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Behaviour of Marine Pollutants Studied by Radioecological Methods

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by

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SUMMARY

The large scale testing of nuclear weapons in the fifties and the beginning of the sixties injected substantial quantities of radioactive substances to the atmosphere. These pollutants had not earlier been present in the environment, and intensive studies began in order to follow their behaviour. From these so-called radioecological studies much has since been learned about atmospheric processes, water movements, soil properties and biological systems.

In the last seven years methods for studying the marine environment, in particular the dispersion of waterborne radioactive pollutants have been developed under a programme sponsored by the Nordic Council of Ministers. The main sources of radioactive tracers in these studies have been nuclear reprocessing plants in Western Europe, but fallout from nuclear weapons testing and discharges from nuclear power plants in Scandinavia have also been studied.

The methods and some of the results from these studies have wider applications than those confined to radioactive pollution. It appears that some of the experiences may be used in the study of pollution of the marine environment in general. This report intends to demonstrate such perspectives on the basis of the knowledge acquired in previous Nordic work.

The application of radioecological studies on other pollutants may be accomplished by a number of different approaches.

- addition of radioactive tracers to the system to be studied (as an example, radioactive europium has been added to eels, in order to study their life cycle in nature).
- making use of radiotracers already present in the environment (e.g. using radiocesium and technetium from reprocessing plants in order to trace water movements).
- deriving analogue methods to those developed for radioactive isotopes (e.g. using bioindicators for non radioactive substances such as heavy metals).

As an example of the last approach, it can be mentioned that models have been set up to describe environmental pollution around a given source by means of bioindicator systems, preferentially using mussels and seaweed. Thus local, regional and semiglobal models for the dispersion of waterborne pollution in the North Atlantic region are now available. With proper adjustments these models are also valid for waterborne pollutants other than radionuclides. The models may then be considered to give conservative estimates of the concentrations of any waterborne pollutant in the marine environment.

The main advantage of radioecological methods in environmental studies is that the fate of various elements and the rate of various processes can be assessed without an increase in the concentrations of the elements to potentially toxic levels. This is done by means of models, which are validated by actual measurement. From such models it is possible to calculate the doses to man and thus to evaluate the impact of the pollution in question.

Such an evaluation is relatively straightforward for radioactive substances because here a direct proportional relationship between radiation dose and health hazard is assumed. For non radioactive pollutants such a relationship is often difficult (or impossible) to establish. Nevertheless some of the methods and the philosophy used within radiological protection could be applied to pollution problems in general. One aspect is the added knowledge about the actual path of pollutants through different foodchains ("critical pathways"). Another outcome could be the identification of those remedial measures which yield the greatest reduction in health hazard relative to cost.

This report contains 5 chapters. Chapter 1 contains a number of examples where the techniques and the results from Nordic marine radioecological studies may be used. Thus it is shown how it is possible to calculate the dispersion of waterborne pollu-

tion from the North Sea to the fishing grounds at Lofoten, the Faroe Islands, Iceland and Greenland. It is also demonstrated how the transport and dilution of pollution from the "Øresund Metropol" (Copenhagen/Malmö) may be traced in the Cattegat, and how the pollution of the Baltic Sea by North Sea water may be followed.

Chapter 2 deals with bioindicators i.e. organisms, which have been used for the monitoring of radioactive pollutants. It is shown how it is possible to use such systems (e.g. seaweed) along with a specially developed model for the monitoring of liquid discharges.

Chapter 3 shows how the radioactive discharges from the British reprocessing plant, Sellafield have been used to trace water masses in the North Atlantic region. It has been possible to follow the effluent over distances of nearly 10.000 km in the ocean. On this basis dilution factors and transit times for pollution in the northern North Atlantic can be calculated.

Chapter 4 gives a number of examples of the application of radioecological methods in general ecology and in pollution studies. These methods have e.g. been used for studying the production of organic matter (carbon-14), microchemical processes, bioturbation in sediments, and metabolism in various organisms.

The last chapter contains a proposal for the actual transfer of methodology from radioecology to other fields. As a first step a seminar is suggested with participants from science, authorities and industry, together with pollution experts from inside and outside the Nordic countries. The next step would involve the formulation of proposals for projects, followed by implementation.

Sammanfattning (Summary in Swedish)

De storskaliga kärnvapenproven under 50- och början av 60-talet medförde en spridning av betydande mängder radioaktiva ämnen till atmosfären. Denna typ av förorening, som inte tidigare uppträtt i miljön, medförde intensiva studier av dessa ämnens beteende. De s k radioekologiska studierna har lett till ett ökat vetande om atmosfäriska processer, vattentransporter, markegenskaper och biologiska system.

Under de senaste sju åren har metoder utvecklats för studier av den marina miljön och speciellt spridningen av de vattenburna radioaktiva föroreningarna genom ett program, som erhållit stöd av Nordiska Ministerrådet. De huvudsakliga källorna till de radioaktiva spårämnena har varit upparbetningsverk i västra Europa men även nedfallet från kärnvapenproven och utsläppen från kärnkraftverk i Skandinavien har studerats.

Metoderna och några av resultaten från dessa undersökningar har en vidare tillämpning än det, som begränsas av radioaktivitet. Erfarenheterna kan utnyttjas för studier av allmänna föroreningar i den marina miljön. Denna rapport, som baseras på de erfarenheter, som vunnits i det nordiske arbetet, är tänkt att demonstrera sådana aspekter.

Tillämpningen av radioekologiska studier för andra förorenande ämnen kan genomföras på skilda sätt.

- tillsats av radioaktiva spårämnen till det system, som skall undersökas (t ex märkning av ål med radioaktivt europium för att studera beteende, tillväxt, mm i naturen).

- utnyttja redan existerande radioaktiva spårämnen i miljön (bl a radiocesium och teknetium från upparbetningsverk för studier av vattentransporter).

- tillämpning av metoder analoga till de, som utvecklats för radioaktiva isotoper (bl a användningen av bioindikatorer för icke radioaktiva substanser som tungmetaller).

Som exempel på den sista ansatsen kan nämnas modeller, som utvecklats för att beskriva föroreningar i miljön från en given källa genom ett system av bioindikatorer, främst musslor och blåstång. Lokala, regionala och semiglobala modeller för spridning av vattenburen förorening i Nordatlanten är nu således tillgängliga. Med lämpliga justeringar gäller dessa modeller även för andra föroreningar än radionuklider. Modellerna ger därmed ett konservativt mått på koncentrationen av varje annan vattenburen förorening i den marina miljön.

Den huvudsakliga fördelen med radioekologiska metoder i miljö studier är att de enskilda ämnenas öde och de olika processernas hastighet kan bestämmas utan att koncentrationerna av ämnena behöver ökas till potentiellt toxiska nivåer. De modeller, som tillämpas, kan verifieras med directa mätningar. Från dessa modeller är det möjligt att beräkna doser till människan och därmed uppskatta verkan av den aktuella föroreningen.

En sådan utvärdering kan göras tämligen direct för radioaktiva ämnen då man här kan räkna med ett direkt, proportionellt förhållande mellan stråldos och hälsorisk. För icke radioaktiva föroreningar är ett sådant förhållande ofta svårt (eller omöjligt) att slå fast. I vilket fall bör dock metoder och filosofi, som tillämpas inom strålskyddet, även kunna tillämpas på allmänna föroreningsproblem. En aspekt är den ökade kunskapen om den väg en aktuell förorening tar genom olika näringskedjor ("critical pathway"). Ett annat utfall kan vara möjligheten att identifiera sådana åtgärder, som innebär största möjliga reduktion av hälsorisk i relation till kostnad.

Denna rapport innehåller 5 kapitel. Kapitel 1 ger ett antal exempel där teknik och resultat från de internordiska radioekologiska studierna kan tillämpas i marin miljö. Sålunda har möjligheterna påvisats att beräkna spridningen av vattenburen förorening från Nordsjön till fiskeområdena vid Lofoten, Färöarna, Island och Grönland. Här påvisas också möjligheterna att spåra transporten och utspädningen av föroreningar från Öresundsmetropolen (Köpenhamn/Malmö) till Kattegatt och hur man kan följa föroreningar från Nordsjön til Östersjön.

Kapitel 2 behandlar bioindikatorer, dvs organismer, som har använts för monitoring av radioaktiva ämnen. Möjligheten påvisas att använda sådana system (t ex havsalger) samman med en speciellt utvecklad modell för rutinuppföljning av vätskeformade utsläpp.

Kapitel 3 visar hur de radioaktiva utsläppen från det brittiska upparbetningsverket vid Sellafield kan användas för att spåra vattenmassorna i den nordatlantiska regionen. Det har varit möjligt att följa utsläppen över en distans av nära 10.000 km i havet. På basis av detta kan utspädningsfaktorer och transporttider beräknas för föroreningar i norra Atlanten.

Kapitel 4 ger ett antal exempel på tillämpningar av radioekologiska metoder i allmän ekologi och föroreningsstudier. Sådana metoder har bl a använts för studier av produktionen av organiskt material (kol-14), mikrokemiska processer, bioturbation i sediment och ämnesomsättning hos olika organismer.

Det sista kapitlet innehåller förslag till överföring av metoder från radioekologi till andra aktuella områden. Som ett första steg föreslås ett seminarium med deltagare från vetenskap, myndigheter och industri, tillsammans med experter på föroreningar inom och utom de nordiska länderna. Nästa steg innebär en formulering av förslag till projekt för genomförande.

1. POSSIBLE PRACTICAL APPLICATIONS

In this chapter some possible practical applications of radioecological results and methods for aquatic pollution management in general are summed up.

Chapter 2, 3 and 4 give further examples and details.

Pollutant transport from the North Sea to the Arctic

The Sellafield discharges have led to calculation of transport times and transfer factors from the point discharge at Sellafield to several water bodies, e.g. the North Sea, the Baltic, the west Norwegian waters, the Barents Sea, the East Greenland Current and even the West Greenland Current. The results are calculated for elements that behave approximately conservatively*) (Technetium and Caesium), and can be used directly for other conservative substances. As an example it can be mentioned that a conservatively behaving substance well mixed in the North Sea will appear at the fishing grounds at Lofoten, West Norway, within a year later at approximately half the concentration, and at the south west Greenland fishing banks approximately 5-6 years later in 1-2% of the North Sea concentration (see chapter 3). The currents transferring pollutants from Northern Europe to Greenland do not directly approach Iceland and the Faroe Islands. The fishing banks at these countries will therefore generally be less effected by water-born pollutants from Europe than Green-

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*) Conservative: i.e. elements that are transported fully dissolved in the water without being sedimented, evaporated or degraded. Technetium is ideally conservative, whereas caesium shows a certain sedimentation.

land waters, although the latter are farther away than the former. For point discharges to the North Sea, as e.g. a chemical waste dumping, an oil-platform release or a river discharge the pollutant concentration should be calculated after dilution with the volume of the North Sea in order not to overestimate the resulting concentrations in other water bodies.

If sedimentation constants and degradation rates are known the results will even be applicable for non-conservative pollutants.

Pollution of the Baltic with North Sea effluents

The Baltic environment is more susceptible to pollutant discharges than most other marine environments. One reason for this is that the water turnover rate is low. The mean residence time has been calculated to 25 years based on current measurements and on radionuclide studies (^{137}Cs and ^{90}Sr). It is of interest for the Paris Convention[+)] as well as the Helsinki Convention[++)] to calculate mass balances of pollutant transports between the Baltic and the North Sea. This transit between the convention areas can be studied more in detail when making use of radiotracers from the nuclear industry (cf. chapter 3).

Transport and dilution of pollutants from the "Øresund Metropol"

Using ^{60}Co-discharges from the Barsebäck Nuclear Power plant as a tracer, patterns of transport and dilution from the Sound northwards into the Cattegat has been investigated (cf. chapter 3).

+) Convention on the prevention of marine pollution (of the North Sea) from land-based sources.

++) Convention on the protection of the marine environment of the Baltic Sea area.

For pollutants discharged on the Swedish side above the distinct density gradient, the results can be used directly provided the substances in question are not less conservative than the discharged ^{60}Co. Also non conservative pollutants can be included by applying the relevant sedimentation constants and biodegradation rates. The results will even be applicable for discharges at other locations, e.g. from the Copenhagen area, if the initial dilution is determined e.g. by conventional short range dye or tracer techniques. As an example based on the Barsebäck discharges it can be stated as a conservative estimate that if 1000 kg of a pollutant were released monthly into the Sound surface waters, approximately 0.01 mg/m^3 would be found in Kattegat 100 km north of the release, or in other words, the monthly release would be diluted in 100 km^3 or 10^{11} m^3 of water.

Biotechnology in monitoring of pollutants from industrial sources

Marine bioindicators, especially mussels and seaweeds, are in use in monitoring not only radionuclides but also stable metals, chlorinated hydrocarbons and petroleum hydrocarbons.

A model describing the expected level of radionuclides in a bioindicator as a function of discharge and month of discharge (cf. chapter 2), developed and tested on basis of laboratory and field work performed during the Nordic radioecology projects, is directly applicable to other pollutants monitored by bioindicators. The model has been verified for radionuclides in Fucus vesiculosus at the Ringhals nuclear power site, and if the appropriate rate constants for accumulation, loss and growth are included it can be applied for other substances and for other bioindicators as well in addition to estimating expected concentrations the model will indicate the period of time after discharge, when the pollutant is still measur-

able in the bioindicator. It will thus be helpful in planning appropriate sampling intervals and when deciding whether observed variations in the bioindicator response should be explained by deviations from reported discharge rates or by natural variations.

The results obtained during this work has improved the usefullness of bioindicators to be a highly valuable quantitative tool in monitoring radioactive nuclides, and the same principles could easily improve the usefullness of bioindicators for non-radioactive pollutants as well.

2. BIOINDICATORS

Introduction

This chapter deals with a special branch of biotechnology, namely the use of living organisms in monitoring pollutants.

In this context bioindicators are organisms whose contents of a certain pollutant are used to indicate the level of that pollutant in the sampling area. (Dahlgaard, 1981). In ecotoxicology the word bioindicator is also used in other connections which are not considered here.

Bioindicators are used to monitor the transport of certain pollutants, their distribution and time trends. They have several advantages above water measurements two of which will be mentioned here: - due to accumulation, the detection limits for several substances are lovered considerably, and - due to slow loss rates, fluctuating levels are integrated over a certain period of time.

Both these effects appears from Figure 2.1, where monthly liquid discharges of ^{65}Zn from a nuclear power plant are compared with the measured values of ^{65}Zn in the seaweed Fucus vesiculosus. It appears that due to accumulation the amount of ^{65}Zn in one kg of seaweed equals the amount in several thousands of liters of water. Furthermore it is seen that the high concentrations observed just after a high discharge decrease more slowly than the decrease in discharges (the integration effect).

Seaweeds and Mussels

In the studies aimed at evaluating bioindicators for monitoring of radioactive discharges from nuclear power plants, Fucus vesiculosus and the soft parts of the common mussel, Mytilus edulis,

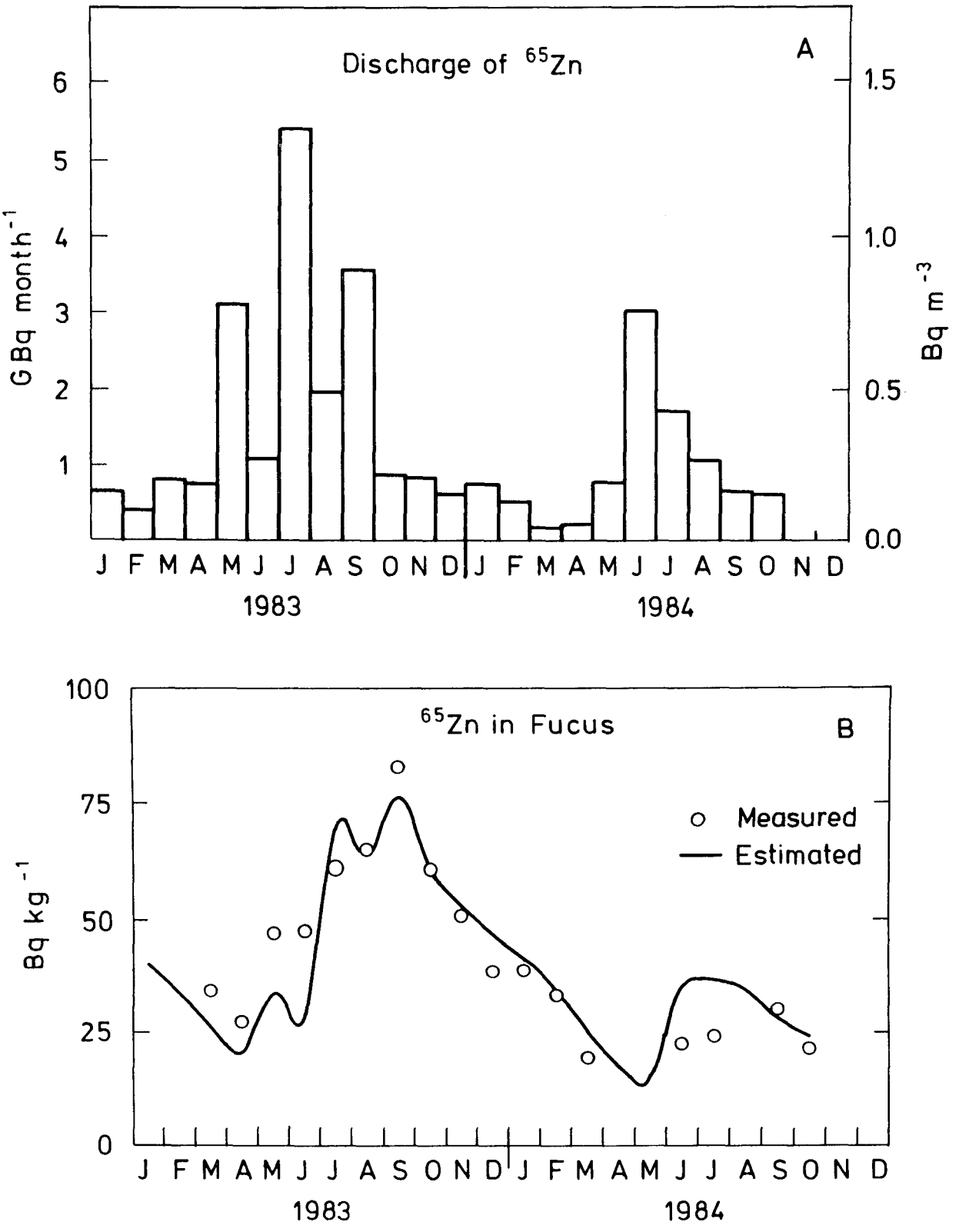


Fig. 2.1. Monthly discharge of ^{65}Zn from Ringhals (A) and modelled and measured concentrations of ^{65}Zn in Fucus vesicolusus sampled 2.3 km from the discharge (B). The scale on the discharge values is given both as total discharge (GBq month^{-1}) and equivalent concentrations in water (Bq m^{-3}) at the seaweed sampling site.

have been chosen as the favoured species. Their great cumulative capacity and their wide distribution and abundance at all seasons have supported this choice.

Fucus vesiculosus, the well known "bladder wrack", is found in huge masses in shallow waters along most marine and brackish water coasts in Northern Europe. Due to a special chemical composition, and physiology, Fucus vesiculosus and some related brown algae accumulate several elements to high concentrations. Fucus vesiculosus plants normally obtain an age of 3-5 years.

Mytilus edulis, the common edible mussel, is likewise very widespread in marine and brackish coastal waters. The mussels act as a filter for huge amounts of water retaining particles down to approximately 1 µm. Thus, a relatively small mussel with 200 mg dry weight in its soft parts will typically filter 50 liters of water a day. This behaviour results in a special sensitivity for pollutants associated with particles.

Fish and shellfish

In the Nordic countries the consumption of seaweed and mussel is low. In monitoring programmes aimed at protecting man it would therefore be desireable to include fish and crustaceans.

Unfortunately however, many commercially important fish species are so mobile that their value in monitoring is very limited. Also, for several radionuclides and metals, the concentration in fish is so low that the measurements are difficult to perform. However, by including surveys of pollutant levels in fish and shellfish in a seaweed- or mussel-programme, the relevant ratios between fish and the bioindicators can be estimated. It is thus possible to use one of the bioindicators for routine-monitoring and subsequently estimate the order of magnitude for the level in fish. This method has been used in assessing the transport of ^{60}Co from Barsebäck nuclear power plant to man via fish (cf. Boelskifte 1985).

Concentrations factors

Table 2.1 shows some examples of concentration factors estimated for Fucus and Mytilus. Here, the concentration factor is expressed as the amount of pollutant in one kg of dry bioindicator divided by the amount of the same substance in one liter of water. These values are approximately 5 times higher than similar values on a fresh weight basis. Ideally, concentration factors should be given on fresh weight basis, but due to variations in the water content, values based on dry weight are less variable. For some elements effects of salinity and season have been observed, while for others unexplained differences between different locations have been found. It is probable that the chemical state of the elements plays a role (see chapter 4). The table will therefore essentially give the order of magnitude to which some elements are concentrated in the bioindicators relative to water.

Table 2.1. Concentration factors on dry weight basis (Bq kg^{-1} dry weight/Bq l^{-1} of water) for several elements in the seaweed Fucus vesiculosus and soft parts of the mussel Mytilus edulis. The data can vary with environmental parameters e.g. salinity and location. Values on fresh weight basis will be approximately 5 times lower.

All values in thousands

	^{99}Tc	^{137}Cs	^{239}Pu	^{241}Am	^{60}Co	^{54}Mn	^{65}Zn	^{110m}Ag	^{51}Cr
Fucus	100	0.2	10	10	70	80	300	400	5
Mytilus	0.5	0.1	2	4	25	15	350	300	-

Due to the very low concentration of radionuclides in the sea it is generally necessary to utilize much bigger samples than in studies of stable metal or other pollutants. Often 1-2 kg are needed for one measurement. It is usually more difficult to sample and prepare 1 kg mussel soft parts than several kilos of seaweed, and Fucus shows higher concentration factors than Mytilus soft parts for several elements. Therefore, Fucus has mostly been used in the Nordic studies, whereas the use of Mytilus is internationally more widespread.

Loss rates, growth rates, and time-integration

Not only the concentration factor is of importance for the use of a bioindicator. The rate of loss of pollutant from the bioindicator as described by the biological half life and the growth rate of the organisms giving rise to a dilution of the pollutant concentration with newly formed biomass, are responsible for the so-called time integration. If the loss and the growth is slow, the integration time is long, and if the loss and/or the growth is fast, the bioindicator will only "remember" a high pollutant level for a short time after the level in the water has gone down.

Figures 2.2 and 2.3 illustrate some differences in loss rates between different elements and between mussels and seaweed. In the mussel experiment a distinct seasonal variation was seen. During winter, the loss stopped completely, whereas it started again next summer. In the seaweed experiment, several elements were retained almost completely (Zn, Co, Mn, Tc, Ag) whereas Europium was lost very fast. In the mussel experiment, Europium and Americium, which are chemical analogues, were lost very slowly. It is very important to consider such differences when interpreting bioindicator results and when designing bioindicator programmes.

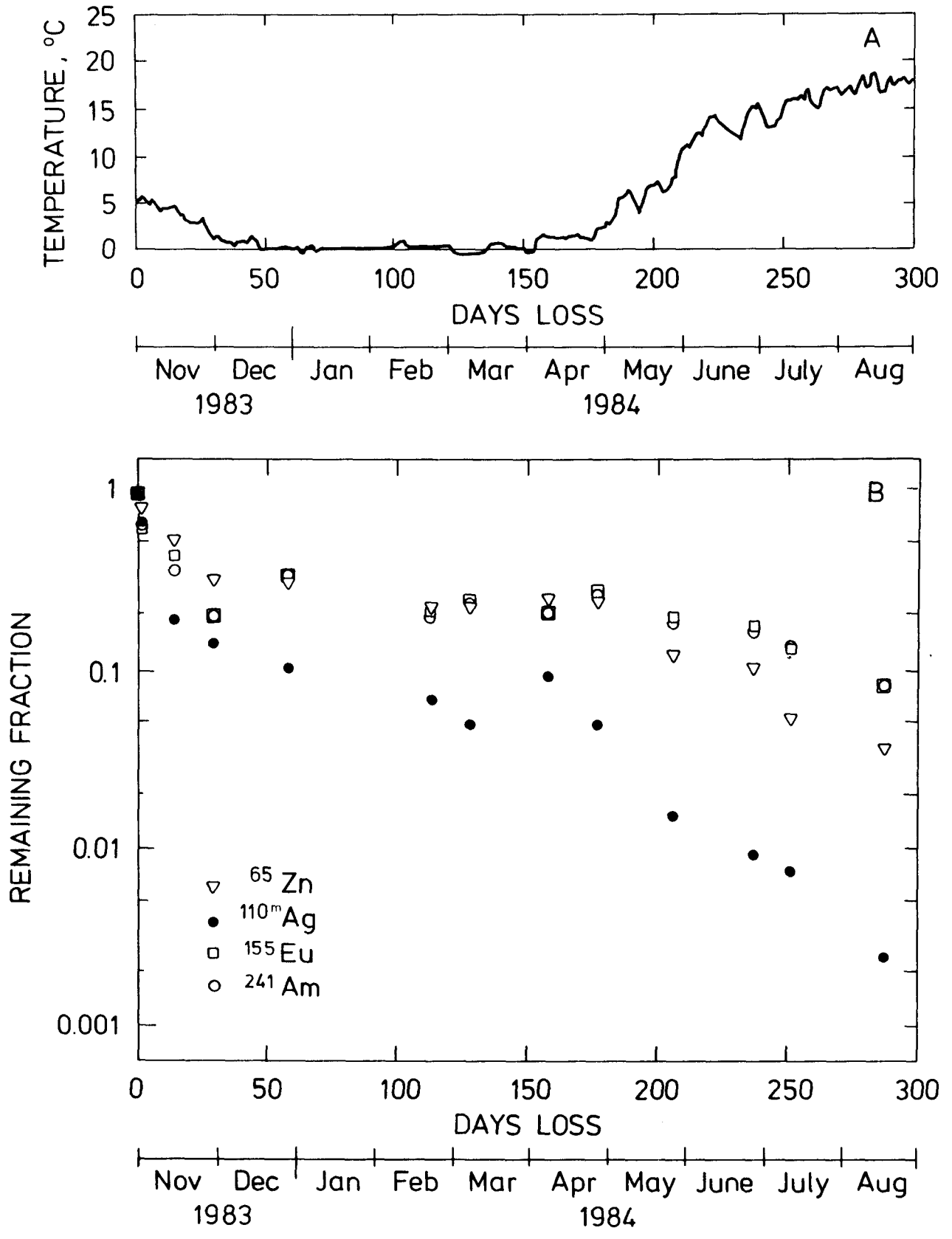


Fig. 2.2. Loss of activity from mussels (Mytilus edulis) in the cooling water intake channel at Forsmark on the coast of the Bothnian Sea after experimental contamination in the laboratory (B), and temperatures in the water during the experiment (A).

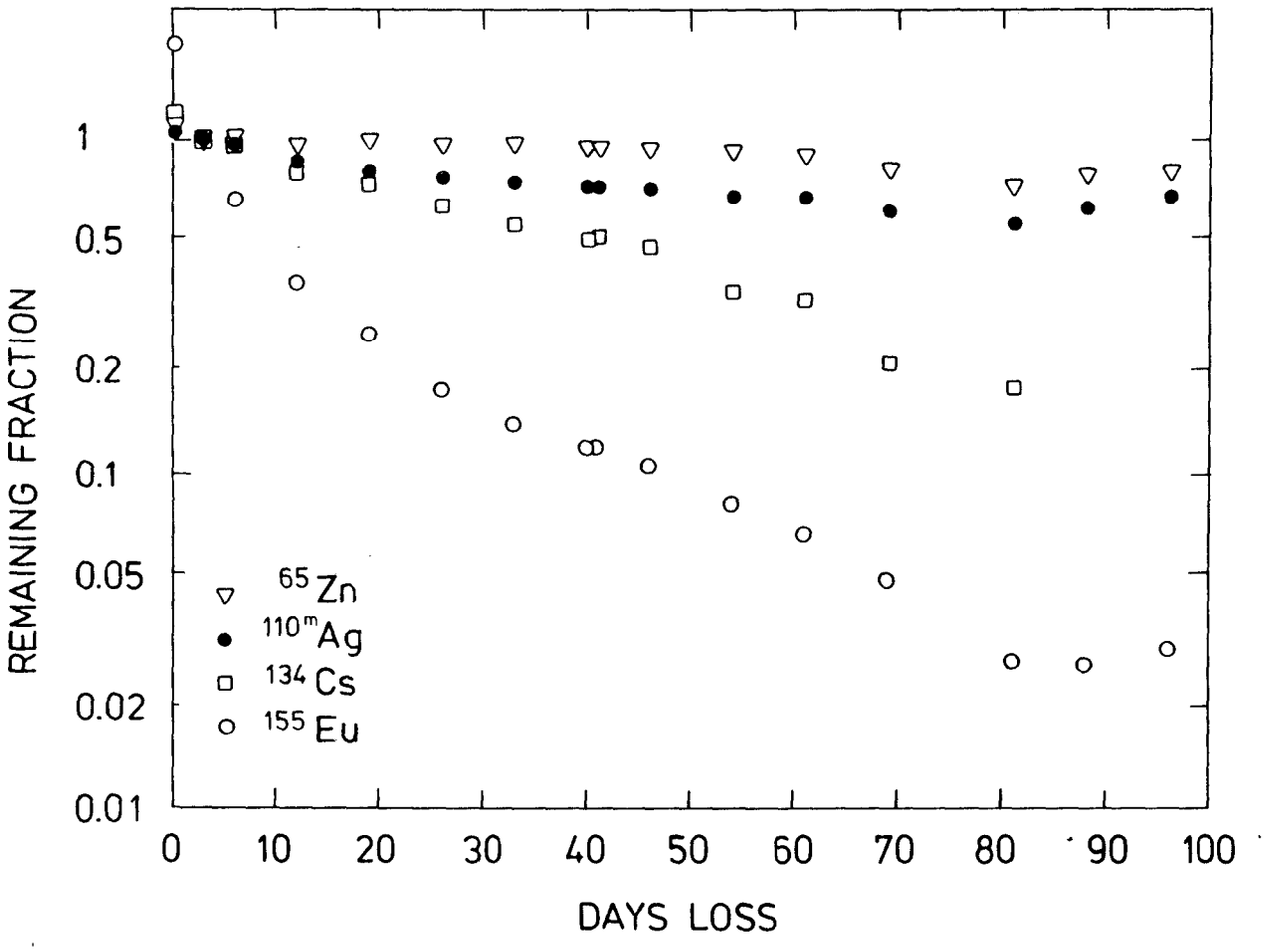


Fig. 2.3. Loss of activity from whole Fucus vesiculosus seaweed plants after experimental contamination in the laboratory.

A model describing the function of bioindicators: SENSI

The bioindicator response is influenced by seasonally varying environmental parameters such as light, temperature, salinity etc. The parameters affect accumulation rates, loss rates and growth rates. A model is now available to describe the accumulation and loss of several radionuclides by Fucus vesiculosus as a function of month of discharge and month of sampling. Originally this model was developed to describe the function of Fucus as a bioindicator for the varying liquid discharges of radionuclides from a nuclear power plant. The principle is equally valid for other pollutants. The model utilises information on seasonal variation in rate of accumulation, rate of loss, and growth

rates, and for the radionuclides, physical half lives. The estimated values in Figure 2.1 were calculated on the basis of monthly discharge values by means of SENSI. It appears that for the summer months when dilution due to growth of the plants, and loss of elements from the bioindicators is at its highest, the bioindicator integrates the pollutants over a shorter time than in winter. This means that a peak discharge in the autumn of some elements can be monitored in Fucus till next spring, whereas a spring peak discharge may be detectable for a few months only.

Monitoring and trends

From the information mentioned above, it is possible to design a monitoring programme near a discharge in such a way that no major discharges of e.g. metals or radionuclides can pass by unattended provided adequate information exists on the behaviour of the discharged substances in relation to the indicator. As variations in bioindicator behaviour is not only restricted to reproducible seasonal changes, it will of course always be desireable to have reliable discharge reports, as is the case for peaceful nuclear installations. Relating the monitoring values to discharge values can then give important information to be used in case of an accidental uncontrolled discharge and in monitoring substances from diffuse, uncontrolable sources.

In order to distinguish trends in the pollutant level from variations due to e.g. season and from unexplained stochastic variations between years it is very important to establish reliable time-series of observations over several years. Long time series have now been established for a few substances, mainly radionuclides, and similar work with other pollutants would be of significant help in environmental protection work.

3. DISPERSION STUDIES OF MARINE POLLUTION

Introduction

The use of radioactive tracers is a well-known methodology within science and medicine. When radiotracers are to be deliberately used in environmental studies the authorities first need to give their permission. If the work is limited to the use of relatively small amounts of short-lived radionuclides, permission will usually be granted. This limitation implies that it generally will not be possible to follow the dispersion of pollutants by radiotracers in the marine environment over long distances and over many years.

In the last decades authorized discharges from nuclear installations have, however, given new possibilities for carrying out radiotracer studies in the marine environment. In the following, some of the results obtained during a four-year Nordic study (Aarkrog, 1985) are presented and the perspectives for marine pollution studies in general are indicated.

The Sellafield tracer experiment

British Nuclear Fuels Limited operates the Sellafield (formerly Windscale) reprocessing plant in the UK. The authorized annual discharges from this installation (decay-corrected to 1983) have been about 3.3 PBq ^{137}Cs (1 PBq = 10^{15} Bq), 0.073 PBq ^{134}Cs, and 0.34 PBq ^{90}Sr during the period 1974-1981. Furthermore approximately 0.04 PBq ^{99}Tc have been discharged annually during the seventies. In recent years these radionuclides have been detected in nearly all parts of the northern North Atlantic. (Casso & Livingston, 1984, Aarkrog et al., 1983, Holm et al., 1983) The activity may be used to give important information on the dispersion of waterborne pollutants in the marine environment.

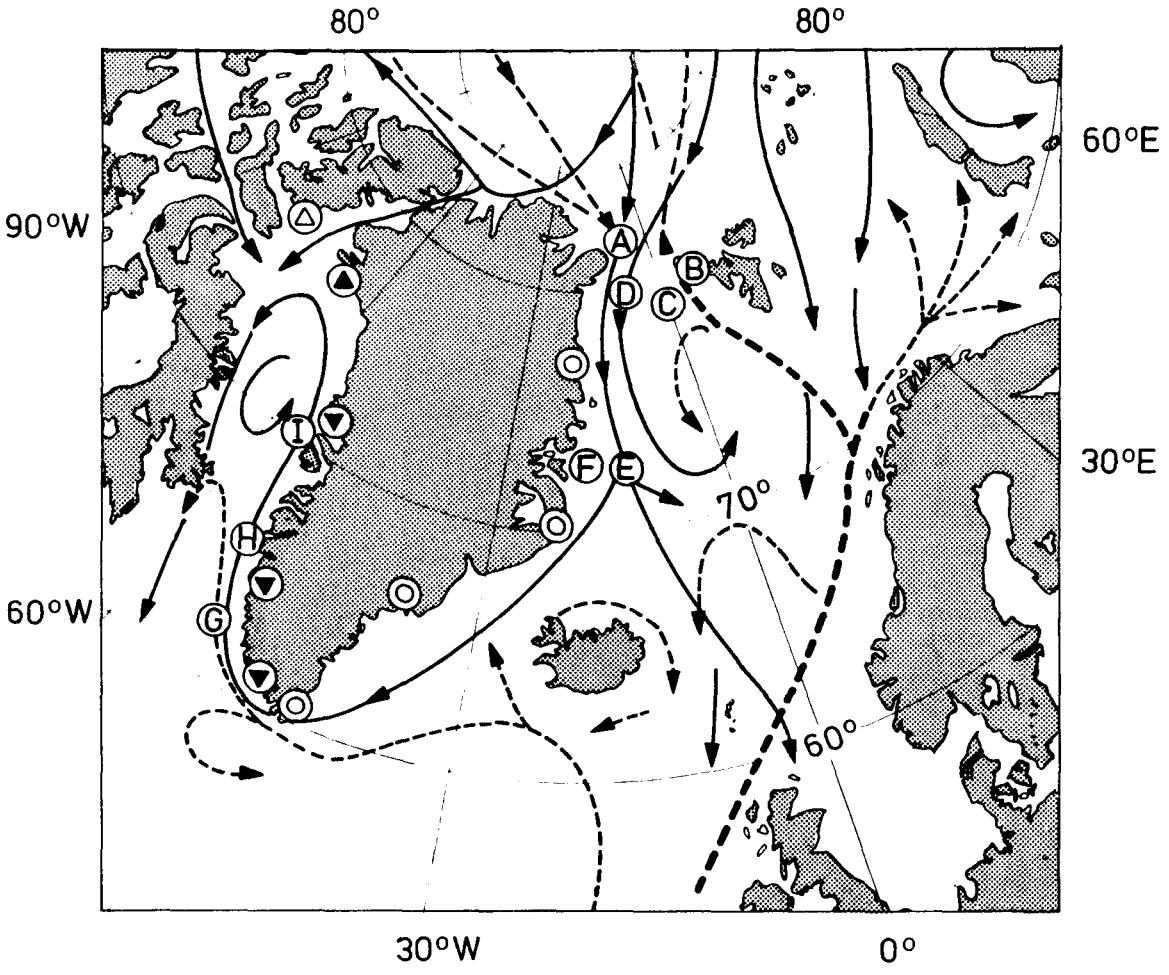


Fig. 3.1. Main sea current in the North Atlantic and Arctic Oceans (dotted lines: warm, solid lines: cold) (After Rey, 1982). Furthermore, the map shows sampling locations for seawater and Fucus around Greenland. Sellafield discharges the radioactive wastes to the Irish Sea. From there they flow northward along the British west coast, around Scotland, then turns southward into the North Sea, where they run eastward. Some of the activity enters through the Skagerrak, the Danish Straits, and the Baltic Sea. But the main part turns northward and continues with the Norwegian Atlantic Current along the Norwegian west coast. Reaching northern Norway a branch of the current turns northward as the West Spitzbergen Current. The remainder goes eastward where it follows the Russian north coast. The West Spitzbergen Current enters the Arctic Ocean through the Fram Strait between Svalbard and North-East Greenland. From the Arctic Ocean emerges the East Greenland Current, which runs southward along the Greenland east coast. At Kap Farvel it turns northward after mixing with the Irminger Current coming from the southeast and turns into the West Greenland Current. This current follows the Greenland west coast up to Baffin Bay where a branch turns westward joining the southward heading Labrador Current. The remaining part of the West Greenland Current continues up into the Nares Strait between North West Greenland and Ellesmere Island in Canada.

It has been possible to follow the radioactive tracers from Sellafield the entire way to Thule in North-West Greenland, i.e. over a distance in the order of 10 000 km along the current (see Fig. 3.1.)

Transfer factors

The transfer of a radionuclide (or another pollutant) from a source to a sample collected at a given location is expressed by the so-called transfer factor.

This factor is defined as the ratio of the infinite time integral of the radionuclide concentration in the sample to the total discharge of this radionuclide from the source. In other words if a source has discharged A Bq and the infinite time integral in seawater at the location is B Bq $m^{-3}a$ the transfer factor becomes B/A $m^{-3}a$. In the steady-state case, where annual discharges of α Bq a^{-1} and an equilibrium concentration at the location of β Bq m^{-3} are assumed, the transfer factor becomes β/α $m^{-3}a$. It is a prerequisite for making these calculations that there is no other source of the pollutant than the one considered.

In order to determine transfer factors from Sellafield to watermasses in the northern North Atlantic, the radionuclide ^{134}Cs has been used. This isotope was chosen because ^{137}Cs, as well as ^{90}Sr, are present in the North Atlantic as a significant fallout background. This background is not uniform. Higher concentrations are thus found in coastal waters, e.g. the Baltic Sea, and in Arctic waters, e.g. along the East Greenland Coast, than in the open ocean. Cesium-134 does not contribute to the fallout background at present. However, there are two sources of ^{134}Cs: Sellafield and Cap de la Hague in France. The contribution from Cap de la Hague is a few percent of that seen from Sellafield and may thus be neglected at distances remote from the discharge point. However, in the south-eastern part of the North Sea the contribution from Cap de la Hague is noticeable.

Figure 3.2 summarizes the transfer factors from Sellafield to various water bodies in the northern North Atlantic. The transfer factors are determined from ^{134}Cs measurements but we have corrected for radioactive decay of ^{134}Cs. Hence the factors are applicable for any substance that behaves similar to cesium in the marine environment. In other words, if it is assumed that the Irish Sea is polluted with a substance discharged at a rate of 1000 kg per year, one would expect an amount of the pollutant on the order of

$$0.1\text{-}1 \text{ ng } (1 \text{ ng} = 10^{-9}\text{g})$$

in 1 cubic meter of surface water collected at Greenland.

Bioindicators

The concentration of the radiotracers are often very low at distances far from the discharge point. This is, e.g. the case for ^{99}Tc, which is the other radiotracer beside ^{134}Cs which have been studied. Technetium 99 is concentrated in Fucus vesiculosus. The concentration factor is 100 000, i.e. 1 kg Fucus (dry weight) contains the same amount of ^{99}Tc as 100 m^3 of the seawater in which the plant has been growing (cf. also chapter 2).

By this method it has been possible to trace ^{99}Tc from Sellafield the whole way described in fig. 3.1 to Thule in North-West Greenland by measuring on Fucus samples collected along the coastlines where the currents run. Figure 3.3 shows the results of this study.

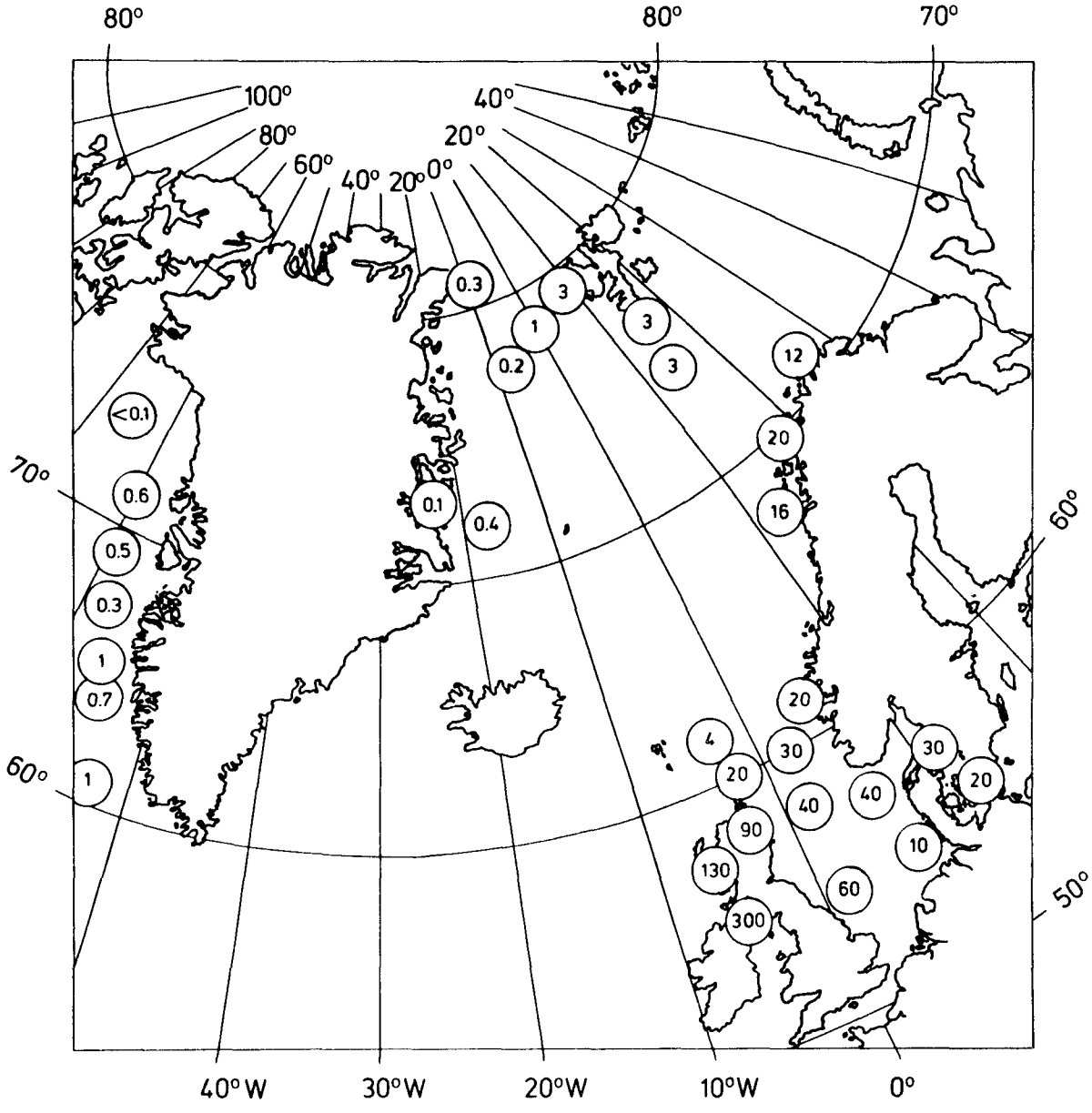


Fig. 3.2. Transfer factors of ^{134}Cs discharged from Sellafield to various surface waters. Unit Bq m^{-3} per PBq a^{-1} (all data decay corrected). The factor of 10 from the German Bight was calculated after correction for contribution of ^{134}Cs from Cap de la Hague in France.

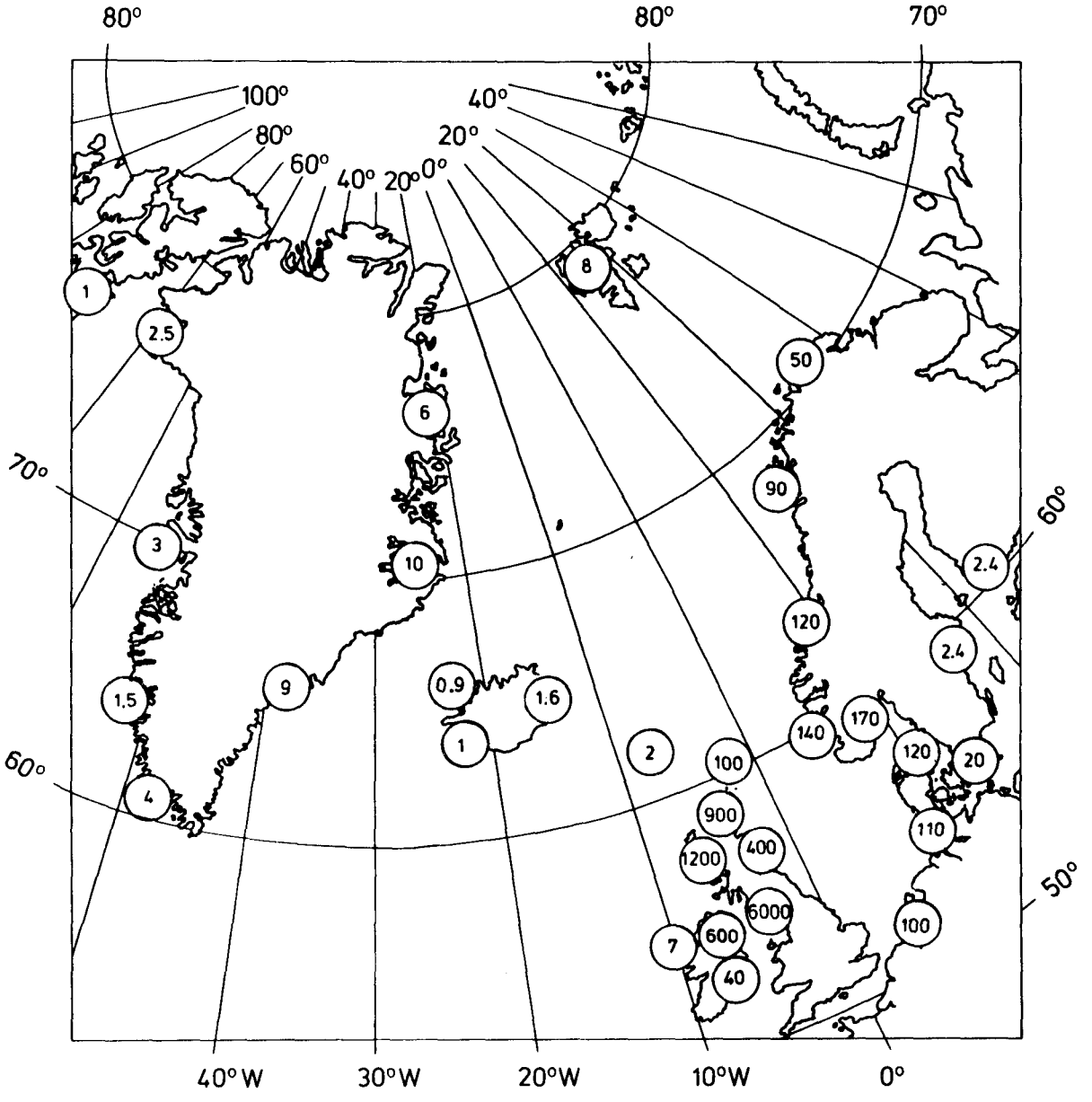


Fig. 3.3. Technetium-99 in Fucus vesiculosus/disticus collected in 1980-1984 (mean values). Unit: Bq kg^{-1} dry weight.

If these results are compared with those obtained from the ^{134}Cs water measurements the agreement between the two methods is surprisingly good. In the North Sea both methods show levels an order of magnitude less than the concentrations found in the North Channel at the outlet from the Irish Sea. In the Danish Straits we find one to two orders of magnitude lower concentrations are found. Along the Norwegian west coast the concentrations are approximately 5%; at Svalbard the levels have decreased to 1%, and finally around Greenland we find approximately 1 o/oo of the North Channel values.

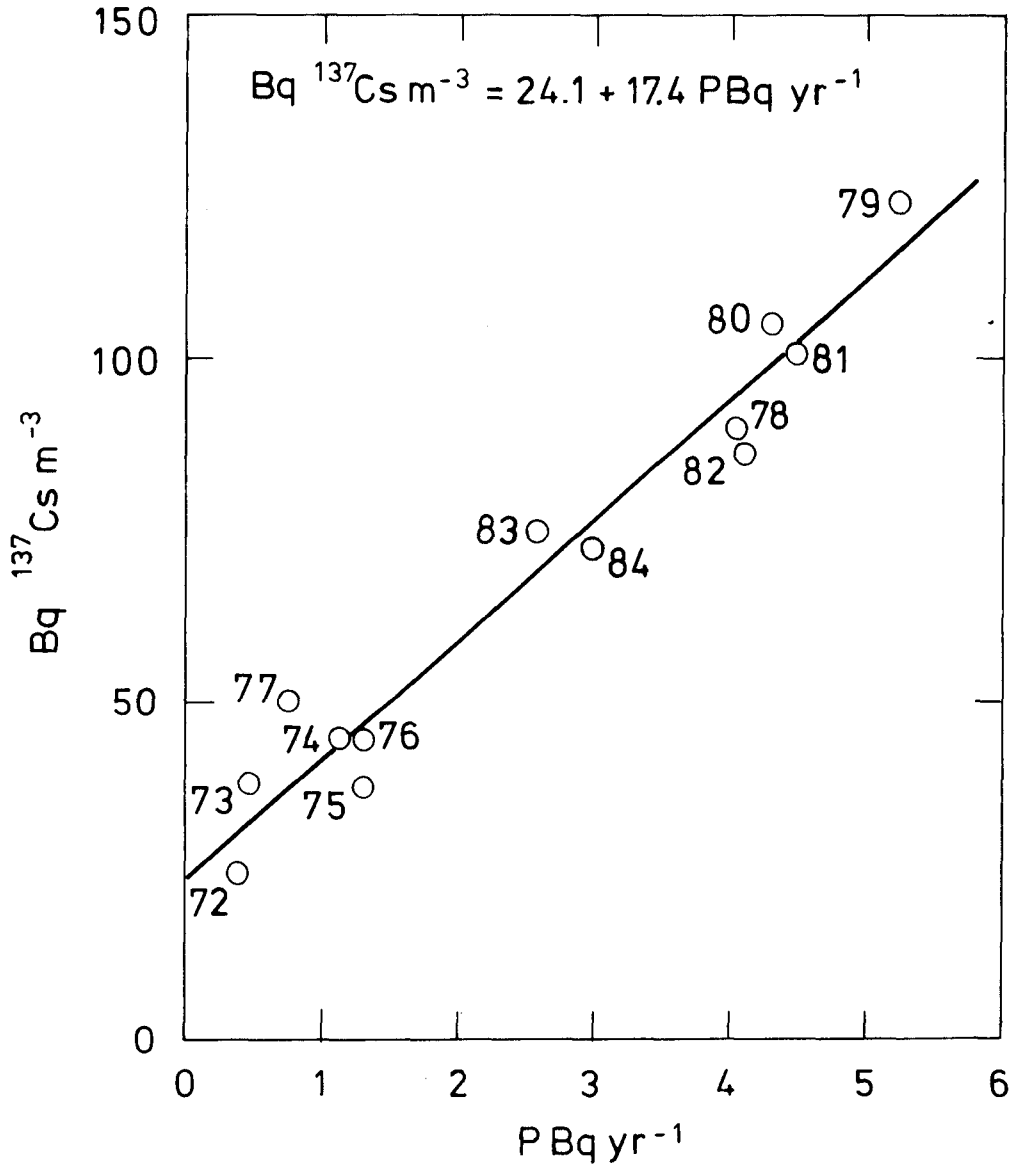


Fig. 3.4. Cesium-137 concentration in "North Sea water" (35 o/oo salinity) in the Danish Straits related to discharges from Sellafield 4 years earlier.

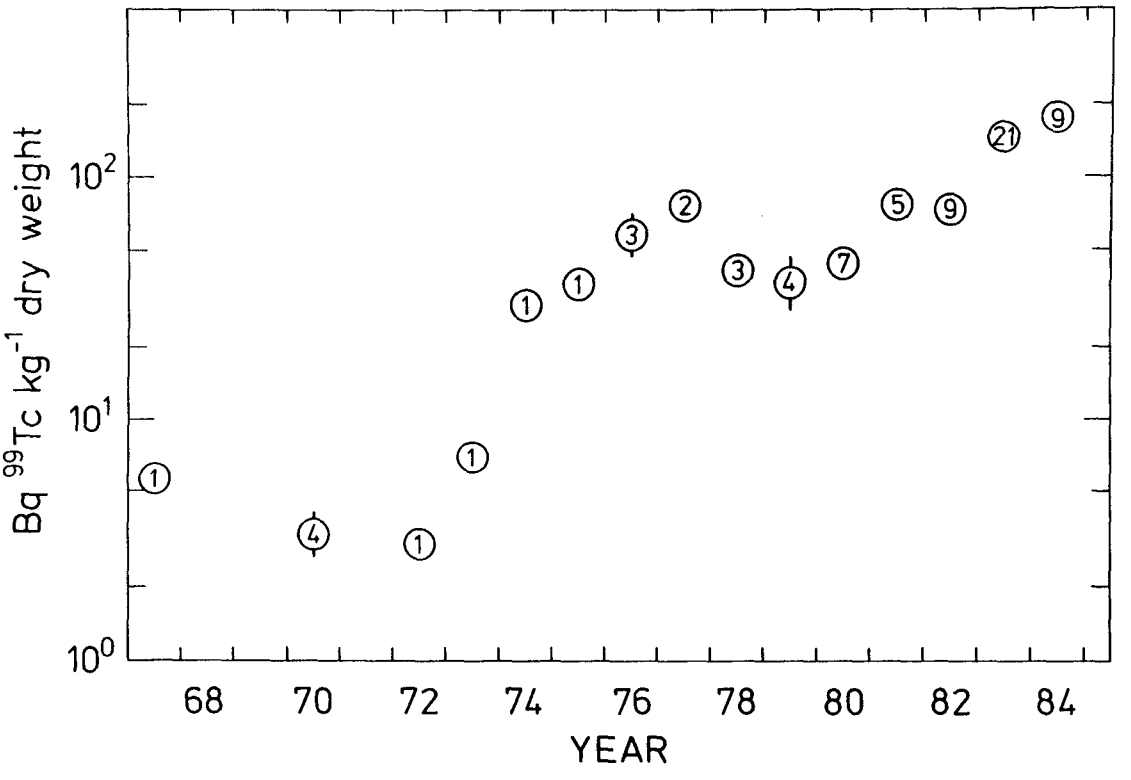


Fig. 3.5. Annual mean values of ^{99}Tc in Fucus vesiculosus collected in the Cattegat 1967-1984. The number of samples and ± 1 standard error of the mean are indicated.

Transit times

Since 1972 ^{137}Cs concentrations have been measured in the Danish Straits. From these measurements and from the discharge data from Sellafield a transit time of 4 years from Sellafield to the Danish Straits have been found (see Fig. 3.4.)

This information may be used to determine at what time enhanced ^{99}Tc discharges began from Sellafield. In Fig. 3.5 we notice a marked increase in the ^{99}Tc concentrations of Fucus collected in the Danish Straits in 1974. Hence it is concluded that increased discharges of ^{99}Tc began in 1970.

Along the Greenland east coast the ^{99}Tc levels in Fucus showed an increase in 1977 (cf. Fig 3.6) It is concluded that the transit time from Sellafield to the East Greenland Current is approximately 7 years. This estimate is in agreement with a previous

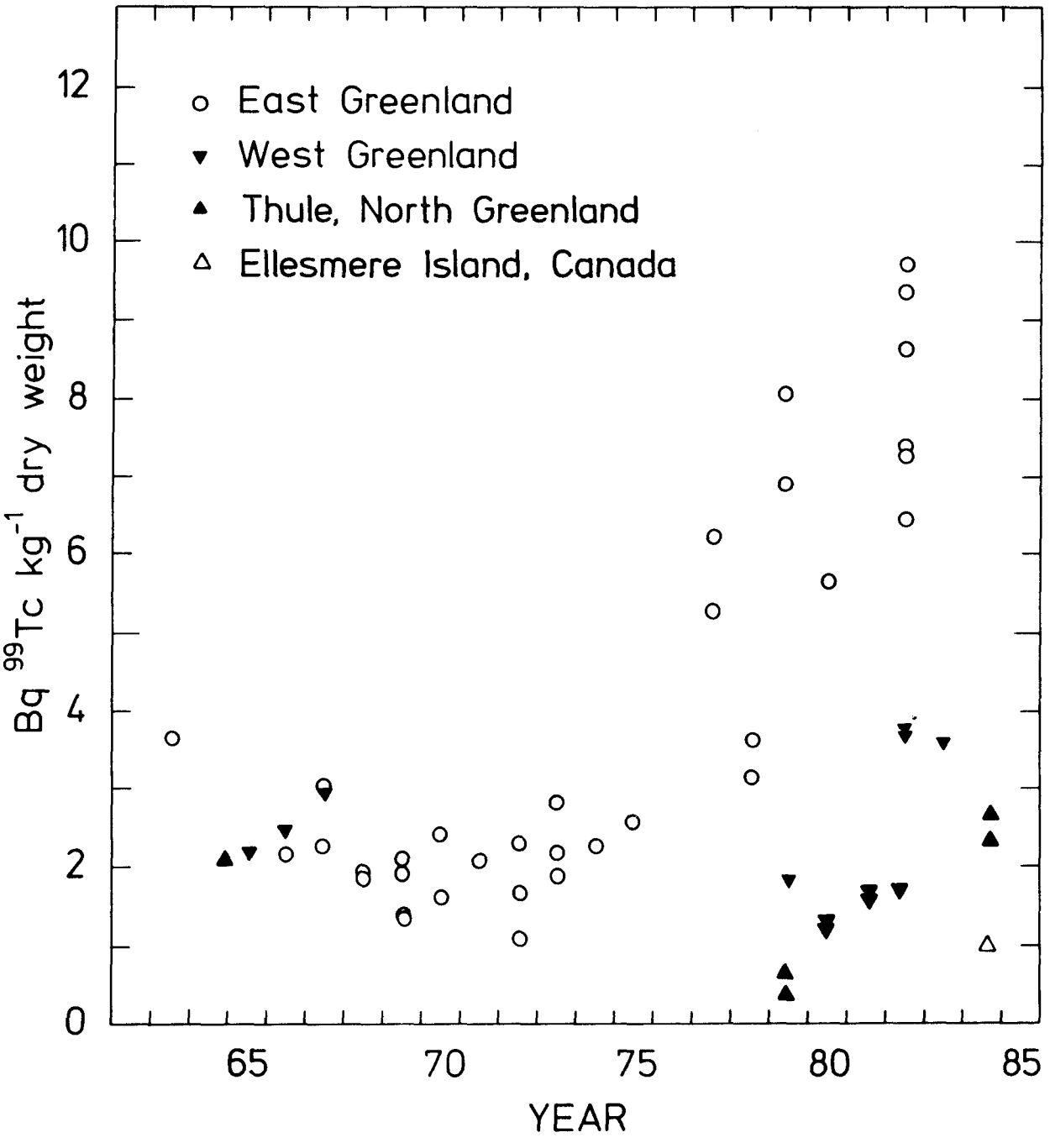


Fig. 3.6. Technetium-99 in Fucus vesiculosus/disticus collected in Greenland 1963-1984. (For sampling locations cf. Fig. 3.1)

one based upon the appearance of ^{134}Cs in the East Greenland waters. The current velocity around Greenland is on the order of 0.1 m s^{-1}. Hence it will take 1-2 years to pass from North-East Greenland anti-clockwise to North-West Greenland. The transit time from Sellafield to West Greenland thus becomes 8-9 years. It takes approximately 3 years for the effluent from

Sellafield to reach the central part of the North Sea.

From the above, certain conclusions can be drawn with respect to transfer of pollutants. The North Sea is a main recipient for waterborne waste from Western Europe. The transit time from the North Sea to the fishing grounds in West Greenland is estimated to 5 to 6 years. Furthermore, a waterborne contaminant is expected to be diluted by a factor of approximately 100 when passing from the North Sea to West Greenland surface waters.

Dispersion of pollutants in the Danish Straits

The Nordic bioindicator study has in particular dealt with a study of the discharges of ^{60}Co from the Swedish nuclear power plants at Barsebäck and Ringhals (Boelskifte, 1985, Nilsson, 1981). As a bioindicator for this radionuclide Fucus, which concentrates ^{60}Co from the seawater by a factor of 10^4-10^5 mainly depending upon salinity has been used. It has been found that the dispersion of ^{60}Co discharged from Barsebäck follows the expression

$$C = K \cdot A^{-\beta}$$

where C is the concentration of ^{60}Co in Fucus in Bq kg^{-1}

A is the distance in km from Barsebäck

β is a constant: 1.4

K is a constant: approx. 1200 in recent years

A diffusion-simulation model based on auto regressive models estimated on current velocity data was set up. From this a regression as shown above has been found with β = 1.3, i.e. in good agreement with the empirical value found from Fucus.

Furthermore, it has been seen that if the dilution equation found above is applied to the Danish Straits a mean residence time for the water in agreement with that obtained from waterflow studies i.e. of ~ 0.2 year has been found.

From the Sellafield tracer study it has been found that slightly less than 1% of the discharges from Sellafield enter the Baltic

Sea, i.e. 0.03 PBq ^{137}Cs per year in recent years. This can thus be calculated in spite of the fact that the additional ^{137}Cs cannot be measured since the total inventory of ^{137}Cs from fall-out in the Baltic Sea is approximately 0.35 PBq ^{137}Cs.

4. THE APPLICATION OF RADIOECOLOGICAL METHODS IN GENERAL ECOLOGY AND POLLUTION STUDIES

Introduction

The radioecological experiences gained during recent decades have generated a number of general principles of value for the surveillance of pollutants. Naturally occurring radionuclides are available for studies in the environment and those from man-made sources like weapon-test fallout and the operation of nuclear installations.

The possibility to trace circulation and transport of water masses over large sea areas is one example mentioned in the previous chapters. Others are the transport, distribution and interaction of elements in biota at the specimen, population and ecosystem levels which can be followed both in the field and by laboratory experiments.

The main advantage of radioecological methods is the possibility to study the fate of elements and rate of processes. This can be done in field and laboratory experiments without a heavy increase in the concentrations of the studied elements, e.g. trace metals.

Some examples of application

During recent decades natural and artificial radionuclides have been used in various areas and for various purposes.

The important use of natural radioactivity for dating materials can be mentioned here (Libby 1955) or the applications of nuclear techniques in hydrology (IAEA 1968).

The numbers of studies of alpha-emitting nuclides and some of their daughter products in the marine environment have increased recently. An overview of the applicability of the natural-

ly occurring nuclides is given by Pentreath (1984), addressing such problems as the determination of rates of transfer in particulate matter, deposition rates, bioturbation rates, etc.

In the past 30 years isotopic tracers have been used in ecological studies, especially low-energy isotopes of elements naturally occurring in all living systems. The extensive use of radioactive carbon (^{14}C) for measuring organic production was introduced by Steemann Nielsen (1951) and the same method has been used, among others, by Sorokin (1966) for studies of the nutrition of aquatic animals.

Studies are presented by Bryan et al. (1966) where results from laboratory experiments on rates of uptake and factors affecting them have been combined with results from field work at Windscale (Sellafield), from fallout studies and from stable isotope analyses.

The radiotracer experiment approach in ecology has since then developed extensively and a series of methods and experimental designs for radioecological studies in the marine environment are given, e.g. by IAEA (1970, 1975a, 1975b, 1979).

The use of radioactive isotopes to measure the transfer of materials in aquatic food chains is discussed by Conover and Francis (1973), demonstrating some of the sources of error identified until then. They also noted that ecologists still seemed to be largely unaware of the available literature despite its obvious applicability.

The possible contribution of radioecology to marine productivity studies is presented by Preston et al. (1972) but now based on studies of fallout radionuclides and those introduced to coastal waters, especially ^{137}Cs. The studies involve sediment transport and biological re-concentration as well as movements of water, later discussed in detail by, among others, Aarkrog et al. (1985). The use and limitations of such studies

and the concept of the "biologically available" fraction in relation to trace elements in sea water is presented.

Techniques for determination of physico-chemical forms and concentration of trace elements in natural waters are given by Salbu (1984). The studies are based on sensitive nuclear techniques with special emphasis on transformation processes and chemical speciations, which are essential for the transport, mobility and bioavailability of elements. It is stated that radioactive tracers and fractionation techniques are most valuable tools for the investigation of microchemical processes, e.g. the chemistry of colloids and their role as transporting agents, important for the understandng of dynamics and for modelling purposes.

Sediment chemistry and the potential mobility of the various elements deposited are important aspects of contamination of the aquatic environment. The significant role of bioturbation on sediment properties and biogeochemical processes is demonstrated by Evans (1982) (using ^{137}Cs and ^{60}Co as tracers) and by Kershaw (1985).

Environmental application aspects are given by Pentreath (1977) in a comprehensive review of radionuclides in fish. The potential information which can be extracted from radioecological studies is discussed in detail.

Of special interest is the proposed transfer of radiological techniques - the use of kinetic data and the estimation of body burden - which have been little used in other fields. Thus some of the basic equations used for the turnover of the radionuclides by the biota could be applied to the troughput of other chemicals. For example in equation (1), the function which describes the physical decay of the radionuclide could be substituted by one which describes the breakdown or metabolism of a compound, in conjunction with additional exponential functions to describe the build-up of the metabolite.

Equation 1 (Pentreath 1976): $\frac{dC_t}{dt} = KC_{ss} - (K+\lambda)\ C_t$

where C_t = concentration in the organisms at time t
K = excretion coefficient
C_{ss} = assymptotic value
λ = decay constant of the radionuclide

Radioecology is essential in the radiological assessment. The dose assessment for man and organisms is, however, highly dependent on a suitable modelling. This emphasis on models has widely contributed to the general development of time-independent models as well as time-dependent (dynamic) models, of interest also for stable elements. Oceanographic models for the dispersion of wastes is given, e.g. by GESAMP (1983). Another example is a model for accumulation of radionuclides in marine organisms given by Evans (1984). An example is also given in chapter 2 above.

Some Scandinavian literature surveys can be added to the list of examples. Pärt (1977) in Swedish deals with the significance of the physico-chemical states for the uptake and turn-over of trace metals and their radioactive isotopes in aquatic organisms.

A literature survey on concentration factors for stable metals and radionuclides in fish, mussels and crustanceans is given by Neumann (1985) in Swedish with a shorter English version.

The few examples presented, of which many are chosen to demonstrate an early approach, give evidence of the applicability of radioecological metods in various fields within biology and ecology. The literature today is voluminous in spite of the fact that many physiologists, toxicologists, ecologists, etc., not yet have taken advantage of the possibilities at hand.

Discussion

The behaviour of artificial radionuclides in the environment is closely related to the behaviour of the corresponding stable elements. The interaction, and especially the importance, of the physico-chemical states of the nuclides is stressed by several authors, since they affect the availability for both biological accumulation and the behaviour in relation to sediments.

Radionuclides and stable trace elements are present in different physico-chemical forms - simple ions, molecules, hydrolysis products, polymer colloids, pseudocolloids or incorporated or sorped to inorganic or organic particles. The studies of physico-chemical forms gives information on microchemical phenomena which cannot be derived from macrochemistry. It is also clear that interaction occurs with "foreign" components like micro-algae, clays, etc., which influences the size distribution patterns by sorption, complexing/chelating or aggregation/dispersion processes. The demands for better understanding of the transformations, kinetics and reversibilities open up a large field of research in the future.

The main entrance of radionuclides into the ecosystem at the lower trophic level, e.g. the planktonic or benthic algae, is directly from the surrounding water and thus highly dependent on the physico-chemical form. The transfer to other compartment of the ecosystem, e.g. fish, is mainly through the food chains. Once accepted at the lower trophic level, especially by absorption, it seems probable that the behaviour of the radionuclide and its stable counterpart along the pathways will be comparable in a more general way.

This is also the case regarding the fate of nuclides once they are digested and accepted by an individual organism for internal distribution to various organs, etc.

It is stated by, e.g. Pentreath (1977) that the use of radionuclides in studying the uptake of a number of chemicals, particularly the heavy metals, is already established and should lead to improved methods of toxicity testing by rapidly indicating their internal fate. There are also many aspects of fish physiology and fisheries biology in which the further application of radionuclides and utilization of data that have been derived would be of considerable gain.

There is a comprehensive literature in radiobiology in the sense of effects of radiation on individuals, organs, etc. Sublethal and lethal effects on individuals have been observed only in experiments with extremely high radiation doses compared to those occurring in the environment. At the population or ecosystem levels no hazardous effects of radiation have been described (IAEA 1976, Woodhead 1984).

It should be emphazised that the fate of various elements easily can be followed, e.g., on the cellular or organ level as well as on population and ecosystem levels at very low concentrations of the radionuclides.

Advantages given by radioactive tracer methods in biology and ecology are thus their ability to describe the fate of various elements and the rate of various processes without an increase in the concentrations of the studied element to potentially toxic levels.

Data on concentration factors, ratios, transfer factors and distribution factors (Kd) used in a time-independent model will always be of relevance for a description of equilibrium concentrations in a steady-state situation (IAEA, 1985).

The various contaminants, however, often appear in the environment in gradients or in pulses conditioned by variations in the discharges. To this should be added, for example the seasonal predispositions of organisms. Such a situation contradicts a

steady-state assumption and equilibrium conditions. Therefore, the development of dynamic models such as compartment models, in which the element in each compartment is expressed as a function of time and the transfer between compartments are represented as rates is of special value.

5. CONCLUSION

The radioecological experience gained mainly during the last two decades is today limited to a relatively small group of experts. It would be an advantage to spread it to a broader audience in order to make the methods available for the surveillance of pollution, either in field tests, in control routines or in further research.

One main goal is to more generally apply a methodology involving critical pathways and critical organisms in the study of the ecosystem, including man. This approach could be useful as a routine procedure for hazardous substances and their daughter products, like PCBs, DDT, oil products, as well as heavy metals.

A typical example is the management of the mercury problem, where dose to man sometimes is estimated on the basis of a theoretical fish, constructed from information on people's feeding habits and covering a variety of fish species, rather than on an exclusive diet by a critical group on a single fish species containing high levels of mercury.

A number of fields of application have been outlined in this report. Thus, by the methods described above it should be possible to predict transit times and dilution factors from the main recipient of waterborne pollution in northern Europe: The North Sea, to pollution sensitive areas such as the Baltic Sea and Arctic waters. Furthermore, transport and dilution of pollutants from the "Øresund Metropol" could be described applying the authorized discharges of radioactive tracers from the nuclear power plant Barsebäck. Also, the model developed for the function of bioindicators (seaweed) could be adapted for the monitoring of marine pollution with heavy metals.

Other fields of application certainly exist. They may most conveniently be identified in a seminar with the participation

of representatives from science, responsible authorities and industry, and pollution experts from inside and outside the Nordic countries. It is thus suggested that such a seminary would be a practical first step in the endavour to use the radioecological experience in other fields. The seminar should, firstly provide

input from radiological experts, for example on methods for estimation of dose over time and the tools used: measurement techniques, design of appropriate experiments, models, etc.

Subsequently,

useful fields of application may be identified through discussions between representatives from national authorities, industry and radiological experts.

The role of the seminar would thus be to provide a forum for the exchange of ideas and for technology transfer.

In a subsequent step, methods could be introduced on a project level, involving, contacts, e.g. at the national level, between authorities and industries concerned, for the inclusion of "radioecological" techniques in control programmes or between authorities and science for biological effect studies, and other special projects.

Radioecological methods can thus be expected to contribute to a better control of pollution of Nordic waters and to a more reliable estimate of the consequences of this pollution. This would form an improved basis for decision making within environmental protection in the Nordic countries. In this sence the proposed programme of action may also be of interest for the Nordic committee of Environment protection.

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