RADIONUCLIDES IN THE OCEANS

Inputs and inventories

COORDINATORS

PIERRE GUÉGUÉNIAT, PIERRE GERMAIN and HENRI MÉTIVIER

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Inputs and Inventories

Coordinators:

P. Guéguéniat, P. Germain and H. Métivier

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Foreword

This book is a collection of papers given on the first day of the international symposium *Radionuclides in the Oceans* (RADOC 96-97). This symposium was organized by the Nuclear Safety and Protection Institute (IPSN) and the Ministry of Agriculture, Fisheries and Food (MAFF). It was divided into two parts. The first part, « Inventories, Behaviour and Processes », took place at Octeville-Cherbourg, France (7-11 October 1996) and dealt with the same themes as the 1987 Cherbourg symposium (Radionuclides as a tool for oceanography) and the 1991 Norwich symposium (Radionuclides in the study of marine processes). The second part, « Impacts on Man and the environment », will take place at Norwich and Lowestoft (7-11 April 1997), and will cover the themes of radiological and environmental protection and modelling.

Ten years after Chernobyl, after the French decision to end nuclear weapon testing in the Pacific Ocean, after the end of the OECD-NEA Coordinated Research and Environmental Surveillance Programme related to low-level waste dumping in deep sea, and one hundred years after the discovery of radioactivity, IPSN has considered it useful to assemble the available information on artificial radioactivity levels in the seas and oceans. The objective is to address scientific and public concerns about the use of the sea as a « waste repository », and to answer regarding the radioactive contamination of the seas and oceans. Therefore, international experts have been invited to describe and quantify, during the first day of the symposium (Part I), the inputs and inventories of artificial radionuclides released in seas and oceans by civil and military activities.

In the different chapters of this book, radionuclides are studied in geographical areas of different size, and varying physical and biological features. Therefore, some presentations deal with oceans taken as a whole (Atlantic, Pacific and Indian oceans). Some sources of radionuclides, such as atmospheric fallout, both before and after Chernobyl, have a large scale impact, whereas others, such as marine dumping sites, sites for nuclear weapon tests, and damaged submarines provide more localised inputs of artificial radionuclides.

Others presentations focus on of particular interest seas from the point of view of specific radionuclide sources and the particular processes which govern radionuclide behaviour. The Channel, the North Sea and the Irish Sea are quite shallow and receive, directly or indirectly a small fraction of the wastes released by reprocessing plant wastes. The Baltic Sea has been strongly affected by Chernobyl, and is subject to a high salinity pressure gradient. The Black Sea has also been marked by Chernobyl and is strongly anoxic. The Mediterranean Sea receives an input of radionuclides from the Rhone River outflow into the western basin and has exchanges with the Atlantic Ocean. Dumped radioactive wastes, radionuclide inputs from the Ob and Yenisei river basins, radionuclides from European reprocessing plants transported by ocean currents, and radionuclide transport by ice must be considered in the Arctic, Barents, Kara and Laptev Seas.

It appears that such a comprehensive review of all the available data has not been made in recent years and it is the aim of this book to provide it.

The production of this book was made possible by the collaborative efforts of the international experts in providing, within a tight schedule, the inputs requested by IPSN; they deserve our gratitude. Our thanks also go to J.F. Hémidy, Director of the « École d'Ingénieurs de Cherbourg », and J. Lemière of the « Institut Universitaire de Technologie, Cherbourg – Manche », who have very kindly made available their premises for the *Radionuclides in the Oceans* Symposium. We also would like to thank M. Masson, D. Boust, P. Bailly du Bois, D. Maro for their review of the manuscripts and proofs, as well as Mrs C. Leconte, S. Le Cavelier, Mrs I. Le Corre who have devoted much efforts to the organization of this event.

We are very grateful to the Minister of the Environment, Mrs Corinne Lepage, who has accepted to open the session devoted to inputs and inventories of radionuclides in seas and oceans.

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Summary

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Radionuclides in the Atlantic Ocean: A Survey

F. Nyffeler, A.A. Cigna,
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The knowledge of the distribution of radionuclides in the ocean is gradually being updated through a continuous flow of data, ranging from the periodical monitoring records issued by national authorities to the results of local and ocean scale scientific cruises, carried out within the framework of large international programs. This paper tries to summarize and to update the state of our knowledge regarding the Atlantic Ocean, by scaling some prominent features. We consider successively the ocean scale distribution of radionuclides from bomb testing fallout, compared with the natural background, the regional inputs from land-based sources and from the adjacent seas. Finally, the controlled or uncontrolled dumping practice and the accidents like the Chernobyl case are also taken into account to estimate the collective doses to the world population.

1.1. Introduction

Anthropogenic radionuclides have been introduced to the Atlantic Ocean from a variety of sources since the advent of the nuclear age. The sources include fallout from atmospheric nuclear weapon's tests, nuclear waste dumping/disposal activities and accidents, in particular, the Chernobyl accident in April 1986. The temporal and spatial nature on these inputs has affected greatly the past, present and future distributions of the various radionuclides involved. The natural radionuclides resulting from the non-nuclear industry (*e.g.*, the fertilisers plants) should also be considered in comparison with those of primordial or of cosmogenic origin. However, the natural radionuclides are ubiquitous in the ocean in significant concentrations, and the anthropogenic contribution to the natural stock is not expected to be comparatively relevant.

Three categories of man-made radionuclides can be noted according to their time and space scales.

a) *All ocean inputs:* this applies chiefly to fallout from atmospheric nuclear weapon's tests which took place in the late 50's and early 60's in both the North and South Atlantic. About 3/4 of the Atlantic input was to the North Atlantic and 1/4 to the South Atlantic. In each hemisphere, the delivery was maximal at mid-latitudes and minimal at the equator and at the poles. The input of ^{238}Pu from the burn-up of the U.S. SNAP 9A satellite over the Southern Hemisphere in 1964 was also widely introduced to the Atlantic but, in this case, the latitudinal distribution was reversed, with the large fraction of the input going into the South Atlantic.

b) *Point sources and regional inputs:* these inputs include the point sources from nuclear fuel reprocessing discharges such as Sellafield (UK) and Cap de la Hague (France). Because it was randomly distributed, the fallout to Western European coastal waters after the Chernobyl accident enters the same category of inputs. In all three cases, the soluble fraction of the radionuclides introduced were largely transported into the Arctic and sub-Arctic seas.

c) *Dumping and accidents:* these include the planned dumping of packaged low level nuclear waste at a variety of sites and by several countries, the accidental losses in deep water of nuclear powered submarines, as well as the disposal of reactors with their fuel in the shallow water of the Northern Seas by the Former Soviet Union (FSU). In all these cases, any releases which have occurred have lead to extremely localized contamination to date.

For each of these categories, the type of radionuclides released, their isotopic ratios and the ratio between species can be used to determine their origin. The ratios also provide new opportunities to gain an understanding of processes as the large scale transport associated with the global ocean circulation, the vertical mixing, the biological accumulation and the short-circuit mechanisms. The wide spectrum of half-lives of radionuclides allows to use them for a variety of scientific applications, from the local study of the sediment and the processes in the seabed to the changes in the ocean scale circulation over decades. Some of these issues have been addressed in the proceedings of a previous symposium on the use of radionuclides in the study of marine processes (Kershaw *et al.*, 1991). The aim of the present paper is to review and update the actual level of knowledge by putting emphasis on some prominent features associated with the three categories of inputs indicated above.

1.2.   Field Measurements and Databases

Measurements of man-made radionuclides have been performed throughout the North East Atlantic and the Arctic since the 1960's. The most comprehensive suite of measurements in the Atlantic was made in 1972/1973 as

part of GEOSECS, the « Geochemical Ocean Sections program » (National Science Foundation, 1987). The next two large programmes were, « Transient Tracers in the Ocean » (TTO) and the « South Atlantic Ventilation Experiment » (SAVE) in 1981 and 1987-1989 respectively. In addition, numerous data reports and review papers were issued by the German Hydrographic Institute (Kautsky, 1980; 1987), the Obninsk labora- tory, Russia (Vakulovskii *et al.*, 1985), the Woods Hole Oceanographic Institution, USA (Livingston, 1988), the Lowestoft laboratory, UK, (Kershaw and Baxter, 1995), the Bedford Institute of Oceanography, Canada (Smith *et al.*, 1990), Lund University, Sweden (Roos and Holm, 1993), the Risoe National Laboratory, (Aarkrog *et al.*, 1983; 1986; 1987; Dahlgaard, 1984; 1994; 1995a) and the French IPSN-CEA laboratory at Octeville (Guégéniat *et al.*, 1995). During the past decade, programs aiming at improving the use of the national radiological data bases available in the riparian countries of the northern seas have been launched under the aegis of the European Community (CEC, 1990) and of the IAEA (Povinec, 1994).

Radioactive waste was dumped in the north-east Atlantic until 1982 and the operations have been supervised by the NEA (Nuclear Energy Agency). In 1981, the Agency set-up the CRESP programme (Co-ordinated research and Environmental Surveillance Programme), to increase knowledge of the processes controlling the transfer of radionuclides in the environment (NEA, 1982), which provided a significant amount of original site specific data.

The disposal of radioactive waste in the shallow Arctic areas have recently triggered concerns, which resulted in a temporarily boosted scien- tific effort. The inventory of the radioactive waste disposed in the Arctic region by the FSU has been reported in the so-called « White Report » (Yablokov *et al.*, 1993), and the consequences of such dumping operations are presently assessed within the framework of the IASAP programme (Sjoeblom and Linsley, 1995). As an outcome, a significant amount of site specific data are presently accumulated in the polar areas, which are of vital environmental concern (Strand and Holm, 1993).

1.3. The Ocean Scale Distribution of Radionuclides

Other than in the Arctic and sub-Arctic seas and their southern deep water overflows, virtually all of the anthropogenic signal throughout the North and South Atlantic derives from fallout from atmospheric nuclear weapon's tests. Hence the ocean scale distribution of anthropogenic radionuclides in the Atlantic is, in reality, that of the fallout signal (plus the ^{238}Pu from the satellite power source). The fate of this signal is determined by a number of factors, *viz.* the original input pattern, physical mixing of affected water masses, chemical properties with respect to particle and biological removal, and radioactive decay.

1.3.1.   Principal Radionuclides

The principal nuclides involved fall into two broad categories based on their behaviour in respect to association with particles:
a) soluble, b) reactive. Also, for convenience, we shall break down the discussion into other components, a) surface waters, b) the ocean water column and c) the sediments.

1.3.1.1.   Soluble Radionuclides

These include ^{137}Cs, ^{90}Sr and ^{3}H. ^{14}C is also largely a soluble tracer but has both bomb-produced and pre-bomb cosmogenic origins. ^{129}I has been shown to serve as a soluble tracer of nuclear fuel reprocessing discharges but its large scale fallout distributions have not been studied. The first four have been extensively studied in the Atlantic in several large programs and other studies. They have found wide use as tracers of ocean circulation and used in model validation as well as in studies of physical processes such as deep sea ventilation. The fission products, ^{137}Cs and ^{90}Sr, have been shown to maintain the same constant ratio of about 1.5 to each other (Bowen *et al.*, 1974) throughout the Atlantic Ocean (and as such characterised their input ratio in fallout) – other than the slight ratio change with time associated with their slightly different half-lives – ^{90}Sr ($T_{1/2}$ = 28.8 years), ^{137}Cs ($T_{1/2}$ = 30.0 years).

1.3.1.2.   Reactive Radionuclides

The principal reactive radionuclides studied in the Atlantic resulting from fallout are the Pu isotopes, ^{238}Pu, ^{239}Pu and ^{240}Pu, and ^{241}Am. In the water column, in addition to studies of their basic behaviour in the ocean, they have been used to study the nature of the scavenging process by which metals are incorporated into the pool of sinking particulates, fractionated from other soluble species and eventually transferred to the ocean sediments. In sediments, as well as providing an index of the current extent of removal from the water column, the transuranic elements have advanced the interpretation of bioturbation, the biologically driven physical mixing process in marine sediments.

1.3.2.   Horizontal Distribution and Penetration

GEOSECS and TTO provided the most extensive opportunity to observe the fallout transient's distribution and penetration throughout Atlantic waters.

Unfortunately, only ^{3}H and ^{14}C were measured, as resources for fallout studies had become scarce. The distribution patterns of ^{3}H, especially, can be used to infer the approximate evolution of the distribution of ^{137}Cs and ^{90}Sr. This approximation follows from the fact that the ratio of ^{3}H to ^{137}Cs or ^{90}Sr increased over time - during the input to the ocean - because of the continuing input of ^{3}H from the troposphere due to hydrological recycling, including re-evaporation of terrestrial freshwater sources (Livingston, 1985). Currently in the mid-90's, the World Ocean Circulation Experiment (WOCE) is completing a survey of the Atlantic Ocean as part of the WOCE Hydrographic Program (WHP). Again this will provide ^{3}H and ^{14}C data, amongst other tracers, and can be used only to approximate the evolution of ^{137}Cs and ^{90}Sr.

Other than these large programs, information for the large scale evolution of fallout radionuclides in the Atlantic has to be pieced together from individual studies on a variety of short cruises by many laboratories. There is much more information in the two decades following the arrival of fallout in the ocean than in recent years, so the long term evolution is not well described.

1.3.3. The Vertical Distribution of the Radionuclides in the Atlantic

1.3.3.1. Surface Waters

In surface waters, the variation in the Atlantic in space and time is reasonably well characterised due to the larger number of surface water samples which have been collected and analysed. The situation for ^{137}Cs has been reported very recently as part of an IAEA Co-ordinated Research Program report (MARDOS) (Povinec, 1994). By inference, this can be used to derive ^{90}Sr distributions also – by dividing ^{137}Cs concentrations by 1.5. The key data sets showing the variation of the N/S distributions of surface ^{137}Cs concentrations over time come from the GEOSECS Atlantic expedition in 1972/1973 (National Science Foundation, 1987), a Polish cruise from Europe to Antarctica in 1977-1978 and a Swedish Antarctic expedition in 1988-1989 (Holm *et al.*, 1991). Other data sets were provided by several other MARDOS program participants.

Inspection of these data confirms the expected reduction in surface water concentrations over time due to both radiogenic decay and by physical mixing. What is interesting, however, is that the overall pattern of the N/S distribution, with concentration maxima in the mid-latitudes on both hemispheres, has been largely maintained – presumably because of the dominant large circulation gyres in both the North and South Atlantic Ocean. The total change in Atlantic surface ^{137}Cs concentrations, from the

maximum values observed after the fallout input in the mid-60's to the values reported for 1990 in the MARDOS report, can be derived from comparison of the MARDOS data with data from the RIME report (National Academy of Sciences, 1971). For this comparison, RIME report of ^{90}Sr concentrations were converted to ^{137}Cs by multiplication by the factor of 1.5 mentioned above (Bowen *et al.*, 1974). In brief, concentrations have declined by a factor of 7. When radioactive decay is considered, the decline by mixing is about 3.5.

A similar approach was used by Holm *et al.* (1991), who contrasted the change in surface Pu concentrations from 1972 (Geosecs) to 1988 (Swedish expedition). They noted a 4-5 fold decline in surface Pu concentrations over this period. An even greater change can be presumed to have occurred from the concentrations associated with ^{137}Cs maxima in the high fallout years. Derived Pu concentrations from the RIME report data (converting from ^{90}Sr by multiplying by 0.018), the ratio of ^{239,240}Pu/^{90}Sr in fallout (Harley, 1975), suggest a decline in surface Pu from the mid-60's to 1988 of a factor of 31. As noted by Holm *et al.* (1991), the more rapid decline in surface Pu concentrations is in consequence of its removal on particles which sink out of the surface layer. Much of this removal is subsequently released to subsurface, intermediate and deep waters through demineralisation.

1.3.3.2. The Ocean Water Column

As mentioned above, there are no large scale measurements of fallout nuclides in the Atlantic after Geosecs (other than ^{3}H or ^{14}C). The evolution of the fallout transient has to be inferred from the scattered stations occupied subsequent to Geosecs or, in the case of the soluble species, from the evolution of the ^{3}H signal as mapped by the TTO, SAVE and WOCE programs. The situation is much worse in the South Atlantic – where the data for subsurface fallout concentrations other than Geosecs are minimal.

Ostlund (1985) noted several features of the fallout transient traced by ^{3}H, between 1972 (Geosecs) and 1981 (TTO). In intermediate waters and deep waters, the signal had significantly deepened and moved south due to ventilation processes – as illustrated by changes in the 0.2, 1 and 2 isolines of tritium concentration (expressed in TU81N – tritium units as at 1981). In addition, tritium could be detected as far south as 26°N in the southward flowing deep western boundary current fed from dense water overflows from the Greenland and Norwegian Seas (see Fig. 1.1.).

It was on the TTO expedition that the first evidence of Sellafield ^{137}Cs was observed in this dense overflow water as far south as Labrador in the boundary current (Livingston, 1988). Other Sellafield components, such as ^{90}Sr, ^{129}I and ^{99}Tc must also characterize this stream, but were not determined at all then or detected.

The fallout of Chernobyl was scattered very unevenly over the area of the Baltic Sea (HELCOM, 1989). The range of Cs–137 concentration measured in May and June 1986 was between 40 Bq m^{-3} (Arkona Sea) and 5 200 Bq m^{-3} (northern coastal area of the Gulf of Finland). In general the concentrations of radionuclides were lowest in the southern parts of the Baltic Sea and in the Bothnian Bay. Most contaminated sea water areas were in the Bothnian Sea, in the Gulf of Finland and in the Mecklenburg Bight. A substantial difference was obvious between coastal and open sea areas, so that the concentrations were higher near the coast (STUK, 1987). The activity ratio Cs–134/Cs–137 after the accident was measured to be in the range of 0.5 that corresponded to the theoretical value for the fuel of Chernobyl NPP (IAEA, 1986b).

By the end of 1988, the Caesium concentrations were in great extent smoothed out due to mixing processes in the Baltic Sea and river water input. Nevertheless, Cs–137 contents essentially vary in the different sea areas. This can be seen in Figure 10.4. which illustrates the Cs–137 distribution in surface water in 1989. In 1991 in the surface waters of the eastern part of the Baltic Proper, the Cs–137 content varies from 100 to 130 Bq m^{-3}, being (120 ± 10) Bq m^{-3} on average.

An insignificant increase of Sr–90 and alpha–emitting plutonium isotopes activity concentrations were observed during the first year after the accident in those parts of the Baltic Sea where high deposition occurred. In general the Pu–239,240 concentrations in the Baltic Sea are very low, in average less than 10 mBq m^{-3}. The radioactive contamination of the Baltic Sea after the Chernobyl accident has been dominated by the caesium isotopes 134 and 137 in its typical and fairly constant activity ratio of 0.54. This activity ratio gives the opportunity to identify the Chernobyl signal also in the outflow from the Baltic, *e.g.* in the Norwegian Coastal current.

10.3. Temporal Evolution of the Chernobyl Contamination in the Baltic Sea

Figure 10.3. shows the temporal trend of the Cs–137 and Sr–90 activity concentration in surface water at a position in the Belt Sea (Schleimündung) from 1970 to 1995. The first maximum of Cs–137 in 1980 is due to the maximum annual discharge at Sellafield in 1975 revealing the time of transport of contaminated water to this position between four to five years. The concentrations of Cs–137 changed dramatically after the deposition of the Chernobyl fallout. The seasonal variation with increasing levels in spring and decreasing levels to autumn and winter are explained by the penetration of higher contaminated surface water from the northern Baltic due to higher fresh water input by snow melting and wet deposition in spring and, consequently, higher output of surface water from the Baltic. The opposite

effect takes place during autumn and winter. However, the highest levels were recorded at this position in 1991. Since then the activity concentration of Cs–137 shows a decreasing trend.

The initial contamination of the surface layer penetrated rapidly into deeper waters in those areas with the lack of the stable halocline. In 1991 in the deep water layer of the Baltic Proper, the caesium concentrations were still lower than in surface water. For example at the Gothland Deep the Cs–137 activity concentrations in the surface and deep water were 120 and 25 Bq m^{-3}, respectively.

During the following years after 1986, the contamination of the water in the Gulf of Finland have decreased more rapidly than those of the Bothnian Sea. This is due to both the river water run–off from Neva, Luga, and Narva from less contaminated land areas and higher water exchange rates between the Gulf of Finland and the Baltic Proper than between that one of the Bothnian Sea. The contaminated water moved into southern direction, which lead to increasing Cs–134 and Cs–137 activity concentrations in initially less contaminated areas .

The distribution of Cs–137 in May 1994 both, in surface water and in vertical profiles is shown in Figure 10.5. The highest values are mostly still measured at the surface but the contamination has penetrated significantly into deep layers. This is valid in particular for those areas with less pronounced halocline, *e.g.* Northern Baltic Proper and Bothnian Sea.

The Cs–137/Salinity relation of a survey in May 1994 is displayed in Figure 10.6a. This figure reveals the mixing of water between different areas contaminated at mainly two different levels:

• A significant high and large range contamination of water with low salinity which may be related to coastal areas in the Bothnian Sea and,
• the deposition on the central Bothnian Sea and Baltic Proper.

The Cs–137/Salinity correlation for the values from the Bothnian Sea is not well established indicating that the Cs–137 concentration covers a large range over a small salinity distribution ($r = -0.19$). Concentrations of Cs–137 in surface water of the Bothnian Sea are still higher than compared to other parts of Baltic Sea with values above 110 Bq m^{-3}. The correlation, however, in the Baltic Proper and, in particular in the western Baltic and Belt Sea is well established:

$$A_{Cs-137} = 125.1 - 4.17 \times S \qquad (r = -0.88)$$

While the concentrations of Sr–90 increased only slightly in 1986 but in a short period they returned to pre–Chernobyl level (at about 20 Bq m^{-3}). The Sr–90 activity concentration A_{Sr-90} is less strongly dependent on the salinity (Fig. 10.6b.):

$$A_{Sr-90} = 17.907 - 0.304 \times S \qquad (r = -0.87)$$