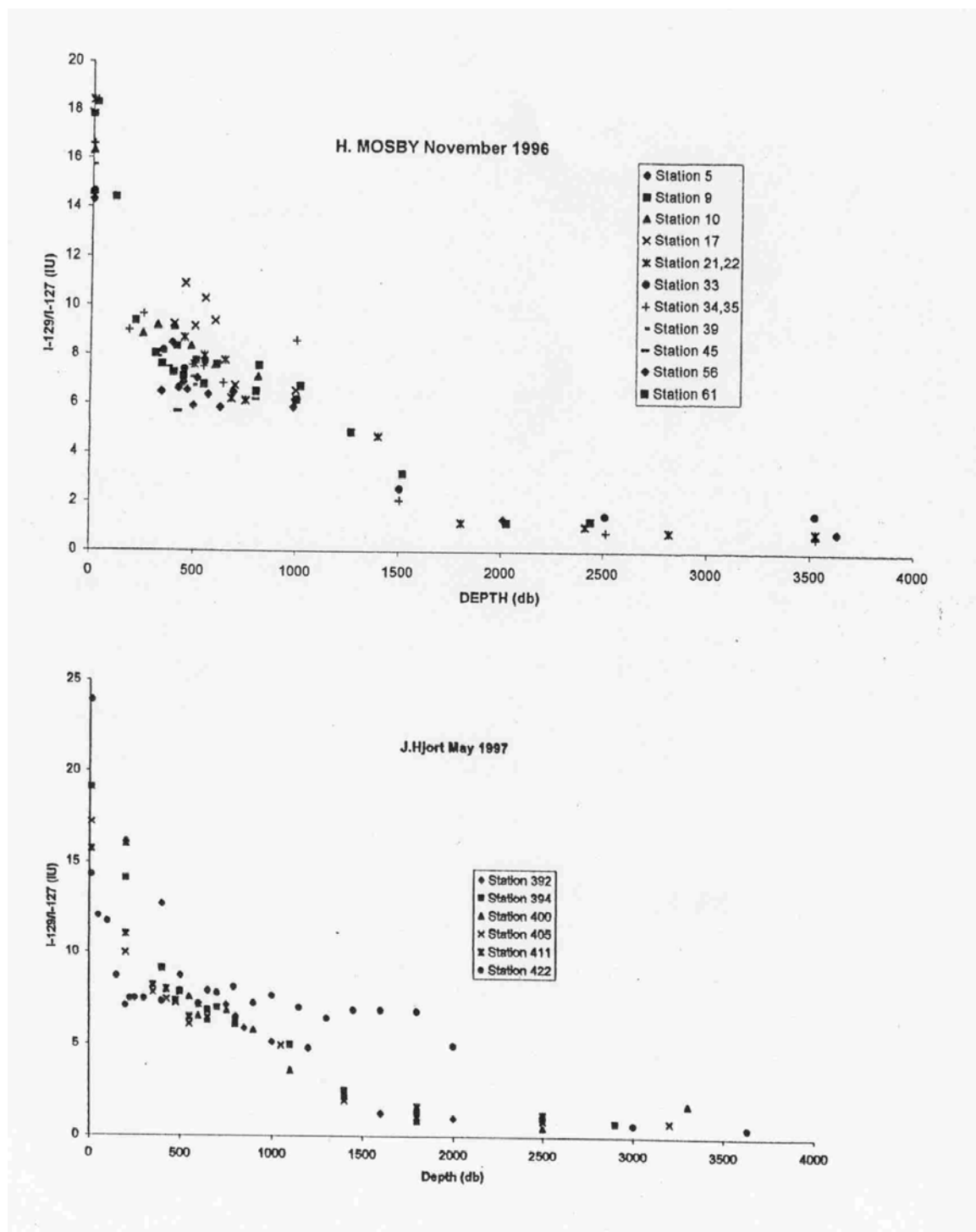
**Figure 26**  $^{129}\text{I}$  concentrations ( $10^{-12} \text{ g l}^{-1}$ ) in surface seawater around Denmark



**Figure 27**     Distribution of  $^{129}\text{I}/^{127}\text{I}$  ratios in seawater samples ( $\text{IU} = 10^{-10}$ ) taken along the Norwegian coast in 1993



**Figure 28** Depth profile of  $^{129}\text{I}/^{127}\text{I}$  (IU= $10^{-10}$ ) in the Greenland Sea



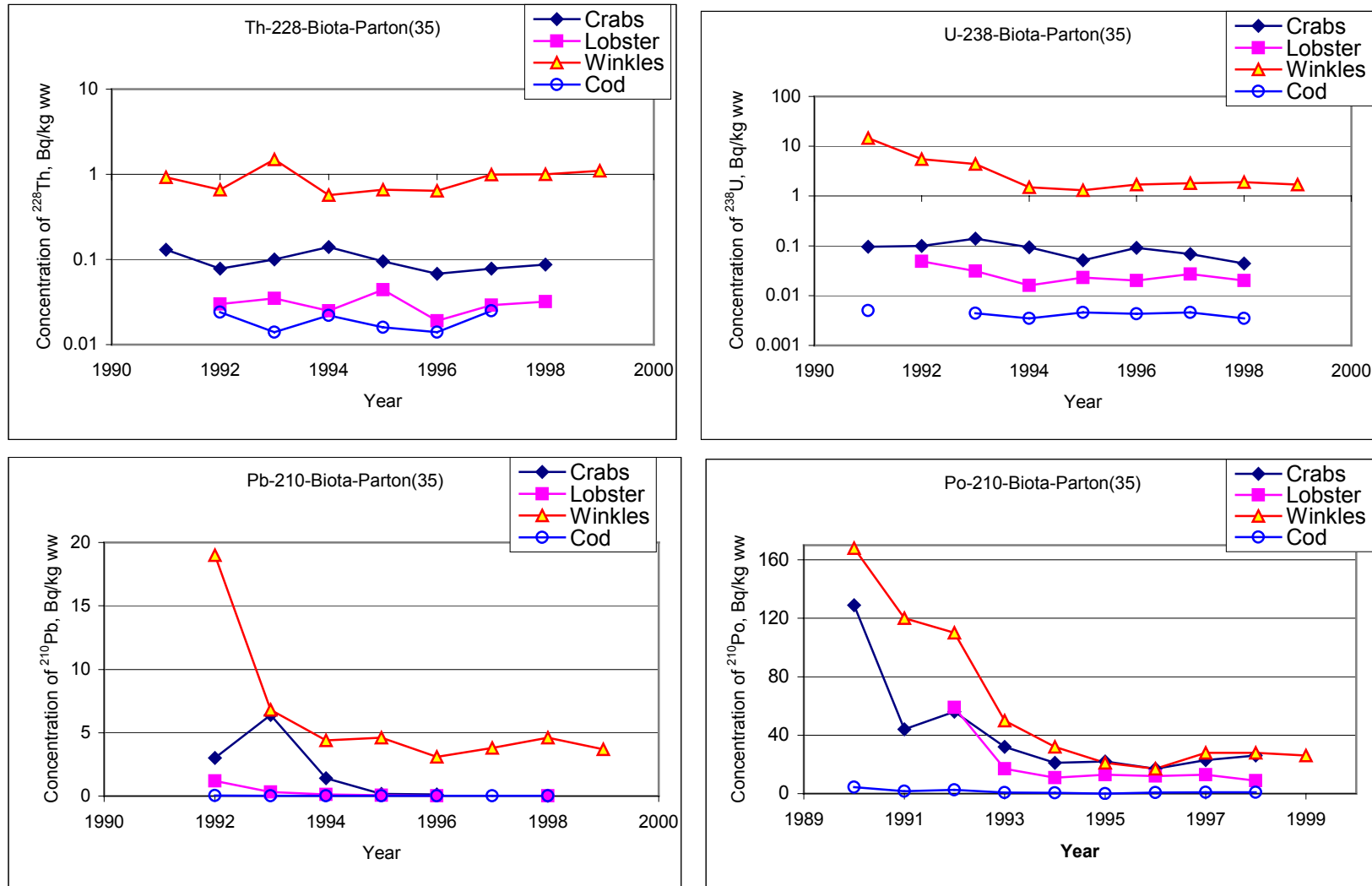
**Figure 29** Temporal variation of naturally occurring radionuclides in biota

Figure 29 (cont'd)

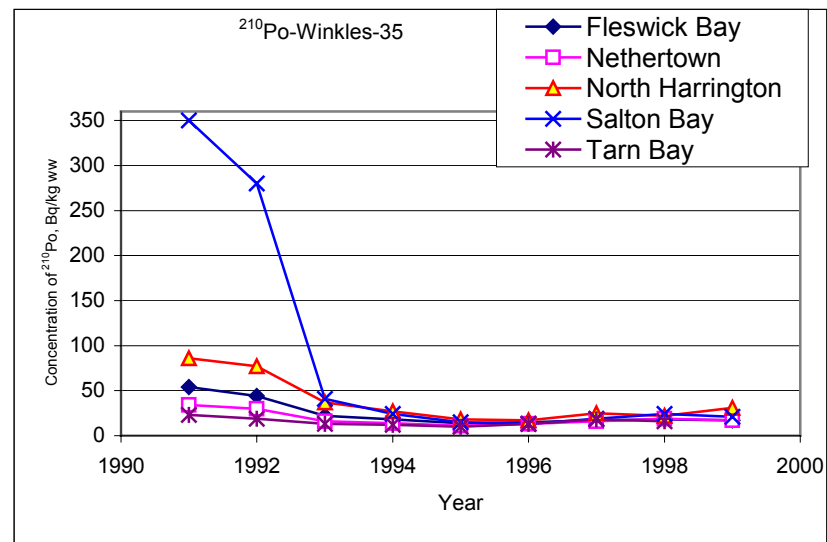
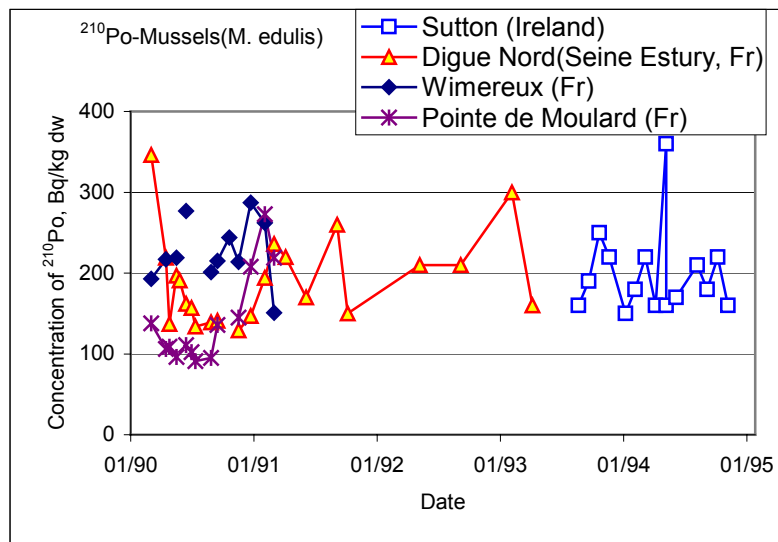


Figure 30  $^{137}\text{Cs}$  in surface waters of European seas (1976-1980).

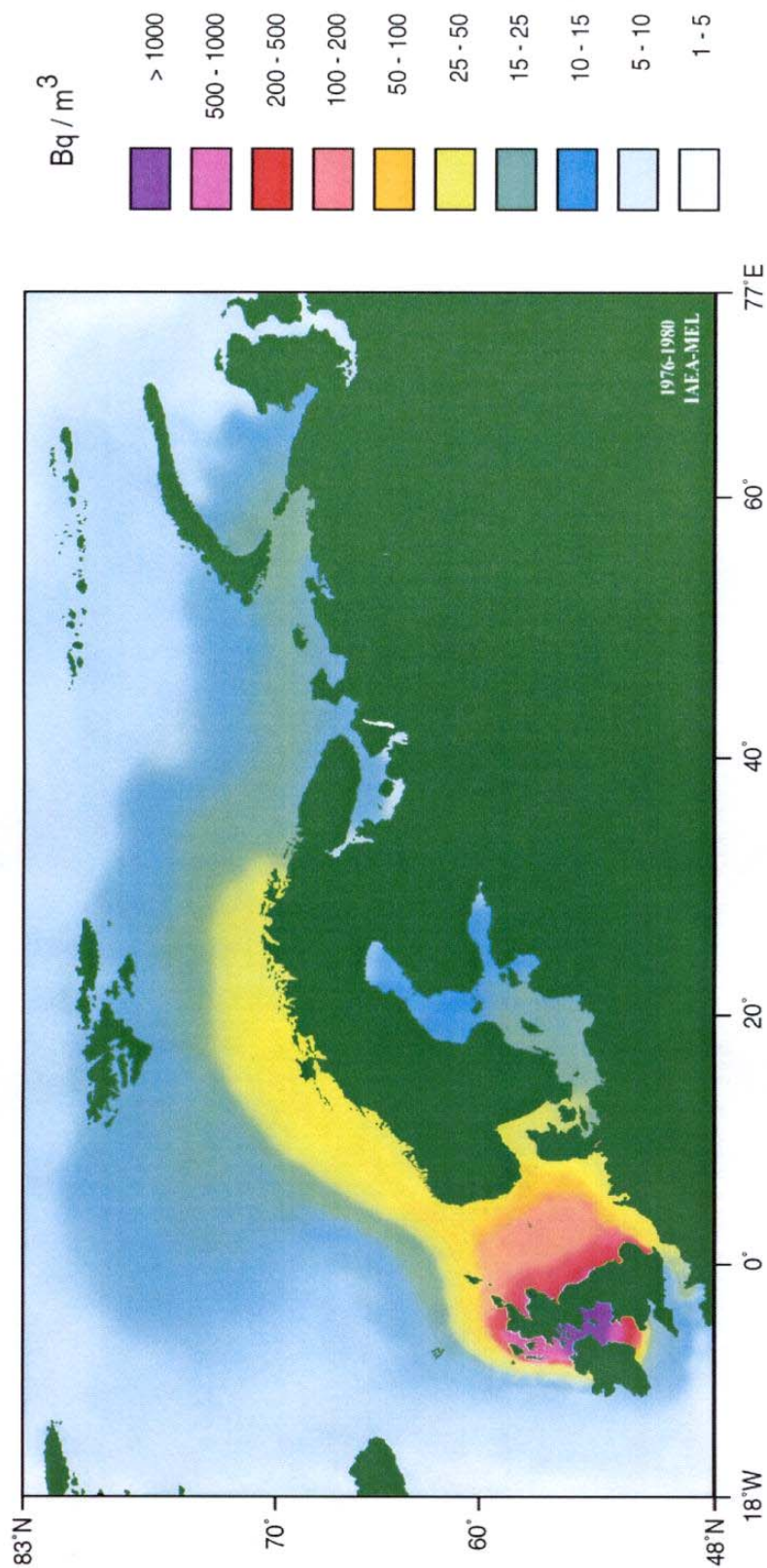


Figure 31  $^{137}\text{Cs}$  in surface waters of European seas (1981-1985).

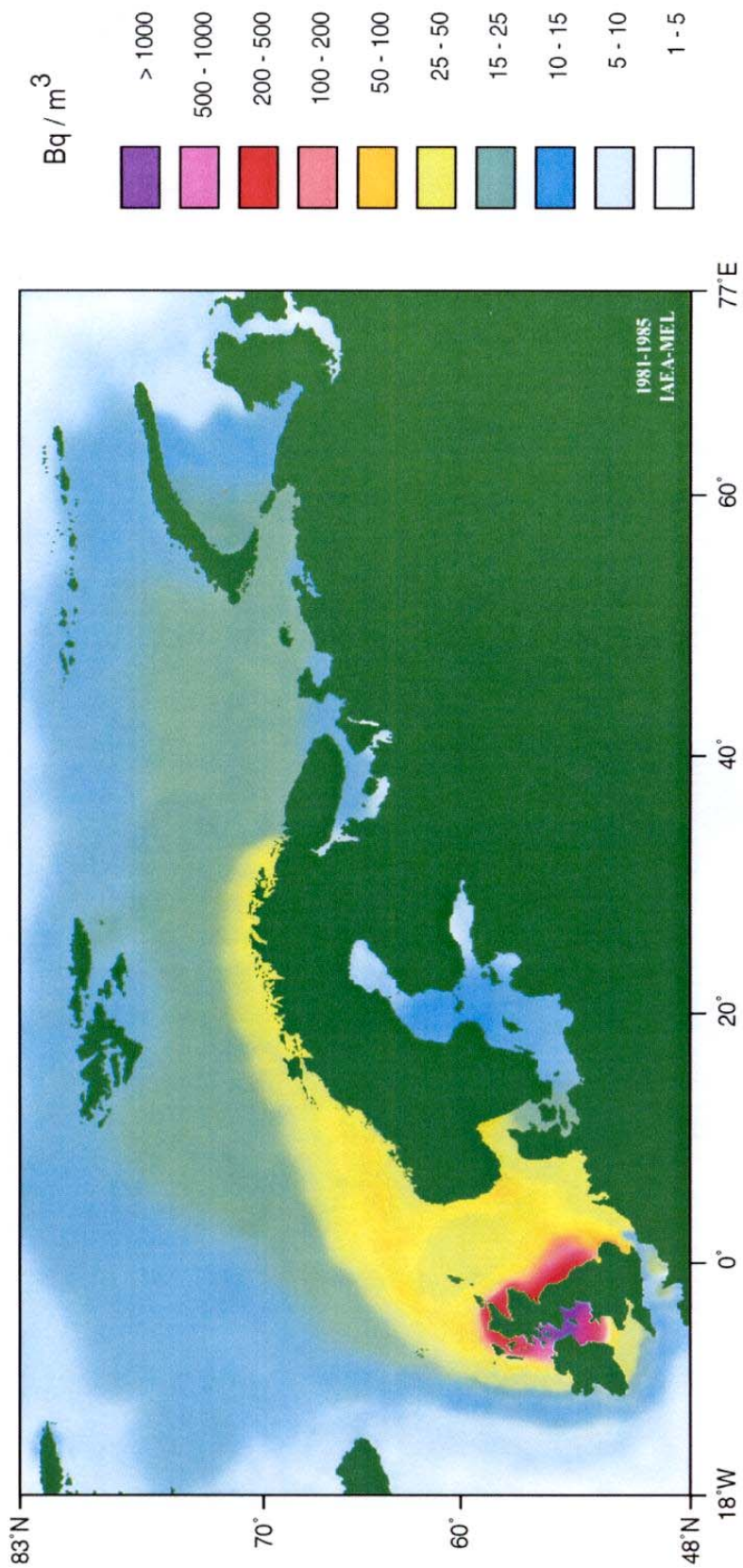




Figure 32  $^{137}\text{Cs}$  in surface waters of European seas (1986-1990).

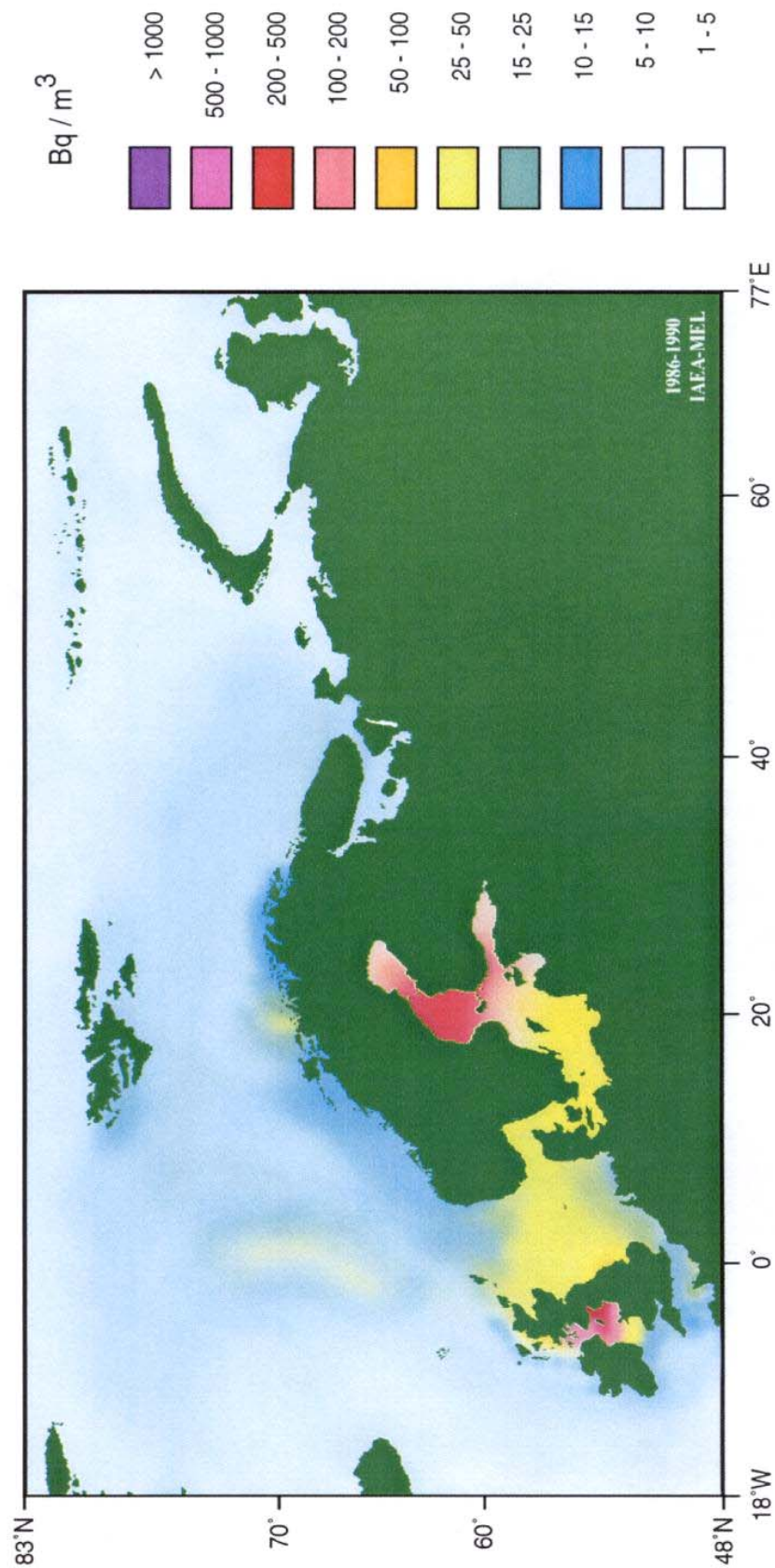
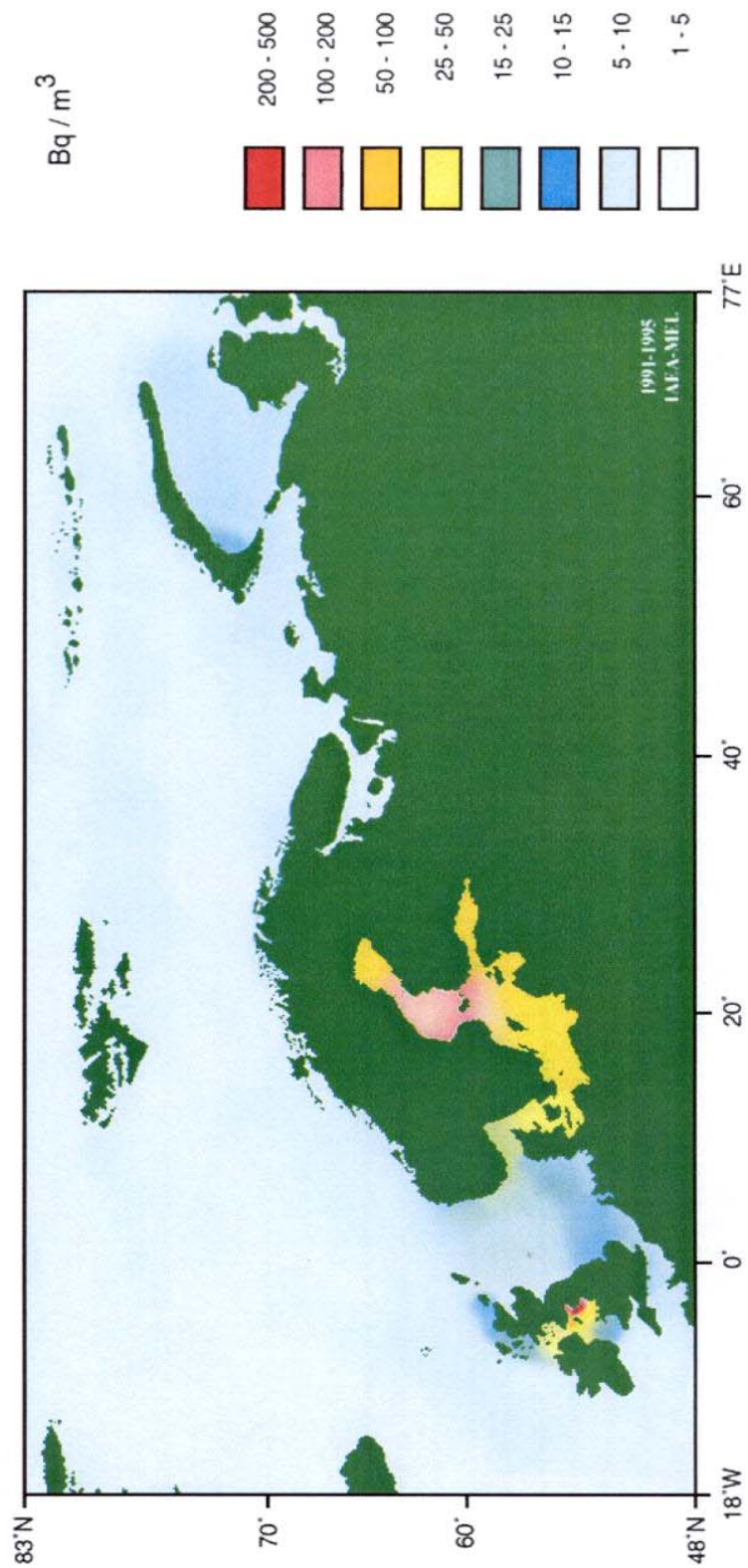




Figure 33  $^{137}\text{Cs}$  in surface waters of European seas (1991-1995).





**Abstract:**

The MARINA II study provides information on radioactive discharges to the North East Atlantic, on radionuclide concentrations in various marine environmental media and an assessment of their impact on humans and marine biota.

Compared to the mid 60's, by the end of the 1990s, the overall civil nuclear and other anthropogenic inputs of radioactivity into the North East Atlantic have decreased by several orders of magnitude for alpha- and beta-emitters and for tritium. The maximum levels were reached in the 1960s and early 1970s. Over the same time period this resulted in reductions in radionuclide concentrations in the marine environment and consequently in reductions in the individual doses to members of critical groups and in collective doses to the public.

Since the mid-1980s, the main contribution to discharges of beta-activity into the OSPAR region is from nuclear reprocessing plants (Sellafield and Cap de la Hague) while the discharges of alpha-activity have been dominated by the phosphate industry and by oil and gas production in the North Sea. As a result of the activities discharged and the higher biological effectiveness of alpha radiation, phosphate and oil production currently are the major contributors to collective dose to the population of the European Union from industrial activities. Other sources (e.g. production and application of radiopharmaceuticals, discharges from shipyards servicing nuclear submarines in the UK, historic dumping of wastes at sea and accidental releases other than Chernobyl) are comparably negligible.

Over the period 1988 to 1999 effective doses to critical groups in the Sellafield area show no downward trends partly due to the impact of remobilisation of plutonium isotopes from historic discharges, which are contained in the sediments of the Irish Sea, leading to relatively stable plutonium concentrations in sea water and thus in seafood. In addition, since 1994 the treatment of historic liquid wastes led to increased discharges of technetium-99, adding to the exposure. For the Cap de la Hague area there is a decreasing trend in effective doses since 1988. For the OSPAR region in general the doses to critical groups follow the same decreasing trend as the environmental radionuclide concentrations.

Doses to non-human biota due to industrial activities are low (order of magnitude of natural background level). Based on today's knowledge detrimental effects to populations of marine biota are not expected.

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