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***Worldwide marine radioactivity
studies (WOMARS)
Radionuclide levels in oceans and seas***

Final report of a coordinated research project



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International Atomic Energy Agency

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FOREWORD

The International Atomic Energy Agency's Marine Environment Laboratory in Monaco has completed a four year Coordinated Research Project (CRP) on Worldwide Marine Radioactivity Studies (WOMARS). The primary objective of the CRP was to develop an understanding of the present open ocean distribution of radionuclides in the water column and sediment and thus predict the radiological impact to be addressed, as well as to encourage and support marine radioactivity studies in Member States by methodological assistance and total analytical quality management. The project was designed with the intention of reviewing and contributing to scientific knowledge of the processes which affect radionuclide distributions and the sources which have introduced radionuclides to the world ocean.

The specific objectives of the CRP were to: identify the major sources of anthropogenic radionuclides in the world ocean; develop present knowledge of the distributions of key radionuclides (^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$) in water and sediment of the world ocean; and study the development of radionuclide concentrations in water with time using good quality historical data (e.g. from the GEOSECS programme of the mid-1970s and new data sets collected recently).

Two research coordination meetings were held (IAEA-MEL Monaco, 1999; Woods Hole Oceanographic Institution, Woods Hole, USA, 2000) during which the objectives of the CRP, the organization, data compilation, evaluation and synthesis were discussed. The presentations and discussions covered both the development of average radionuclide concentrations in surface water and in water profiles over the last twenty years. A. Aarkrog of Denmark chaired the Research Co-ordination Meetings. The Scientific Secretary was P. P. Povinec.

The topics covered were: Atlantic and Indian Oceans and their marginal seas (Working Group 1); Pacific Ocean and its marginal seas (Working Group 2); Marine Processes (Working Group 3).

The success of the CRP was due to the active collaboration of the chief scientific investigators and the participating institutions as well as numerous other marine institutions which provided radionuclide data for storage and evaluation for the project in the framework of the development of the IAEA's Global Marine Radioactivity Database (GLOMARD). The IAEA would like to express its gratitude for the information provided and for the very fruitful cooperation.

The IAEA officer responsible for this publication was P.P. Povinec of the IAEA Marine Environment Laboratory, Monaco.

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SUMMARY

This publication summarizes the results of the Coordinated Research Project (CRP) on Worldwide Marine Radioactivity Studies (WOMARS) carried out by the IAEA's Marine Environment Laboratory in Monaco. It provides the most comprehensive information on levels of anthropogenic radionuclides in the world ocean. Three anthropogenic radionuclides — ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ were chosen as the most representative of anthropogenic radioactivity in the marine environment, comprising beta-, gamma- and alpha-emitters which have the highest potential contribution to radiation doses to humans via seafood consumption. Although the ocean contains the majority of the anthropogenic radionuclides released into the environment, the radiological impact of this contamination is low. Radiation doses from naturally-occurring radionuclides in the marine environment (e.g. ^{210}Po) are on the average two orders of magnitude higher.

The results confirm that the dominant source of anthropogenic radionuclides in the marine environment is global fallout. The total ^{137}Cs input from global fallout was estimated to be 311 PBq for the Pacific Ocean, 201 PBq for the Atlantic Ocean, 84 PBq for the Indian Ocean and 7.4 PBq for the Arctic Ocean. For comparison, 5 about 40 PBq of ^{137}Cs was released to the marine environment from Sellafield and Cap de la Hague reprocessing plants. The Chernobyl accident contributed about 16 PBq of ^{137}Cs to the sea, mainly the Baltic and Black Seas, where the present average concentrations of ^{137}Cs in surface water were estimated to be about 60 and 25 Bq/m³, respectively, while the worldwide average concentration due to global fallout is about 2 Bq/m³.

For the purposes of this study, the world ocean was divided into latitudinal belts for which average radionuclide concentrations were estimated. Further, where available, time trends in radionuclide concentrations in surface water were studied and mean residence times of radionuclides in these areas as well as in the world ocean were estimated. The results confirm similar mean residence times for ^{90}Sr and ^{137}Cs in surface water, 28 ± 3 years and 13 ± 1 year for $^{239,240}\text{Pu}$.

Changes in radionuclide concentrations in water profiles with time in the North Atlantic and Pacific Oceans were also studied. A clear decrease of radionuclide concentrations in surface water was observed due to transport of radionuclides to medium water depths.

The results obtained in the framework of the WOMARS CRP provide the most complete data set available to Member States on levels of anthropogenic radionuclides in the marine environment. The results will be used as the international reference source on the average levels of anthropogenic radionuclides in the marine environment so that any further contributions from nuclear reprocessing plants, radioactive waste disposal sites, nuclear bomb test sites and possible nuclear accidents can be identified.

1. INTRODUCTION

Radionuclides have been released to the environment from a multiplicity of sources, both planned and accidental. Given that more than 70% of the surface of the Earth is ocean, it is not surprising that much of these releases now reside there. The largest ocean, the Pacific, has been a major repository for these releases — in part because of its sheer size, and in part due to localized releases there.

There has been a growing number of known sources of anthropogenic radionuclides in the marine environment. They include global nuclear fallout following atmospheric weapons tests, the Chernobyl accident, discharges of radionuclides from nuclear installations, dumping of nuclear wastes into the world's oceans and seas, nuclear submarine accidents, contributions from nuclear weapons tests sites, loss of nuclear weapons and radioactive sources, and satellite burnup.

Due to variations in the source inputs and subsequent dispersion, mixing and transport of radionuclides in the world's oceans and seas, the marine environment is labelled with radionuclides differing from one region to another according to the predominant source there.

The main global source of radionuclides in the marine environment is fallout from nuclear tests performed in the atmosphere. In some regions, like the Irish, Baltic and Black Seas, the concentration of ^{137}Cs in the marine environment depends on the input due to discharges from reprocessing facilities and from the Chernobyl accident, and in these regions the evolution of its concentration is quite dynamic.

The interest in, and relevance of knowledge of the behaviour of radionuclides in the ocean derives from a variety of issues. Firstly, the fate of radionuclides needs to be well understood to provide the basis for an assessment of adverse environmental or human health consequences, real or perceived. Then, this accumulated knowledge provides a critical basis for rapid assessment of the impact of future releases — especially unplanned ones. These include accidents involving radionuclide releases from such sources as coastal nuclear facilities, nuclear waste disposal sites or from maritime transport of nuclear fuel or nuclear wastes. Finally, radionuclides are powerful tracers providing basic insights into a variety of oceanic processes. For example, plutonium as a particle active radio-element, is readily incorporated into several compartments of the ocean carbon cycle. Because of the relatively well defined temporal and spatial aspects of its introduction to the ocean, its movement within the ocean provides many insights into a large number of processes in the oceanic water column, and in biological and sedimentary systems.

Assessment of marine radioactivity in a given region therefore requires knowledge of the source terms and an understanding of oceanic processes. The deposition of radionuclides from these sources is unevenly distributed over the world ocean. The input function of global fallout in the world ocean, mainly due to nuclear weapons tests carried out in the sixties, can be estimated from worldwide monitoring mainly performed on land. On the other hand, discharges from nuclear fuel reprocessing plants or dumping of liquid and solid radioactive wastes have generally had a more local character, although soluble radionuclides have been transported over long distances by prevailing currents.

In order to estimate radionuclide inputs from local sources, radionuclide distributions in the world's oceans and seas must be better known so that results from scientific expeditions to nuclear waste dumping sites and/or other sources of anthropogenic marine radioactivity can be reviewed objectively, comparatively and comprehensively. This requires a detailed study

of the present distributions and inventories of the main radionuclides of interest (^3H , ^{14}C , ^{90}Sr , ^{137}Cs and Pu isotopes) in the world's oceans and seas as well as possible predictions of their concentrations in the near future.

As a contribution to these investigations, in 1995 the International Atomic Energy Agency's Marine Environment Laboratory (IAEA-MEL) commenced a seven year project "Research on Worldwide Marine Radioactivity" with the generous support of the Science and Technology Agency (STA) of the Government of Japan. As part of this project a four year (1998–2001) Coordinated Research Project (CRP) entitled "Worldwide Marine Radioactivity Studies (WOMARS)" was carried out.

The objectives of the CRP were to :

- (i) Identify the major sources of anthropogenic radionuclides in the world ocean,
- (ii) Develop present knowledge of the distributions of key radionuclides (^{90}Sr , ^{137}Cs and Pu isotopes) in water and sediment of the world ocean,
- (iii) Study the development of radionuclide concentrations in water with time using good quality historical data (e.g. from the GEOSECS programme of the mid-1970s and new data sets collected recently).

The main aim of the CRP was to develop an understanding of the present open ocean distribution of radionuclides in the water column and sediment and thus predict the radiological impact to be addressed. Further, to encourage and support marine radioactivity studies in Member States by methodological assistance and total analytical control management. The project was designed to review and contribute to scientific knowledge of the processes affecting radionuclide distributions and the sources which have introduced radionuclides to the world ocean.

Three anthropogenic radionuclides (^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$) were chosen as the most important and radiologically typical for beta-, gamma- and alpha-emitters. They are the most abundant anthropogenic radionuclides of each class present in the marine environment and can lead to the highest radiation doses to humans and marine biota.

In this publication, the state of knowledge of the distribution and behaviour of radionuclides in the world ocean and indications of the changes which have evolved over time are discussed. It will not constitute a comprehensive review of all studies involving radionuclides in the ocean but will try to cover the most relevant aspects. The studies cited generally provide reference to a related work and thus fuller details should be sought in the quite large literature base on this topic.

The CRP participants collected large amounts of data from their respective regions of interest as well as from other regions, consideration being given to the entire coverage of the oceans of the world. In addition, specific tasks were assigned to collect information from countries not participating in the CRP so that these data could also be included in the Global Marine Radioactivity Database (GLOMARD), which is under development in the Radiometrics Laboratory at IAEA-MEL.

The information collected is of high scientific value and represents a reference source on average radionuclide levels in the marine environment. The present report summarizes the results obtained in the framework of the CRP.

2. INPUT OF ANTHROPOGENIC RADIONUCLIDES INTO THE WORLD OCEAN

The world ocean covers an area of $361.11 \cdot 10^6$ km² or 70.8% of the total surface of the Earth [1]. Hence, the ocean is a major recipient of anthropogenic radionuclides released to the environment by atmospheric as well as aquatic pathways.

The oceans may receive radioactive fallout directly from the atmosphere and indirectly as runoff from rivers. Radionuclides may, however, also be discharged directly into the ocean as liquid waste or from dumped solid wastes. Some radionuclides will behave conservatively and stay in the water in soluble form, whereas others will be insoluble or adhere to particles and thus, sooner or later, be transferred to marine sediments.

The three main sources of anthropogenic radionuclides are nuclear weapons testing in the atmosphere, the Chernobyl accident and water-borne discharges from nuclear reprocessing plants, notably Sellafield in the UK and Cap de la Hague in France [2]. Other sources e.g. sea dumping of nuclear waste, routine discharges from nuclear power plants, sunken nuclear submarines, satellite losses, lost nuclear weapons and the use of radioisotopes in medicine, industry and science, contribute orders of magnitude less to the contamination of the world ocean than the three major sources.

The aim of this chapter is, on the basis of the most recent information [2] to present an estimate of the input of anthropogenic radionuclides to the world ocean, and to calculate the inventories of these radionuclides as of the year 2000. As ⁹⁰Sr and in particular ¹³⁷Cs are the main contributors to the present radiation doses from man-made sources in the environment [3], the focus is on these two radionuclides.

2.1. Nuclear weapons testing

Atmospheric nuclear test explosions have contaminated the Earth globally with radioactive debris. The weapons tested from 1945 to 1951 were fission weapons in the 20–100 kilotons TNT range. They produced mainly tropospheric fallout, i.e. the debris remained below the tropopause and was not dispersed globally, but was deposited around the latitude band of the test site. However, in 1952 the United States and in 1953 the former Soviet Union (FSU) tested their first thermonuclear devices (megatons TNT range) and fallout from these explosions occurred worldwide. In the following years and up until the early sixties the major inputs of radionuclides from nuclear weapons testing occurred at Novaya Zemlya Island in the Arctic Ocean (80.89 Mt fission yield) and at Bikini Atoll in the Pacific Ocean (42.2 Mt fission yield). Other important locations in this respect have been Enewetak and Johnston Atolls in the North Pacific (26 Mt), Christmas Island in the Indian Ocean (15.5 Mt), Lop Nor in China (12.2 Mt), Mururoa and Fangataufa Atolls in the South Pacific (6.1 Mt) and Semipalatinsk in Kazakhstan (3.7 Mt).

The distribution of global fallout (based on data from [2]) is shown in Fig. 1. The data are from a worldwide network of land-based measuring stations for ⁹⁰Sr in fallout. The global fallout pattern first of all reflects the atmospheric transport of debris from the stratosphere to the troposphere with maximum transfer at the mid-latitudes. The global precipitation pattern and the locations of the test sites also influence the global distribution of weapons fallout. The significantly lower amount of fallout in the Southern hemisphere is due to the relatively low number of test explosions there, but also to the limited atmospheric exchange between the northern and southern stratospheres.

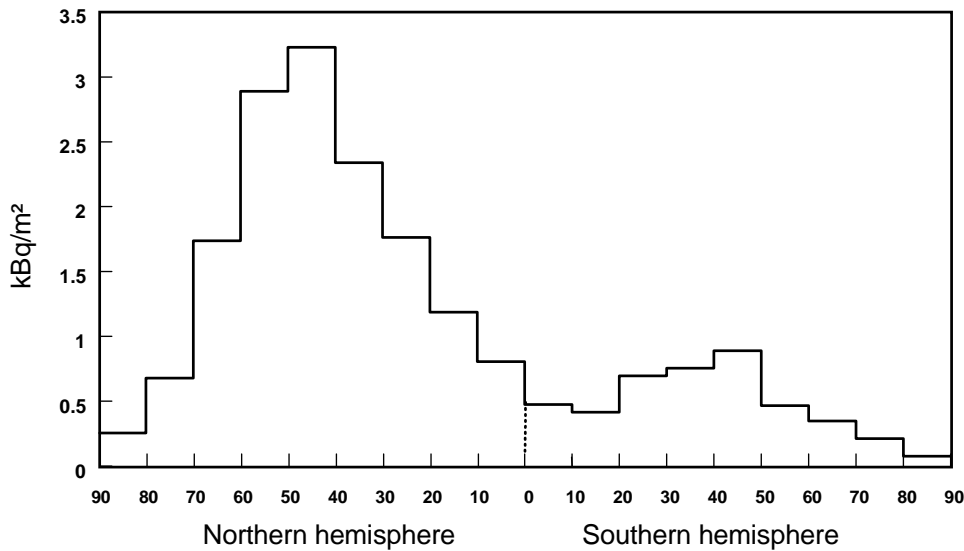


FIG. 1. Integrated deposition density of ⁹⁰Sr (kBq/m²) (cumulative deposition is 0.40 x integrated in 2000) [2].

Table I shows the inputs of ⁹⁰Sr and ¹³⁷Cs from global fallout to the various parts of the world ocean based on fallout over land. The ¹³⁷Cs data were obtained from ⁹⁰Sr data by multiplying by 1.6 [2]. The deposition densities (Bq/m²) in the various latitude bands were from [2] and the ocean areas were obtained from [1]. Figure 2 shows the relative input of ⁹⁰Sr from global fallout to the various parts of the world ocean. The relative areas of the four oceans compared to the total area of the world ocean are for the Pacific, Atlantic, Indian and Arctic Oceans: 50%, 26%, 20% and 4%, respectively. Hence, the Indian and Arctic Oceans have received less input of ⁹⁰Sr from global fallout than could be expected from their respective areas, whereas the Atlantic has received about 25% more than expected. The discrepancies are explained by the lower fallout to the Arctic and Southern hemisphere than to the temperate regions of the Northern hemisphere (see also Fig. 3 where the latitudinal distribution of ⁹⁰Sr input in PBq is shown). In this context it should be noted that present radionuclide inventories in the Arctic Ocean are higher than those expected from direct input due to transport from temperate latitudes in the North Atlantic via the Norwegian Coastal Current.

In order to obtain the actual (year 2000) inventories in the world ocean, the values in Table I should be decay-corrected by a factor of 0.40. The same decay factor has been used for both ⁹⁰Sr and ¹³⁷Cs.

UNSCEAR [2] has, based on the fission yields of the nuclear weapons tested, reported the global release (PBq) of those radionuclides which have been globally dispersed in atmospheric nuclear testing. Table II shows those still present in the environment.

TABLE I. INPUTS FROM GLOBAL FALLOUT (PBq)

Summary of ^{90}Sr input to the world ocean from global fallout

Latitude	Arctic Ocean	Atlantic Ocean	Indian Ocean	Pacific Ocean	Total	Ocean area 10^{12}m^2
N 90°-60°	4.6	10.2	0.0	1.3	16.1	17.2
N 60°-30°	0.0	57.3	0.0	71.6	128.9	46.8
N 30°-0°	0.0	30.8	13.3	65.8	109.9	90.6
S 0°-30°	0.0	10.3	14.6	26.6	51.5	98.0
S 30°-60°	0.0	15.4	22.5	25.8	63.7	88.2
S 60°-90°	0.0	1.7	2.1	3	6.8	20.4
Total	4.6	125.7	52.5	194.1	376.9	361.1

Summary of ^{137}Cs input to the world ocean from global fallout

N 90°-60°	7.4	16.3	0.0	2.1	25.8
N 60°-30°	0.0	91.7	0.0	114.6	206.2
N 30°-0°	0.0	49.3	21.3	105.3	175.8
S 0°-30°	0.0	16.5	23.4	42.6	82.4
S 30°-60°	0.0	24.6	36.0	41.3	101.9
S 60°-90°	0.0	2.7	3.4	4.8	10.9
Total	7.4	201.1	84.0	310.6	603.0

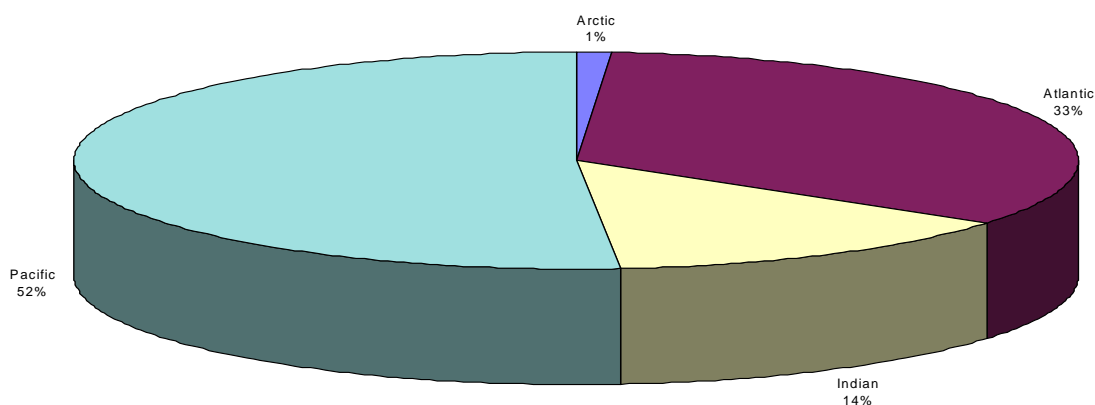


FIG. 2. Relative input of ^{90}Sr from global fallout to the world ocean.

The inputs to the ocean for ^{90}Sr and ^{137}Cs were taken from Table I. The other radionuclides were calculated from ^{90}Sr and their release ratios to ^{90}Sr . As ^{241}Pu decays to ^{241}Am with a half-life of 14.35 years, most of the ^{241}Pu deposited in the ocean (and elsewhere) has already decayed to ^{241}Am .

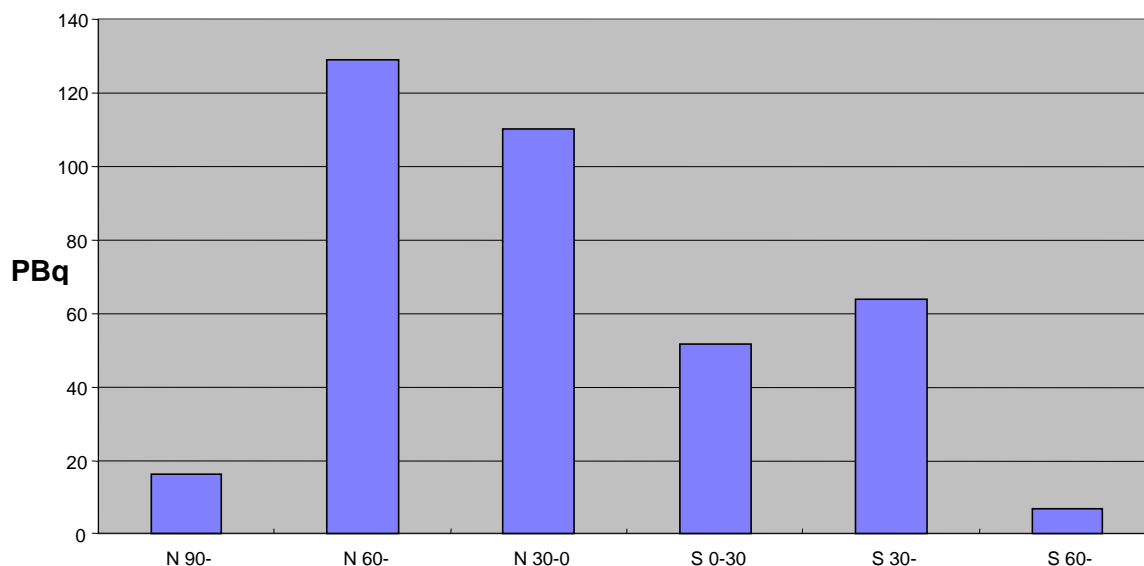


TABLE II. GLOBAL FALLOUT RADIONUCLIDES STILL PRESENT IN PBq-QUANTITIES IN THE YEAR 2000

Nuclide	Half-life (year)	Global release (PBq)	Release ratio to ^{90}Sr	Input to ocean (PBq)	Ocean inventory 2000 (PBq)
^3H	12.33	186000	299	112693	13300
^{14}C	5730	213	0.3424	129	128
^{90}Sr	28.78	622	1	377	151
^{137}Cs	30.07	948	1.52	603	251
^{239}Pu	24110	6.52	0.0105	4.0	4.0
^{240}Pu	6563	4.35	0.007	2.6	2.6
^{241}Pu	14.35	142	0.2283	86	13.7

According to [2] the total fission yield of atmospheric nuclear test explosions is 189 Mt (TNT). The production estimate of ^{90}Sr is 3.88 PBq Mt^{-1} . Hence, the total production of ^{90}Sr from nuclear weapons testing in the atmosphere is 733 PBq. However, 29 Mt (corresponding to 113 PBq ^{90}Sr) were not globally dispersed, but were deposited as local fallout close to the test sites. Hence, only 160 Mt or 622 PBq ^{90}Sr became global fallout. Since about 2–3% of ^{90}Sr decayed before deposition, the total amount deposited is calculated to be 605 PBq ^{90}Sr . From measurements of the global deposition of ^{90}Sr (Fig. 1), the global input from nuclear weapons testing in the atmosphere is calculated to be 604 PBq ^{90}Sr . It may thus be concluded that measurements and calculations from fission yields agree.

As nearly all local fallout (28 Mt fission) occurred at the US test sites at Bikini and Enewetak Atolls in the Pacific Ocean (0–30°N latitude belt), it is assumed that this part of the Pacific Ocean may have received about 113 PBq ^{90}Sr besides the 66 PBq ^{90}Sr (see Table I) received from global fallout. However, the 113 PBq may not be fully dispersed in sea water as some of it is imbedded in minerals (corals). Hence, it is difficult to estimate to what extent the inventory of ^{90}Sr in the world ocean has increased from this source. It is even more difficult to

estimate the contribution from local fallout for the other radionuclides mentioned in Table II. Only actual environmental measurements in this part of the Pacific Ocean can give an indication of the possible contamination to the world ocean from local fallout at Bikini and Enewetak.

Local fallout from the Novaya Zemlya test site has probably not contributed significantly to the radionuclide inventories in the ocean. According to [2], the debris from these tests went almost entirely (96%) to the stratosphere. Less than 0.5% was distributed as local and regional fallout and the remaining part was deposited as tropospheric fallout.

2.2. Nuclear reprocessing

Reprocessing is carried out to recover U and Pu from spent fuel for reuse in reactors. The major reprocessing plants are in the United Kingdom (Sellafield), France (Cap de La Hague) and Japan (Tokai). Sellafield has been the major source of radioactive contamination of the marine environment. In the 1970s British Nuclear Fuel Ltd., Sellafield (earlier Windscale), began to substantially increase its authorized water-borne radioactive discharges to the Irish Sea. These discharges, first of all of ^{137}Cs , but also other radionuclides, notably plutonium isotopes, americium and technetium, have become measurable in most parts of the NE Atlantic and to a large extent in the Arctic Ocean. Table III and Figs 4 and 5 show the annual liquid discharges of ^{137}Cs and ^{90}Sr from Sellafield and Cap de La Hague, respectively. Furthermore, the decay-corrected amounts (to year 2000) are shown. While the ^{90}Sr discharges from Cap de la Hague amounted to 20% of those from Sellafield, the discharges of ^{137}Cs were less than 3% of the corresponding Sellafield releases. Apart from the 40 PBq ^{137}Cs and 6.5 PBq ^{90}Sr released from the two European reprocessing plants, 126 PBq ^3H , 1 PBq ^{99}Tc and 0.015 PBq ^{129}I contribute to the contamination of the ocean at present and the NE Atlantic (30-90°N) in particular. Whereas the input of ^3H from reprocessing is negligible compared with that from global fallout (see Table II), Sellafield and La Hague are major sources to the oceanic inventories of ^{99}Tc and ^{129}I . These two very long lived radionuclides (half-lives: 213,000 and 1,570,000 years, respectively) will remain in the marine environment long after all the other radionuclides mentioned in this context (Table II) have disappeared.

2.3. The Chernobyl accident

Approximately two thirds of the ~100 PBq ^{137}Cs released by the Chernobyl accident in 1986 were deposited outside the former Soviet Union [4]. Although most of this activity fell over land, a significant part went to the sea. In particular, the European marginal seas received a substantial amount of the Chernobyl debris. The Baltic Sea was the most contaminated. The total inventory from the accident was calculated to be 4.5 PBq ^{137}Cs [5]. Since Chernobyl, the Baltic Sea has been a main source of fresh inflow of ^{137}Cs to the NE Atlantic Ocean.

The Black Sea received 2–3 PBq ^{137}Cs from the Chernobyl accident [6]. The outflow from the Black Sea is the main source of “new” ^{137}Cs in the Mediterranean Sea. However, the inflow is less than the decay of the present ^{137}Cs Chernobyl Mediterranean Sea inventory of 3–5 PBq. The Mediterranean Sea also received direct fallout from the accident. From German and British measurements it has been estimated [7], that the North Sea received 1.2 PBq ^{137}Cs from Chernobyl. The Chernobyl inventory in the NE Atlantic was estimated at about 6 PBq ^{137}Cs . In the summer of 1987, surface sea water samples collected in the Greenland, Norwegian and Barents Seas and off the west coast of Norway and the Faroe Islands, all contained Chernobyl-derived ^{137}Cs .

TABLE III. RADIOACTIVE DISCHARGES FROM SELLAFIELD AND CAP DE LA HAGUE, 1970–1998 IN TBq

Year	<i>Sellafield</i>				<i>Cap de la Hague</i>			
	^{137}Cs	^{90}Sr	$^{137}\text{Cs ac}$	$^{90}\text{Sr ac}$	^{137}Cs	^{90}Sr	$^{137}\text{Cs ac}$	$^{90}\text{Sr ac}$
1970	1200	230	600	112	89	2	45	1
1971	1300	460	665	229	243	8	124	4
1972	1289	562	675	286	33	16	17	8
1973	770	280	413	146	69	19	37	10
1974	4100	390	2249	208	56	52	31	28
1975	5230	466	2936	255	34	38	19	21
1976	4289	381	2464	214	35	20	20	11
1977	4480	427	2634	245	51	36	30	21
1978	4090	597	2460	351	39	70	23	41
1979	2600	250	1601	151	23	56	14	34
1980	2970	352	1871	217	27	29	17	18
1981	2360	277	1522	175	39	27	25	17
1982	2000	319	1320	207	51	86	34	56
1983	1200	204	810	135	23	142	16	94
1984	434	72	300	49	30	110	21	75
1985	325	52	230	36	29	47	21	33
1986	18	18	13	13	10	69	7	49
1987	12	15	9	11	8	57	6	42
1988	13	10	10	7	9	40	7	30
1989	29	9	22	7	13	29	10	22
1990	24	4	19	3	13	16	10	13
1991	15	4	12	3	6	30	5	24
1992	15	4	12	3	3	18	2	15
1993	22	17	19	14	4	25	3	21
1994	14	29	12	25	11	16	10	14
1995	12	28	11	25	5	30	4	27
1996	10	16	9	15	2	11	2	10
1997	8	37	7	34	3	4	3	4
1998	8	18	7	17	3	3	3	3
Total	38837	5528	22911	3195	961	1106	566	744

Note: **ac** is the actual activity by the year 2000.

The total Chernobyl ^{137}Cs input to the world ocean is estimated at about 16 PBq. It has been assumed here that the input of ^{90}Sr to the sea was negligible. The inputs of ^{137}Cs were estimated at 10 PBq in the 30–60°N latitude belt and at 6 PBq in the 60–90°N belt. Nearly all ^{137}Cs went to the N Atlantic and Arctic Oceans. The relative input to the ocean thus became significantly less than that from nuclear weapons fallout, because the Chernobyl accident was a tropospheric event, first of all contaminating the surrounding European land areas.

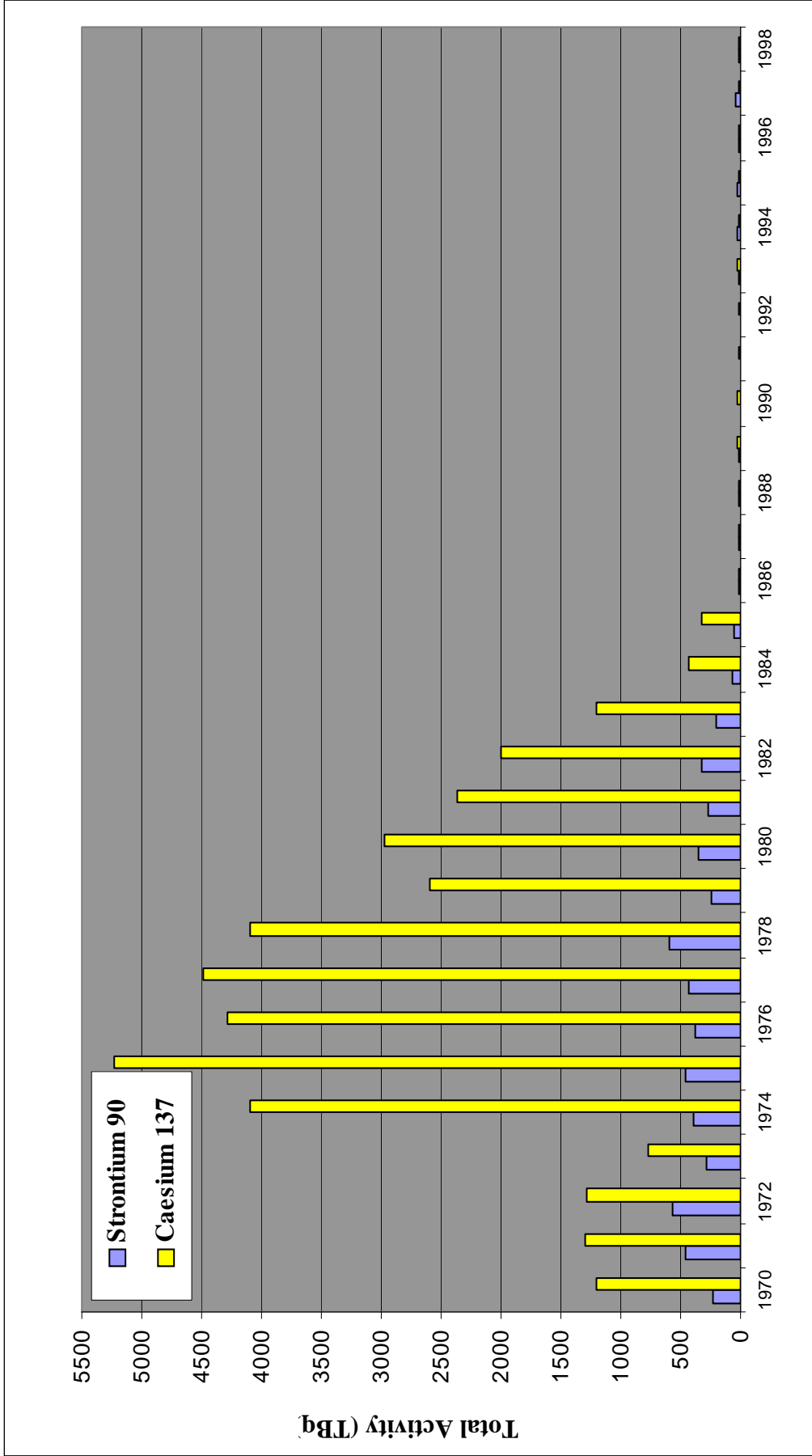


FIG.4. Radioactive discharges from Sellafield, 1986-1998 in TBq.

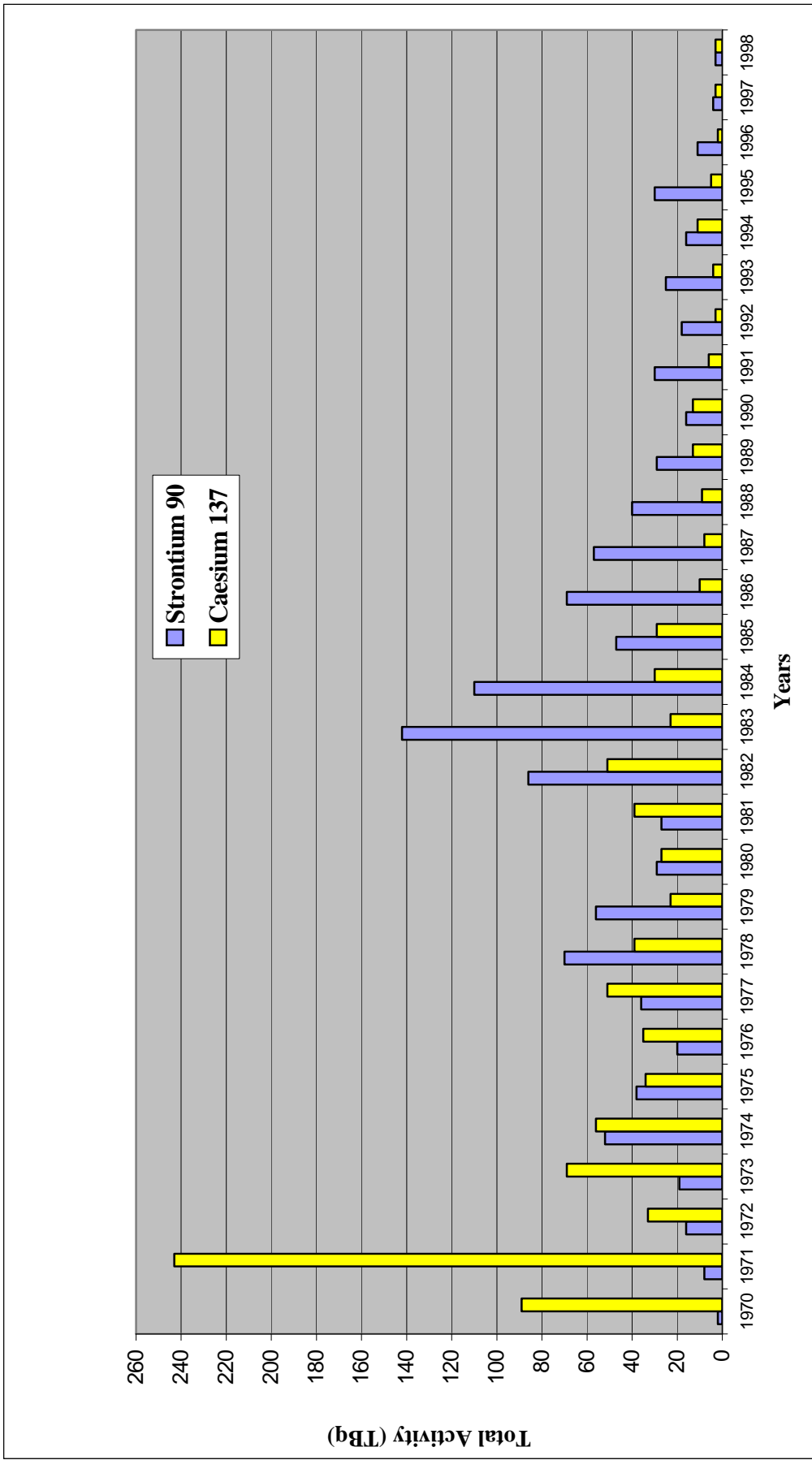


FIG. 5. Radioactive discharges from Cap de la Hague, 1970–1998 in TBq.

2.4. Other nuclear accidents

A few nuclear accidents have released radionuclides directly into the sea. Most accidents have, however, involved releases to the atmosphere. The marine environment has thus received only indirect contamination either from atmospheric deposition or through runoff from land.

The two major nuclear accidents in 1957, Kyshtym and Windscale, are not believed to have contributed significantly to the contamination of the marine environment.

In 1968 a US B-52 aircraft crashed during an emergency landing on sea-ice 11 km west of Thule Airbase in NW Greenland. A similar accident had occurred two years previously at Palomares in SE Spain [8], but the radioactive debris from that accident had mainly been deposited on land. At Thule, the marine environment became contaminated with about 1 TBq $^{239,240}\text{Pu}$ [9].

A number of nuclear submarines, American as well as Russian, have been lost in the world ocean. The most recent loss was the Russian submarine "Kursk" in the Barents Sea in August 2000. This submarine was recovered in 2001 and no contamination of the sea occurred. Another loss was the Soviet "Komsomolets" submarine, which sank at a depth of 1700 m at Bear Island in the eastern part of the Norwegian Sea. The activity in the wreck is estimated at 2.8 PBq ^{90}Sr and 3 PBq ^{137}Cs , and the nuclear warheads may contain 16 TBq $^{239,240}\text{Pu}$ [10]. Close to the "Komsomolets", slightly enhanced levels of ^{137}Cs have been measured in sea water, but the sunken nuclear submarines may be considered only as potential sources to radioactive contamination of the ocean.

Some satellites are nuclear powered. On a few occasions such satellites have burned up in the upper atmosphere and have thereby contributed to the contamination of the ocean. This was the case in 1964 when a SNAP-9A nuclear power generator aboard a US satellite re-entered the atmosphere in the Southern hemisphere [11]. The generator contained 0.6 PBq ^{238}Pu . Since the accident, seawater samples from the Southern hemisphere have shown an enhanced $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratio, compared to ocean water from the Northern hemisphere.

2.5. Sea dumping

Sea dumping in the western world was carried out on a national basis, from the late 1940s to the mid-1960s, mainly by the US in the Atlantic and Pacific Oceans and by the UK in the NE Atlantic. In 1967, the European Nuclear Energy Agency (later NEA) organized an international operation where about 0.3 PBq solid waste was deposited at a depth of 5 km in the eastern Atlantic Ocean. Similar international operations continued until 1982. At that time about 0.7 PBq α activity, 42 PBq β activity and 15 PBq tritium had been dumped in the N Atlantic [7, 12]. The Coordinated Research and Environmental Surveillance Programme Related to Sea Disposal of Radioactive Waste "CRESP" has assessed the radiological impact of the NEA dumping activities [13]. Minor releases of Pu from the dumped waste have been measured at the dumpsite.

The former Soviet Union (FSU) disposed of radioactive waste for over three decades in the Kara and Barents Seas. The total amount of waste dumped in the Arctic Seas was estimated to be approximately 90 PBq [14]. The International Arctic Seas Assessment Project (IASAP) study [15] has estimated that the inventory in 1993/94 had decayed to 4.7 PBq. Monitoring has shown [10], that the releases from the dumping sites are small. Model calculations have demonstrated that the total collective dose over the next 1000 years to the

world's population from the Russian dumpsite is in the order of 10 man Sv [15]. This is 300 times less than the corresponding dose from the radioactive waste dumped in the NE Atlantic [13].

2.6. Use of radioisotopes

Radioisotopes are used in medicine, industry and research. Those most widely used as unsealed sources are usually short lived, e.g. ^{99m}Tc and ^{131}I and are thus of no importance as sources to worldwide marine contamination. Hence, although the global usage of ^{131}I in nuclear therapy is approximately 0.6 PBq, it does not result in a measurable inventory in the world ocean.

Longer lived radionuclides such as ^3H and ^{14}C are used in research, but releases to the environment are very low and compared to the oceanic inventories from global fallout, of no significance.

2.7. Hypothetical and potential inputs

During 1949–1951, medium and low level liquid radioactive waste from the Soviet nuclear establishment, Mayak in the South Urals was released into the Techa River. In total 100 PBq were discharged. ^{90}Sr and ^{137}Cs contributed 11.6% and 12.2%, respectively [16]. Most of the activity was retained in a cascade of reservoirs in the upper reaches of the Techa River, which is part of the Ob River system. Although the distance from Mayak to the outlet of the Ob River in the Arctic Ocean is about 2000 km, it cannot be ruled out that ^{90}Sr which is more mobile than ^{137}Cs , may have reached the Arctic Ocean around 1950. The contamination may be estimated to be in the order of 1 PBq [17].

Since 1951, Lake Karachay at Mayak has been used for disposal of radioactive waste. The lake has received 3.6 EBq ^{137}Cs and 0.74 EBq ^{90}Sr [18]. Lake Karachay is a potential source for future contamination of the Ob River system and thus of the Arctic Ocean. The Krasnoyarsk Mining and Chemical Complex is another hypothetical source to radioactive contamination of the Arctic Ocean. Other nuclear installations in the FSU (and elsewhere) should also be kept in mind in this context. In e.g. Murmansk and Vladivostok, radioactive waste is stored in the military naval bases and at remote locations, also along the Siberian coastline lighthouses are powered by ^{90}Sr generators. Each generator contains in the order of 10 PBq ^{90}Sr and in the event of an accident is a potential source of environmental contamination. A ^{90}Sr generator was lost in the Sea of Okhotsk, but no contamination has been reported.

2.8. River runoff

Radionuclides deposited in the terrestrial environment may be transported to groundwater, lakes and rivers, and later to the sea. Runoff is thus a secondary transfer of atmospheric radioactivity to the sea. About 9% of the ^{90}Sr inventory on land will be removed by runoff [19]. For ^{137}Cs the figure is about 2% [20].

As the total deposits of global fallout ^{90}Sr and ^{137}Cs are 622 PBq and 948 PBq, respectively, and as 377 PBq ^{90}Sr and 603 PBq ^{137}Cs were deposited in the sea, the terrestrial input has been 245 PBq ^{90}Sr and 345 PBq ^{137}Cs . Furthermore, the total deposition from Chernobyl was ~10 PBq ^{90}Sr and ~85 PBq ^{137}Cs [2]. Of this, 10 PBq ^{90}Sr and 69 PBq ^{137}Cs fell over land.

The global runoff of ^{90}Sr is thus calculated as: $0.09(245+10) = 23 \text{ PBq}$ and that of ^{137}Cs becomes: $0.02(345+69) = 8 \text{ PBq}$. By the year 2000 these inputs would have decayed to about $9 \text{ PBq } ^{90}\text{Sr}$ and $3 \text{ PBq } ^{137}\text{Cs}$. In other words runoff may increase the ^{90}Sr inventory in the world ocean by 6% and the ^{137}Cs inventory by 1–2%, which is insignificant when other uncertainties are taken into consideration. Furthermore, radionuclides transported by runoff will often be bound to particulates and thus be sedimented rather than transferred to biota. Hence, runoff is not believed to influence the marine pathway ingestion doses to man significantly.

2.9. Summary of inputs

The total input from all sources of ^{90}Sr to the world ocean has been 383 PBq (377 PBq from global fallout and 6.5 PBq from reprocessing). The input of ^{137}Cs was 659 PBq (604 PBq from fallout, 40 PBq from reprocessing plants) and 16 PBq from Chernobyl. To these figures should be added local inputs in the equatorial N Pacific, which may be in the order of 100 PBq for each of the two radionuclides. Table IV summarizes the present (year 2000) inventories of ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ in the world ocean from all major sources, including local fallout.

TABLE IV. RADIONUCLIDE INVENTORIES IN THE YEAR 2000 IN THE WORLD OCEAN (PBq)

Arctic & Atlantic Oceans						Indian & Pacific Oceans			World
Latitude	Global fallout	Local fallout	Repro-cessing	Chernobyl	Subtotal	Global fallout	Local fallout	Subtotal	ALL SOURCES
^{90}Sr									
N 90°-30°	28.8	0.0	3.9	0.0	32.7	29.2	0.0	29.2	61.9
N 30°-0°	12.3	0.0	0.0	0.0	12.3	31.6	22.5	54.1	66.5
S 0°-30°	4.1	0.0	0.0	0.0	4.1	16.5	22.5	39.0	43.1
S 30°-90°	6.8	0.0	0.0	0.0	6.8	21.4	0.0	21.4	28.2
TOTAL	52.1	0.0	3.9	0.0	56.0	98.6	45.0	143.6	199.7
^{137}Cs									
N 90°-30°	46.1	0.0	23.5	11.5	81.1	46.7	0.0	46.7	127.8
N 30°-0°	19.7	0.0	0.0	0.0	19.7	50.6	36.0	86.6	106.3
S 0°-30°	6.6	0.0	0.0	0.0	6.6	26.4	36.0	62.4	69.0
S 30°-90°	10.9	0.0	0.0	0.0	10.9	34.2	0.0	34.2	45.1
TOTAL	83.4	0.0	23.5	11.5	118.4	157.8	72.0	229.8	348.2
$^{239,240}\text{Pu}$									
N 90°-30°	1.3	0.0	0.0	0.0	1.3	1.3	0.0	1.3	2.5
N 30°-0°	0.5	0.0	0.0	0.0	0.5	1.4	1.0	2.4	2.9
S 0°-30°	0.2	0.0	0.0	0.0	0.2	0.7	1.0	1.7	1.9
S 30°-90°	0.3	0.0	0.0	0.0	0.3	0.9	0.0	0.9	1.2
TOTAL	2.3	0.0	0.0	0.0	2.3	4.3	2.0	6.3	8.6

It has been assumed that the inputs of local fallout from the US Pacific tests were $113 \text{ PBq } ^{90}\text{Sr}$, $181 \text{ PBq } ^{137}\text{Cs}$ and $2 \text{ PBq } ^{239,240}\text{Pu}$. It was further assumed that this fallout was equally divided between latitude bands N 30-0° and S 0-30°. ^{90}Sr and ^{137}Cs had by the year 2000 decayed to 45 PBq and 72 PBq , respectively.

The major inputs from global fallout occurred in the late fifties and early sixties, reprocessing discharges peaked in the last half of the seventies and nearly all of the fallout from Chernobyl was deposited in 1986. There has been some redistribution of the radionuclides in the ocean due to both advection and mixing processes. Water-borne releases from reprocessing in western Europe and global fallout have thus been dispersed to the Arctic Ocean and contamination from the Baltic Sea, due to the Chernobyl accident, has reached the North Atlantic and Arctic Oceans.

The NE Atlantic is the part of the world ocean most contaminated by radionuclides because of ^{137}Cs input from all three major sources: global fallout, reprocessing and Chernobyl. The lowest input of anthropogenic radionuclides has been to the waters surrounding the Antarctic continent. In these southern parts of the Pacific, Indian and Atlantic Oceans, the concentrations of e.g. ^{137}Cs are about 40 times lower than those found in the NE Atlantic [21].

Although the ocean contains most of the anthropogenic radionuclides released into the environment, the radiological impact of this contamination is low. Radiation doses from naturally-occurring radionuclides in the marine environment (e.g. ^{210}Po), are on the average two orders of magnitude higher [21].

3. PROCESSES AFFECTING RADIONUCLIDE CONCENTRATIONS AND INVENTORIES IN THE OCEANS

A wide range of radionuclides has been released into the oceans. Their distribution in space and time can be quite complex, but is always related to four general processes: the input function/source, radioactive decay, biogeochemistry, and oceanic processes.

A brief review of each of these processes is made below and provides examples and references to studies that incorporate these processes into the interpretation of environmental radionuclide distributions and inventories. It suffices to say that in most instances, considerable ancillary, non-radiochemical data are needed to interpret ocean radionuclide distributions. Thus the most basic studies require a knowledge of local oceanographic conditions in addition to the physical properties of the radionuclide of interest. This review will focus primarily on the man-made, or anthropogenic radionuclides, though considerable effort has been spent in ocean sciences understanding naturally-occurring radionuclides, such as the uranium/thorium decay series. Insights into the expected fate of anthropogenic radionuclides can often be gained by comparisons between appropriate natural and anthropogenic radionuclides.

3.1. Radionuclides as oceanographic tracers

The most fundamental properties needed to understand marine radionuclide distributions are the half-life of the radionuclide, and information on its source. While those isotopes with the shortest half-lives (<days - months) often are of greatest radioecological concern (^{131}I , $t_{1/2} = 8.0$ days), radionuclides with longer half-lives pose a greater potential for redistribution, bioaccumulation, and storage within the oceans and underlying sediment. Also useful with respect to half-lives, is the ability to identify different radionuclide sources, due to the presence of shorter-lived isotopes in any particular sample. For example, the presence of ^{134}Cs ($t_{1/2} = 2.07$ yrs) in surface ocean water shortly after the Chernobyl accident, allowed for the separation of Cs derived from the more recent 1986 source and prior sources of Cs such as weapons testing fallout which by the 80s, had essentially zero ^{134}Cs [22].

TABLE V. CHARACTERISTIC ISOTOPIC RATIOS COMMONLY USED TO IDENTIFY ANTHROPOGENIC RADIONUCLIDE SOURCES IN THE MARINE ENVIRONMENT

Source	$^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio
Global weapons testing fallout	0.18
Nevada test site	0.03
Pacific test sites	>0.30
Novaya Zemlya/Barents Sea	<0.03
Mururoa	<0.06
Source	$^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio in 1986
Global weapons testing fallout	0
Chernobyl fallout	0.5
Source	$^{137}\text{Cs}/^{90}\text{Sr}$ activity ratio
Global weapons testing fallout	1.5 (in 1962, peak fallout year)
Chernobyl fallout	5 (in 1986, year of release)
Sellafield discharges	11 (in 1975, peak discharge year)

If decay-corrected to April 26, 1986, $^{134}\text{Cs}/^{137}\text{Cs}$ ratios of 0.5 were found to be characteristic of the source and subsequent fallout (Table V [23]). Similar examples of using short lived radionuclides to pinpoint a source can be found from the early weapons testing years (e.g. ^{141}Ce and ^{144}Ce in deep ocean sea cucumbers [24]).

All radionuclide data are reported with a reference date, generally either the sampling date or date of release, so that changes in the reported distributions can be attributed to environmental, rather than decay processes. Sources of anthropogenic radionuclides have been reviewed elsewhere (this report, Chapter 2, [25, 26]).

Many of the anthropogenic radionuclides from both fallout and local sources have been used by oceanographers as "tracers" of marine processes. Of great interest, for example in ocean sciences, has been the distribution of fallout ^{14}C and tritium as tracers of the rates of ocean mixing (see below). It should also be noted in this regard that fallout delivery to the Northern hemisphere was much greater than to the Southern hemisphere, thus for the world ocean, there are generally lower activities of all anthropogenic radionuclides in sea water, sediment and biota south of the equator. Also, due to local fallout from weapons tests conducted in the Pacific, the inventories of Pu for example are higher than in soil samples at comparable latitudes (Fig. 6). In terms of local sources, there has been considerable use of Sellafield-derived ^{137}Cs and ^{90}Sr (Fig. 7), and Cap de la Hague-derived ^{129}I to trace water movement in the North and Baltic Seas and into the Arctic Ocean [27, 28]. An additional category of local source input to the ocean is found in indirect input from rivers draining into the ocean. An example of this would be delivery of ^{90}Sr from the Chernobyl reactor site via the Dnepr river to the Black Sea (Fig. 8 [29]).

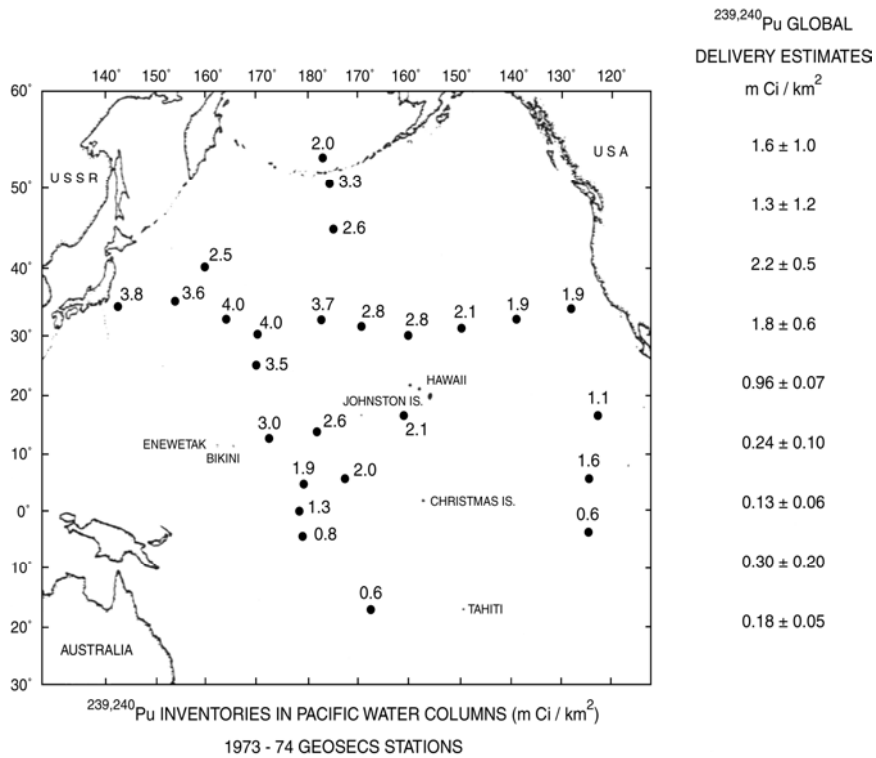


FIG. 6. ^{239,240}Pu inventories in Pacific water columns (mCi/km²) from GEOSECS [30]. Note that the water column inventories in the NW Pacific are higher than those expected from global fallout delivery (column on the right), due to local fallout from testing at Enewetak and Bikini.

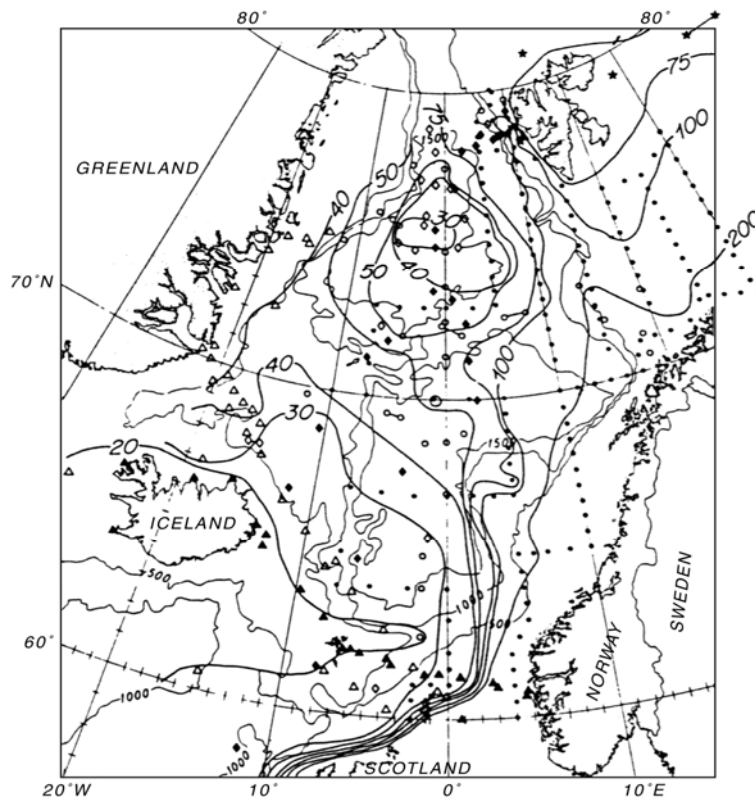


FIG. 7. Surface distributions of ¹³⁷Cs (disintegrations per minute per 100 kg as of 1 January 1981) in the Norwegian-Greenland Seas. Samples were collected in 1981 (dark symbols) and 1982 (light symbols). Note highest values north of Scotland due to Sellafield discharges [27].

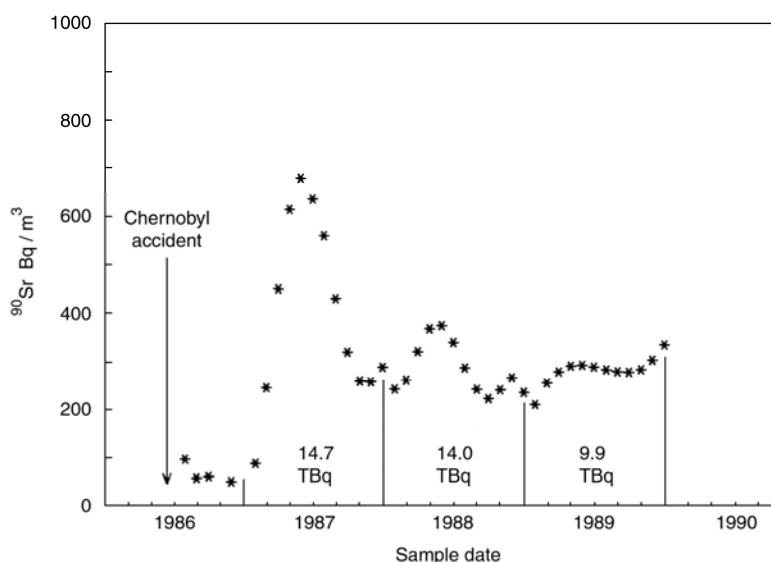


FIG. 8. Average ^{90}Sr concentrations in the Dnepr river near the Black Sea. Total fluxes per year are also indicated. Note large increase in ^{90}Sr concentrations after Chernobyl [29].

3.2. Fate of conservative vs. particle-reactive radionuclides

Once released into the ocean, the fate of each isotope is largely tied to its chemical properties. Broadly speaking, the anthropogenic radionuclides are divided into:

- (i) conservative, or
- (ii) non-conservative or so-called particle-reactive radionuclides.

Conservative tracers are defined as being highly soluble in sea water, thus their redistribution upon entering the ocean is determined largely by physical processes related to ocean mixing and diffusion. Common examples of conservative radionuclides include ^{137}Cs , ^{90}Sr , ^{14}C , tritium, ^{129}I . Particle-reactive elements undergo more rapid removal from the ocean due to the affinity of these elements for natural particle surfaces and the general rain of sinking particles in the ocean which removes these particle-bound isotopes to the deep sea and underlying sediment. Examples include $^{239,240}\text{Pu}$, ^{106}Ru and ^{144}Ce .

A cause of confusion in marine radionuclide studies is that, depending upon the local ocean conditions and the time-scales of interest, different radionuclides can be used as both conservative and non-conservative tracers. For example, Cs is commonly used as a conservative tracer. However particle fluxes are considerably higher in coastal and more productive waters than in the open ocean, hence, ^{137}Cs can be more readily scavenged in the coastal ocean and once bound to clays, can be used in sediment as a tracer of particle mixing and sediment accumulation. While Pu is generally thought of as a non-conservative tracer, its distribution on time-scales of years is largely controlled by physical mixing especially in the open ocean. Contrasting chemical behaviour of reduced Pu (more particle-reactive) and oxidized Pu (more soluble), leads to more rapid and complete removal of Pu from anoxic waters such as the deep Black Sea [31], thus in this case local geochemistry leads to conservative vs. non-conservative behaviour. Not only chemical, but also the physical form in which a radionuclide is delivered may determine its fate. For example, it has been shown that local fallout from land-based testing has delivered Pu to the oceans that was more rapidly removed to deep ocean sediment than its stratospheric-based Pu counterpart. It was hypothesized that this was due to the incorporation of Pu into melted silicate sands from the

Nevada Test Site (Figs 9 & 10) [32]. In this case, the measurements of $^{240}\text{Pu}/^{239}\text{Pu}$ isotopic ratios were used to identify the different Pu sources (Table V).

3.3. Variations in activity ratios

Since many anthropogenic radionuclides are introduced into the oceans in constant, or known local ratios, the change in ratio with time or location can often be used to deduce the extent and rates of particle reactivity for a given pair. For example, $^{239,240}\text{Pu}/^{137}\text{Cs}$ ratios in sea water decreased rapidly, due to removal of Pu onto particles and subsequent vertical sinking of these Pu-bearing particles (Fig. 11). Thus, by the time of the GEOSECS programmes in the 1970s, surface water Pu/Cs ratios in the central Pacific of <0.002 had been found, and in vertical profiles of both radionuclides this ratio increased to >0.02 . Pu was found throughout all depths to 6000 m in at least low levels, while ^{137}Cs was not detected in deep water [30].

Another way in which activity ratios might be observed to change is in a two dimensional sense, with enhanced removal of particle-reactive tracers occurring at ocean margins. Since surface mixing times between ocean gyres and margins is in the order of years to decades, horizontal transport can bring two radionuclides from the central basin in fallout proportions to ocean boundaries, but the more particle-reactive species would be enriched in coastal sediment (and depleted in coastal water) due to enhanced scavenging. With time, there would be a continued separation and removal of slightly particle-reactive species in ocean margins, thus leading to an increase in time, for example, in the Pu/Cs ratio in coastal sediment as Pu continues to be scavenged relative to Cs.

In addition to vertical and horizontal fractionation of conservative and non-conservative elements in sea water, there is also the possibility of post-depositional mobility of one or another species, depending upon geochemical conditions. One example, is the continued release of Cs and Pu from the contaminated sediment at the Marshall Islands (Fig. 12). On the other hand, it is known that in highly reducing marine sediments, pore water concentrations of NH_4 increase and under these conditions Cs becomes more mobile [33, 34] and thus more diffusive. This is evidenced by Cs being found deeper in sediment than Pu delivered at the same time, and its loss to overlying waters.

3.4. Variations in inventory

As might be expected from the discussion above, variations in total inventory (per volume of water, sediment, or per area) are often important in order to learn more about the behaviour of radionuclides in the oceans. Most useful in this regard is some knowledge of delivery patterns (in space and time) and total supply per unit area of the ocean (assuming fallout delivery). Soil samples provide particularly useful information for determining average delivery rates, relative source ratios and inventories of many fallout radionuclides in each latitudinal band [35]. Due to higher than expected Pu (and Cs, Sr) inventories in sea water and sediment, it was possible to deduce, for example, that the N Pacific was impacted by local fallout from US weapons testing at the Pacific Proving Grounds [30]. Inventories were also used to examine the strength of boundary scavenging along the coast for $^{239,240}\text{Pu}$ and naturally-occurring excess ^{210}Pb (Fig. 13) [36].

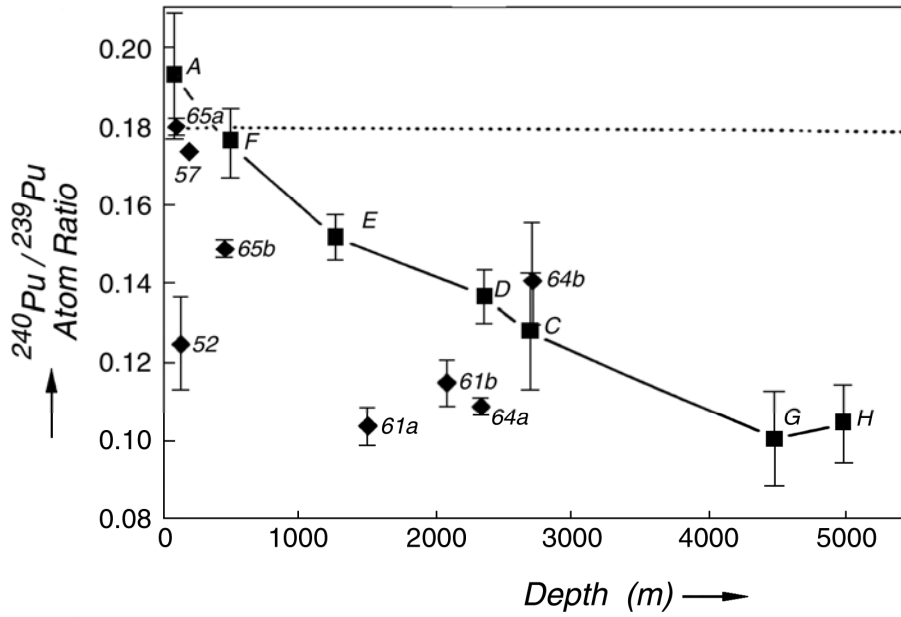


FIG. 9. $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios in sediments vs. depth in the NW Atlantic. Squares were sampled in 1983-1985, while diamonds refer to archived samples (year of sampling indicated [32]).

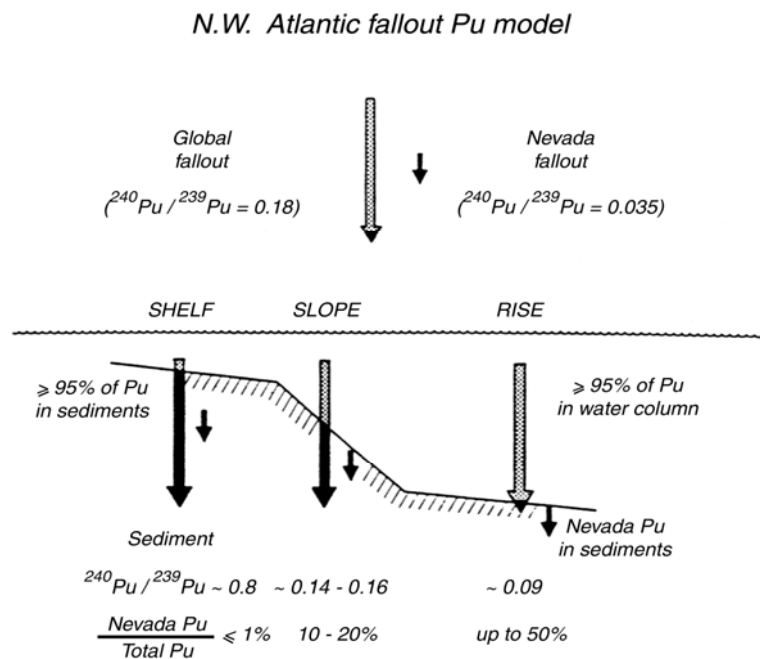


FIG. 10. Fallout model used to identify two sources of fallout to the NW Atlantic, namely Nevada fallout with a low $^{240}\text{Pu}/^{239}\text{Pu}$ ratio, but high efficiency for removal into sediments, and global fallout with a $^{240}\text{Pu}/^{239}\text{Pu}$ ratio of 0.18, and low removal to sediments, except in coastal regions [32].

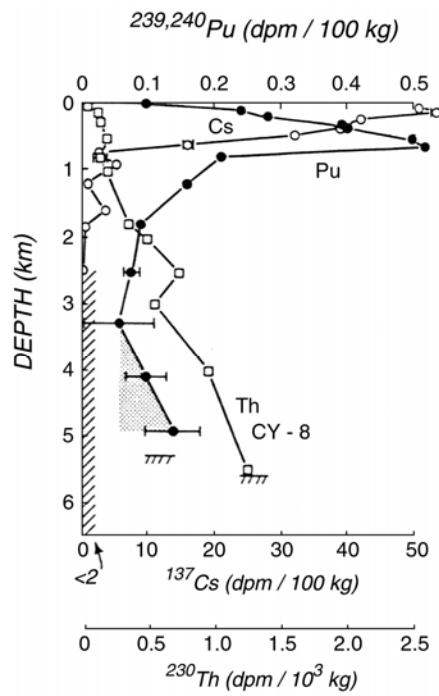


FIG. 11. Vertical profiles of $^{239,240}\text{Pu}$, ^{137}Cs and ^{230}Th from GEOSECS station G225 in the N Pacific. Note low Pu/Cs ratios at the surface due to Pu scavenging, higher Pu/Cs at subsurface Pu maximum, and no detectable Cs below about 2 km, while Pu is found at all depths. This separation with depth is due to the relative difference between the conservative nature of Cs in the oceans, and the particle-reactivity of Pu [26].

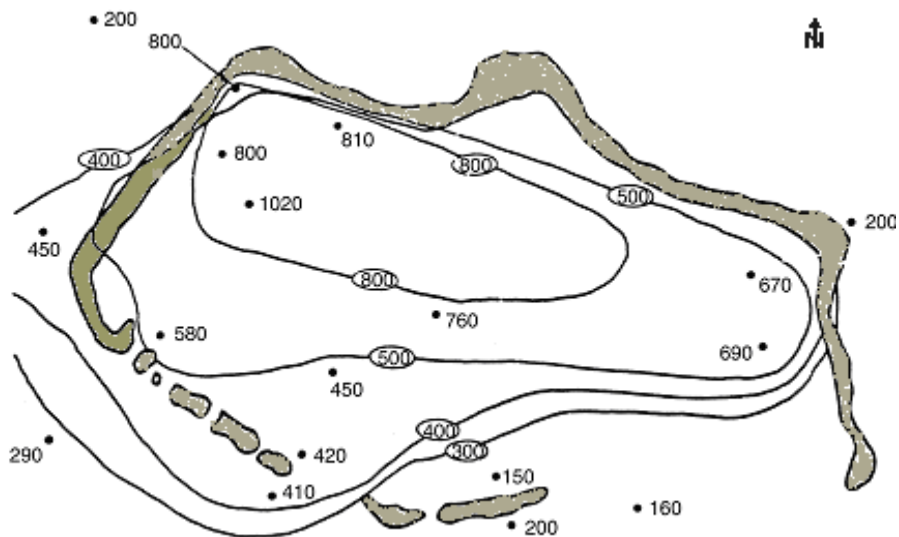


FIG. 12. Surface ocean concentrations of ^{137}Cs (fCi/kg) in Bikini lagoon in 1972. Note highest concentrations associated with the testing site and apparent source of Cs to surrounding waters due to release from coral sediments [37].

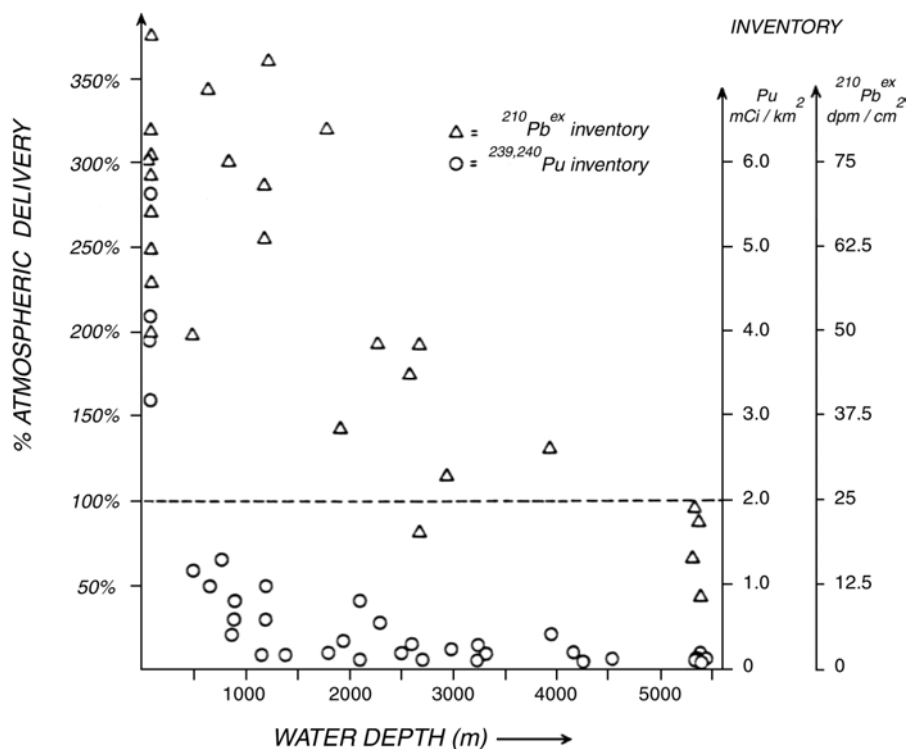


FIG. 13. Sediment inventory vs. water depth for $^{239,240}\text{Pu}$ and $^{210}\text{Pb}^{\text{excess}}$ in the NW Atlantic. The dashed line at 100% represents expected delivery of both isotopes [36].

Variability in local sedimentary inventories also tells us something about sediment redistribution processes, those which could result from storms, or as fine grained sediment are "focused" or otherwise concentrated in specific physical settings (canyons, deep basins) or are lost due to slumping along ocean margins or steep inclines. Often, the integrity of deep ocean box cores can be checked by the presence/absence of fallout Pu or excess natural ^{210}Pb . If there is core top loss during sampling for example, the uppermost layers which include the recent fallout signal, would be missing. In general, particle-reactive radionuclides inventories continue to increase in marine sediment, whereas ocean water concentrations and inventories decrease due to both dilution and scavenging losses.

4. PACIFIC OCEAN

4.1. Overview

The major source of anthropogenic radionuclides in sea water and sediment in the Pacific Ocean and its marginal seas can be traced to global and local fallout from nuclear weapons testing. Other sources have introduced relatively smaller amounts of radionuclides into the Pacific Ocean.

Past measurements of concentrations of ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ in surface water, the water column and sediment from regions of the North and South Pacific Ocean are reported. A comprehensive collection of published references on these radionuclides in the Pacific environment is attached (ANNEX IV). The data for concentrations were extracted from these references as well as from various institutional sources and has been compiled in the GLOMARD database.

The different regions of the Pacific Ocean were chosen on the basis of known ocean current systems (Fig. 14), the location of nuclear test sites, the availability of recent data and, for some, the probability of a relatively uniform distribution of radionuclides.

Radionuclide concentrations measured since 1971 in surface water from 14 regions of the Pacific Ocean and its marginal seas (Fig. 15) are presented in a series of figures. Regression analyses of the results provide best estimates of effective half-lives for radionuclides in surface water. The computed values are used to predict concentrations expected in surface water in the year 2000 which are compared to the most recently measured values in the areas concerned and are shown in the related tables. The remaining parts of this chapter contain tables of data and figures showing recently measured inventories of ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ in the water column and in sediment from regions of the NW Pacific. These results are compared with previous measurements made in the same area to assess changes in inventory over time.

4.2. Radionuclides in the Pacific Ocean

The behaviour of ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ in the Pacific Ocean has been discussed in many papers (see ANNEX IV) and in a more recent review [38]. The most extensive studies were carried out during the 1973–1974 GEOSECS programme that provided the first comprehensive dataset on the lateral and vertical distributions of ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ in Pacific waters [30]. Traditionally, several Japanese institutes, as well as many other research groups, have been active in investigating these radionuclides, especially in NW Pacific water and sediment, e.g. [39–51]. A third group of investigations was concerned with nuclear weapons testing sites in the Marshall Islands, e.g. [52–54] and French Polynesia, e.g. [55–58].

The behaviour of plutonium in the Pacific Ocean will be discussed in more detail as this element has received the greatest attention. The state of knowledge of the behaviour of Pu in the Pacific Ocean has evolved slowly due to the intrinsic complexity of the many factors involved and to the economic and logistical constraints on its study in the world's largest ocean. Three of the main factors relevant to Pu behaviour are:

- (i) The physical and chemical form of plutonium as it enters the ocean — frequently source-dependent,
- (ii) The basic physico-chemical properties of plutonium e.g. oxidation state, speciation and the ability to form complexes, and the resultant phase partition between particles and solution,
- (iii) The physical, chemical and biological processes that act on plutonium in the Pacific Ocean in a heterogeneous manner — both geographically and over seasonal, annual and multi-annual time-scales.

The major input of plutonium to the Pacific Ocean both quantitatively and geographically, resulted from atmospheric testing of nuclear weapons. A significant but smaller input of plutonium, derives from the global fallout of a ^{238}Pu source in a navigational satellite (SNAP-9A) that burned up over the Indian Ocean in 1964 [35]. Other inputs, e.g. from nuclear waste disposal, isolated underwater nuclear tests or land-based releases, are relatively minor in comparison and tend to have rather localized effects. The primary source

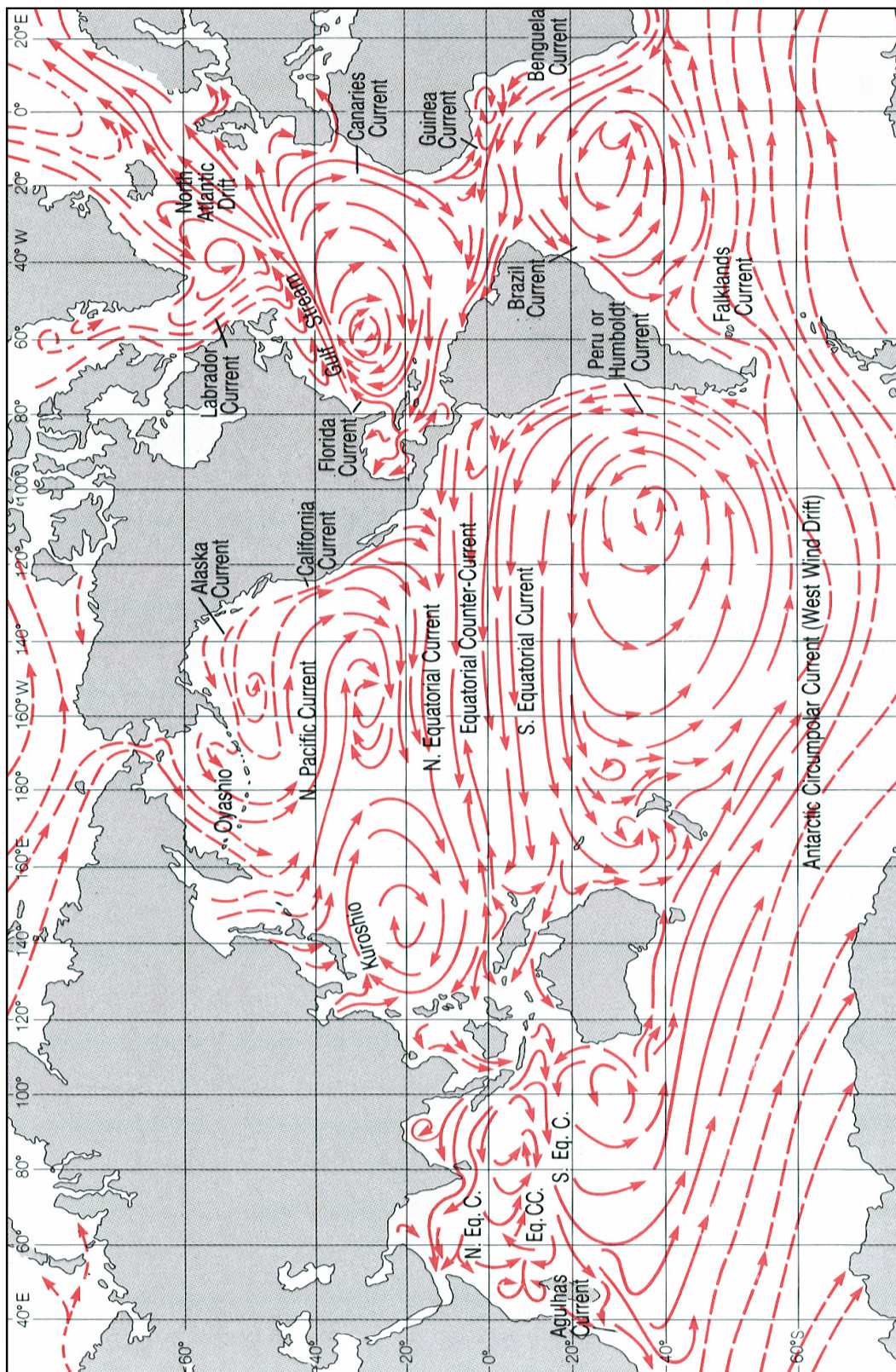


FIG. 14. The global surface current system (adapted from *The Open University* [59]). Cool currents –dashed arrows; warm currents –continuous arrows. The map shows average winter (Northern hemisphere) conditions.

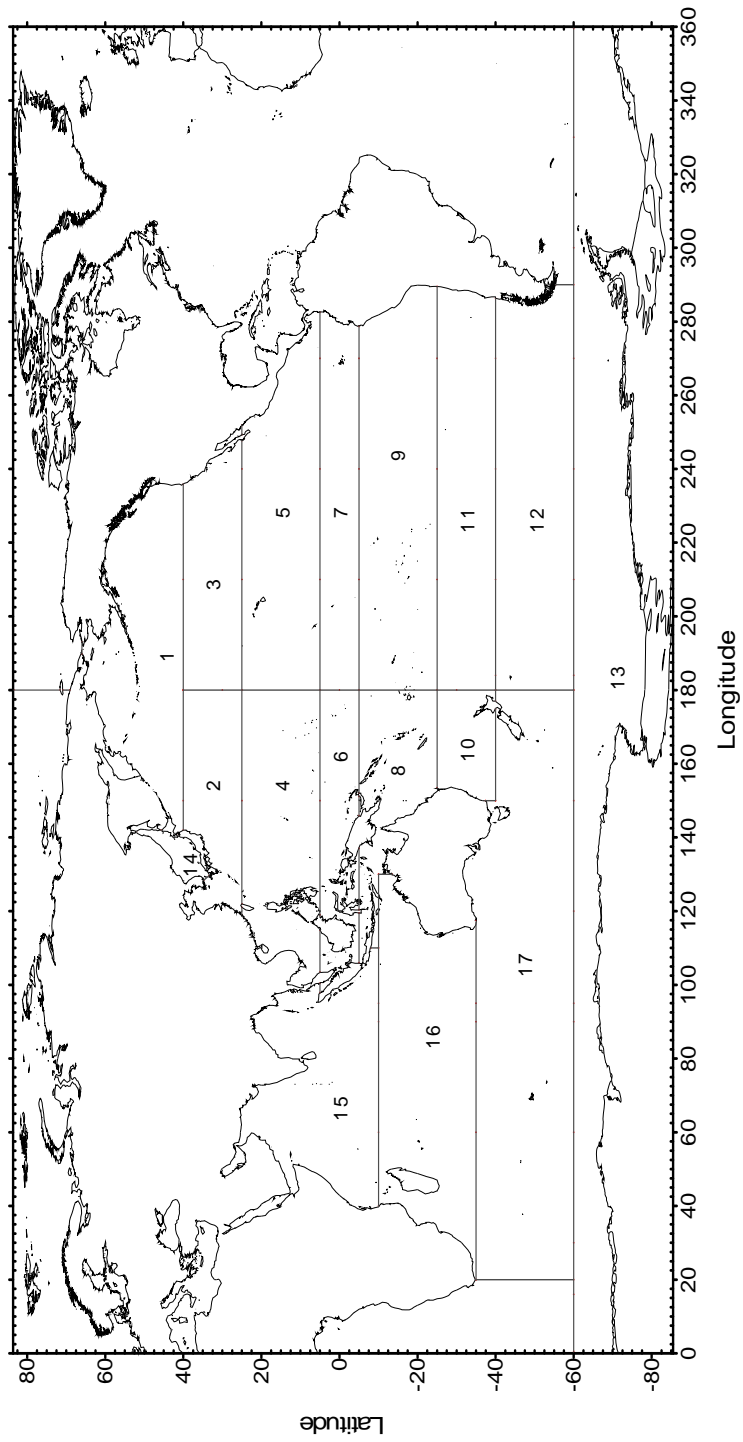


FIG. 15. Latitudinal boxes in the Pacific and Indian Oceans.

from atmospheric nuclear weapons testing is fallout distributed globally in the stratosphere and subsequently delivered to land and ocean surfaces through the troposphere. This fallout pattern has a strong latitudinal dependence with maxima at mid-latitudes and minima towards the equator and poles [25, 35]. In addition, since the majority of atmospheric nuclear weapons tests were conducted in the Northern hemisphere, fallout delivery to the Earth's surface has been substantially larger than in the Southern hemisphere.

The second and very substantial input of plutonium to the Pacific Ocean was derived from the extensive series of atmospheric nuclear weapons tests carried out by the US weapons testing programme at the Pacific Proving Grounds at Bikini (11°36'N, 165°22'E) and Enewetak (11°30'N, 162°20'E) Atolls between 1946 and 1958 [54]. A total of 66 tests were conducted at these atolls. Many of these tests were conducted at or near ground level and thus much of the resultant radioactivity was delivered to the troposphere with substantial fallout to regional ocean areas. Surface tests conducted over coral islands or over shallow water produced highly localized input [60] because large quantities of soil were incorporated in the ensuing fireball and cloud. In these cases different types and sizes of high specific activity particles were formed consisting of mixtures of partially hydrated CaO, Ca(OH)₂ and CaCO₃ formed by high temperature vaporisation and subsequent condensation processes [61]. Therefore, in addition to the injection of fallout debris into the stratosphere, these surface-based tests have produced significant quantities of local, or so-called tropospheric fallout. Such fallout remains within the troposphere, from which it is rapidly deposited. Depending upon local wind conditions and wet and dry deposition patterns, tropospheric fallout was deposited preferentially within the vicinity of the test site [62]. In the years that followed testing, contaminated particles deposited in lagoon and surrounding slope and basin sediment formed a reservoir and source term for the marine environment through solubilization and water transport. The mean exchange rate of Enewetak lagoon water is approximately one month and estimates for Bikini lagoon water vary between 30 and 140 days [63]. Other Pacific tests were conducted by the US and the UK at Christmas Island (1°52'N, 157°20'W) and by the USA at Johnston Atoll (17°00'N, 163°30'W). However, these were conducted at high altitude and are unlikely to have contributed significantly to local tropospheric fallout in comparison to the Enewetak and Bikini tests. France also conducted an extensive series of atmospheric tests at Mururoa and Fangataufa Atolls (around 22°S, 135°W) [57]. Most of the 46 tests and trials were conducted between 1966 and 1974 at altitudes of 200 to 700 m. In consequence of the much lower yields than of the US tests at Bikini and Enewetak, and the altitude of detonation, it is anticipated that the delivery of local fallout from the French tests to the surrounding ocean areas must have been of much smaller significance.

Inventories of ^{239,240}Pu in excess of global fallout levels persisting in Pacific atoll sediment are continuously mobilised into solution and provide a source term to the surrounding ocean. Analyses of grab and core samples collected from Enewetak Atoll in 1972 were used to give an estimate of the total sediment inventory for plutonium of 44.4 TBq [52]. The estimated plutonium inventory for Bikini sediment was 54.4 TBq. Using residence times of lagoon water and the average soluble inventories, it was estimated that the annual discharge of ^{239,240}Pu from Enewetak and Bikini Atolls is 0.10 and 0.12 TBq, respectively. Thus, the cumulative export to the equatorial Pacific Ocean over the past 40 years has been about 8 TBq. Typical present-day ¹³⁷Cs/^{239,240}Pu inventory ratios in the contaminated Marshall Islands range between 1.8 and 19. This implies that a maximum of about 2 PBq of the present Pacific Ocean inventory was derived from ¹³⁷Cs initially deposited on or around the lagoons. This represents less than 2% of the total ¹³⁷Cs deposited in the N Pacific Ocean from global fallout. By comparison, the total ^{239,240}Pu inventory in Mururoa and Fangataufa lagoon sediment was estimated to be about 25 TBq [58]. For comparison, the ¹³⁷Cs inventories in

sediment in both lagoons are only about 0.9 TBq [57]. Using the radionuclide residence time of 100 days for Mururoa and 30 days for Fangataufa, the annual release rate for $^{239,240}\text{Pu}$ from Mururoa and Fangataufa lagoon sediment to the open ocean was estimated to be about 0.01 TBq [57]. Hence, the total cumulative export to the ocean from this source over the past 30 years would be about 0.3 TBq.

A considerable amount of work has been carried out over the last 40 years with respect to the fate and behaviour of Pu in the Pacific Ocean. For example, [26] included an extensive section on many aspects of Pu in the Pacific in a major review of the aquatic behaviour of Pu. Robison and Noshkin [54] have produced an informative review of environmental impact assessment studies around the US test sites at Bikini and Enewetak, and the IAEA [57] addressed similar issues around the French test sites at Mururoa and Fangataufa.

The basic property controlling the oceanic behaviour of Pu is its tendency to associate with particulate phases. This however varies considerably, depending on speciation (e.g. oxidation state), physico-chemical form and density of particles. The higher oxidation states (V, VI) would have substantially less particle association than the lower (III, IV), as discussed in [26]. This was specifically discussed with respect to open Pacific waters in a paper by Nelson et al. [64]. Buesseler [65] discussed the influence of physico-chemical forms on Pu oceanic behaviour — noting especially, in the Pacific, the difference between calcium-rich particles produced following surface tests in the Pacific and the smaller (sub-micron) iron-rich particles introduced to the stratosphere from atmospheric nuclear weapons tests. Finally, particle density is a major influence on the oceanic behaviour of Pu. Estuarine, coastal and slope sediment have been shown in many studies to receive large amounts of scavenged Pu from the large population of biogenic and inorganic particles present in shallower, near-shore locations. In contrast, open ocean oligotrophic settings (covering most of the global ocean) have been shown to exhibit patterns of Pu distributions wherein the largest part of the delivered Pu is still resident in the water column (e.g. [30, 66]).

In the Pacific, these generally complex patterns are further complicated by substantial amounts of Pu delivered there, in addition to global fallout, from tropospheric local fallout from the Bikini and Enewetak test series (such as the "Mike" test in 1952 at Enewetak). Buesseler [65] has noted that the Pu signal from these tests can be distinguished on the basis of the much higher proportion of ^{240}Pu relative to ^{239}Pu (see also Chapter 3). Observed oceanic $^{240}\text{Pu}/^{239}\text{Pu}$ ratios of up to 0.36 are contrasted with the value of 0.18 that generally has represented the global fallout average. Pu from this local fallout, with its characteristic isotopic signature, has physico-chemical characteristics that lead to its much faster removal from the ocean surface and transfer to the deep ocean compared to Pu from global fallout. It is also true that, at the locations where these high ratios were observed, the Pu inventories in the water column, or total of water plus sediment, are substantially in excess of global fallout values. The distribution of this excess, both geographically and vertically in the N Pacific is highly consistent with its originating from a local fallout source because of enhanced biological removal and the high $^{240}\text{Pu}/^{239}\text{Pu}$ signature. The geographical distribution agrees with that expected from regional input in the vicinity of the test sites, subsequently redistributed by the Western N Pacific gyral circulation and the Equatorial Current and Counter-Current systems (Fig. 14). The evidence from vertical distributions can be drawn from two sets of observations. Firstly, one can look at the distribution of stations that show elevated Pu concentrations in near-bottom water. Again, the GEOSECS data [30] show deep-water Pu maxima in the NW Pacific to be well correlated with inventories exceeding global fallout levels. This has also been seen in other data sets — always subject to data limitations, such as undersampled deep water or analytical discrepancies. Secondly, it is useful to examine the geographical distribution of locations where data permit an assessment of the proportion

of Pu inventories between the water column and underlying sediment — or even between upper and deep ocean water inventories. A higher proportion of the total Pu inventory was found at stations of high sediment Pu inventories in the western N Pacific (up to 16%) compared to the station north of Hawaii (2%). Taken together, these observations seem to point to the existence, in the Pacific, of a second source of Pu (in addition to global fallout), derived from the ground level nuclear tests in the 1950s, which has been shown to be associated with particle scavenging and transport processes at rates substantially greater than the rates typically seen for global fallout Pu in open ocean regions.

4.3. Distribution of radionuclides in surface water and sediment

For the study of the distribution of radionuclides in sea water and sediment, the Pacific Ocean has been divided into different latitudinal boxes (Fig. 15) on the basis of known ocean current systems, the location of nuclear test sites, the availability of recent data and, the probability of a relatively uniform distribution of some radionuclides. Twelve boxes have been delineated in the Pacific Ocean and one box has been attributed to the Southern Ocean. The Sea of Japan has been given a separate box because of its oceanographic specificity and importance in marine radioactivity studies. Box 1 (north of 40°N) is the subarctic Pacific Ocean, where the largest fallout rates of ^{137}Cs and ^{90}Sr over the Pacific Ocean were observed in the 1960s [2]. Boxes 2 and 3 (25°–40°N) are upstream and downstream of the Kuroshio Extension, respectively, where the water masses of the Kuroshio and Oyashio Currents mix. Boxes 4 and 5 (5°–25°N) are downstream and upstream of the North Equatorial Current, which is a typical oligotrophic ocean current corresponding to the subtropical gyre (Box 5 also includes the California Current). These boxes include the Pacific Proving Grounds nuclear weapons tests sites (Bikini, Enewetak, Johnston Atolls and others), especially important for plutonium [65, 67]. The subtropical region of the western N Pacific was affected by local fallout as opposed to the subtropical region of the eastern N Pacific. Boxes 6 and 7 (5°S–5°N) are downstream and upstream of the South Equatorial Current, respectively. The boundary between boxes 4, 5 and 6, 7 is approximately at the Equatorial Counter-Current. Box 7 includes the Equatorial upwelling region. Boxes 8 and 9 (5°–25°S) are downstream and upstream of the South Equatorial Current, respectively. The boundary between boxes 6, 7 and 8, 9 approximately corresponds to the South Equatorial Counter-Current. Box 9 includes the French nuclear weapons test sites (Mururoa and Fangataufa). Box 10 (25°–40°S) corresponds to the Tasman Sea. Box 11 (25°–40°S) is the mid-latitude region of the South Pacific. Boxes 12 (40°–60°S) and 13 (below 60°S) include the Southern Ocean and the Antarctic Circumpolar Current.

4.3.1 Surface water

Figures 16 to 19 show ^3H , ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ surface water data from the GLOMARD database for the period 1991–1995, for which average radionuclide concentrations were calculated for the corresponding latitudinal boxes in the Pacific Ocean. Although the density of data is not very high, a clear latitudinal effect of increasing average radionuclide concentrations from the south to the north can be observed, as expected. The ^3H data have a very good coverage in the central and South Pacific Ocean thanks to the WOCE programme [68]. The average concentrations increase from 13 mBq/L estimated for Box 13, up to 330 mBq/L estimated for the Sea of Japan. ^{90}Sr data have the highest density in the NW Pacific Ocean (similar to ^{137}Cs and $^{239,240}\text{Pu}$) increasing from 0.4 mBq/L from the south to 1.8 mBq/L for the Sea of Japan. ^{137}Cs and $^{239,240}\text{Pu}$ data show an increase from 0.6 mBq/L and 0.8 $\mu\text{Bq/L}$, respectively, in the south to 2.8 mBq/L and 8.5 $\mu\text{Bq/L}$ in the Sea of Japan, respectively.

Except for the fact that the highest average radionuclide concentrations were observed in the Sea of Japan, the data show lower average concentrations than expected in Box 3 for $^{239,240}\text{Pu}$ (however, the data set is very small). Similarly, in the north, Box 1 shows lower average concentrations for ^{137}Cs than expected (however, the geographical distribution of data in this box is heavily concentrated in the NW).

Similar data sets are shown in Figs 20 to 23 for the years 1996–2000. Here, the data are very sparse and they are mainly based on the results obtained in the framework of the WOMARS project. A comparison (where possible) of decay-corrected data shows more or less the same trend in data observed for the years 1991–1995.

Although the data density is very low, it was possible, at least for the NW Pacific Ocean, to develop a distribution chart for ^{137}Cs in surface water using data extracted from the GLOMARD database for the years 1991–2000 (decay-corrected to 1.1.2000 (Fig. 24)).

For a more detailed discussion on radionuclide data in the boxes see Chapter 4.3.

4.3.2. Sediment

A similar set of data was developed for the distribution of ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ in sediment (Figs 25 to 27 for 1991–1995 and Figs 28 to 29 for 1996–2000 (no data are available for $^{239,240}\text{Pu}$)). The data density is even less than for surface water. Only few data are available for the NW Pacific Ocean and the Sea of Japan, mainly obtained in the framework of the WOMARS project. Therefore, a detailed evaluation of sediment data is not possible.

Pacific sediment is, of course, the ultimate sink for the Pu present there. The sheer size and great depth of the Pacific have limited the number of Pacific sediment studies. However, shelf and slope studies have been undertaken on both the eastern and western Pacific margins due to their proximity to coastal laboratories. On the eastern side, Sholkovitz [26] has presented a comprehensive overview of previous work on California basin sediment. Beasley et al. [69] reported on transuranic measurements in shelf sediment off Washington and Oregon. Some data were reported for the western side [70], the East China and Yellow Seas [43] and the Seto Inland Sea [71]. All these studies point to the same major conclusion. As indicated by the general excess inventories over those anticipated from global fallout, Pacific marginal sediment underlying waters with high biological productivity and larger suspended sediment loads are major sinks in Pu removal and sequestration. The additional component from transported local fallout Pu has also to be considered in respect of western marginal sediment — as discussed in Chapter 4.6.2.

The number of open Pacific Ocean sediment Pu studies is smaller than needed to permit a comprehensive estimate of the distribution and inventories residing there — especially for specific points in time. For example, it is unfortunate that the comprehensive GEOSECS sampling programme did not collect sediment cores at the wide distribution of stations occupied. There is another factor that complicates the acquisition of reliable estimates of Pu distributions and inventories, the heterogeneity of the observed Pu distributions to be found — especially in deep-ocean sediment. This is presumably a consequence of the relatively short time, in a geological sense, over which Pu has been arriving at the sea-floor. Presumably, on the scale of hundreds and thousands of years, these distributions will tend to even out — beginning to approach the more homogeneous distributions at a steady state that can be observed for long lived Th isotopes, for example [72]. There, in a series of 17 sediment cores collected in the late 1970s in a single area in the central N Pacific Ocean, north of

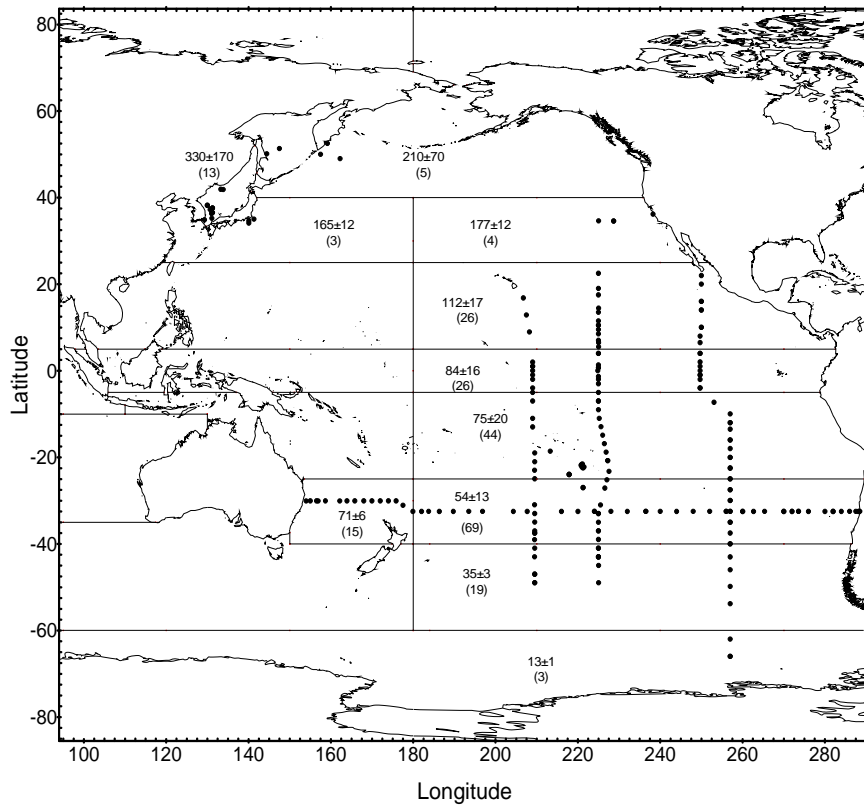


FIG. 16. ^3H in surface water (1991–1995), decay-corrected to 01.01.2000, in mBq/L.

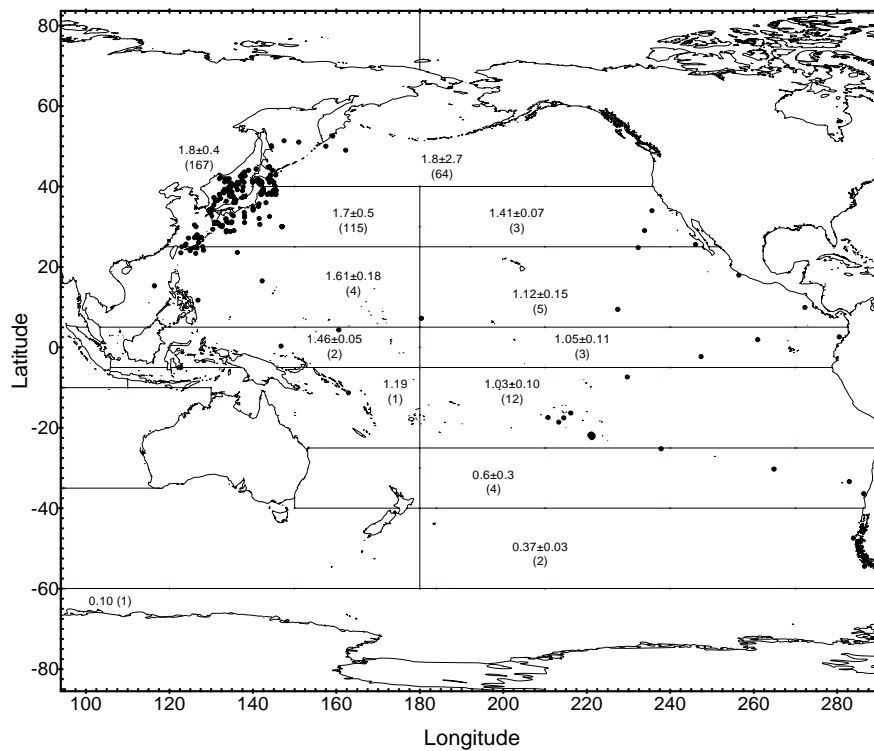


FIG. 17. ^{90}Sr in surface water (1991–1995), decay-corrected to 01.01.2000, in mBq/L.

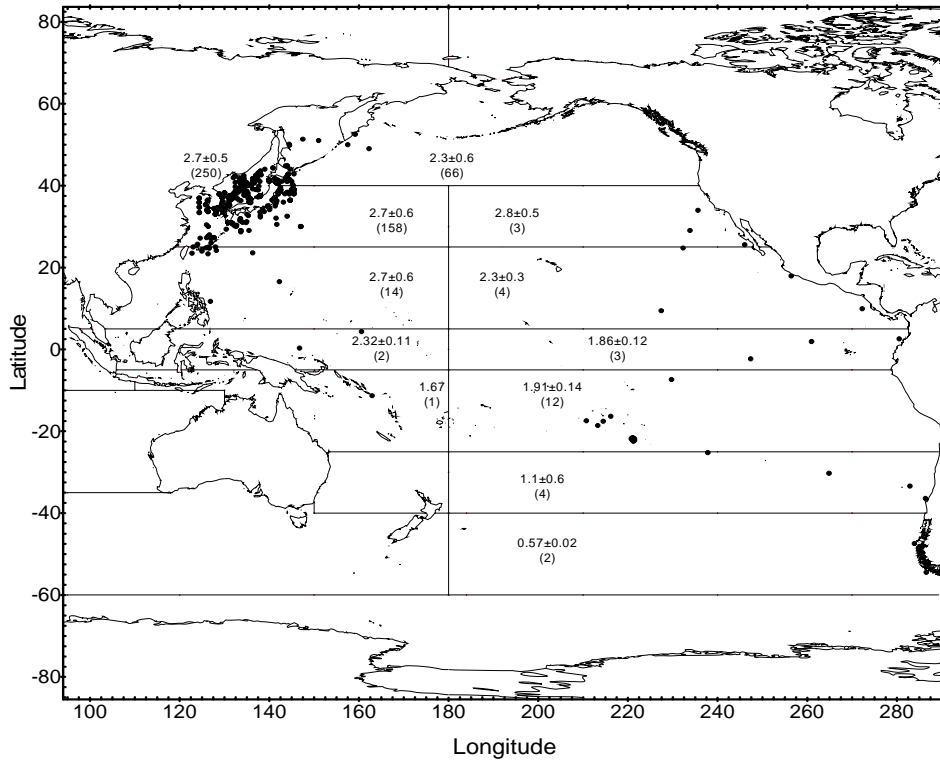


FIG. 18. ^{137}Cs in surface water (1991-1995), decay-corrected to 01.01.2000, in mBq/L.

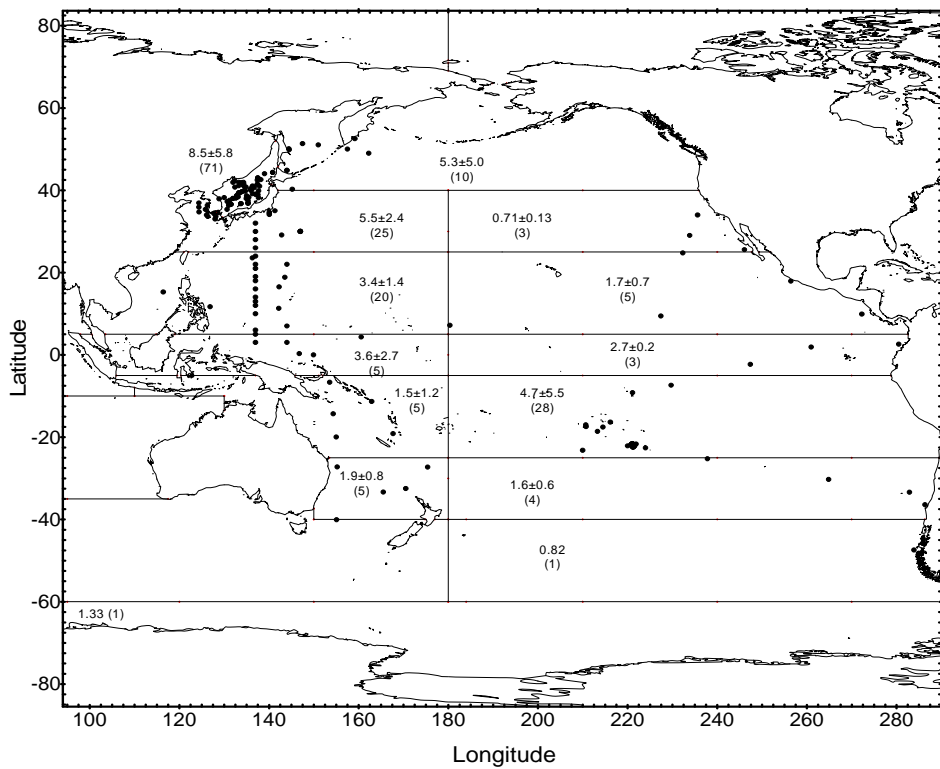


FIG. 19. $^{239,240}\text{Pu}$ in surface water (1991-1995) in $\mu\text{Bq/L}$.

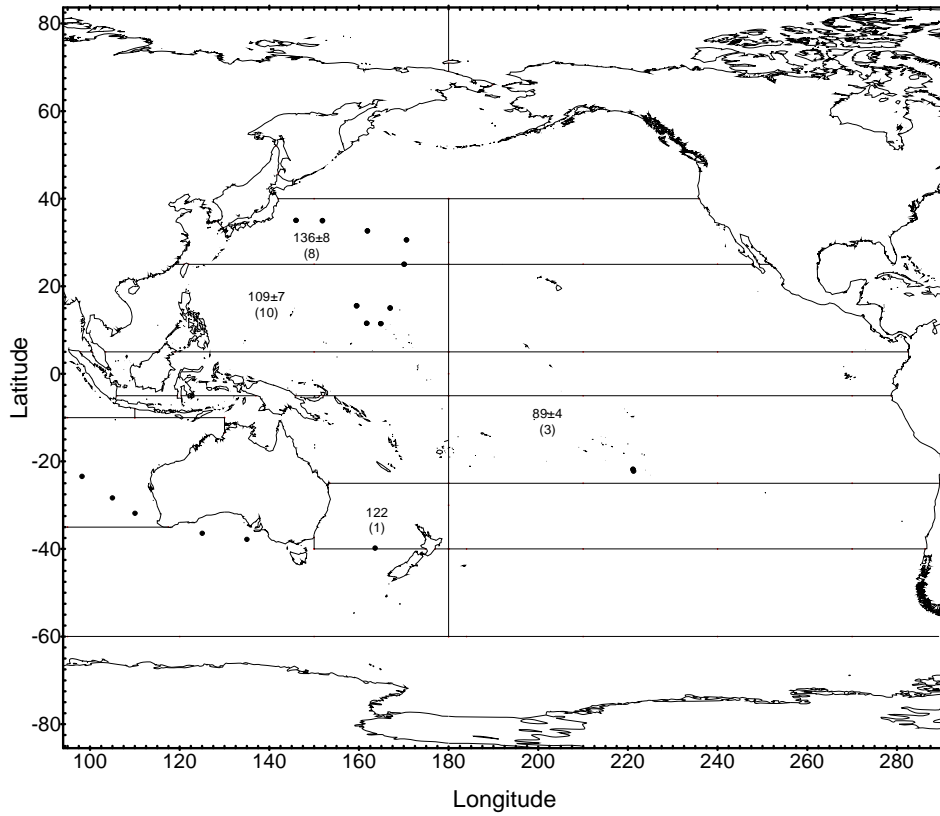


FIG. 20. ^3H in surface water (1996–2000), decay-corrected to 01.01.2000, in mBq/L.

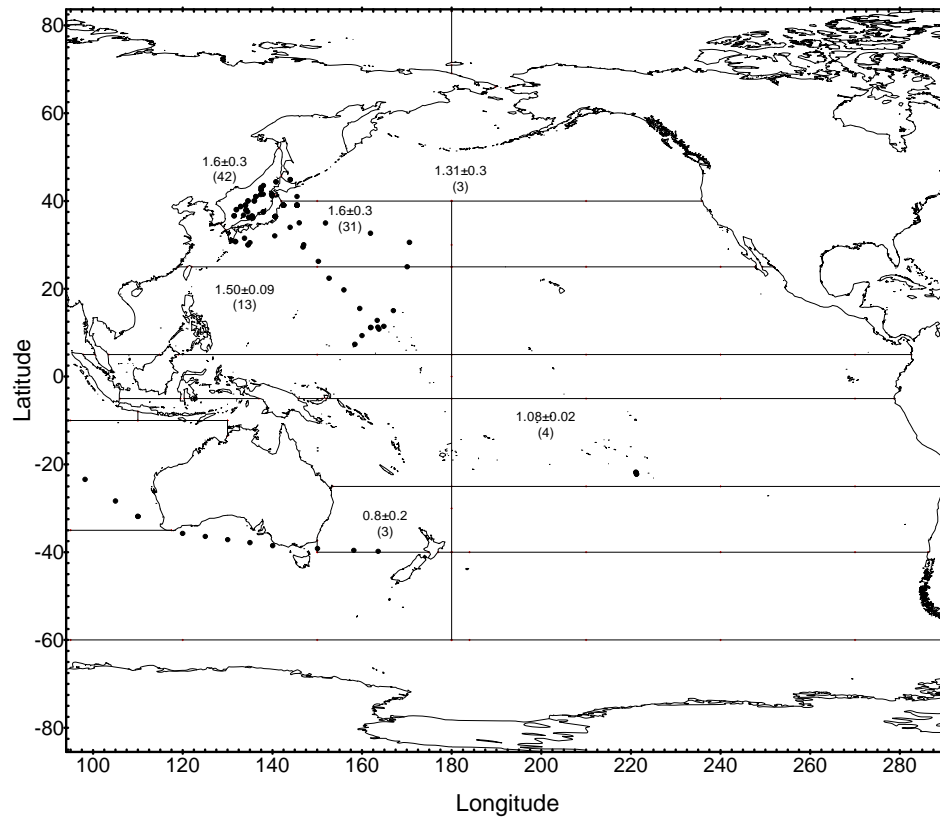


FIG. 21. ^{90}Sr in surface water (1996–2000), decay-corrected to 01.01.2000, in mBq/L.

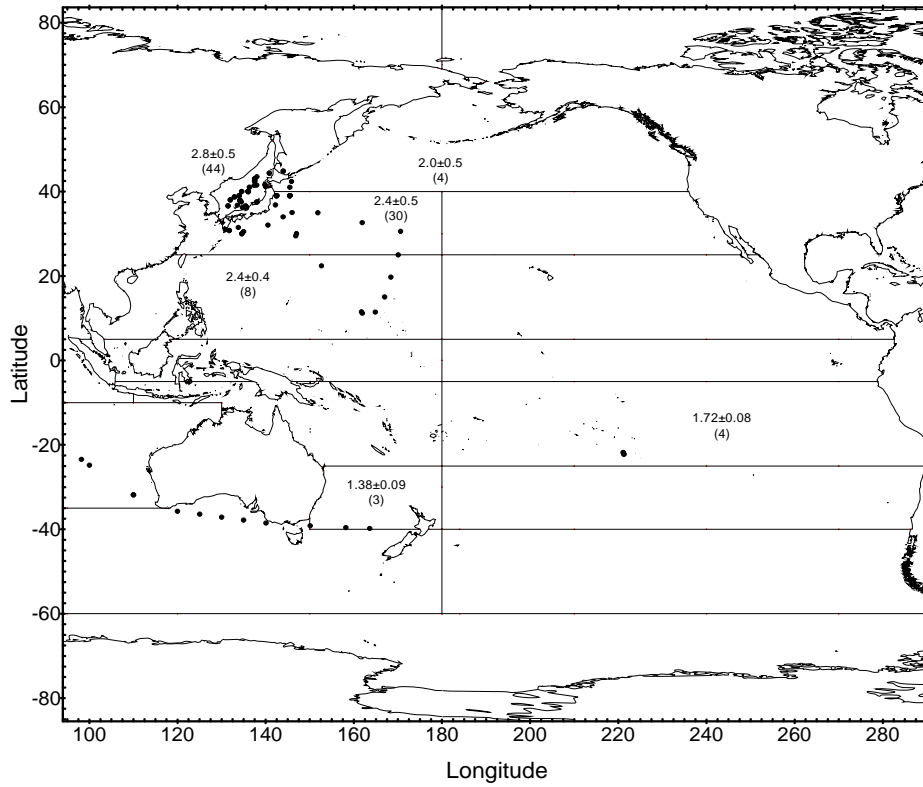


FIG. 22. ^{137}Cs in surface water (1996–2000), decay-corrected to 01.01.2000, in mBq/L.

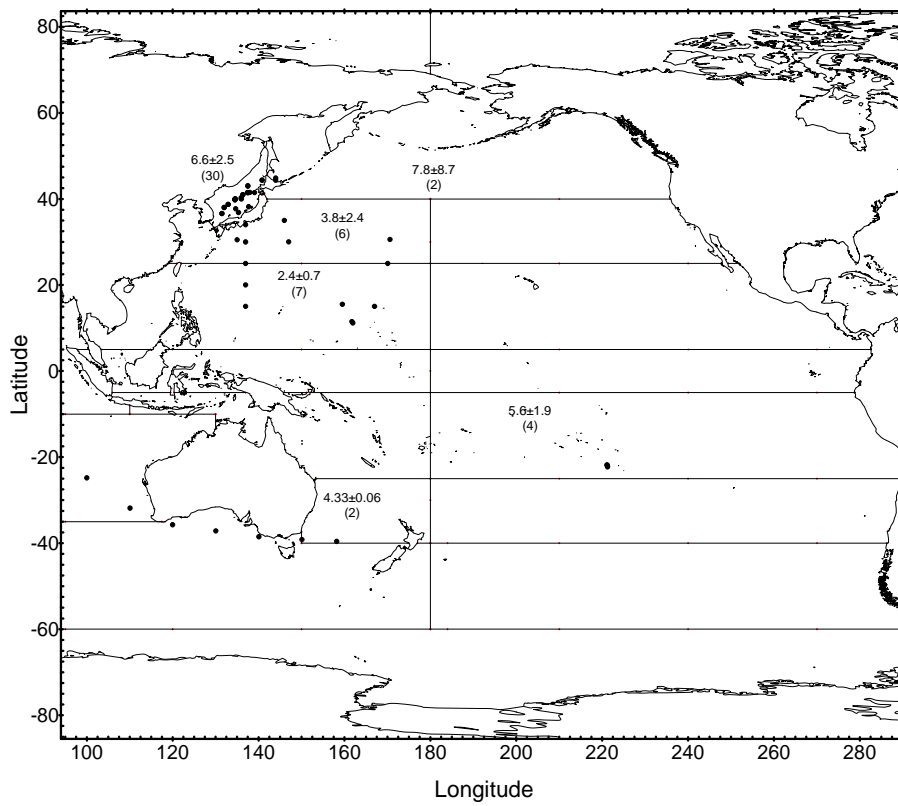


FIG. 23. $^{239,240}\text{Pu}$ in surface water (1996–2000), in $\mu\text{Bq/L}$.

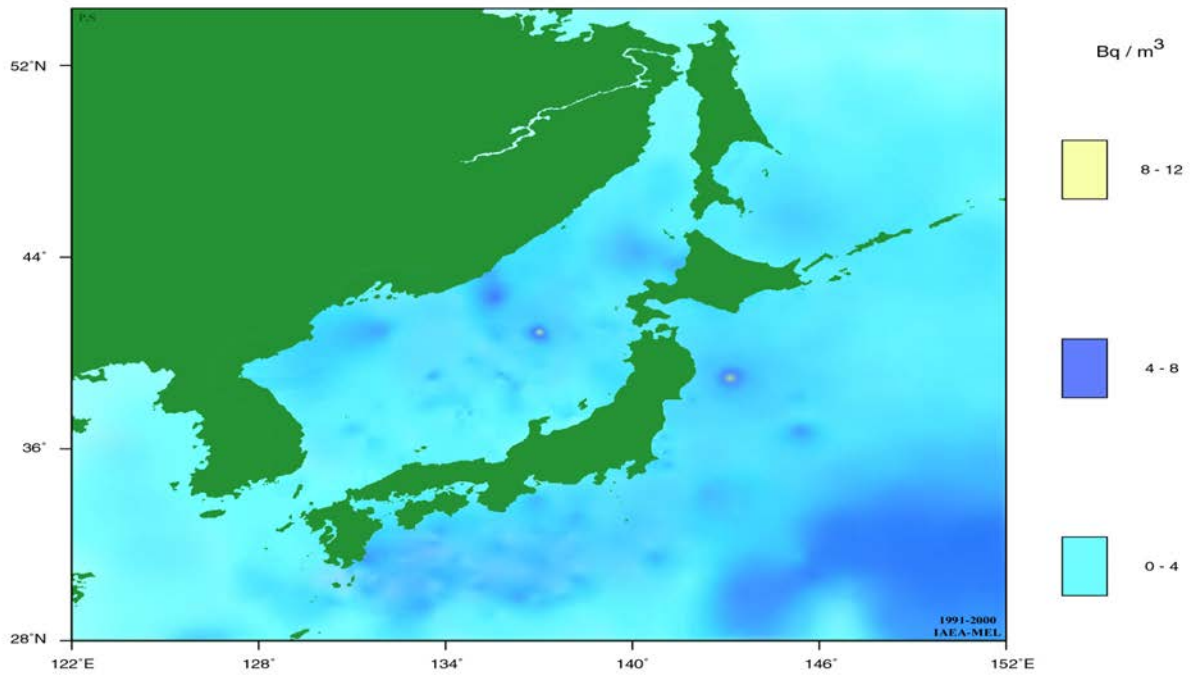


FIG. 24. ¹³⁷Cs in surface water of the NW Pacific Ocean (1991–2000).

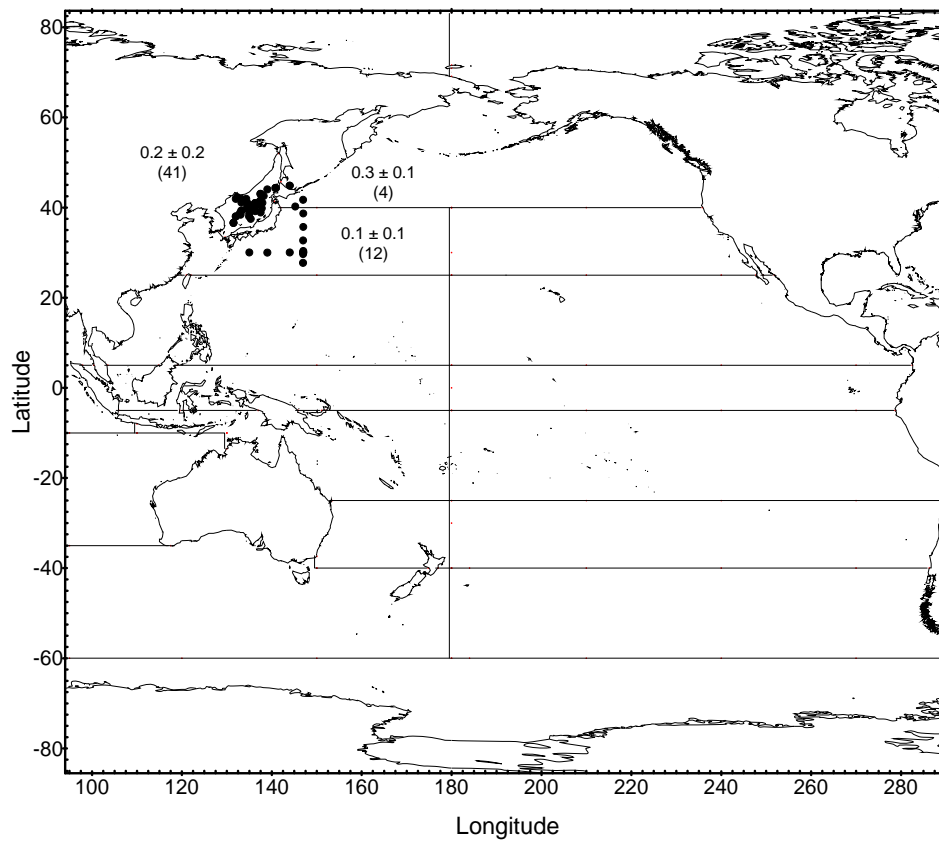


FIG. 25. ⁹⁰Sr in NW Pacific surface sediment (1991–1995), decay-corrected to 01.01.2000, Bq/kg dw.

Hawaii, Bowen reported variations of around a factor of 10 in the observed Pu distributions and inventories. In a similar manner, the depths of bioturbation varied considerably — from 5 to 20 centimetres. Although sampling artefacts are always a concern, he reported that this was not a major contributor to the observed heterogeneity as four different sampling devices were used and no clear correlation with device was found. The hypothesis advanced to explain this heterogeneity was based on the linkage, over short time intervals, with the variability in the places of more intense bioturbation — given the relatively small population density of benthic organisms supportable by low fluxes of food via sinking particles.

Even with this limitation, the actual number of places where deep ocean Pacific Pu measurements in sediment have been obtained is quite limited with respect to spatial coverage — and even more so in respect of time. The main data sets built into the Global Marine Radioactivity Database (GLOMARD) at IAEA-MEL [73] were supplied by these Japanese institutions: the Japan Chemical Analysis Center (JCAC), the Marine Safety Agency (MSA) and the Fisheries Agency (FA). IAEA-MEL data from the joint Japanese-Korean-Russian expeditions in 1994 [74] and 1995 [75] were used in the evaluations as well [76, 77]. Further data were obtained from a literature survey covering the western and central N Pacific [41, 42, 44–48, 78–82] as well as the eastern Equatorial Pacific [83]. A significant trend, discussed below, is the frequently observed excesses of Pu, in either absolute or relative (to ^{137}Cs) terms found especially in western regions close to downstream locations around the test sites at Bikini and Enewetak and towards the equator in the eastern Pacific. This will be discussed with reference to the important Pu source from local fallout from these atoll weapons tests.

4.4. Surface water radionuclide time series

This section compares ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ concentrations measured since 1971 in surface water of 14 regions of the Pacific Ocean and its marginal seas in a series of figures. Regression analyses of the results provide best estimates of effective decay constants and effective half-lives for radionuclides in surface water. The computed values are used to predict concentrations in surface water for the year 2000 and are compared to the most recently measured values in the areas concerned. Levels of anthropogenic radionuclides (^{137}Cs , ^{90}Sr and $^{239,240}\text{Pu}$) in surface water of the Pacific Ocean were evaluated using the data stored in the GLOMARD database.

The time series of anthropogenic radionuclides (^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$) during 1971–1998 are examined to estimate their surface concentrations in the year 2000, as anthropogenic radionuclides in surface water in the 1950s and 1960s were mainly controlled by input processes such as global and local fallout [2]. The concentrations of ^{90}Sr with time in surface water for each box decrease exponentially (Figs 30 to 33). Therefore, temporal changes in surface radionuclide concentrations can be expressed by an exponential function over three decades in time. The surface concentrations and effective half-lives of ^{137}Cs , ^{90}Sr and $^{239,240}\text{Pu}$ in each box of the Pacific are calculated from the corresponding regression lines. However, the confidence levels of regression lines vary greatly between boxes because of the different densities of data.

The ^{90}Sr concentrations in surface water in 2000 calculated from the time series or estimated from recent measurements are in the range of 0.4 to 1.5 mBq/L (Table VI). The surface concentration of ^{90}Sr in Boxes 8, 10, 11 and 12 cannot be estimated because of the lack of data (less than 3 values). Most of the surface ^{90}Sr concentration data are crowded in Box 2 (76%). Higher surface ^{90}Sr concentration occurs in the mid-latitude region of the western N Pacific (Box 2). The lowest values appear in regions of high latitude in the South Pacific. The geographical distribution of ^{90}Sr in Pacific surface water in 2000 is homogeneous, as is that of ^{137}Cs .

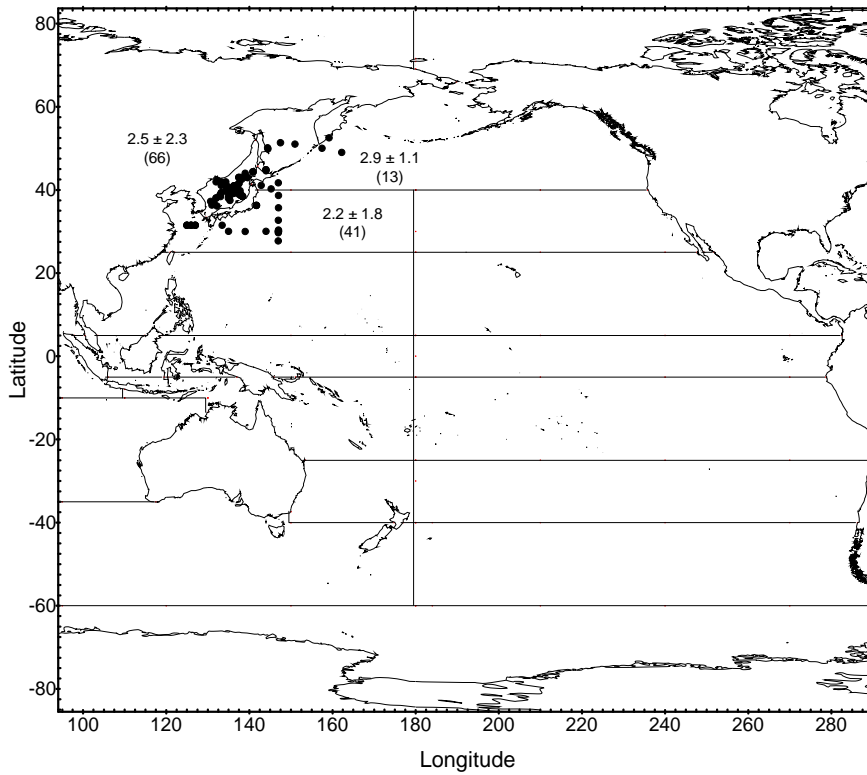


FIG. 26. ^{137}Cs in NW Pacific surface sediment 1991–1995, decay-corrected to 01.01.2000, Bq/kg dw.

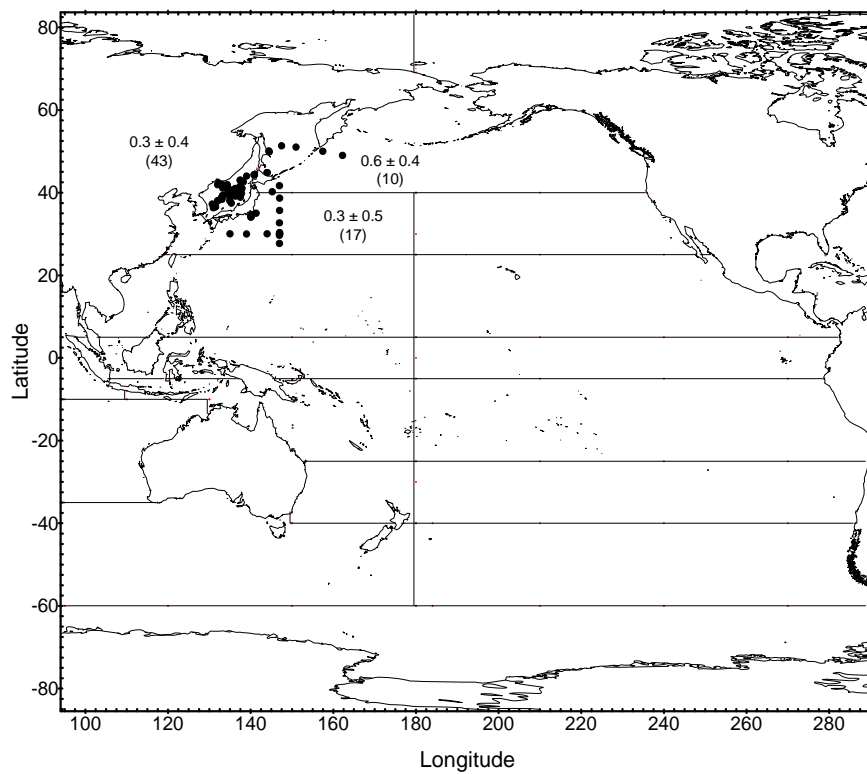


FIG. 27. $^{239,240}\text{Pu}$ in NW Pacific surface sediment (1991–1995), decay-corrected to 01.01.2000, Bq/kg dw.

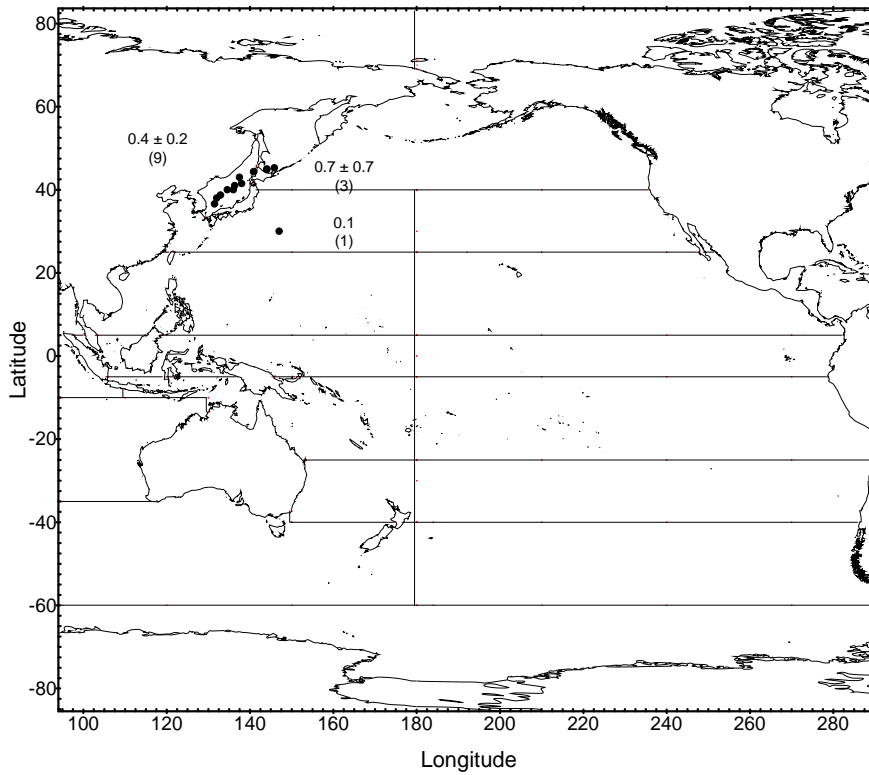


FIG. 28. ^{90}Sr in NW Pacific surface sediment (1996-2000), decay-corrected to 01.01.2000, Bq/kg dw.

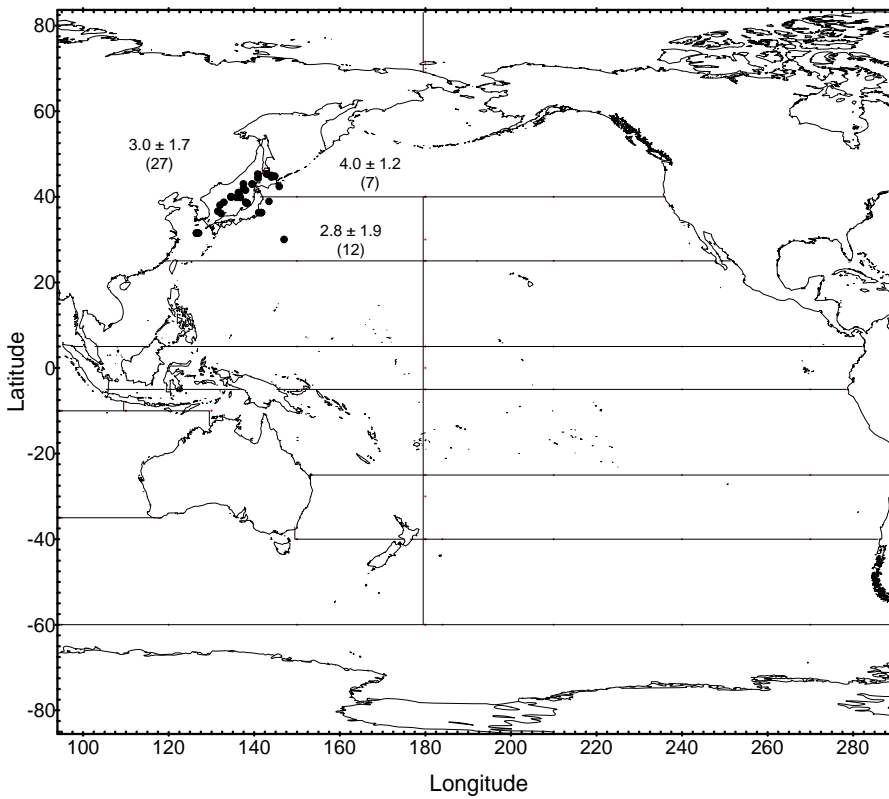


FIG. 29. ^{137}Cs in NW Pacific surface sediment (1996-2000), decay-corrected to 01.01.2000, Bq/kg dw.

The calculated ^{137}Cs concentrations in surface water for each box in 2000, together with the most recent data are shown in Table VII and Figs 34 to 37. The surface concentration of ^{137}Cs in Boxes 8 and 12 cannot be estimated because of the lack of data (less than 3 values). The ^{137}Cs concentrations ranged from 0.7 to about 2.8 mBq/L. Higher surface ^{137}Cs concentration occurs in the eastern Equatorial Pacific (Box 7). Lower values appear in the Tasman Sea (Box 10). The geographical distribution of surface ^{137}Cs in 2000 differed from that observed in the 1960s. The geographical distribution of surface ^{137}Cs in the early 1970s showed maxima in the mid-latitude region of the N Pacific [84], when it was still controlled by the input process, i.e., global fallout. Occurrence of higher surface ^{137}Cs concentration in the eastern Equatorial Pacific may be explained by the south-westward advection of subsurface higher ^{137}Cs concentrations penetrating in the eastern N Pacific and the upwelling of subsurface ^{137}Cs in the Equatorial Pacific [85, 86]. In fact, the subsurface maximum of ^{137}Cs concentrations (near 100 m depth) has been observed in the subtropical N Pacific [30, 46]. It is noteworthy that surface ^{137}Cs is homogeneous in the Pacific compared to surface $^{239,240}\text{Pu}$.

The $^{239,240}\text{Pu}$ concentrations in surface water in 2000 (Figs 38 to 41) are estimated to be in the range of 0.5 to about 3 $\mu\text{Bq/L}$ (Table VIII). Higher surface $^{239,240}\text{Pu}$ concentrations appear in the subarctic Pacific (Box 1) and the eastern Equatorial Pacific (Box 7). Higher surface $^{239,240}\text{Pu}$ concentration in the subarctic Pacific can be explained by rapid recycling of scavenged $^{239,240}\text{Pu}$ due to large vertical mixing in winter, whereas higher surface $^{239,240}\text{Pu}$ in the eastern Equatorial Pacific is supported by upwelling of subsurface enriched $^{239,240}\text{Pu}$ as well as ^{137}Cs . The lower values appear in the mid-latitude region of the eastern N Pacific (Box 3), which is consistent with the higher decrease rate of ^{137}Cs in Box 3. Another point is that estimated surface $^{239,240}\text{Pu}$ concentrations in Boxes 2 and 4 of the western N Pacific are higher than those in Boxes 3 and 5 of the eastern N Pacific. Furthermore, there is only a small difference in surface $^{239,240}\text{Pu}$ concentrations between the mid-latitude regions of the Northern and Southern hemispheres. As a result, the current geographical distribution of $^{239,240}\text{Pu}$ concentrations in surface water of the Pacific is very complex as it is controlled by both biogeochemical and physical processes [30, 49].

Therefore one may conclude that the present concentrations of anthropogenic radionuclides (^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$) in Pacific surface water are controlled by oceanographic processes (horizontal advection, vertical mixing and biogeochemical processes) rather than input processes due to global fallout.

In order to assess the environmental effects of anthropogenic radionuclides, it is important to estimate the effective half-lives (residence times) of ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ in each box. They have been calculated from their corresponding regression lines and the results are summarized in Tables VI to VIII. The effective half-life of ^{137}Cs in surface water in each box ranged from 11 to about 30 years. The shortest effective half-life of ^{137}Cs is observed in the mid-latitude of the eastern N Pacific (Box 3).

TABLE VI. ^{90}Sr EFFECTIVE HALF-LIVES AND CONCENTRATIONS IN SURFACE WATER FOR THE YEAR 2000

Box No.	Effective half-life $T_{1/2} \pm 1\sigma$ (years)	Est. concentration in 2000.01.01 Concentration $\pm 1\sigma$ (mBq/L)	Recent measurements			Decay-corr. value to 2000.01.01 (mBq/L)
			Averaged value (mBq/L)	No of data	Period	
01	16.2 \pm 1.5	1.3 \pm 0.1	1.5 \pm 0.3	3	1996–1997	1.3
02	14.4 \pm 0.7	1.1 \pm 0.1	1.7 \pm 0.4	31	1996–1997	1.6
03	10.1 \pm 0.5	1.0 \pm 0.1	1.7 \pm 0.1	3	1993	1.4
04	15.0 \pm 1.4	1.2 \pm 0.1	1.6 \pm 0.1	13	1997	1.5
05	14.0 \pm 1.4	0.9 \pm 0.1	1.3 \pm 0.2	5	1993	1.1
06	21.4 \pm 2.0	1.5 \pm 0.1	1.7 \pm 0.1	2	1993	1.5
07	19.6 \pm 5.0	1.0 \pm 0.3	1.2 \pm 0.1	3	1993	1.1
08			1.4 \pm 0.2	1	1993	1.2
09	17.9 \pm 1.2	1.0 \pm 0.1	1.2 \pm 0.1	4	1996	1.1
10			0.9 \pm 0.2	3	1998	0.8
11			0.8 \pm 0.4	4	1993	0.6
12			0.4 \pm 0.1	2	1993	0.4
13			0.1 \pm 0.1	1	1994	0.1
14	14.1 \pm 0.6	1.4 \pm 0.1	1.8 \pm 0.3	42	1996–1997	1.6
15			1.1 \pm 0.2	15	1998	1.0
16			1.2 \pm 0.2	7	1998–1999	1.1
17			0.7 \pm 0.4	12	1998–1999	0.7
18	13.3 \pm 1.4	1.6 \pm 0.2	2.4 \pm 0.4	1	1997	2.3
19						
20	5.8 \pm 0.5	1.5 \pm 0.1	2.0 \pm 0.5	7	1997	1.9
21	12.0 \pm 1.7	8.9 \pm 1.3	12.0 \pm 3.1	71	1996–1997	11.1
22	6.9 \pm 0.4	2.9 \pm 0.2	4.3 \pm 1.3	118	1996–1999	4.0
23			49 \pm 84	14	1999	49
24	5.3 \pm 0.7	2.6 \pm 0.3	4.5 \pm 1.6	43	1996–1997	4.1
25	25.8 \pm 3.4	1.5 \pm 0.2	2.2 \pm 0.9	17	1996–1999	2.1
26			22 \pm 4.0	15	1995–2000	17
27			1.8 \pm 0.3	15	1997–1998	1.7
28	13.8 \pm 1.4	0.7 \pm 0.1	1.3 \pm 0.7	17	1996–1998	1.2
29	24.2 \pm 2.1	0.8 \pm 0.1	1.0 \pm 0.1	5	1992–1993	0.8
30	17.3 \pm 8.8	0.3 \pm 0.2	0.5 \pm 0.2	2	1993	0.4

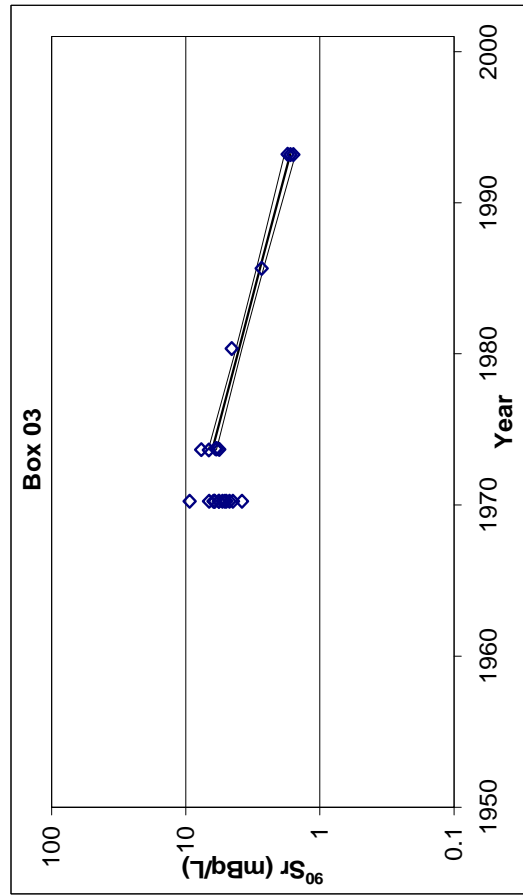
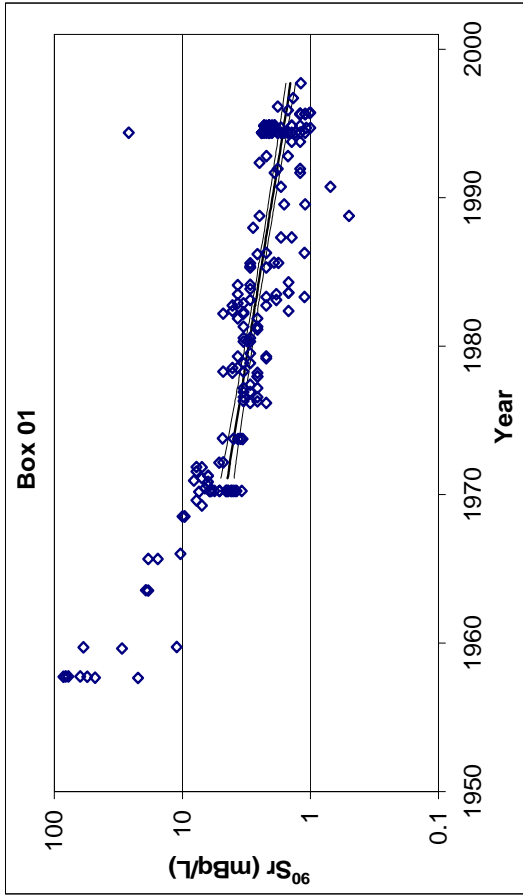
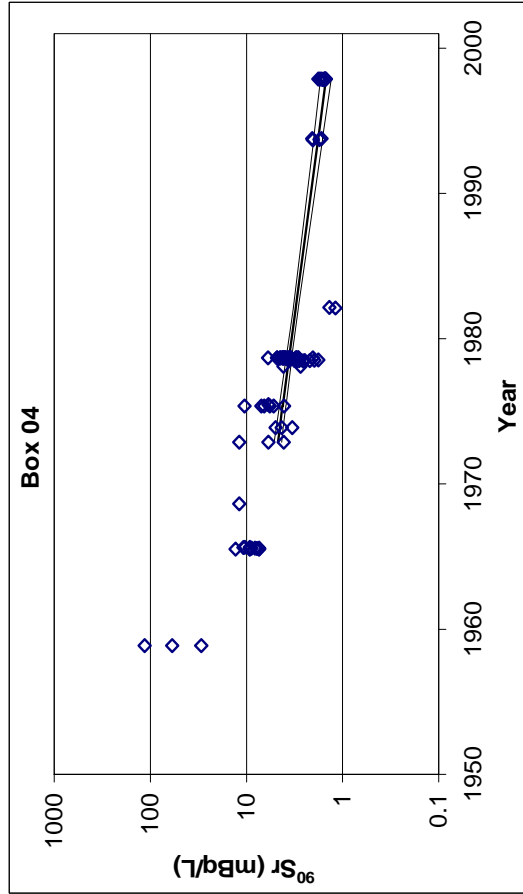
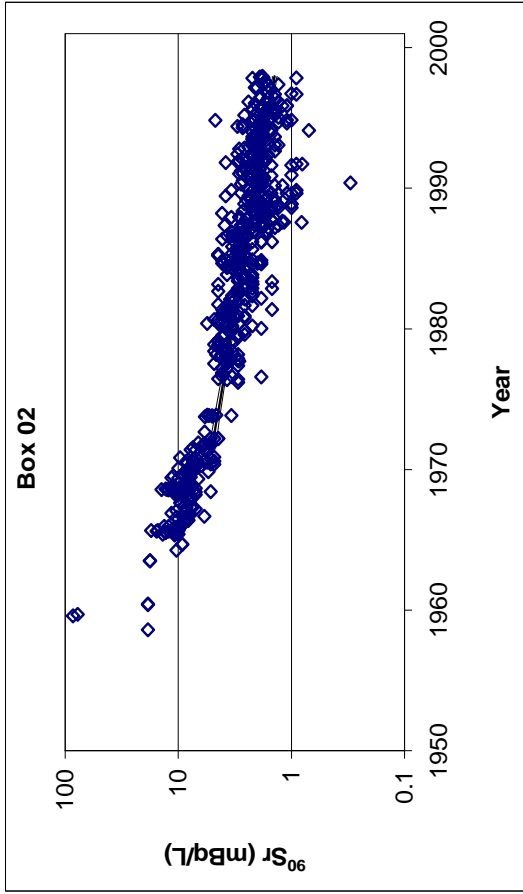


FIG. 30. ^{90}Sr in Pacific surface water.

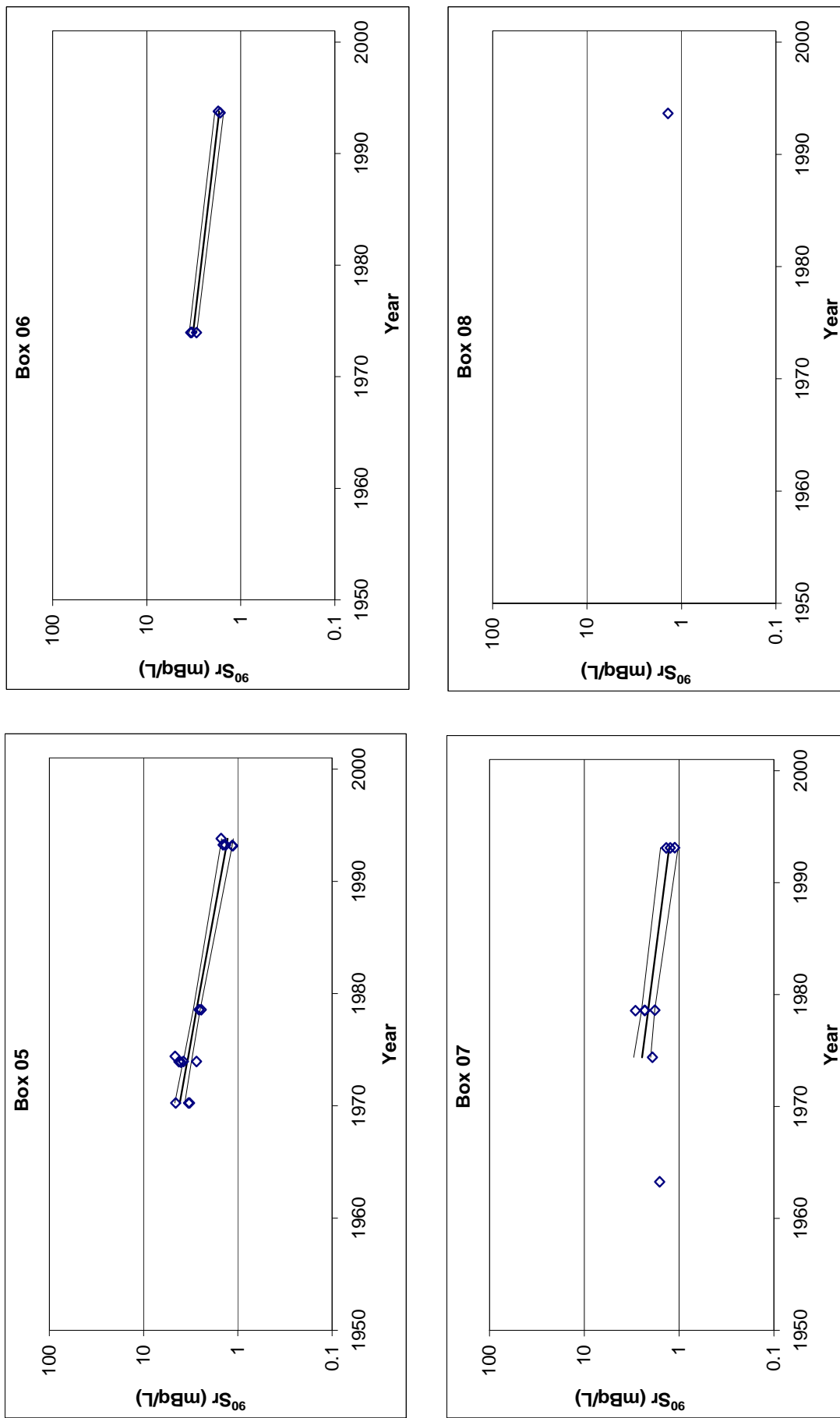


FIG. 31. ^{90}Sr in Pacific surface water.

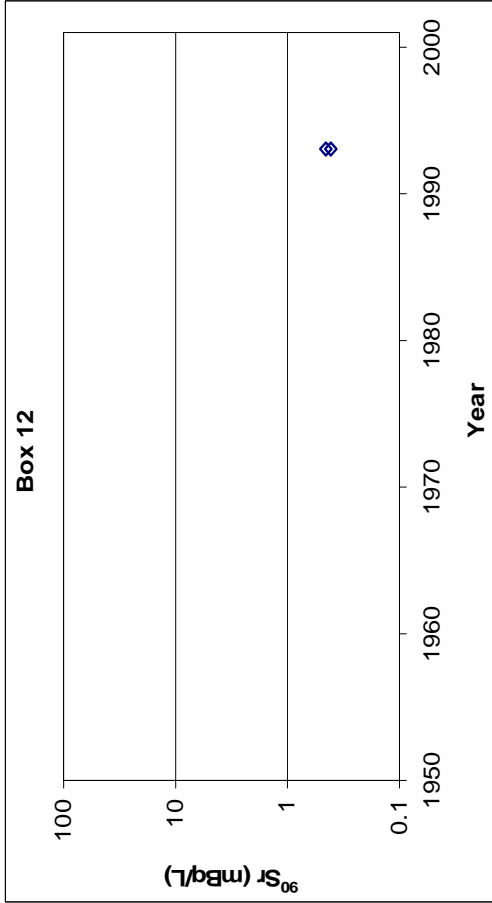
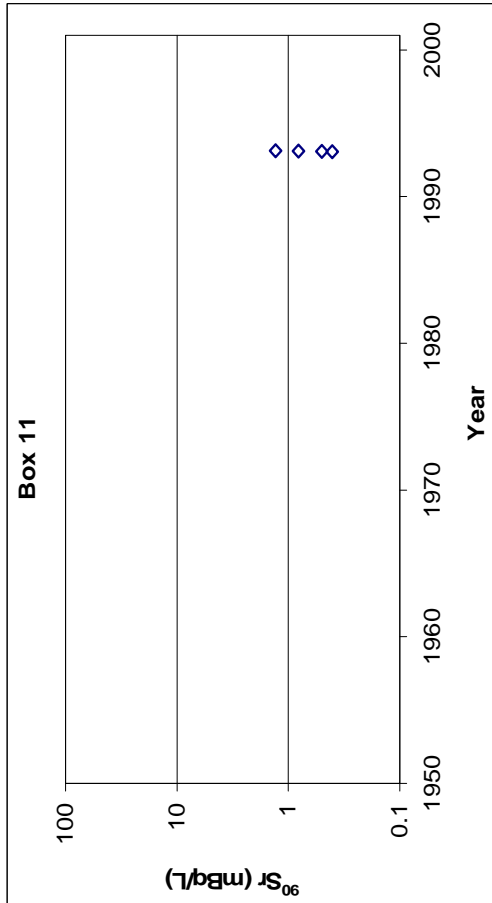
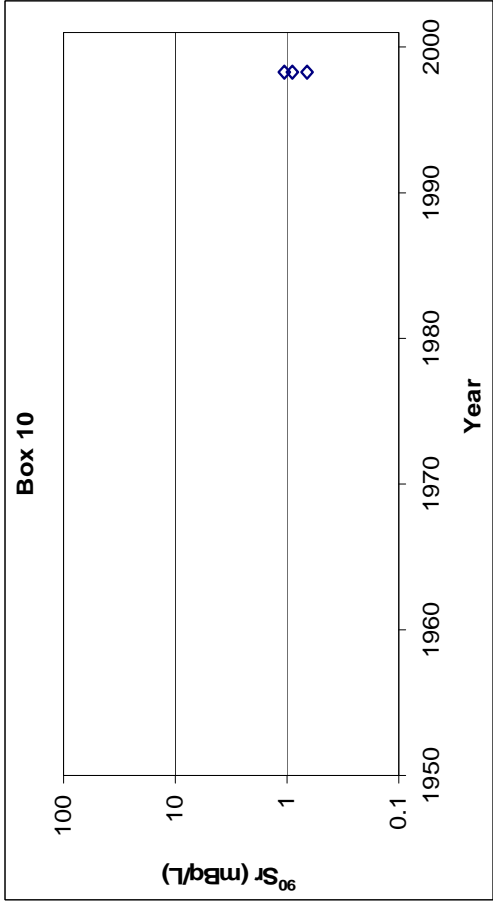
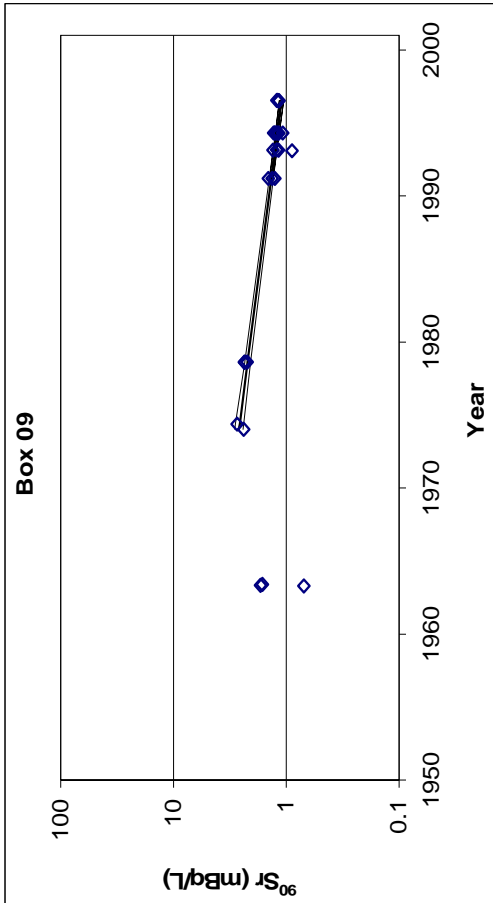


FIG. 32. ^{90}Sr in Pacific surface water.

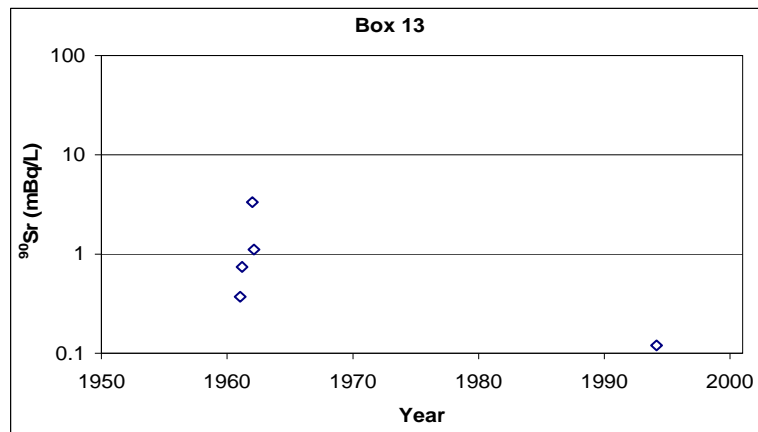


FIG. 33. ⁹⁰Sr in Pacific surface water.

TABLE VII. ¹³⁷Cs EFFECTIVE HALF-LIVES AND CONCENTRATIONS IN SURFACE WATER FOR THE YEAR 2000

Box No.	Effective half-life $T_{1/2} \pm 1\sigma$ (years)	Estimated concentration in 2000.01.01 Concentration $\pm 1\sigma$ (mBq/L)	Recent measurements			Decay-corrected value to 2000.01.01 (mBq/L)
			Averaged value (mBq/L)	Number of data	Period	
01	13.7 \pm 0.8	1.7 \pm 0.1	2.1 \pm 0.5	4	1996–1997	2.0
02	16.5 \pm 0.9	2.3 \pm 0.1	2.6 \pm 0.6	30	1996–1997	2.4
03	10.6 \pm 0.5	1.9 \pm 0.1	3.2 \pm 0.6	3	1993	2.8
04	24.2 \pm 3.1	2.8 \pm 0.4	2.6 \pm 0.4	7	1997	2.4
05	18.4 \pm 4.2	2.7 \pm 0.6	2.7 \pm 0.4	4	1993	2.3
06	29.9 \pm 7.7	2.4 \pm 0.6	2.7 \pm 0.1	2	1993	2.3
07	21.3 \pm 3.0	1.7 \pm 0.2	2.2 \pm 0.1	3	1993	1.9
08			1.9 \pm 0.4	1	1993	1.7
09	28.5 \pm 1.6	1.8 \pm 0.1	1.9 \pm 0.1	4	1996	1.7
10	23.3 \pm 2.6	1.3 \pm 0.1	1.4 \pm 0.1	3	1998	1.4
11	11.9 \pm 2.0	0.7 \pm 0.1	1.3 \pm 0.8	4	1993	1.1
12			0.6 \pm 0.3	2	1993	0.6
13			0.1 \pm 0.1	1	1994	0.1
14	19.0 \pm 0.9	2.5 \pm 0.1	3.0 \pm 0.5	44	1996–1997	2.8
15	20.2 \pm 1.9	1.5 \pm 0.1	1.6 \pm 0.3	17	1998	1.6
16	26.3 \pm 4.9	1.9 \pm 0.4	2.2 \pm 0.3	7	1998–1999	2.1
17	12.7 \pm 7.0	0.7 \pm 0.4	1.1 \pm 0.6	12	1998–1999	1.0
18	13.6 \pm 1.5	2.8 \pm 0.3	15 \pm 3	1	1997	14
19				0		
20	4.2 \pm 0.2	1.9 \pm 0.1	3.8 \pm 2.1	9	1997	3.6
21	14.1 \pm 1.8	49.8 \pm 6.5	66 \pm 20	214	1996–1997	61
22	5.7 \pm 0.3	4.5 \pm 0.2	6.9 \pm 3.1	137	1996–1999	6.5
23			57 \pm 55	28	1999	57
24	6.9 \pm 0.6	3.1 \pm 0.3	4.5 \pm 1.6	44	1996–1997	4.2
25	23.3 \pm 1.8	2.3 \pm 0.2	4.5 \pm 2.9	15	1996–1999	4.2
26	7.3 \pm 1.2	25 \pm 3	30 \pm 14	20	1996–2000	25
27			2.7 \pm 0.4	15	1997–1998	2.6
28	14.9 \pm 1.6	1.3 \pm 0.1	1.8 \pm 0.9	16	1996–1997	1.7
29	23.7 \pm 5.7	1.4 \pm 0.3	1.7 \pm 0.2	5	1991–1993	1.4
30	20.2 \pm 6.3	0.5 \pm 0.1	0.7 \pm 0.2	2	1993	0.6

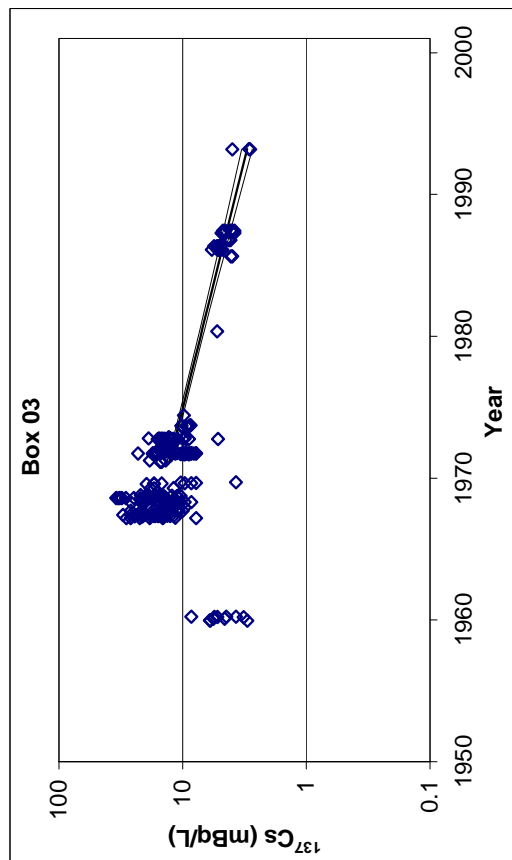
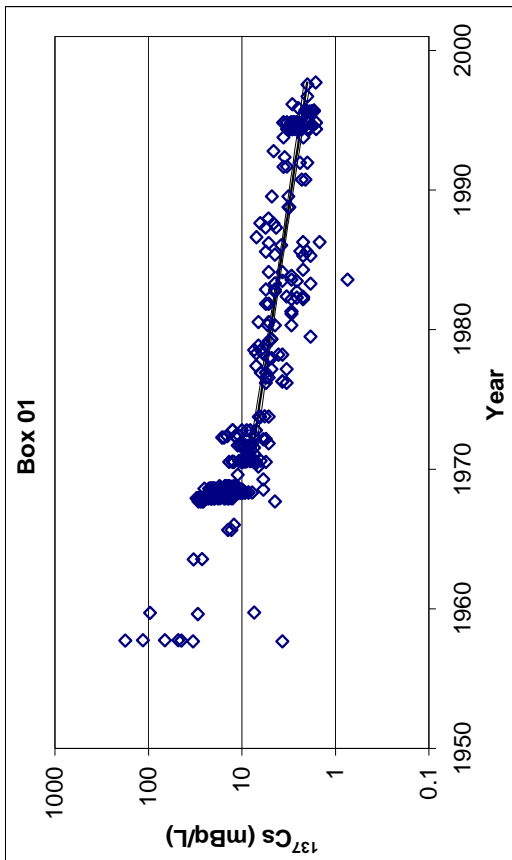
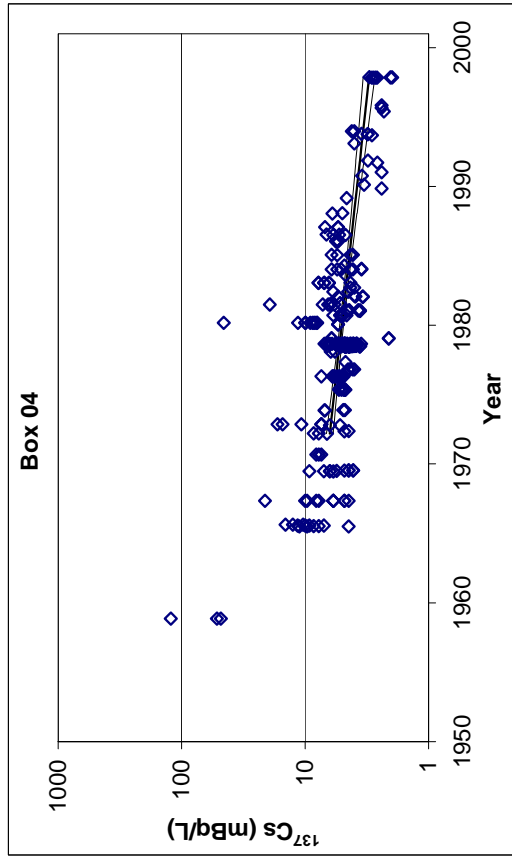
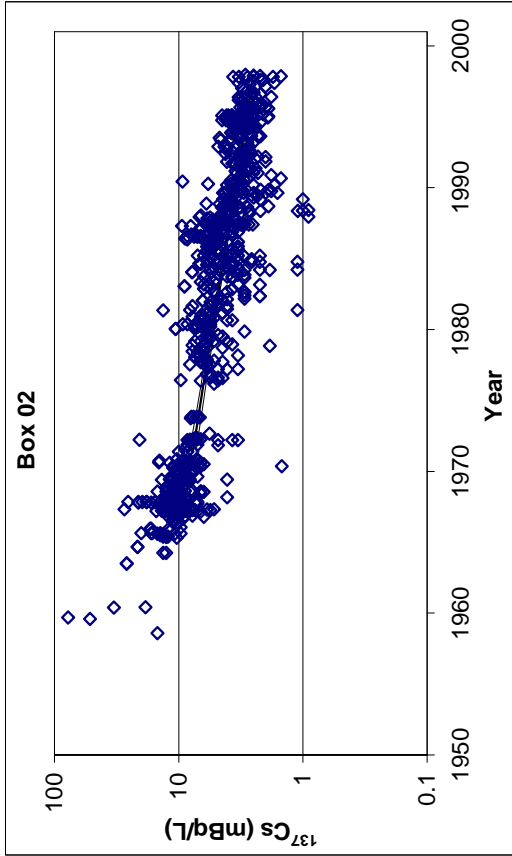


FIG. 34. ^{137}Cs in Pacific surface water.

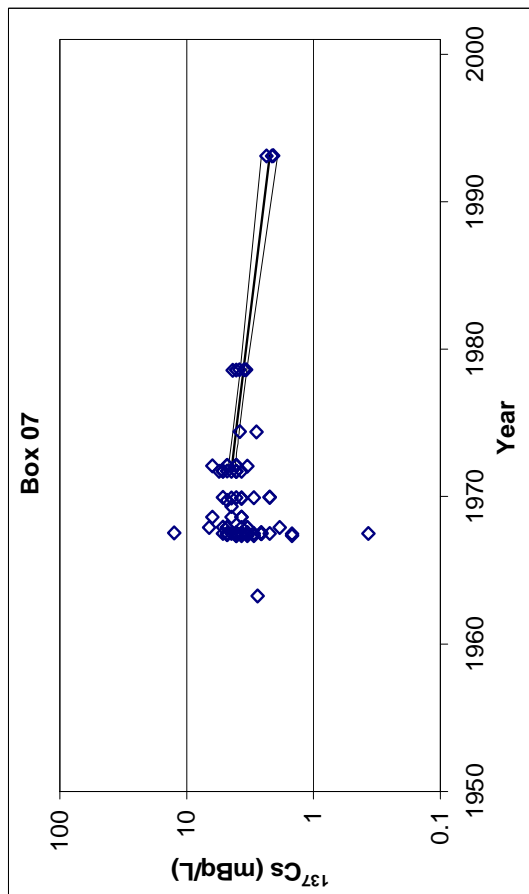
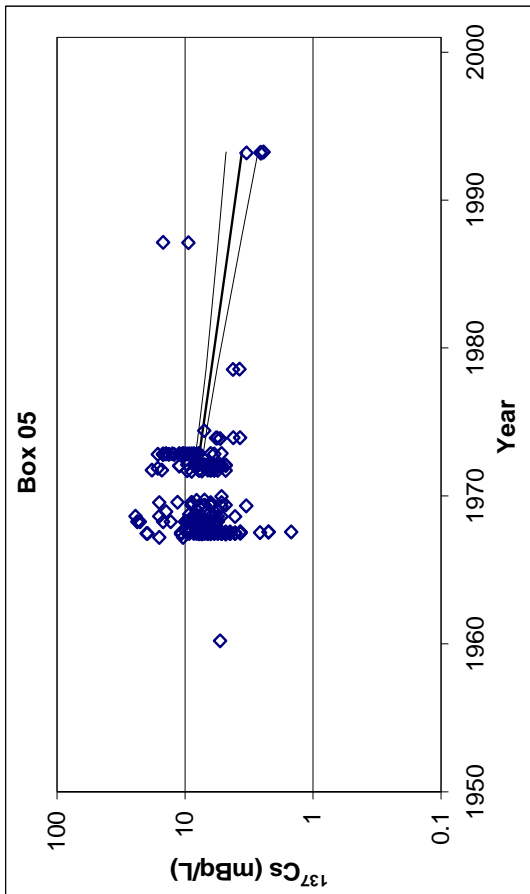
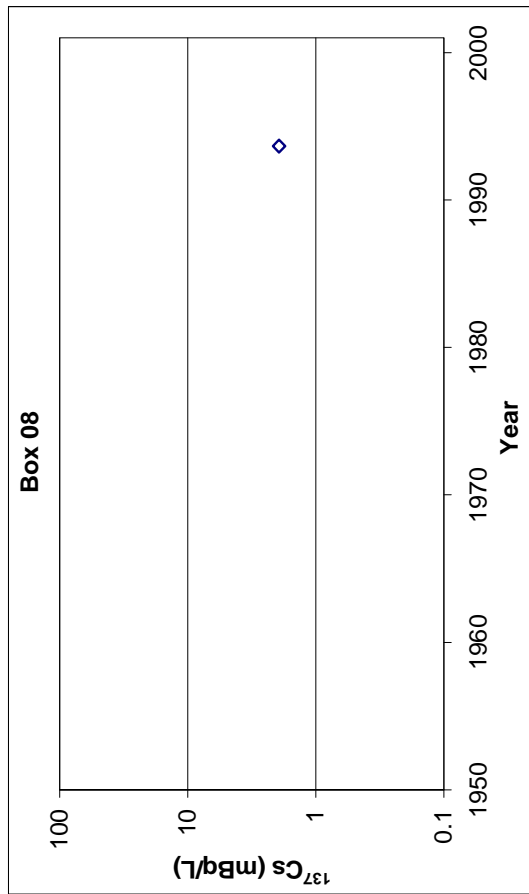
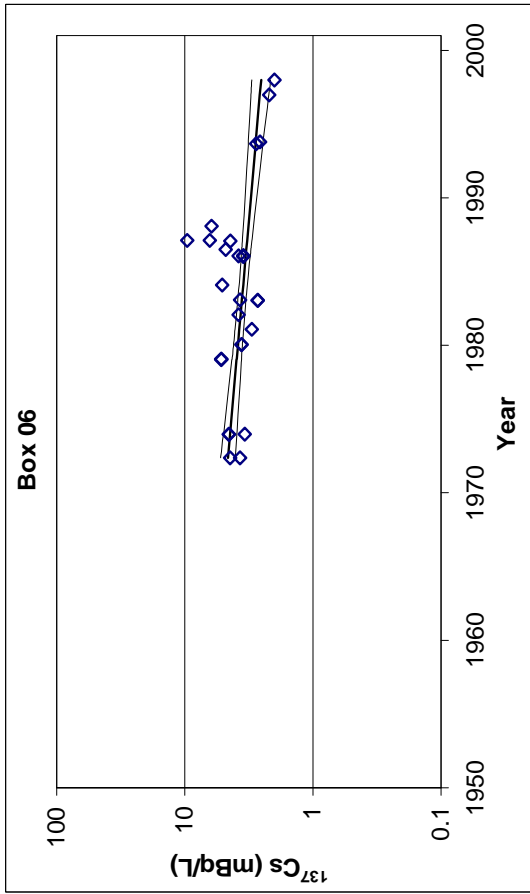


FIG. 35. ^{137}Cs in Pacific surface water.

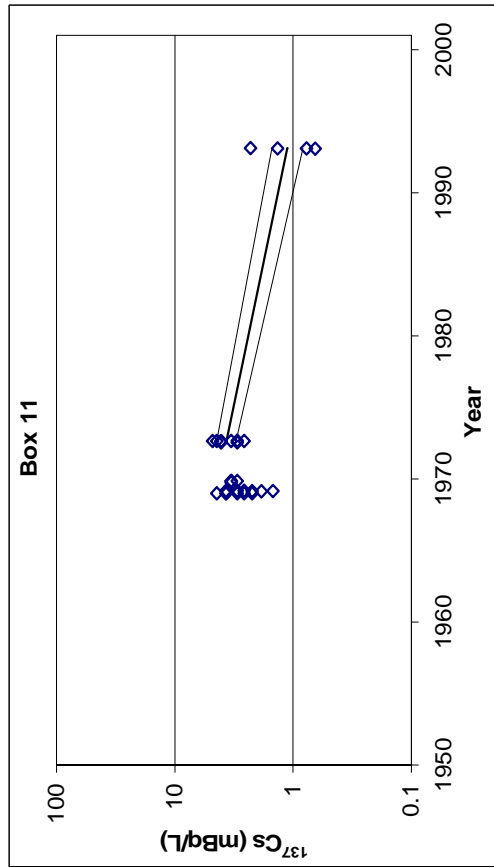
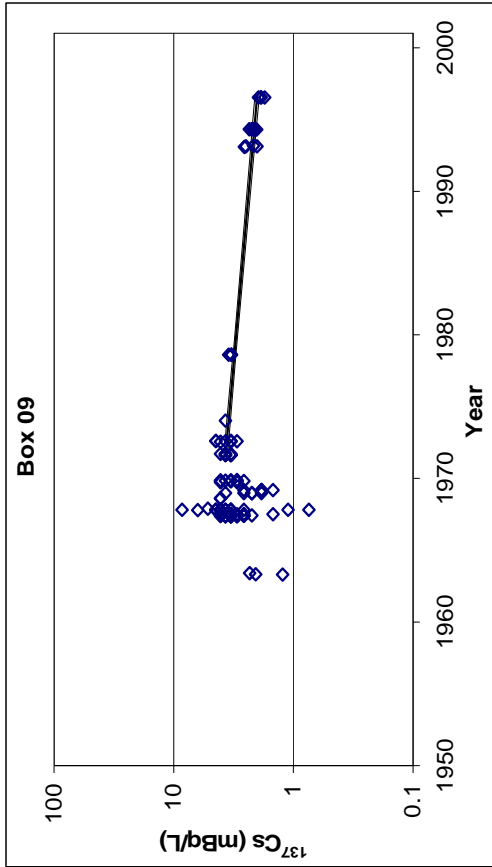
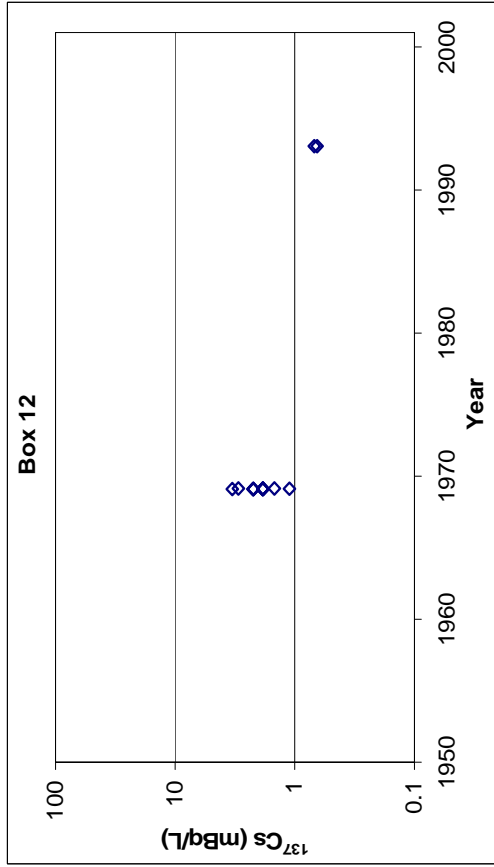
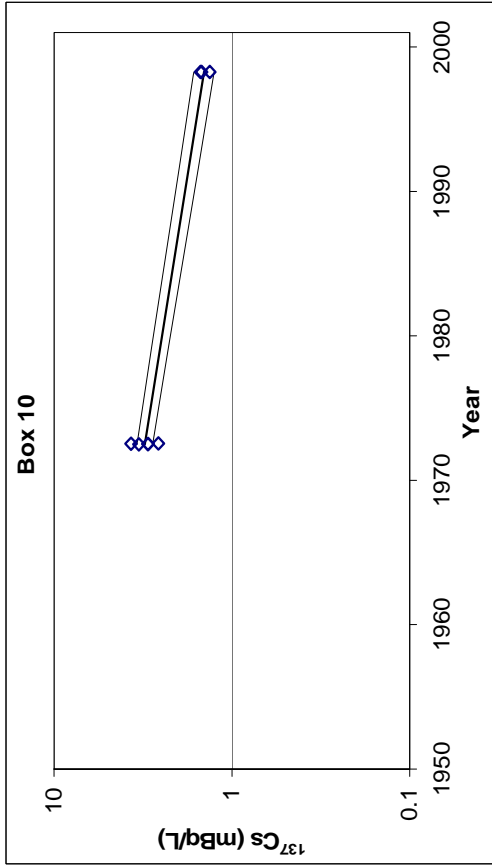


FIG. 36. ^{137}Cs in Pacific surface water.

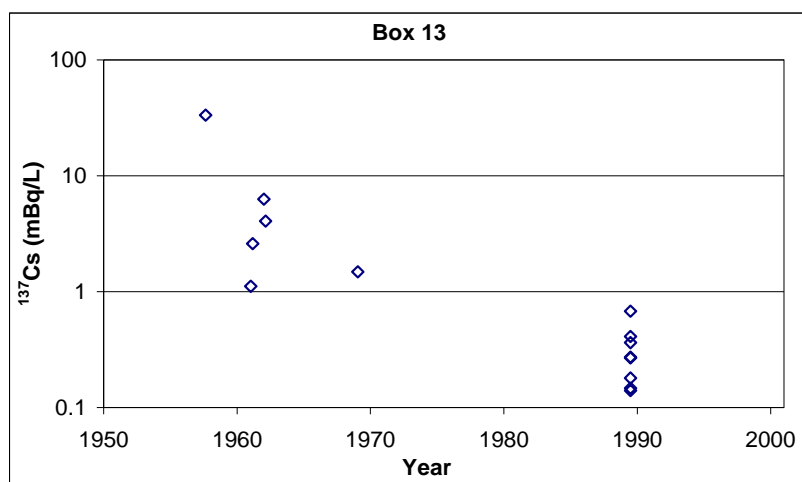


FIG. 37. ^{137}Cs in Pacific surface water.

The longest effective half-life of ^{137}Cs is found in the Equatorial Pacific (Boxes 6 and 7), (which may be due to Equatorial upwelling) and in the eastern subtropical South Pacific (Box 9). The effective half-life of ^{90}Sr in surface water in each box ranged from 10 to 21 years. The shortest effective half-life of ^{90}Sr is observed in the eastern N Pacific (Box 3). The longest effective half-life of ^{90}Sr occurs in the Equatorial Pacific (Boxes 6 and 7), which is the same area as for ^{137}Cs . The effective half-life of ^{90}Sr in seems to be a little less than that of ^{137}Cs , however the data are not precise enough to confirm this. The effective half-life of $^{239,240}\text{Pu}$ in surface water in each box ranged from 5 to about 15 years. The shortest effective half-life of $^{239,240}\text{Pu}$ is observed in the mid-latitude region of the eastern N Pacific (Box 3) as for ^{137}Cs .

These findings suggest that the mid-latitude region of the eastern N Pacific is a sink of anthropogenic radionuclides. The surface water mass in the mid-latitude region of the eastern N Pacific is penetrated under the surface layer of the subtropical gyre. As a result, the subsurface maximum of ^{137}Cs is formed in the subtropical N Pacific [30, 85]. The longest effective half-life of $^{239,240}\text{Pu}$ is found in the eastern Equatorial Pacific (Box 7), as is the case for ^{137}Cs and ^{90}Sr . At present, the eastern Equatorial Pacific seems to behave as a source of anthropogenic radionuclides. The effective half-life of $^{239,240}\text{Pu}$ is generally shorter than that of ^{137}Cs in each box. Plutonium is a particle-reactive radionuclide as opposed to ^{137}Cs i.e. plutonium reacts with biogenic particles in surface water [67, 87], sinks with attached particles, and is remobilised as a result of the biological decomposition of particles in deeper water [49].

In South Pacific surface water, the effective half-lives of the radionuclides investigated appear to be longer than in the N Pacific. When corrected for radioactive decay, surface ^{137}Cs in the South Pacific (except for Box 11) has gradually increased over the past three decades. This may suggest that ^{137}Cs in the N Pacific is transported to the S Pacific, because the major input of ^{137}Cs due to global and local fallout occurred in the N Pacific.

When the ^{90}Sr data for the North, Equatorial and South Pacific are grouped (Table IX), the corresponding mean effective half-lives are 12 ± 1 , (North) 18 ± 2 (South) and 21 ± 2 years (Equatorial Pacific). All Pacific data together give an effective half-life of ^{90}Sr of 16 ± 2 years. ^{137}Cs and $^{239,240}\text{Pu}$ effective half-lives for the whole Pacific Ocean are 16 ± 4 and 7 ± 1 years, respectively.

TABLE VIII. $^{239,240}\text{Pu}$ EFFECTIVE HALF-LIVES AND CONCENTRATIONS IN SURFACE WATER FOR THE YEAR 2000

Box No.	Effective half-life		Est. concentration in 2000.01.01		Recent measurements				
	$T_{1/2} \pm 1\sigma$ (years)		Concentration $\pm 1\sigma$ ($\mu\text{Bq/L}$)		Averaged value ($\mu\text{Bq/L}$)		Number of data	Period	
01	16.7	± 4.3	2.8	± 0.7	7.9	± 8.7	2	1996–1997	
02	9.3	± 1.4	2.2	± 0.3	3.8	± 2.4	6	1996–1997	
03	5.1	± 0.7	0.5	± 0.1	0.7	± 0.1	3	1993	
04	7.2	± 0.7	1.6	± 0.2	2.4	± 0.7	7	1997	
05	8.4	± 1.5	0.9	± 0.2	1.7	± 0.7	2	1993	
06	9.0	± 2.0	1.3	± 0.3	3.6	± 2.7	5	1991–1994	
07	15.1	± 4.2	1.9	± 0.5	2.7	± 0.2	3	1993	
08					1.5	± 1.2	5	1992–1993	
09	12.1	± 3.6	1.6	± 0.5	5.7	± 1.4	4	1996	
10					4.3	± 0.1	2	1998	
11					1.6	± 0.6	4	1993	
12					0.8	± 0.5	1	1993	
13	14.1	± 7.1	1.0	± 0.5	1.3	± 0.7	1	1994	
14	15.7	± 2.6	5.2	± 0.9	6.6	± 2.5	30	1996–1997	
15	21.2	± 7.6	1.5	± 0.5	1.9	± 1.2	13	1998	
16	12.9	± 5.3	2.2	± 0.9	3.0	± 1.5	4	1998–1999	
17	8.6	± 1.1	0.8	± 0.1	1.0	± 0.5	10	1998–1999	
18	13.2	± 1.5	5.0	± 0.5	6.4	± 1.5	24	1995	
19									
20					20	± 12	7	1997	
21	4.9	± 1.4	1.6	± 0.5	3.4	± 2.5	21	1996	
22	24	± 17	11	± 8	15	± 10	33	1997	
23					500	± 400		1994–1999	
24	6.9	± 1.2	9.5	± 1.7	13	± 8	18	1997	
25	12.8	± 2.5	5.1	± 1.0	36	± 16	9	1997	
26					5.3	± 2.3	5	2000	
27					14	± 4	7	1997–1998	
28	8.8	± 0.6	3.1	± 0.2	5.0	± 2.7	17	1996–1998	
29	9.8	± 2.9	1.5	± 0.5	2.8	± 1.3	5	1993	
30	10.7	± 4.9	1.1	± 0.5	1.8	± 0.6	1	1993	

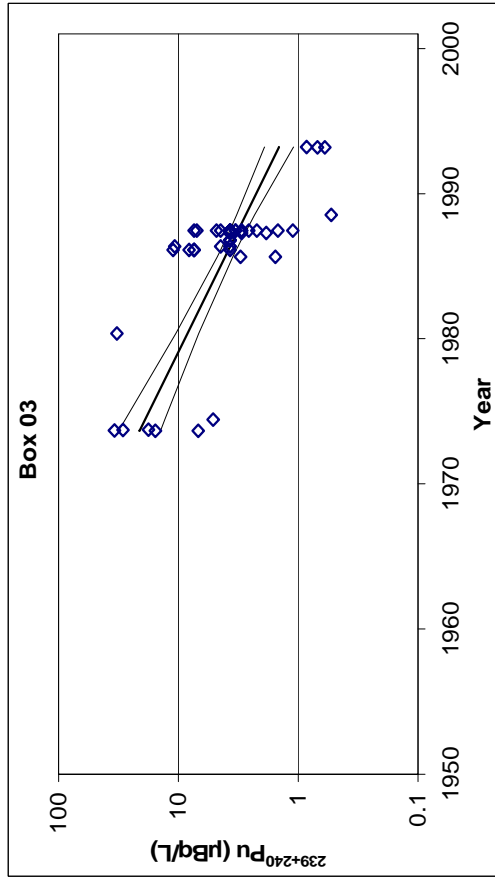
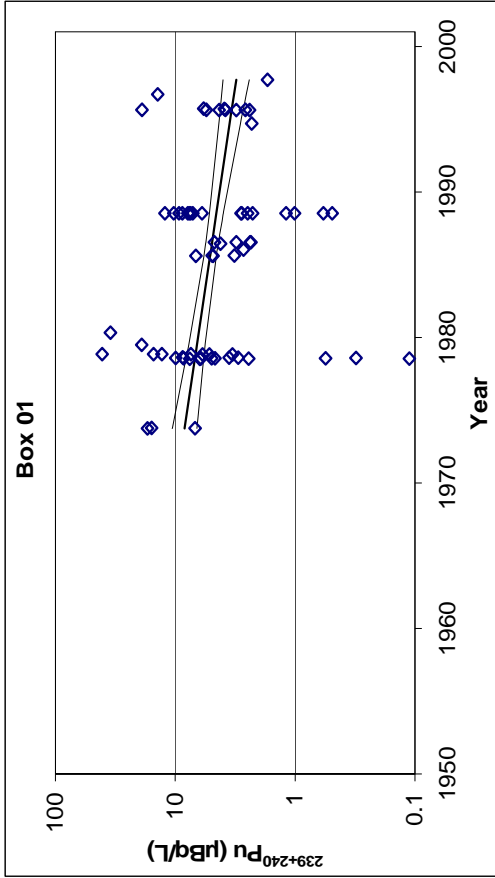
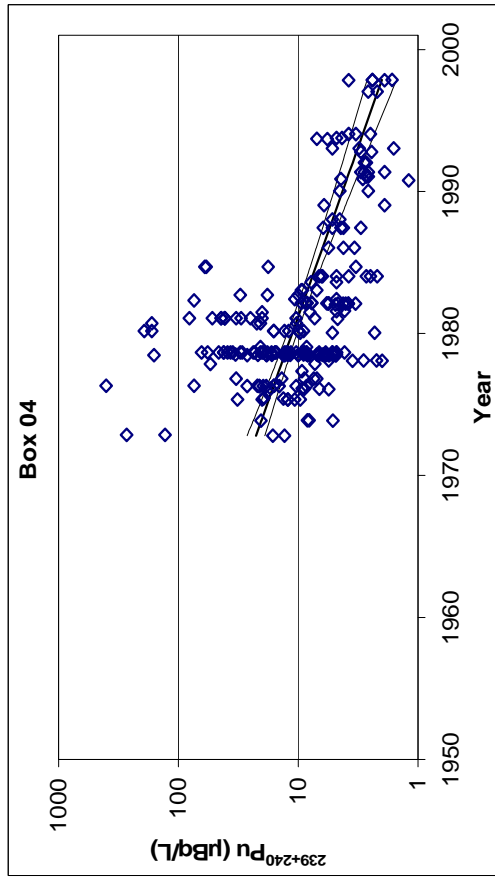
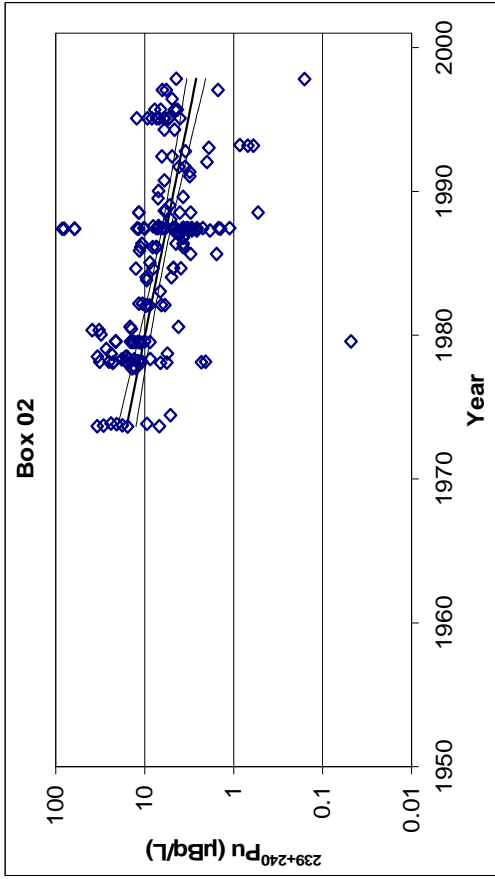


FIG. 38. $^{239,240}\text{Pu}$ in Pacific surface water

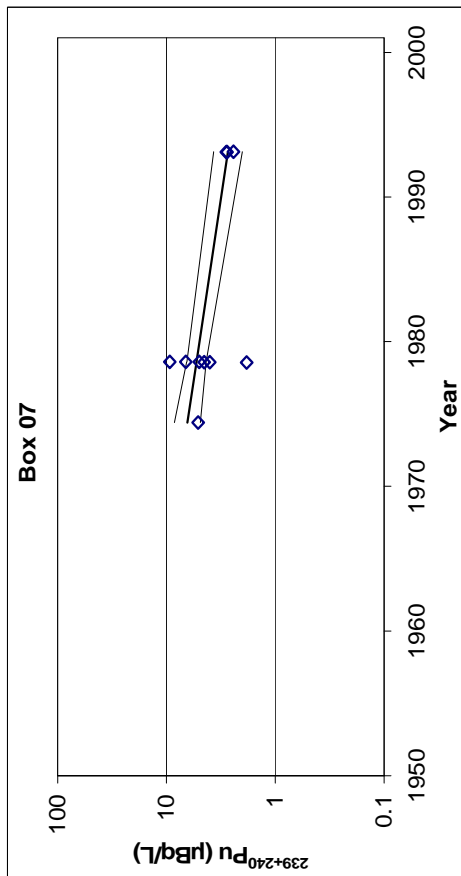
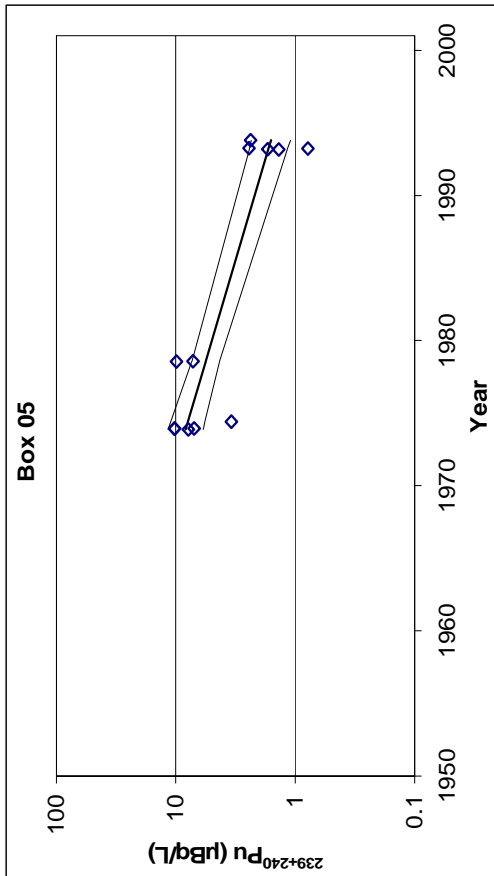
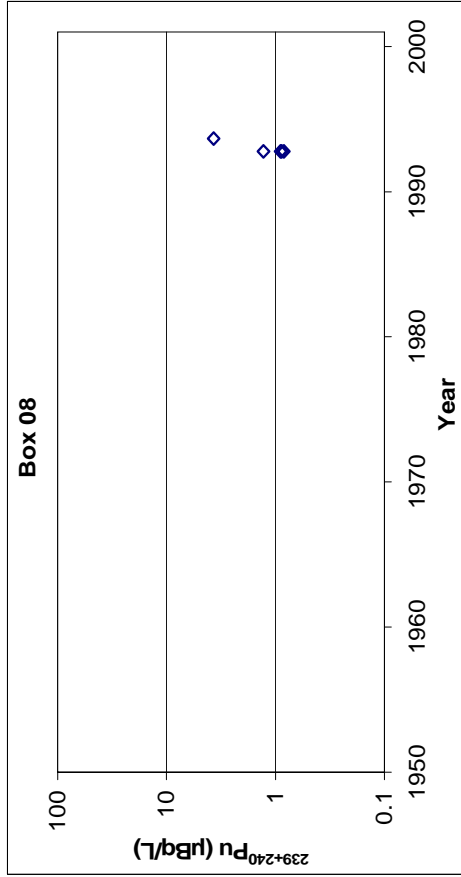
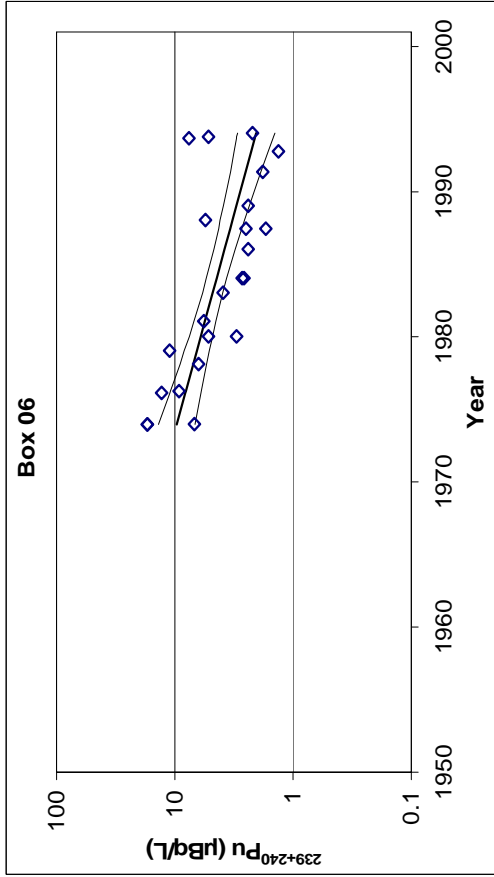


FIG. 38. ^{239,240}Pu in Pacific surface water

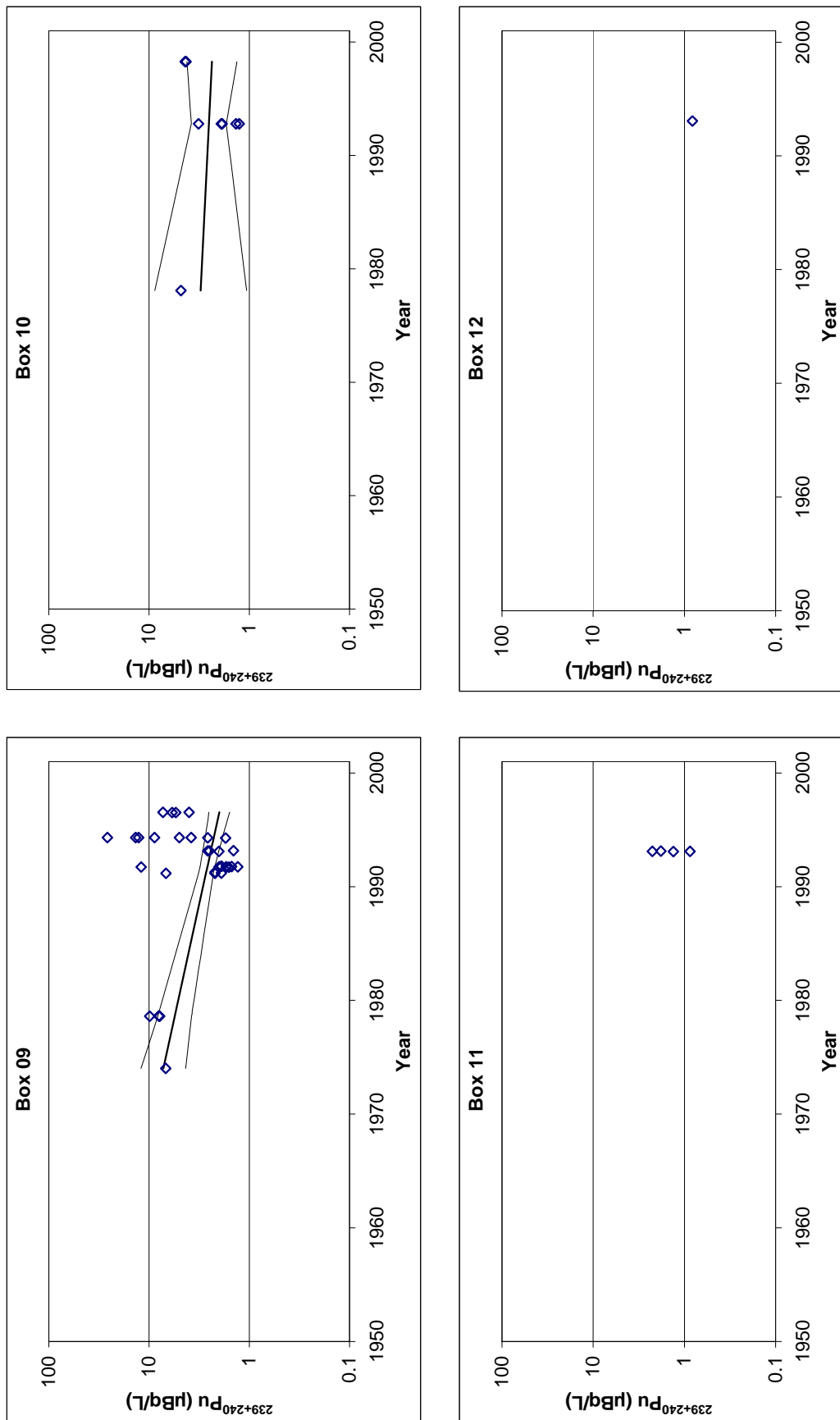


FIG. 40. $^{239,240}\text{Pu}$ in Pacific surface water

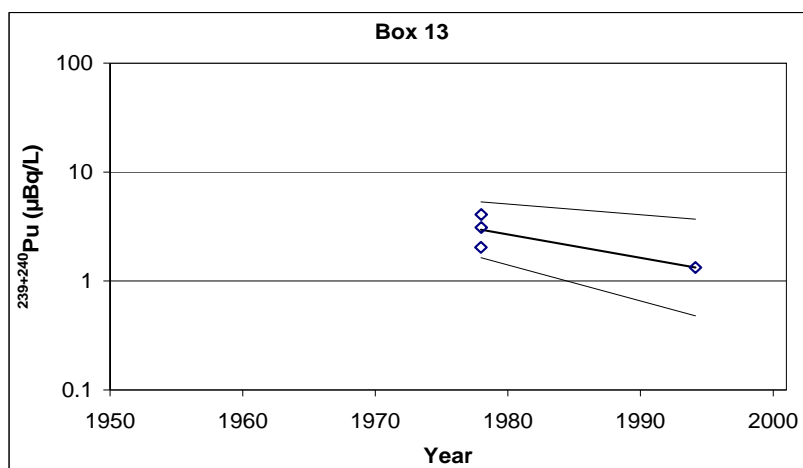


FIG. 41. $^{239,240}\text{Pu}$ in Pacific surface water.

TABLE IX. MEAN EFFECTIVE HALF-LIVES OF ^{90}Sr , ^{137}Cs AND $^{239,240}\text{Pu}$ IN PACIFIC OCEAN SURFACE WATER

Area	Boxes	Effective Half-life $\pm 1\sigma^*$ (years)		
		^{90}Sr	^{137}Cs	$^{239,240}\text{Pu}$
North Pacific	01-05	12.4 ± 1.1	12.7 ± 1.4	6.7 ± 0.9
Equatorial Pacific	06-07	21.1 ± 1.9	28.6 ± 3.0	10.2 ± 2.4
South Pacific	08-12	17.9 ± 1.0	22.2 ± 5.2	12.1 ± 3.6
All Pacific	01-12	15.9 ± 2.4	15.9 ± 4.3	7.4 ± 1.1

* Error weighted averaged values

4.5. Water column radionuclide time series

The primary focus of work to date has been on the distribution and behaviour of ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ in the water column. Bowen et al. [30] have made the most comprehensive study of these radionuclides in open Pacific waters. This study presented and discussed the data obtained at 29 stations in 1973–74 as part of the GEOSECS programme. They covered much of the N Pacific but did not go further south than 19°S . So, as in other studies, the South Pacific has received little attention.

A number of other studies have been made subsequent to the GEOSECS study e.g. [41, 42, 45–48, 51, 78, 81, 82, 88, 89]. These studies provide data to augment the GEOSECS data and to compare possible changes with time in the distributions and inventories in the water column. Some comparative observations are made below.

Most stations in the Pacific Ocean visited in 1973 during the GEOSECS expedition provided ^{90}Sr , ^{137}Cs and Pu water profile data with high spatial and depth resolutions [30]. Therefore, one of the objectives of the IAEA 1997 expedition to the NW Pacific Ocean carried out in the framework of the WOMARS project was to re-visit some of the GEOSECS stations so that the development of radionuclide concentrations in the water column and their distribution could be evaluated with time. A similar comparison is possible with the data

obtained by the KNORR-1978 [82] and Hakuho Maru 1980, 1982 [41, 80] expeditions. The data compiled from these four expeditions, as well as data from other expeditions to the region, enabled the development of time series for studying the behaviour of radionuclides in the ocean with time over the past 24 years. Figure 42 shows the main sampling stations in the NW Pacific Ocean for which the radionuclide time series will be compared. However, the general evaluation of radionuclide data in the Pacific was done using a much larger data set collected and stored in GLOMARD.

Figures 43 to 46 and 47 to 50 show ^{90}Sr and ^{137}Cs profiles, respectively, in water columns of the central NW Pacific Ocean at GEOSECS, Hakuho Maru and IAEA stations (the positions of the sampling stations are shown in Fig. 42). When comparing for instance Figs 43 (^{90}Sr) and 47 (^{137}Cs), it can be seen that both radionuclides show similar behaviour, except that ^{137}Cs concentrations are higher by a factor of about 1.6, which represents the typical global fallout ratio for these radionuclides. There is a remarkable decrease in surface and subsurface concentrations from the GEOSECS to the IAEA expeditions –from about 3 to 2 Bq/m^3 (peak values) for ^{90}Sr (Fig. 43) and from about 4 to 3 Bq/m^3 for ^{137}Cs (Fig. 47).

Oceanic studies of the behaviour of Pu have indicated that, especially in major low productivity oceanic areas, it is clearly not being as rapidly transferred to sediment as more reactive actinides such as Th or Am. Nevertheless, the particulate associations in transport to sediment and retention there, is an important aspect of Pacific Pu studies and a number of papers have addressed this. For example, at least two studies involving sediment traps [89, 90] have served to confirm that, as in other oceans, the flux of sinking biogenic particles fed by biological production in the surface ocean, is the major vehicle for the transport of Pu (or other reactive elements) from the surface to the deep ocean. Clearly, Pu enters into the cycle of organic material that sinks from the surface ocean. On its descent, this material is exposed to oxidation and bacterial degradation processes and both the organic material and its associated elements or radionuclides are to some extent released into sea water. As has been widely recognized, only a very small fraction (in the one per cent range) of material originating at the surface actually arrives at the sea-floor. This process is clearly responsible for the observed deep-water Pacific distributions of Pu below depths ventilated physically from the surface. Furthermore, such material delivered to the sea-floor is available for mixing by benthic organisms into bottom sediment, and can also be released to overlying waters by oxidation prior to burial. The latter process can be expected to produce the bottom Pu maxima for the Pacific deep water above the sea-floor referred to above.

Three major points were made in the GEOSECS study regarding plutonium — and have been confirmed in subsequent work:

- (i) A strong subsurface maximum in Pu concentrations was observed over large areas of the Pacific,
- (ii) Significant increases in Pu concentrations were generally noted in deep water near the sea-floor. (These increases seemed to be restricted to the western and northern stations in the N Pacific region),
- (iii) With the exception of the eastern GEOSECS stations between 30° - 40°N , Pu inventories in the water column substantially exceed those expected from global fallout. Local fallout from Pacific nuclear weapons test sites is proposed as the most likely explanation for this observation.

Figure 51 shows $^{239,240}\text{Pu}$ profiles in water columns of the central NW Pacific Ocean at GEOSECS, Hakuho Maru and IAEA stations (the positions of the sampling stations are shown in Fig. 42). It is clear that after 24 years, the subsurface maximum became much

smaller and less pronounced (a decrease by about a factor of 4) and has moved to deeper water layers (from 450 m to 850 m). Although the initial view of these changes (and as discussed subsequently with respect to Pu inventory change over time) was that they were caused by Pu association with sunken particle fluxes, this view almost certainly needs to be revised in the light of new data for ^{90}Sr and ^{137}Cs . It has been shown that, in the upper 1000 m, the decline in ^{90}Sr and ^{137}Cs maxima and inventories is similar to the Pu changes. This would lead to the conclusion that at least a major part of the observed Pu changes over time was in response to physical circulation in the upper water column that brought advectively into the region water masses bearing significantly lower levels of fallout nuclides.

A similar comparison in the development of the $^{239,240}\text{Pu}$ profile in the water column can be seen in Figs 52 and 53, where data from the GEOSECS, KNORR, and IAEA stations situated west and east of Bikini Atoll, respectively, are compared. The vertical spread in the Pu maxima is much smaller and the recent concentrations have been higher than in Fig. 51. The concentrations and inventories at these stations did not change significantly over 24 years. As these stations are all in the westward flowing North Equatorial Current (with very low biological productivity, oligotrophic conditions), the maintenance of the concentration profiles and inventories in the upper ocean may be related to these water masses recirculating in near-permanent low productivity conditions.

Although no difference is noted in Pu profiles in the upper ocean stations shown in Figs 52 and 53, west and east of Bikini, substantial differences can be found in deep water east of (upstream) and west of (downstream) the atolls. In Fig. 54, various Pu profiles east of the islands are contrasted with others in between and west of the islands. A difference of about a factor of two can be observed for deep-water Pu concentrations in the eastern (broken lines) and western (solid lines) stations visited between 1973 and 1982. Higher deep-water Pu levels were found in 1997 (IAEA station 6, northwest of Bikini). This east/west gradient may have been created initially by the large test fallout patterns. A further explanation that may be applicable could involve the continuous remobilization of Pu to deep water from heavily contaminated material down the steep slopes from the atolls to the sea-floor. As deep-water circulation is slow but heading to the northwest, elevated deep-water concentration levels to the west of the islands could be maintained over time.

On the basis of all available $^{239,240}\text{Pu}$ data in the central NW Pacific Ocean (between 20° – 40°N and 135° – 175°E), a time dependence of the position of the subsurface maximum in the water column has been worked out. This linear dependence (on a semilogarithmic scale) is shown in Fig. 55, documenting a similar trend at all sampling stations between 1973 and 1997. The $^{239,240}\text{Pu}$ maximum moves downward in the water column reaching double the depth in about 40 years. A decrease in the maximum $^{239,240}\text{Pu}$ concentration observed at the central NW Pacific stations with time is shown in Fig. 56. The change of the maximum $^{239,240}\text{Pu}$ concentration in the water column to a half value is reached after 22 ± 8 years. As noted earlier, much of these observed changes may be physically driven.

As discussed, the input function by which Pu was introduced into the Pacific Ocean was mainly two pulse injections into the surface layer in the early 1950s (local tropospheric fallout) and in the early 1960s (global fallout). Therefore, Pu in the water column is in a transient state and its concentrations and inventories should change with time according to oceanographic, geochemical and biological processes that occur in the water column. Bowen et al. [91] proposed that the $^{239,240}\text{Pu}$ maximum they observed in the water column of the Atlantic Ocean could be a result of the bacterial degradation of sinking particles with subsequent release of plutonium.

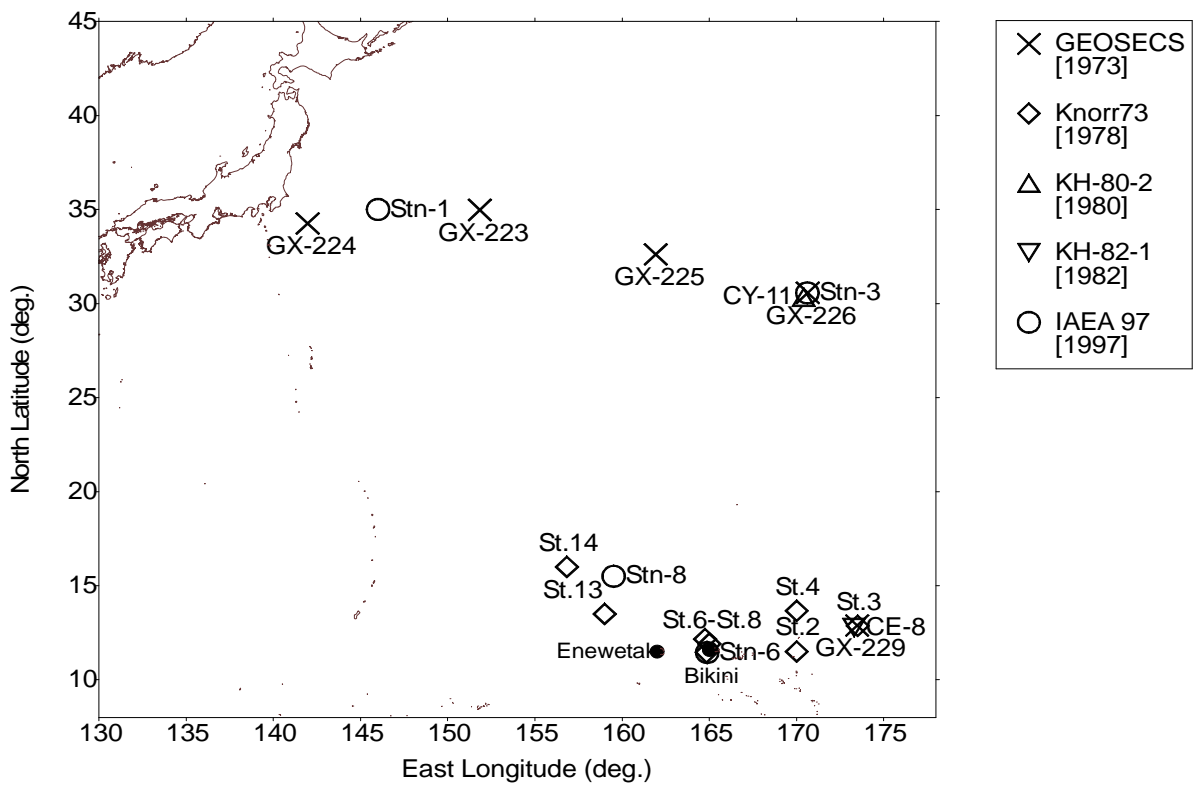


FIG. 42. GEOSECS (1973) [30], KNORR (1978) [82], Hakuho Maru (1980, 1982) [41, 80] and IAEA (1997) [67] sampling stations in the NW Pacific Ocean (data used for the evaluation of water profile time series).

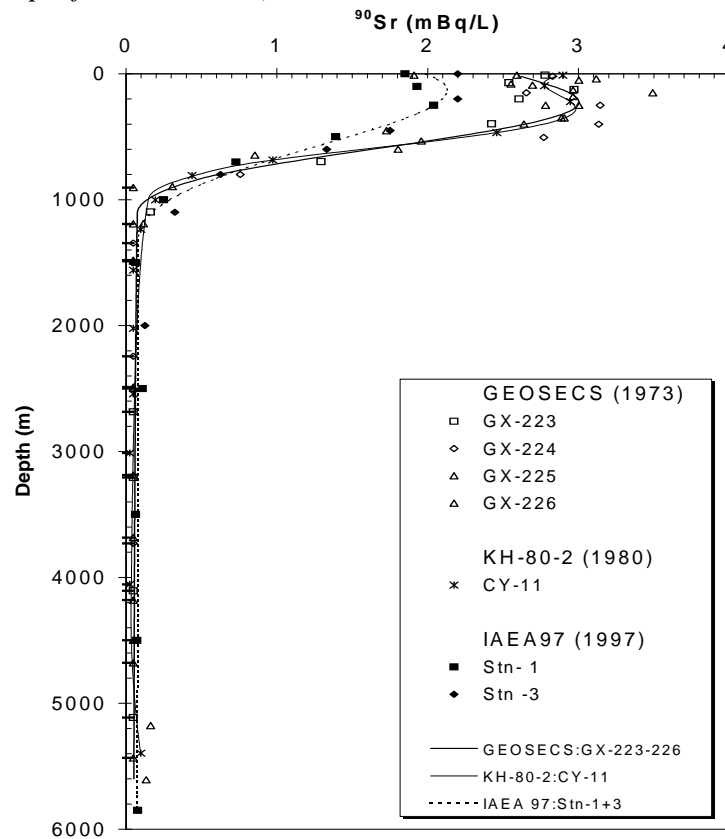


FIG. 43. ^{90}Sr profiles in the water column of the GEOSECS (1973) [30], Hakuho Maru (1980) [41] and IAEA (1997) [67] stations in the central NW Pacific Ocean.

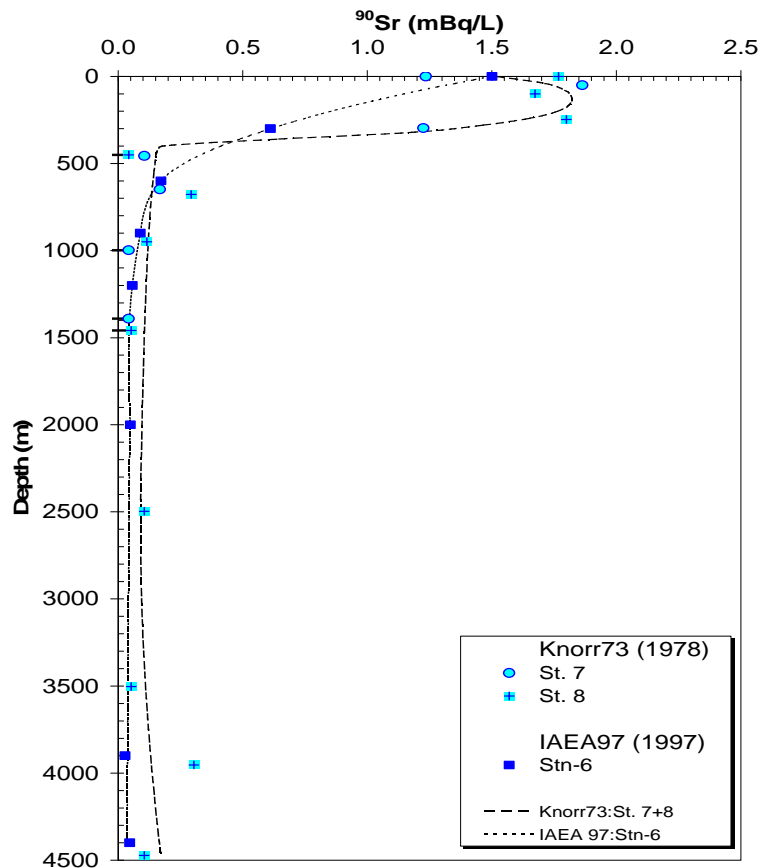


FIG. 44. ^{90}Sr profiles in the water column west of Bikini Atoll.

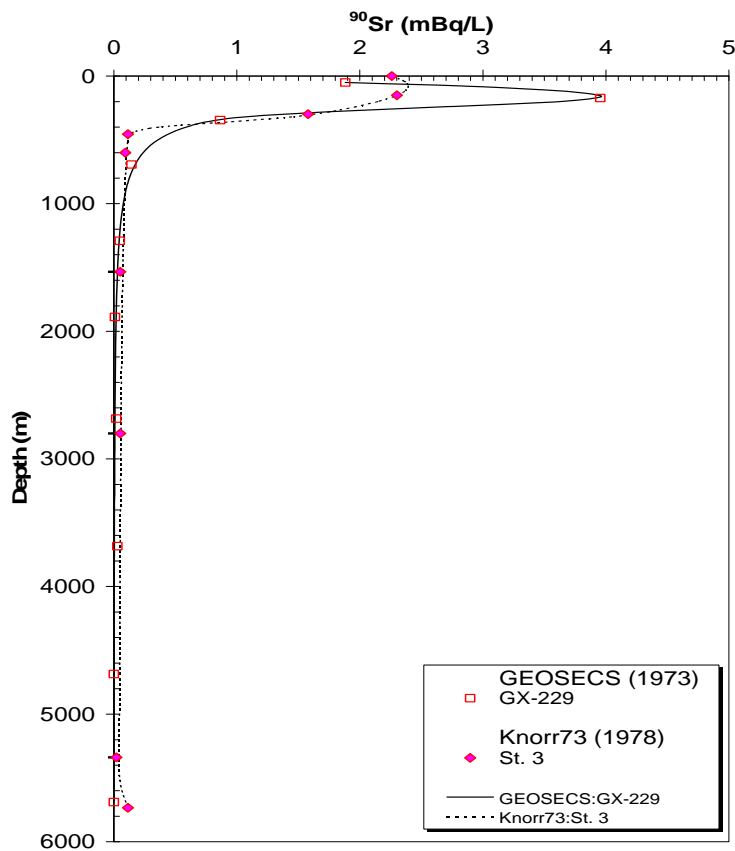


FIG. 45. ^{90}Sr profiles in the water column east of Bikini Atoll.

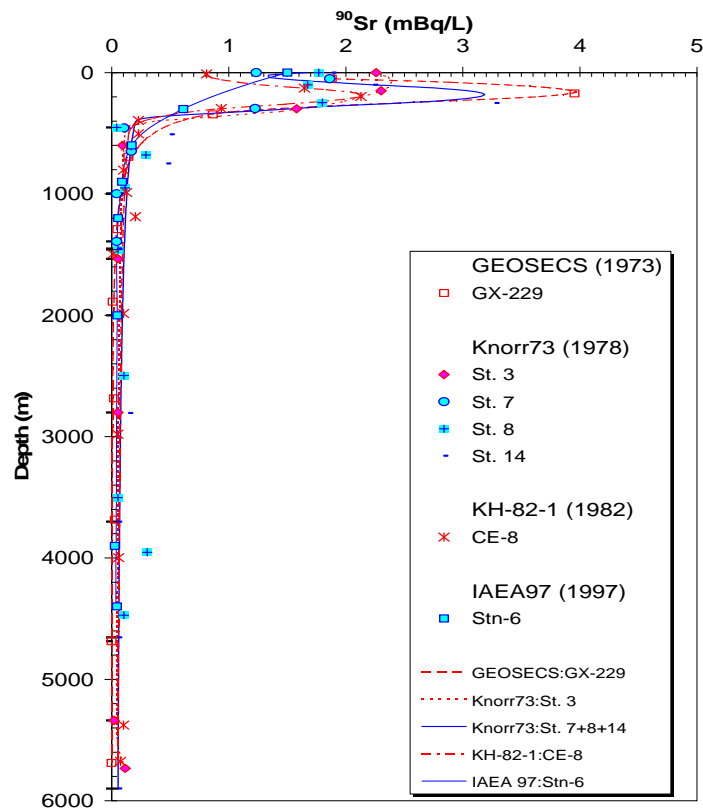


FIG. 46. A comparison of ^{90}Sr profiles in the water column of stations east (broken lines) and west (continuous lines) of Bikini Atoll.

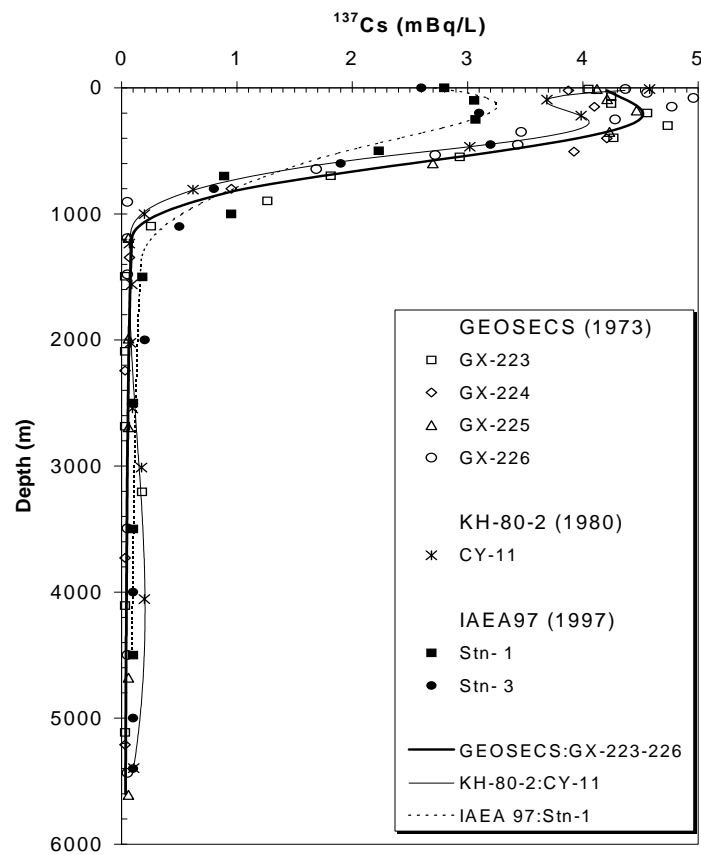


FIG. 47. ^{137}Cs profiles in the water column of the GEOSECS (1973), Hakuho Maru (1980) and IAEA (1997) stations in the central NW Pacific Ocean.

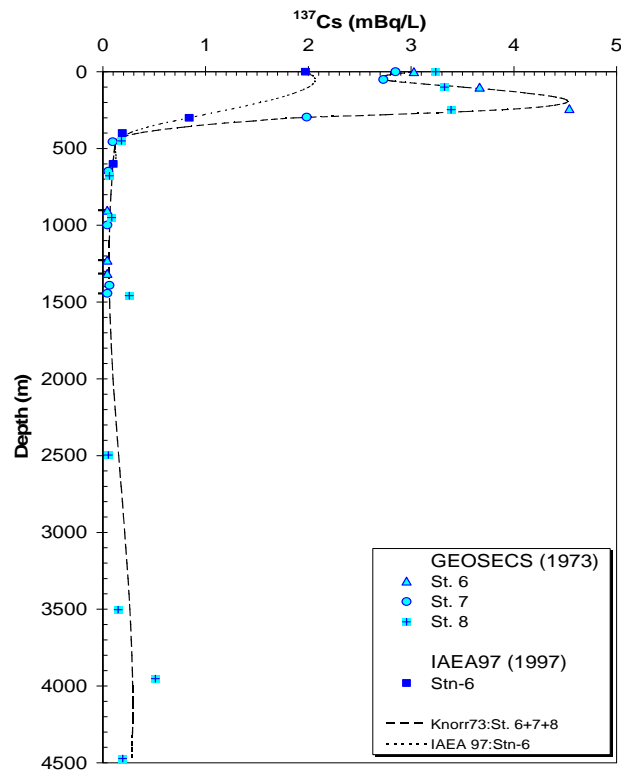


FIG. 48. ^{137}Cs profiles in the water column west of Bikini Atoll.

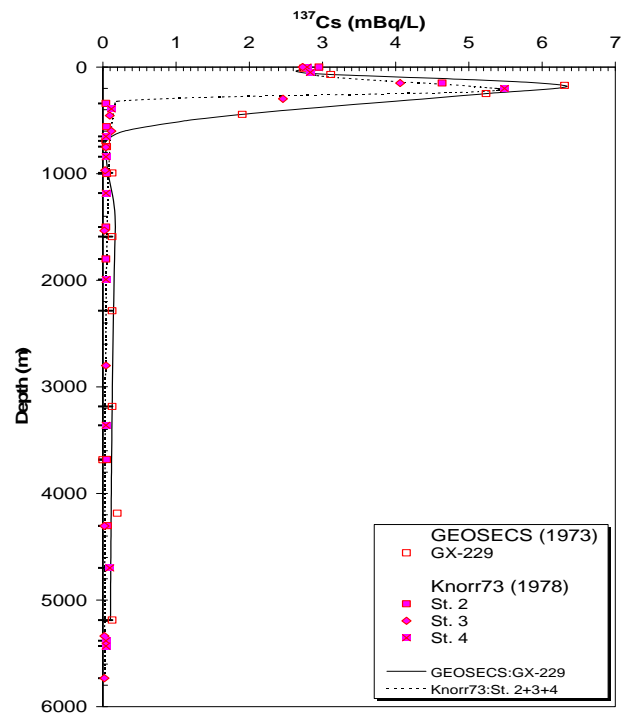


FIG. 49. ^{137}Cs profiles in the water column east of Bikini Atoll.

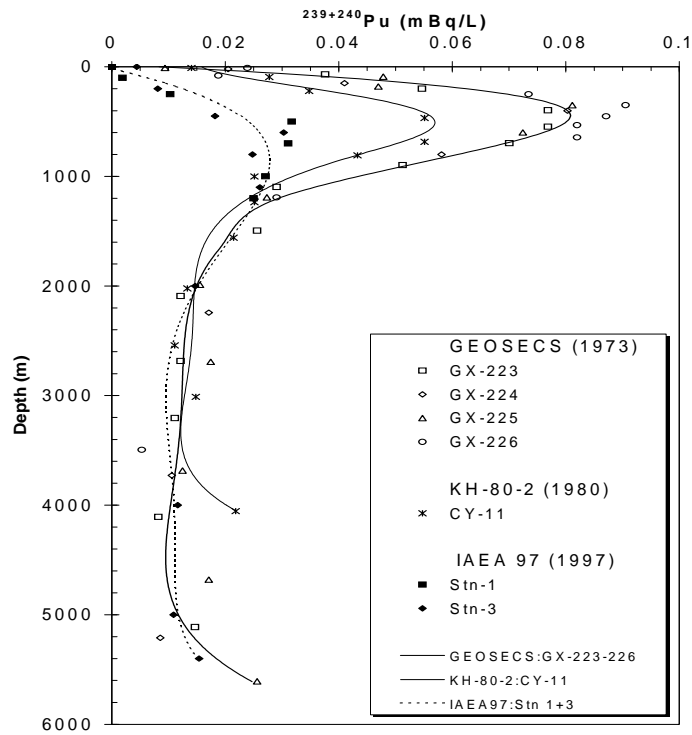


FIG. 50. A comparison of ^{137}Cs profiles in the water column east (broken lines) and west (continuous lines) of Bikini Atoll.

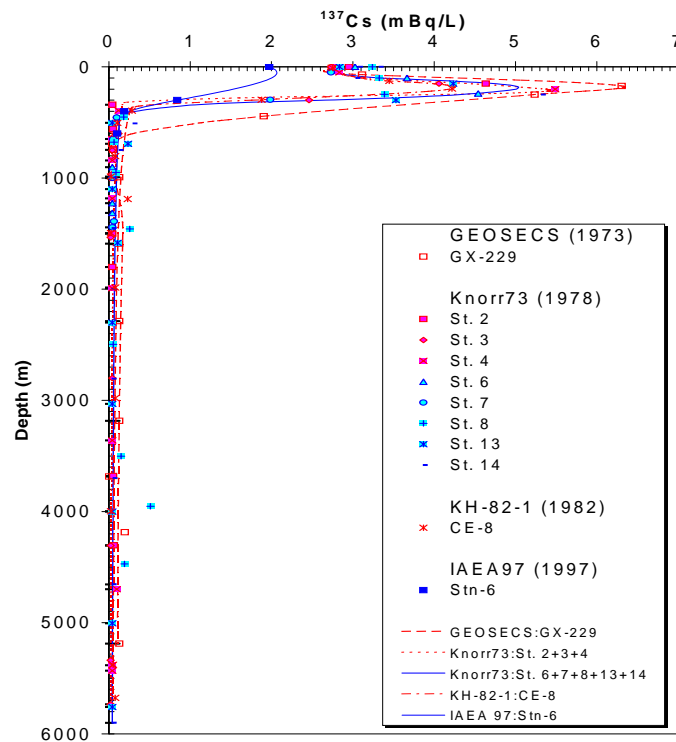


FIG. 51. $^{239,240}\text{Pu}$ profiles in the water column of the GEOSECS (1973), Hakuho Maru (1980) and IAEA (1997) stations in the central NW Pacific Ocean.

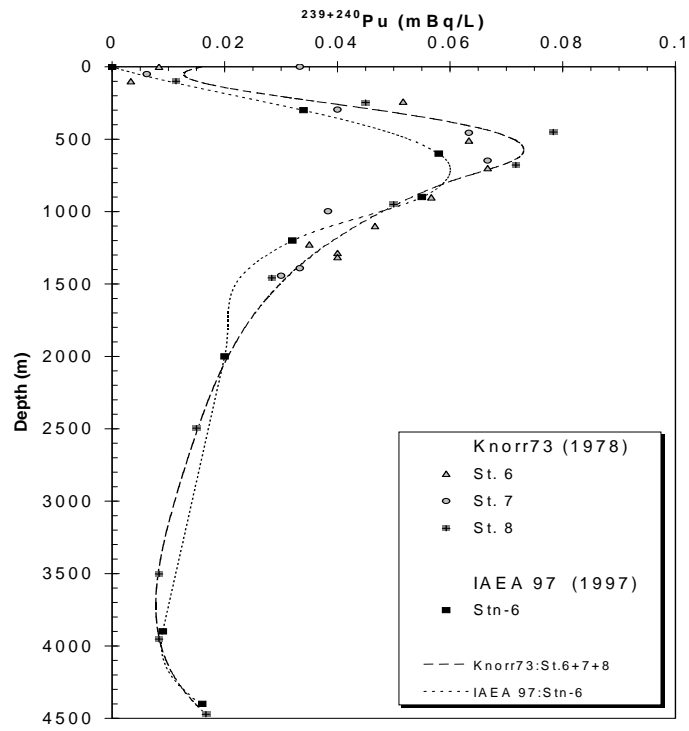


FIG.52. $^{239,240}\text{Pu}$ profiles in the water column of the KNORR(1978) and IAEA (1997) stations west of Bikini Atoll.

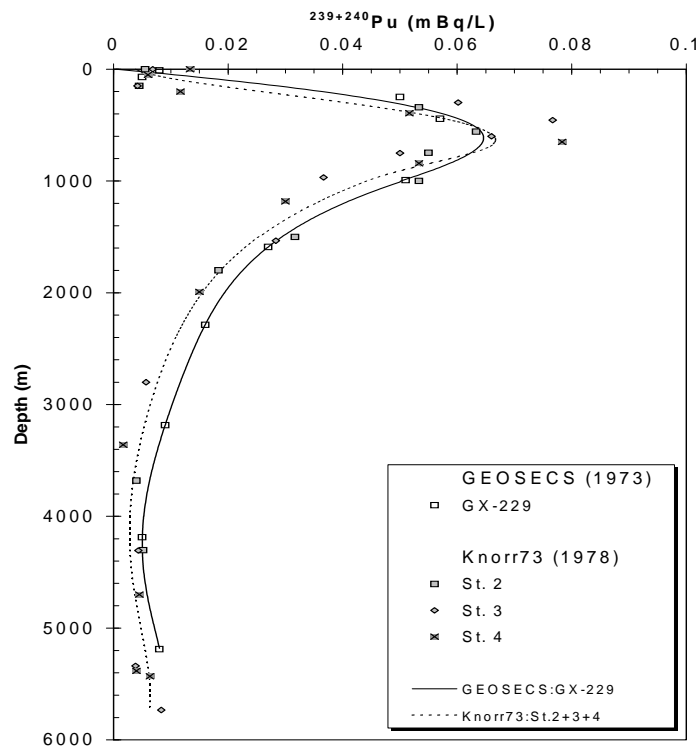


FIG. 53. $^{239,240}\text{Pu}$ profiles in the water column of the GEOSECS (1973) and KNORR (1978) stations east of Bikini Atoll.

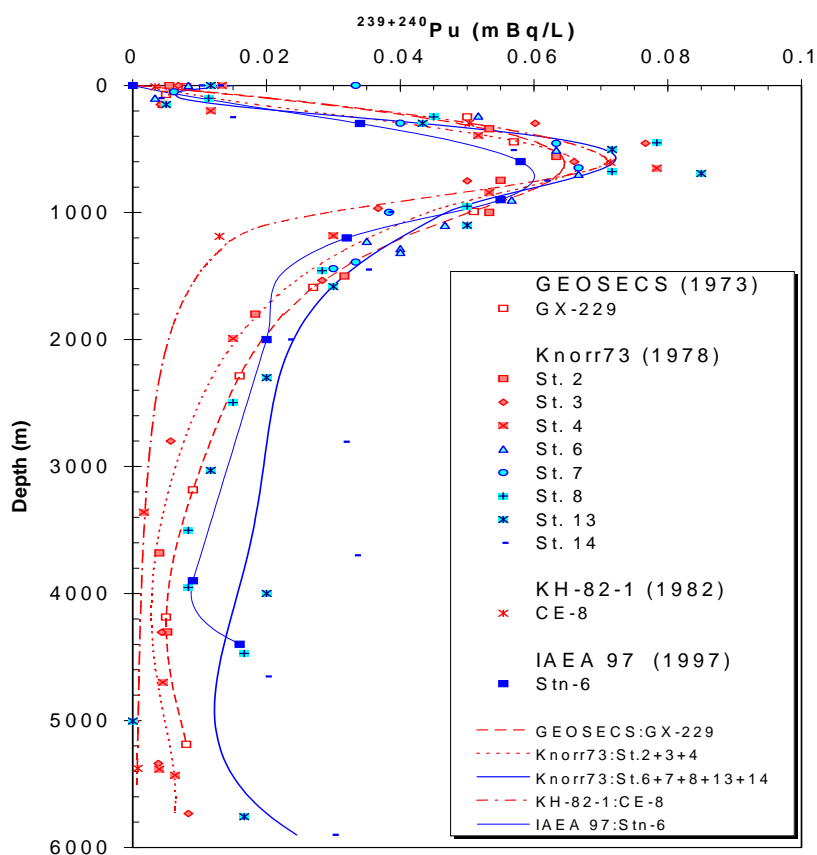


FIG. 54. A comparison of $^{239,240}\text{Pu}$ profiles in the water column of the stations east (broken lines) and west (continuous lines) of Bikini Atoll.

Recently, Hirose [49] developed a simple complexation-scavenging five-layer model to explain vertical $^{239,240}\text{Pu}$ profiles in the N Pacific Ocean. The transport rate constants for dissolved plutonium (as in the case of trace metals) in sea water are expressed as the partition coefficients of plutonium for particulate organic matter and Pu flux in sea water. The model implies that the pattern of the $^{239,240}\text{Pu}$ vertical profile is formed by the process of plutonium being released in inorganic form, following the decomposition of sinking particles by biogeochemical processes with depth. The model was tested using the results of sediment trap experiments [92]. The plutonium residence times estimated by the model for 0–250 m and 0–750 m depth layers are 10 and 33 years, respectively, in agreement with experimental observations. The model has successfully reproduced the vertical $^{239,240}\text{Pu}$ profiles in the water columns as well. However, there are some limitations of the model in connection with physical processes in the water column and chemical forms of plutonium. It has been shown by Nelson et al. [64] (see also [93] for a later review) that the chemical form of plutonium changes with water depth and this would imply that the plutonium partition coefficient will not be constant through the water column either. There are still many uncertainties in Pu aqueous chemistry and therefore, further work is required for a better understanding of plutonium behaviour in the water column. In particular, the role of physical processes needs to be defined.

Another typical feature of the $^{239,240}\text{Pu}$ water profiles in the NW Pacific Ocean shown in Figs 51 and 52 is an elevated $^{239,240}\text{Pu}$ concentration (by about a factor of two) observed in samples taken close to the sea-bottom. This feature has not been observed in water profiles

taken in the NE Pacific Ocean [30]. It has been shown by Buesseler [65] that higher $^{240}\text{Pu}/^{239}\text{Pu}$ ratios associated with tropospheric fallout are found in deeper and bottom water layers of the NW Pacific, as well as in surface sediment. This would indicate that the tropospheric fallout from the Marshall Islands is more rapidly removed from surface water to deeper water and to sediment, causing higher Pu concentrations and higher $^{240}\text{Pu}/^{239}\text{Pu}$ ratios in NW Pacific bottom water and sediment.

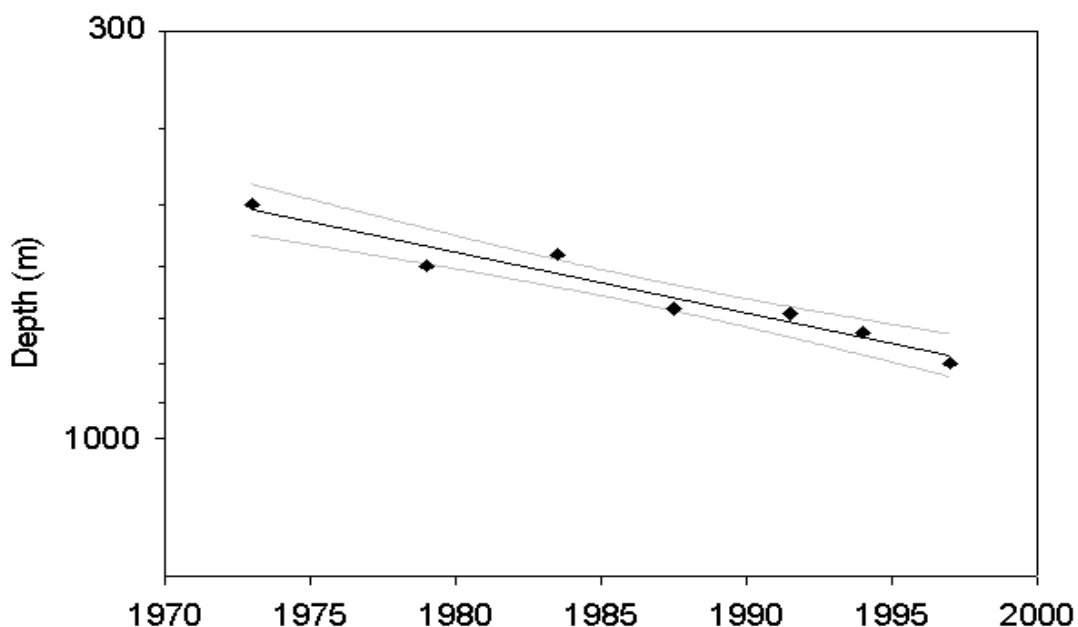


FIG. 55. Movement of a subsurface $^{239,240}\text{Pu}$ maximum in the water column with time (the dotted lines show the 95% confidence interval).

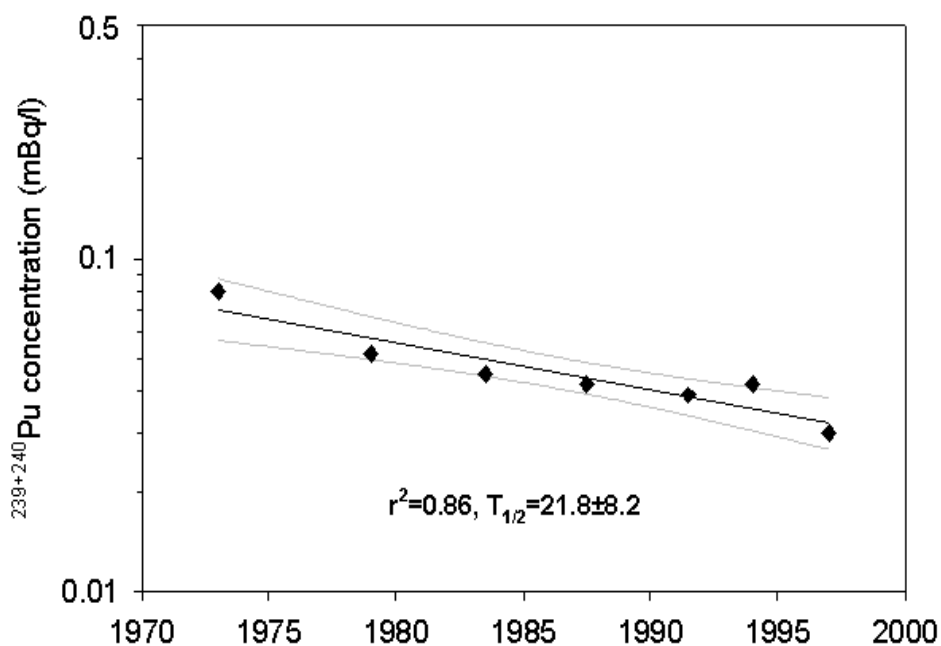


FIG. 56. Dependence of $^{239,240}\text{Pu}$ concentrations in the subsurface maximum with time.

4.6. Radionuclide inventories

4.6.1. Water inventories

The inventory of a radionuclide in a seawater column is calculated by interpolating its concentration measured at each depth:

$$I_w = \frac{1}{2} \left\{ \sum_{i=1}^N (W_{i+1} + W_i) (d_{i+1} - d_i) + 2 W_1 d_1 + 2 W_N (d_B - d_N) \right\} \quad (1)$$

where I_w is the radionuclide inventory in a seawater column (Bq/m^2), N is the number of sampling depths (the depth number is counted downward from the surface to the bottom), W_i is the radionuclide concentration in sea water at depth i (Bq/m^3), d_i is the i -th sampling depth of sea water (m), and d_B is the total depth to the bottom (m). When sea water at a sampling station is collected at the surface, the second term of the right-hand side of the above equation is not taken into account. If the total depth of a sampling station is not given, the third term is also omitted.

^{90}Sr and ^{137}Cs inventories in the water column were compared for GEOSECS and IAEA stations (Figs 43 and 47). As expected, the ^{90}Sr and ^{137}Cs inventories decrease with time from about 3.8 kBq/m^2 to 1.4 kBq/m^2 for ^{90}Sr , and from about 5.5 kBq/m^2 to 2.2 kBq/m^2 for ^{137}Cs .

A change of $^{239,240}\text{Pu}$ inventories in the water column with time has been evaluated for the central NW Pacific Ocean (between 20° – 40°N and 135° – 175°E) and the total water depth between 5000 and 6000 m using data stored in the GLOMARD. The averaged data show that the Pu inventory in water is slowly decreasing with time from about 130 Bq/m^2 to 100 Bq/m^2 (Fig. 57). The rate of decrease of the Pu inventory with time by a factor of two was estimated to be about 50 years, but it is necessary to keep in mind the physical process changes mentioned earlier.

It is clear that the inventories depend on the height of the water column and the geographical position of the sampling station. As the $^{239,240}\text{Pu}$ concentrations below 2000 m are almost constant, to exclude dependence on water depth, the geographical distributions of inventories were compared for 2300 m (the depth of the station east of Kamchatka). As shown in Fig. 58, $^{239,240}\text{Pu}$ water inventories normalized to the depth of 2300 m during the 1990s do not show any latitudinal dependence; they are within the interval of 35 – 53 Bq/m^2 , but the data density is very low. A higher inventory (74 Bq/m^2) observed in the area of the Marshall Islands is believed to be due to specific circulation processes as discussed earlier.

4.6.2. Sediment inventories

The input signal of ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ to NW Pacific Basin sediment should be viewed over time-scales of decades only due to the short delivery time since the introduction of anthropogenic radionuclides into the marine environment. Global fallout patterns show maxima in 1963–1964 corresponding to the weapons testing in the early sixties. Thus, anthropogenic radionuclide penetration due to sediment accumulation alone would be confined to millimetres [83]. The observed penetration depths in the order of centimetres imply that sediment accumulation is not important in governing radionuclide distribution. The stratigraphic record of the upper decimetre of recently deposited deep-sea sediment may be influenced by particle mixing by benthic fauna. Therefore, the depth distribution of anthropogenic radionuclides in bottom sediment has to be interpreted with caution. On the other hand, as sediment is the final depository of radionuclides in the sea, it provides important information on scavenging processes in the water column, on deposition histories of radionuclides and on accumulation processes occurring in sediments.

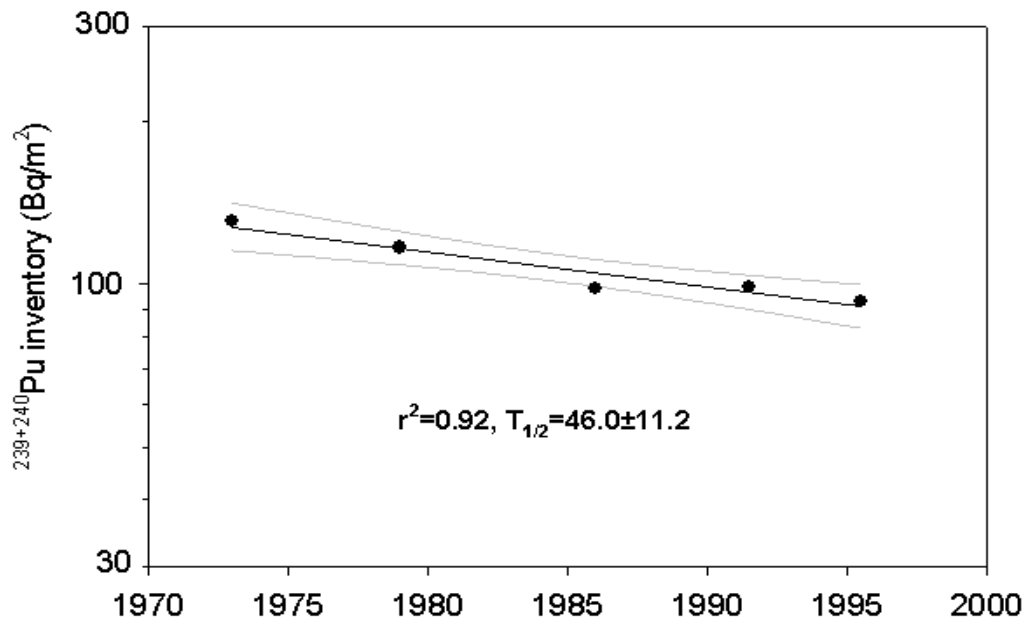


FIG. 57. Changes in $^{239,240}\text{Pu}$ inventories in the water column with time.

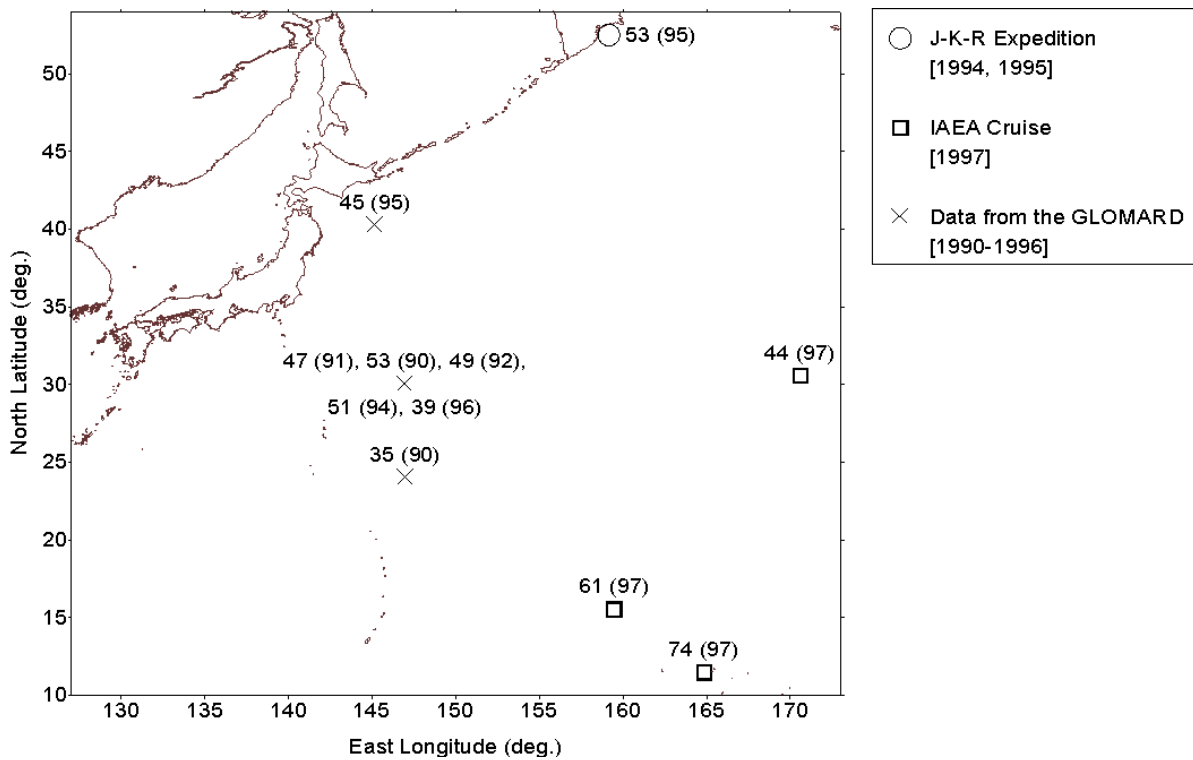


FIG. 58. $^{239,240}\text{Pu}$ inventories (Bq/m^2) in the water column (down to 2300 m) (the year of sampling is shown in parentheses).

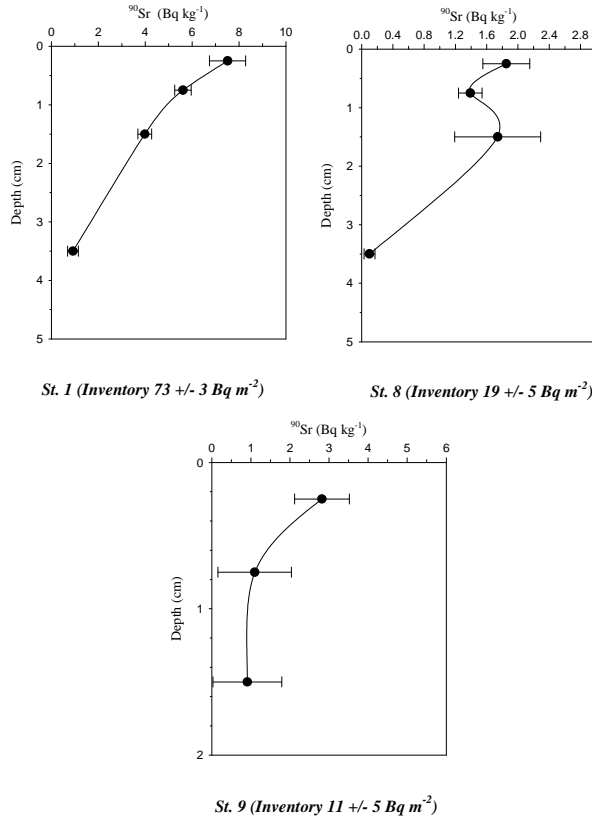


FIG. 59. Depth distribution of ⁹⁰Sr in bottom sediment of the NW Pacific Ocean (IAEA'97).

The data stored in the GLOMARD database show that radionuclide concentrations in sediment cores (usually cores up to a depth of 15 cm were analysed) of the N Pacific Ocean decrease with depth. Some of the undisturbed profiles however, have a peak at a few cm below the surface. The surface radionuclide concentrations are higher in sediment collected in the 1990s than in samples collected in the 1980s. The data show, however, a large variation in radionuclide concentrations between sites.

The inventory of a radionuclide in a sediment core is estimated by summing its concentration in each layer of sediment:

$$I_s = \sum_{i=1}^M S_i D_i (U_i - L_i) \quad (2)$$

where I_s is the radionuclide inventory in a sediment core (Bq/m^2), M is the number of sediment layers (the layer number is counted downward from the surface to the deeper layer), S_i is the radionuclide concentration in layer i (Bq/kg-dry), D_i is the density of layer i (kg-dry/m^3), U_i is the upper boundary of layer i (m), and L_i is the lower boundary of layer i (m).

4.6.2.1. ⁹⁰Sr and ¹³⁷Cs

Published sediment inventories have varied from 11 to 123 Bq/m^2 for ⁹⁰Sr and from 6.9 to 341 Bq/m^2 for ¹³⁷Cs in the NW Pacific Ocean, with low values near the Proving Grounds. Similar values for inventories of ¹³⁷Cs (4–111 Bq/m^2) were reported earlier north of 30°N by [38].

Sediment core samples were collected recently during the IAEA '97 NW Pacific Cruise and the KORDI cruise to the West Caroline Basin [96] (Table X). Depth distributions and

inventories of anthropogenic radionuclide concentrations in bottom sediment from the NW Pacific stations are presented in Figs 59 to 64, respectively.

The ^{90}Sr concentrations in the top 0–0.5 cm surface sediment are 7.5 ± 0.8 , 1.9 ± 0.3 and 2.8 ± 0.7 Bq/kg dw at IAEA stations 1, 8 and 9, respectively. Core top concentrations of ^{90}Sr show a latitudinal trend of increasing values away from the equator, that are similar to ^{137}Cs values in the region. Since ^{90}Sr is more soluble in sea water than ^{137}Cs , it participates in the biological cycle through the formation of celestite (SrSO_4) by acantharians as their structural component [95].

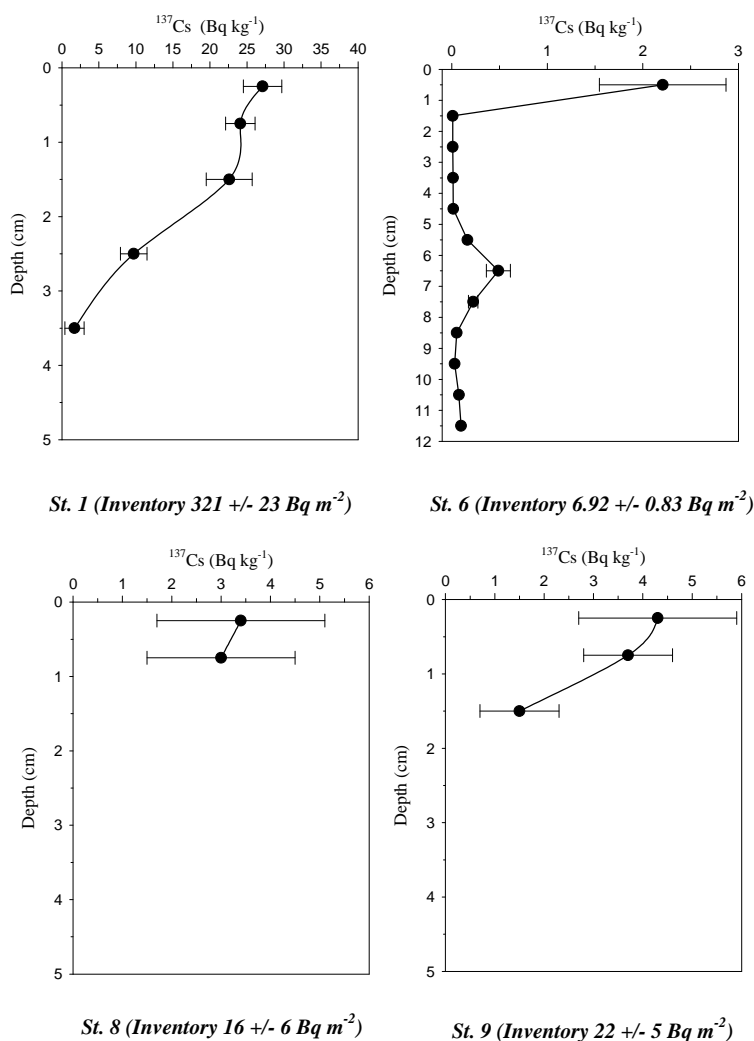


FIG. 60. Depth distribution of ^{137}Cs in bottom sediment of the NW Pacific Ocean (IAEA '97).

The ^{90}Sr distribution is more subject to regional biological influences than that of ^{137}Cs . The penetration depth of ^{90}Sr is 3–4, 0.5–1.0 and 1–2 cm below the sea-floor at stations 1, 8 and 9, respectively. The sediment inventory of ^{90}Sr was estimated based on ^{90}Sr concentration and sediment mass accumulation per unit area and is 72 ± 3 , 19 ± 5 and 11 ± 5 Bq/m 2 at stations 1, 8 and 9, respectively.

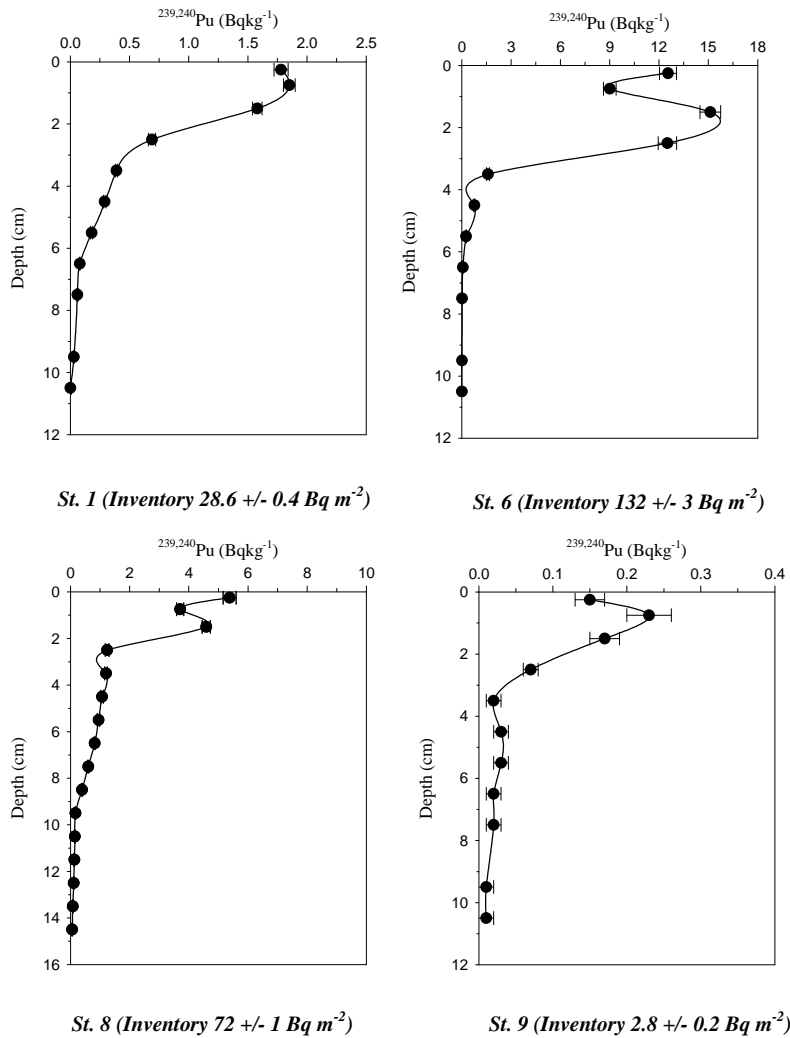


FIG. 61. Depth distribution of $^{239,240}\text{Pu}$ in bottom sediment of the Northwest Pacific Ocean (IAEA'97).

TABLE X. LOCATION OF SEDIMENT SAMPLING STATIONS FROM THE NW PACIFIC BASIN (IAEA) AND WEST CAROLINE BASIN (KORDI)

Station	Sampling date	Latitude	Longitude	Water depth (m)	Remarks
<u>IAEA '97 Pacific Ocean Expedition</u>					
1	24 Oct '97	35°00.065N	145°59.569E	5924	Red clay
6	10 Nov '97	11°27.288N	164°52.413E	4537	Carbonate ooze
8	13 Nov '97	15°30.177N	159°30.536E	5390	Red clay
9	16 Nov '97	22°22.010N	152°40.584E	5600	Red clay
<u>KORDI Cruise TOGA92</u>					
TEQ6	3 Dec '92	5°03.289N	137°01.277E	4629	Red clay
TEQ12	5 Dec '92	2°00.625N	137°00.036E	4157	Carbonate ooze

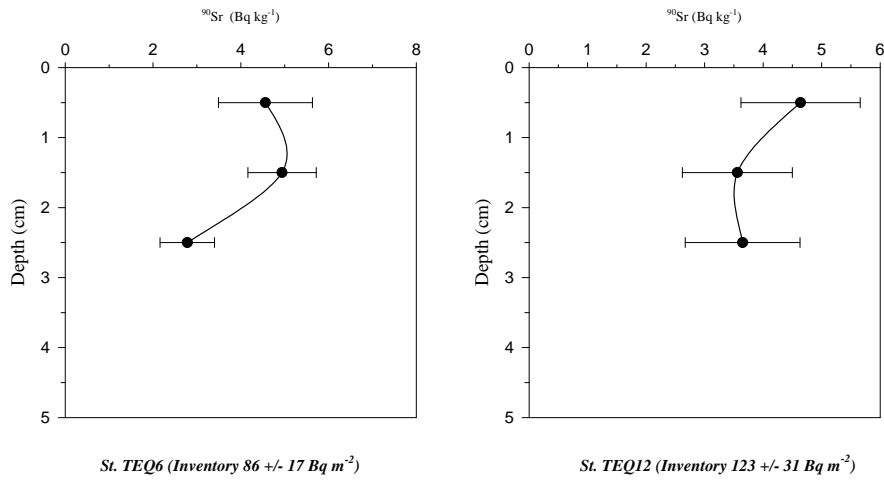


FIG. 62. Depth distribution of ^{90}Sr in bottom sediment of the West Caroline Basin (TEQ9212, 1992).

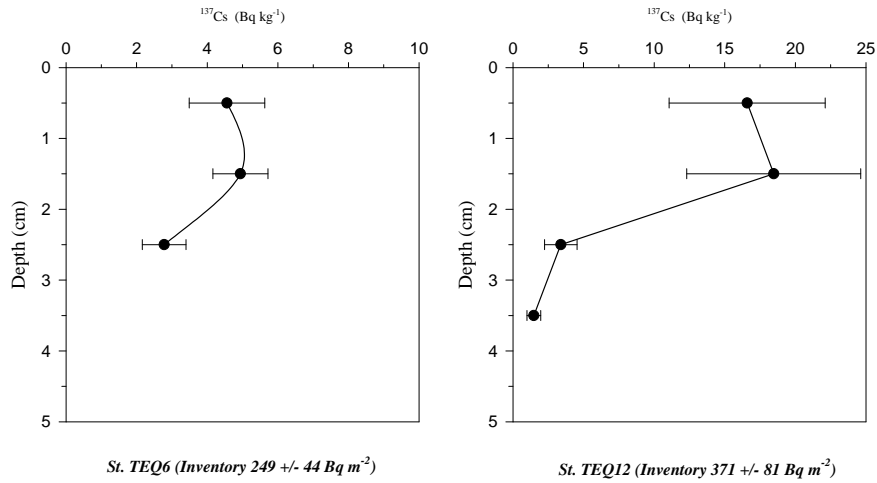


FIG. 63. Depth distribution of ^{137}Cs in bottom sediment of the West Caroline Basin TEQ9212, 1992).

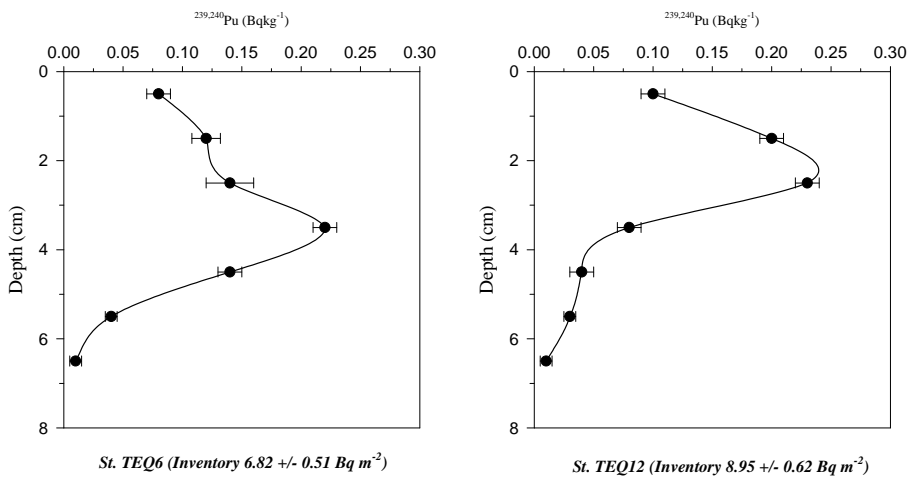


FIG. 64. Depth distribution of $^{239,240}\text{Pu}$ in bottom sediment of the West Caroline Basin TEQ9212, 1992).

In the West Caroline Basin, ^{90}Sr concentrations in the top 0–0.5 cm surface sediment are about 4.6 Bq/kg at stations TEQ6 and TEQ12. The penetration depth of ^{90}Sr is 3–4 cm below the sea-floor at stations TEQ6 and TEQ12. The sediment inventory of ^{90}Sr was estimated based on ^{90}Sr concentrations and sediment mass accumulation per unit area and is 86 ± 17 and 123 ± 31 Bq/m² at stations TEQ6 and TEQ12, respectively (Table XI).

TABLE XI. INVENTORIES OF ANTHROPOGENIC AND NATURAL RADIONUCLIDES IN NW PACIFIC BASIN BOTTOM SEDIMENT (Bq/m²)

Station	$^{239,240}\text{Pu}$	^{137}Cs	^{90}Sr
IAEA 1	28.6 ± 0.4	320 ± 20	73 ± 3
IAEA 6	132 ± 3	6.9 ± 0.8	
IAEA 8	72 ± 1	16 ± 6	19 ± 5
IAEA 9	2.8 ± 0.2	22 ± 5	11 ± 5
KORDI 6	6.8 ± 0.5	250 ± 40	86 ± 17
KORDI 12	8.9 ± 0.6	370 ± 80	120 ± 30

^{137}Cs concentrations in the top 0–0.5 cm surface sediment are 27 ± 3 , 2.2 ± 0.6 , 3.4 ± 1.7 and 4.3 ± 1.6 Bq/kg dw at IAEA stations 1, 6, 8 and 9, respectively. Core top concentrations of ^{137}Cs show a latitudinal trend of increasing values away from the equator, which is the opposite to $^{239,240}\text{Pu}$ in the region. The higher ^{137}Cs concentrations are generally confined to the core top and decrease with depth at all stations. The penetration depth of ^{137}Cs is 3–4, 7–8, 0.5–1.0 and 1–2 cm below the sea-floor at stations 1, 6, 8 and 9, respectively. The sediment inventory of ^{137}Cs was estimated based on ^{137}Cs concentration and sediment mass accumulation per unit area and is 320 ± 20 , 6.9 ± 0.8 , 16 ± 6 and 21 ± 5 Bq/m² at stations 1, 6, 8 and 9, respectively. The activity ratio of $^{239,240}\text{Pu}/^{137}\text{Cs}$ in the core top sediment is 0.07, 5.7, 1.58 and 0.93 at stations 1, 6, 8 and 9. This pronounced latitudinal trend, also reported in earlier studies [30, 83, 90] suggests that local fallout accounts for the high inventories of $^{239,240}\text{Pu}$ and that either less ^{137}Cs was delivered in this fashion (due to its noble gas precursor) or that more ^{137}Cs was released from sinking particles. Nevertheless, the ratios of $^{239,240}\text{Pu}/^{137}\text{Cs}$ at St. 6 still show higher values than the other stations. The direct effect of the test sites decreases with latitude; surface circulation patterns may produce elevated ratios far from the source. Also the interaction of Pu and Cs radionuclides with the sediment particles are different. The distribution coefficient (K_d) is 10^4 – 10^5 for Pu and 10^2 – 10^3 for Cs [94].

In the West Caroline Basin, the ^{137}Cs concentration in the top 0–0.5 cm surface sediment is 10.7 ± 3.6 and 16.6 ± 5.5 Bq/kg dw at stations TEQ6 and TEQ12, respectively. The penetration depth of ^{137}Cs is about 3 cm below the sea-floor at both stations. The sediment inventory of ^{137}Cs is 250 ± 40 and 370 ± 80 Bq/m² at stations TEQ6 and TEQ12, respectively (Table XI).

4.6.2.2. Plutonium

The $^{239,240}\text{Pu}$ inventories in sediment calculated for the N Pacific Ocean from GLOMARD data are shown in Fig. 65. As expected, large variations are observed in $^{239,240}\text{Pu}$ inventories. The $^{239,240}\text{Pu}$ inventory in sediment should grow slightly with time as more plutonium is scavenged onto sediment from the water column. However, it is difficult to

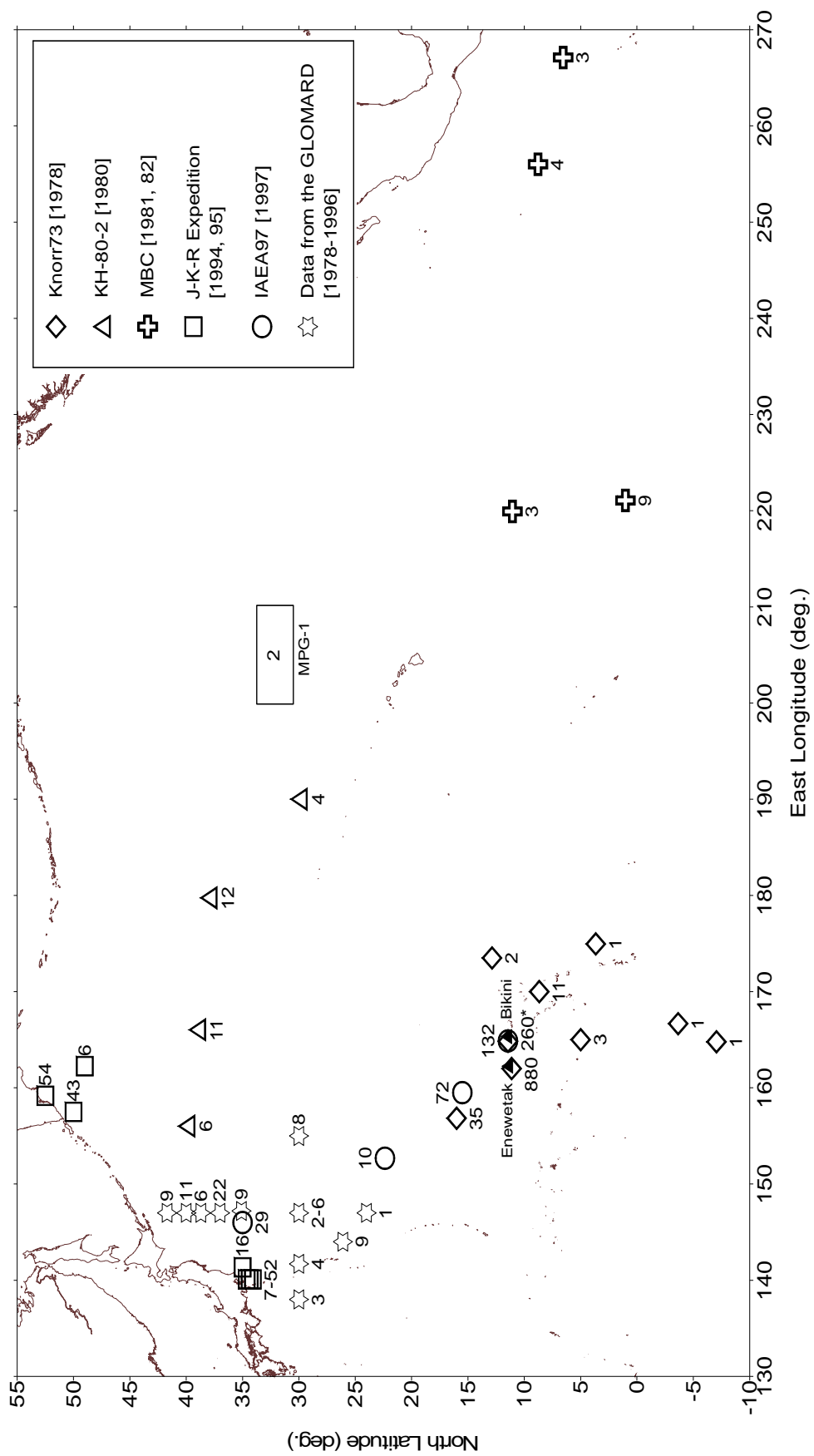


FIG. 65. Sediment $^{239,240}\text{Pu}$ inventories (Bq/m^2). (*The inventory at Bikini Atoll was calculated for 0–4 cm depth only).

establish any time trend because the data are very sparse and large differences have been observed even at stations in near proximity. The spatial distribution of Pu inventories suggests that the inventories decrease from the region of the Marshall Islands to the central NW Pacific (from about 100–900 to 10 Bq/m²), although the data density is very low. A decrease in inventories is also observed from the region of the Marshall Islands to the equator [82]. There is a clear indication that sediment inventories increase by a factor of three to five from the NE to the NW Pacific Ocean, reflecting different Pu sources in the region (the data from the Mid-Pacific gyre (MPG-1), KNORR, and other expeditions). This is in agreement with previous observations [30, 65] that the NW Pacific Ocean is affected by tropospheric fallout from the Marshall Islands area and its transport to the NW Pacific Ocean, while the NE Pacific Ocean shows only global (stratospheric) fallout effects. Cochran [83], analysing data from the MANOP sites (situated in the equatorial region east of the Christmas Islands) found decreasing inventories northwards from the equator and with longitude east as well. This may simply be the result of enhanced Pu removal in the higher-productivity waters centred on the equator in the Eastern Pacific. When the MANOP sediment data are compared with the water column inventory reported at the neighbouring GEOSECS station [30] this pattern fits well. The reported inventory (59 Bq/m²) is nearly ten times that expected from global fallout. These observations, plus the MANOP sediment data, plus their ²⁴⁰Pu/²³⁹Pu ratios (higher than fallout) observed by Buessler [65] seem consistent with a scenario whereby local fallout from Enewetak and Bikini is transported eastwards to the eastern Equatorial N Pacific in the strong N Equatorial Counter-Current and Pu is removed from the surface ocean in the flux of sinking particles from the productive zone on the equator.

A more detailed study of Pu in sediment from IAEA stations shows that in the top 0–0.5 cm core section, ^{239,240}Pu concentrations are 1.78±0.06, 12±1, 5.38±0.22 and 0.15±0.02 Bq/kg dw at IAEA stations 1, 6, 8 and 9, respectively. Core top concentrations of ^{239,240}Pu show a latitudinal trend of decreasing values away from the equator, since atmospheric weapon test sites are located on the equator (Enewetak and Bikini Atolls, 11°N, 162–165°E; Christmas Island, 2°2'N 157°40'W; Mururoa and Fangataufa Atolls, 22°S, 140°W). ^{239,240}Pu concentrations decrease with depth at all stations (Fig. 61). ^{239,240}Pu penetrated to the top 10 cm, top 6 cm, top 15 cm and top 7 cm below the sea-floor at stations 1, 6, 8 and 9, respectively. However, subsurface maxima were observed at stations 1, 6 and 9. ²³⁸Pu concentrations are 0.11±0.01, 0.43±0.10, 0.14±0.02 and 0.06±0.01 Bq/kg dw at stations 1, 6, 8 and 9, respectively. ²³⁸Pu was found in the top 4 cm below the sea-floor. The sediment inventory of ^{239,240}Pu was estimated based on the down core distribution of ^{239,240}Pu concentration and sediment mass accumulation per unit area and is 28.6 ± 0.4, 132 ± 3, 71.8 ± 0.9 and 2.8 ± 0.3 Bq/m² at stations 1, 6, 8 and 9, respectively. The inventories at stations 1, 6 and 8 in the NW Pacific are much higher than those of the NE Pacific, but Station 9 shows inventories similar to the NE Pacific [83]. The highest inventory found at St. 6 is due to local fallout. Cochran [83] reported ^{239,240}Pu concentrations in the core top 0–2 cm section as 0.3–0.5 Bq/kg and its inventory is 2.67 to >8.50 Bq/m² in the east Equatorial Pacific region (140°W–90°W, 1–12°N). However, it is similar to those found in the deep basin of the Sea of Japan [97] and in the NW Pacific off Kamchatka Peninsula [42, 44, 98]. The difference in sedimentary ^{239,240}Pu inventories between stations may be due to the latitudinal influence of local fallout and/or sediment accumulation rates and surface water circulation. Activity ratios of ²³⁸Pu/^{239,240}Pu in bottom sediment varied from 0.01 to 0.06.

In the West Caroline Basin (2–5°N), in the top 0–0.5 cm core section, maximum ^{239,240}Pu concentrations are 0.08±0.01 and 0.10±0.01 Bq/kg dw at stations TEQ6 and TEQ12, respectively. These values are much lower than the rest of the NW Pacific (IAEA '97), probably due to the isolation of the basin by its high topographic relief. The West Caroline

Basin is approximately 4000 to 5000 m deep and is semi-enclosed with rises near Sonsorol Island and the Caroline Islands as well as the Eauripik Rise in the east. These rises, which separate the West Caroline Basin from the western N Pacific, are approximately 2000–3000 m deep and allow Pacific deep water (approximately more than 2000 m depth) and intermediate water (approximately 1000–2000 m depth) to enter the Basin. In the north, the deep sill below approximately 4000 m is located along the Yap Mariana Trench (about 4000 to 8000 m deep), which connects the West Caroline Basin to the open western Pacific Ocean. This strait serves as the only conduit for Pacific bottom water (approximately several hundreds of metres above the sea-floor) to ventilate the West Caroline Basin [99]. Three major currents are important in the West Caroline Basin: the North Equatorial Current and its branch and the Equatorial Counter-Current. The North Equatorial Current is a westbound flow and is observed between 25°N and 5°N near 170–180°E in February, while in August it descends no lower than 10°N. On reaching the western end of the Pacific, the North Equatorial Current comes up against the continental barrier of the Philippines, where it divides into two branches. One branch turns to the south and feeds the eastbound Equatorial Counter-Current [100]. Depth distribution of $^{239,240}\text{Pu}$ concentrations shows subsurface maxima at 3–4 cm and 2–3 cm below the sea-floor. The sediment inventory of $^{239,240}\text{Pu}$ was estimated based on $^{239,240}\text{Pu}$ concentrations and sediment mass accumulation per unit area and is 6.82 ± 0.51 and 8.95 ± 0.62 Bq/m² at TEQ 6 and TEQ 12, respectively. ^{238}Pu concentrations are equal to or less than the analytical detection limit of 0.01 Bq/kg at both stations.

In order to facilitate comparison of the $^{239,240}\text{Pu}$ load at a given site with the different sources of input, the total $^{239,240}\text{Pu}$ inventories have to be calculated, i.e. the sum of the water column and sediment inventories. The $^{239,240}\text{Pu}$ water inventories constitute the greater part of the total inventories, the contributions usually being around 90% except for the station east of Kamchatka, where due to intense biological activity and high scavenging rates, the sediment contribution has reached 50% of the total inventory.

Compared to the estimated global fallout deposition densities [101] the observed total $^{239,240}\text{Pu}$ inventories in the central NW Pacific Ocean show a surplus of about 100%. Of course, after three decades of plutonium input from atmospheric nuclear bomb tests, the geographical distribution of plutonium in the oceans will not be at steady state condition, but will be affected by a complex of processes occurring in the marine environment. Horizontal advection, upwelling and downwelling of water masses and biogeochemical processes in the water column have played an important role in the development of the present distribution of plutonium in the ocean. A plausible explanation for the surplus, which is consistent with previous observations [30, 40, 42, 63, 65] may be the additional input from local fallout transported from the tropics to the N Pacific Ocean.

4.7. Radionuclides in the Far Eastern Seas

4.7.1. Sea of Japan

4.7.1.1. Surface water time series

Levels of anthropogenic radionuclides (^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$) in surface water of the Sea of Japan (Box 14) are evaluated based on the HAM database. The total number of ^{137}Cs , ^{90}Sr and $^{239,240}\text{Pu}$ concentrations in the HAM database are 445, 593 and 126, respectively. Some additional data have been added from the GLOMARD database.

Maps showing the sampling stations for surface water in the Sea of Japan for the period 1991–1995 and 1996–2000 are shown in Figs 66–68.

Time series of ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ during 1971–1998 are examined to estimate their concentrations in 2000. For ^{90}Sr and ^{137}Cs surface concentrations decrease exponentially, whereas surface $^{239,240}\text{Pu}$ concentrations show a slight change over the past two decades (Fig. 69). The surface concentrations of ^{137}Cs and ^{90}Sr in the Sea of Japan in 2000 are estimated to be 2.5 mBq/L and 1.4 mBq/L from the corresponding regression lines, respectively. The surface concentration of ^{90}Sr in the Sea of Japan in 2000 is similar to the level in the western N Pacific (Box 4). The surface $^{239,240}\text{Pu}$ concentration in the Sea of Japan in 2000 is calculated to be 5.2 $\mu\text{Bq/L}$, more than double the highest value estimated for each box in the Pacific. The higher concentration of surface $^{239,240}\text{Pu}$ is explained by rapid cycling of $^{239,240}\text{Pu}$ between the surface and subsurface layers, which is consistent with a small change in surface $^{239,240}\text{Pu}$ concentration [102].

The effective half-lives of ^{137}Cs and ^{90}Sr in surface water of the Sea of Japan are 19 and 14 years, respectively. The effective half-lives of ^{137}Cs and ^{90}Sr are similar to the mid-latitude region of the western N Pacific (Box 4). The result could imply that surface ^{137}Cs and ^{90}Sr in the Sea of Japan are controlled by the Tsushima Current, a branch of Kuroshio Current. The effective half-life of $^{239,240}\text{Pu}$ was estimated to be 16 ± 3 years, which seems to be too long in comparison with Box 4 (7 ± 1 year).

4.7.1.2. Vertical profiles

The vertical profiles of ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ in the Sea of Japan (the locations are given in Fig. 70) are shown in Figs 71–73. The ^{90}Sr and ^{137}Cs concentrations in the water column decrease exponentially with increasing depth [45, 103]. However, the gradient of the decrease rate with depth is smaller than that of the western N Pacific [104]. Especially the vertical profile of ^{137}Cs in the central northern region of the Sea of Japan [50] indicates that relatively high values were observed in the deep layer. This can be explained by physical processes such as vertical mixing in winter [105].

^{90}Sr and ^{137}Cs vertical profiles, which are mainly controlled by physical processes, indicate the penetration of these radionuclides into deep water as well. All these observations suggest that a large part of anthropogenic radionuclides is stored in deep water as vertical mixing in the Sea of Japan is more rapid than in the NW Pacific Ocean.

The vertical profile of $^{239,240}\text{Pu}$ in the Sea of Japan (Fig. 73) shows a surface minimum, subsurface maximum and decreases with increasing depth. Similar vertical profiles of $^{239,240}\text{Pu}$ were observed in the western N Pacific [30, 42, 67] and the Mediterranean [106]. However, the pattern of the vertical $^{239,240}\text{Pu}$ profile in the Sea of Japan is rather broad compared with that in the western N Pacific [102, 103,] which is consistent with the result obtained in the central north Sea of Japan [50]. This observation can be explained by the rapid recycling of plutonium between the surface and subsurface layers due to winter convection.

The higher surface $^{239,240}\text{Pu}$ concentrations found in the Sea of Japan in comparison with the NW Pacific Ocean (Fig. 51) could be explained by winter upwelling of medium-deep (below 500 m) Pu-enriched water [50]. The higher $^{239,240}\text{Pu}$ concentrations at the surface, a rather broad $^{239,240}\text{Pu}$ maximum and higher concentrations observed in deep water suggest recycling on a shorter time-scale than in the NW Pacific.

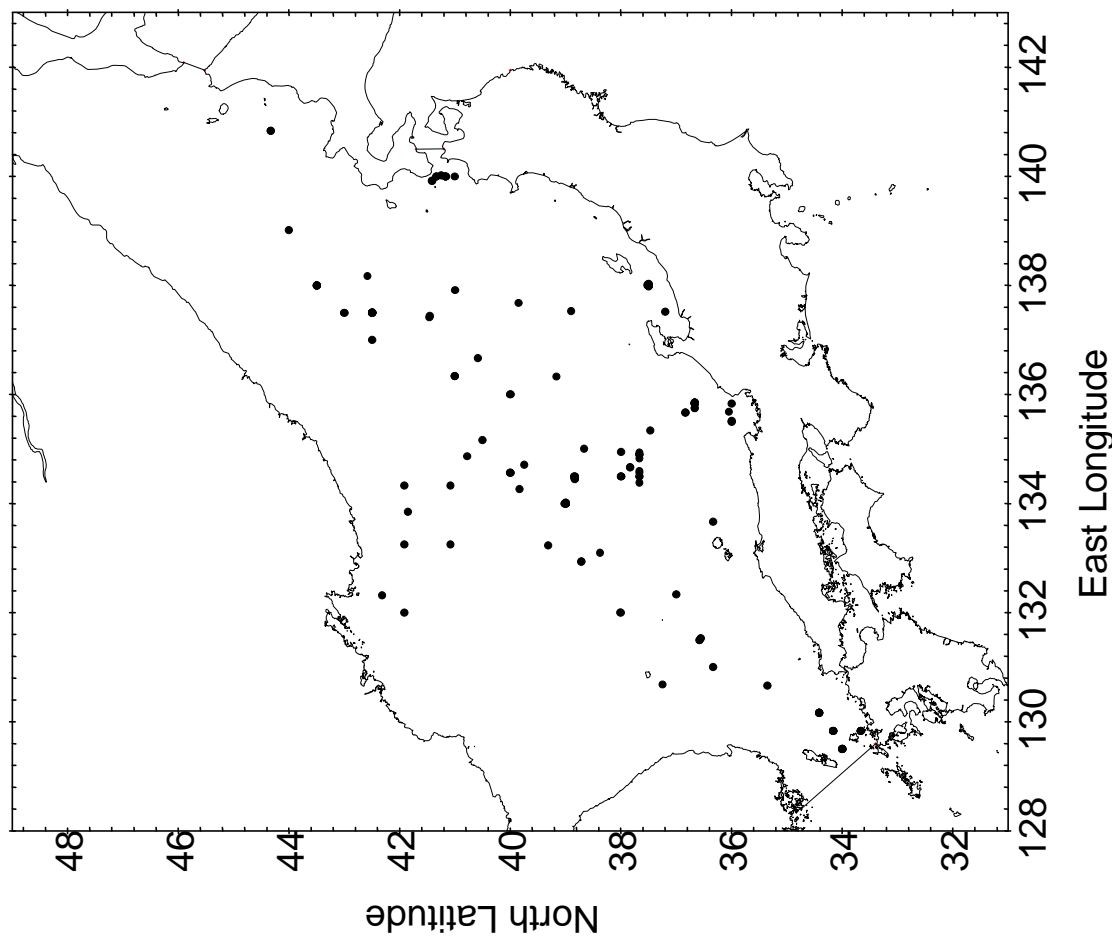
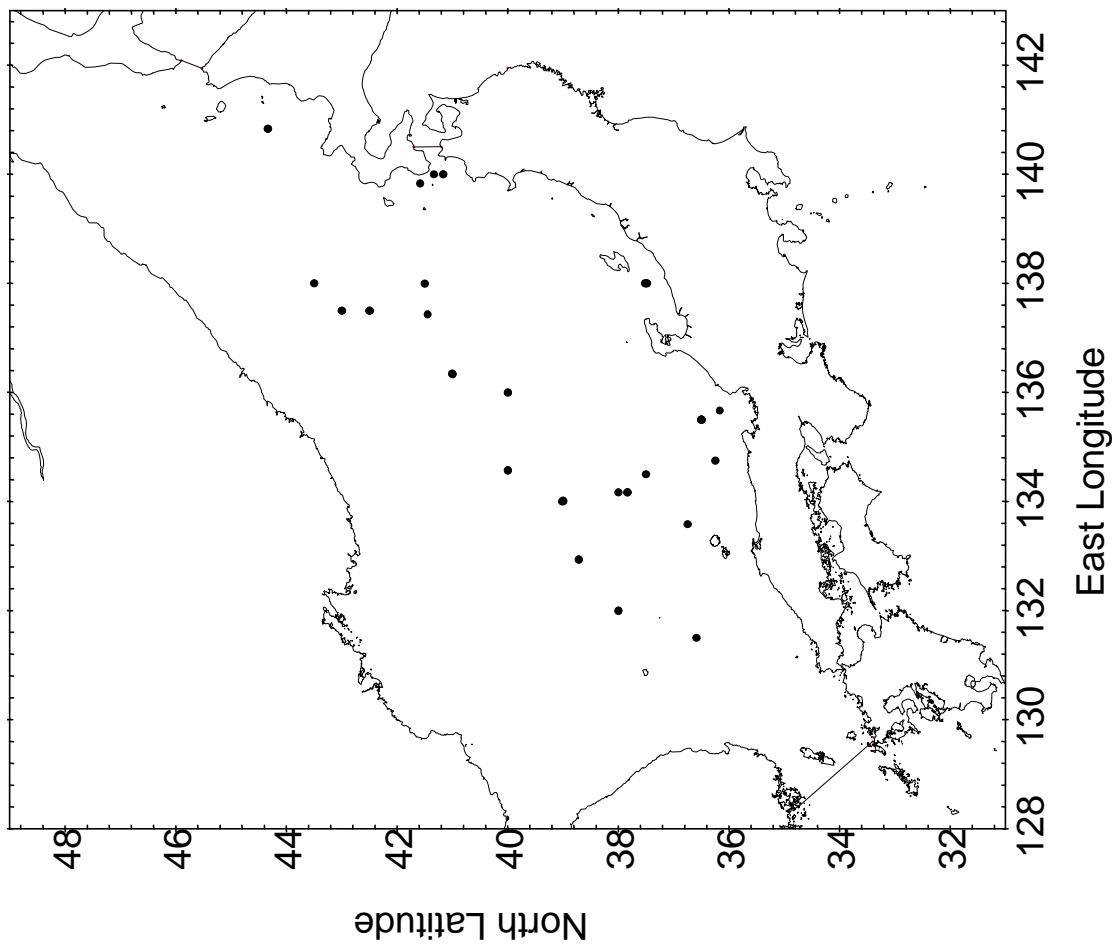


FIG. 66. ^{90}Sr sampling stations in the Japan Sea for the period 1991-1995 (left) and 1996-2000 (right).

For the $^{239,240}\text{Pu}$ inventory in water, a value of 0.12 kBq/m^2 has been obtained, which is higher by about a factor of 2 than expected from global fallout. This estimation is consistent with previous data discussed earlier, explaining the additional input from local fallout transported from the tropics to the N Pacific Ocean and specific oceanographic conditions in the Sea of Japan [50].

4.7.1.3. Sediment

The results for ^{90}Sr show very low concentrations, as expected, because only a small fraction of the ^{90}Sr input will settle with sinking particulates. The $^{137}\text{Cs}/^{90}\text{Sr}$ activity ratios are generally higher than the global fallout ratio.

$^{239,240}\text{Pu}$ concentrations in sediment differ significantly between sampling sites (as do concentrations of ^{137}Cs), the main reason being the higher removal rates in shallow water. Generally, $^{239,240}\text{Pu}$ concentrations in sediment at shallower water depths show higher values, as expected. $^{238}\text{Pu}/^{239,240}\text{Pu}$ activity ratios were the same as the global fallout ratio. The observed inventories are in the range $0.01\text{--}0.06 \text{ kBq/m}^2$ for ^{90}Sr , $0.05\text{--}0.4 \text{ kBq/m}^2$ for ^{137}Cs and $0.001\text{--}0.12 \text{ kBq/m}^2$ for $^{239,240}\text{Pu}$. The variation in the total water and sediment inventories between sampling sites is very small.

4.7.2. The Sea of Okhotsk

^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ concentrations in surface and bottom water show variations between the sampling stations depending primarily on the thickness of the water column. No vertical distributions were available for comparison with the Sea of Japan and/or NW Pacific data. Higher concentrations observed in bottom water could be explained by the high removal rates in productive shallow water. The $^{238}\text{Pu}/^{239,240}\text{Pu}$ activity ratios did not differ statistically from the values expected from global fallout. Similar conclusions could be drawn for $^{239,240}\text{Pu}$ concentrations in sediment. The data for shallow stations in the Sea of Okhotsk show significantly higher sediment inventories than in the Sea of Japan or the NW Pacific. However, the total inventories (water and sediment) did not differ considerably.

4.7.3. The Yellow and East China Seas

The Yellow Sea is a semi-enclosed continental shelf with a surface area of $380 \times 10^9 \text{ m}^2$ and an average depth of 44 m, and is surrounded by the contiguous land mass of China and the Korean Peninsula.

In surface water, dissolved ^{90}Sr concentrations varied from 1.97 to 2.58 mBq/kg with a mean value of $2.34 \pm 0.08 \text{ mBq/kg}$, generally lower in the coastal area than the open sea, and high values were observed in Kyonggi Bay, where the Han River discharges. ^{137}Cs concentrations varied from 1.78 to 3.38 mBq/kg with a mean value of $2.63 \pm 0.21 \text{ mBq/kg}$. The $^{239,240}\text{Pu}$ concentrations varied from 2.17 to 13.35 Bq/kg with an average concentration of $6.11 \pm 0.32 \text{ Bq/kg}$ for 1999 [107].

In bottom sediment, ^{90}Sr concentrations varied from 0.6 to 3.9 Bq/kg with an average of $2.4 \pm 1.8 \text{ Bq/kg}$ in the upper 2 cm layer. ^{137}Cs concentrations varied from 0.1 to 11.2 Bq/kg in the upper 1 cm layer of bottom sediment. $^{239,240}\text{Pu}$ concentrations varied from 0.1 to 0.9 Bq/kg in the upper 1 cm layer. ^{137}Cs and $^{239,240}\text{Pu}$ radionuclides are heavily enriched in the finer fraction of the sediment particle size [107].

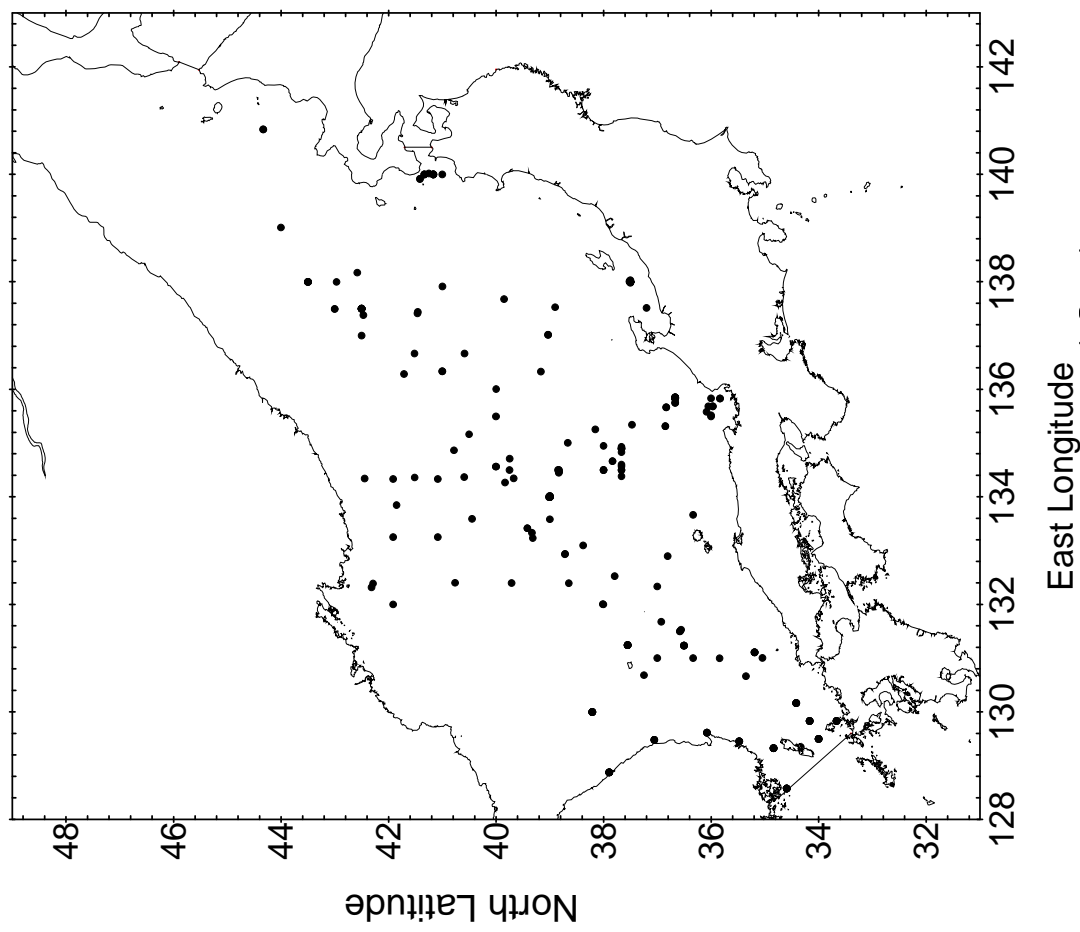
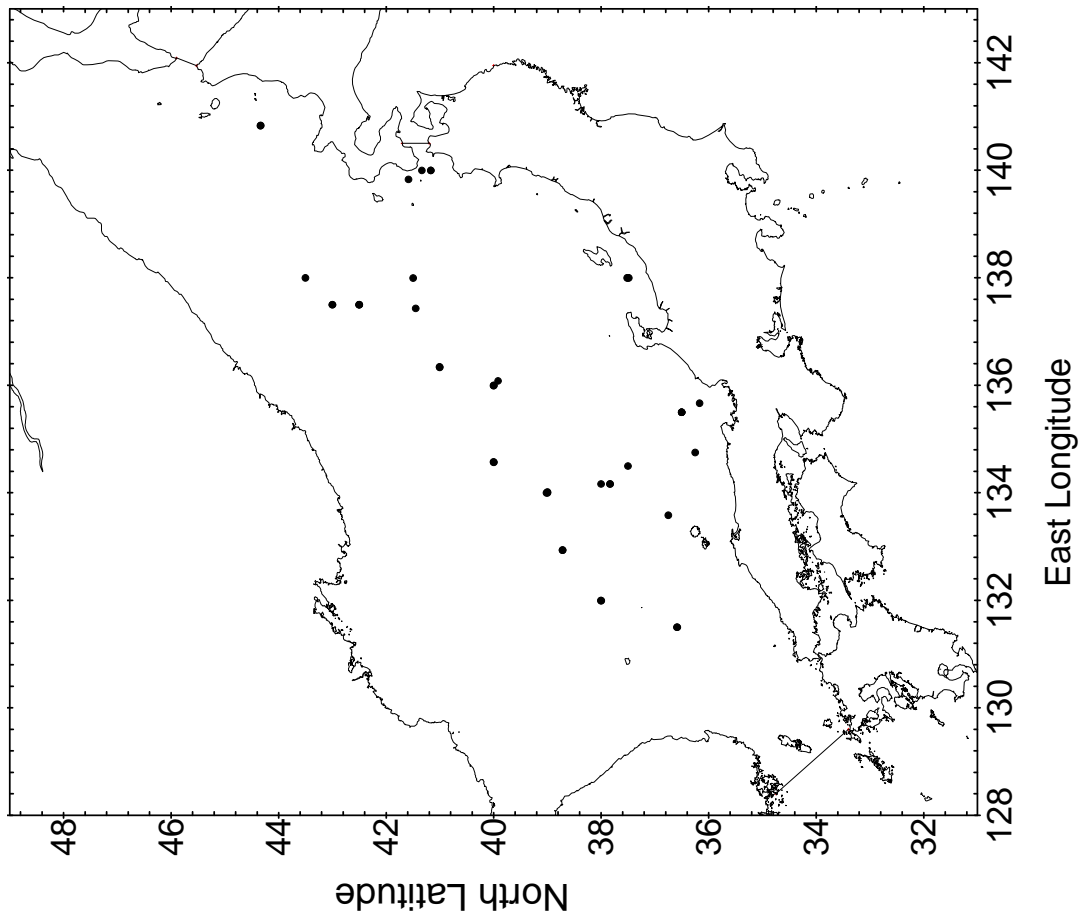


FIG. 67. ^{137}Cs sampling stations in the Japan Sea for the period 1991-1995 (left) and 1996-2000 (right).

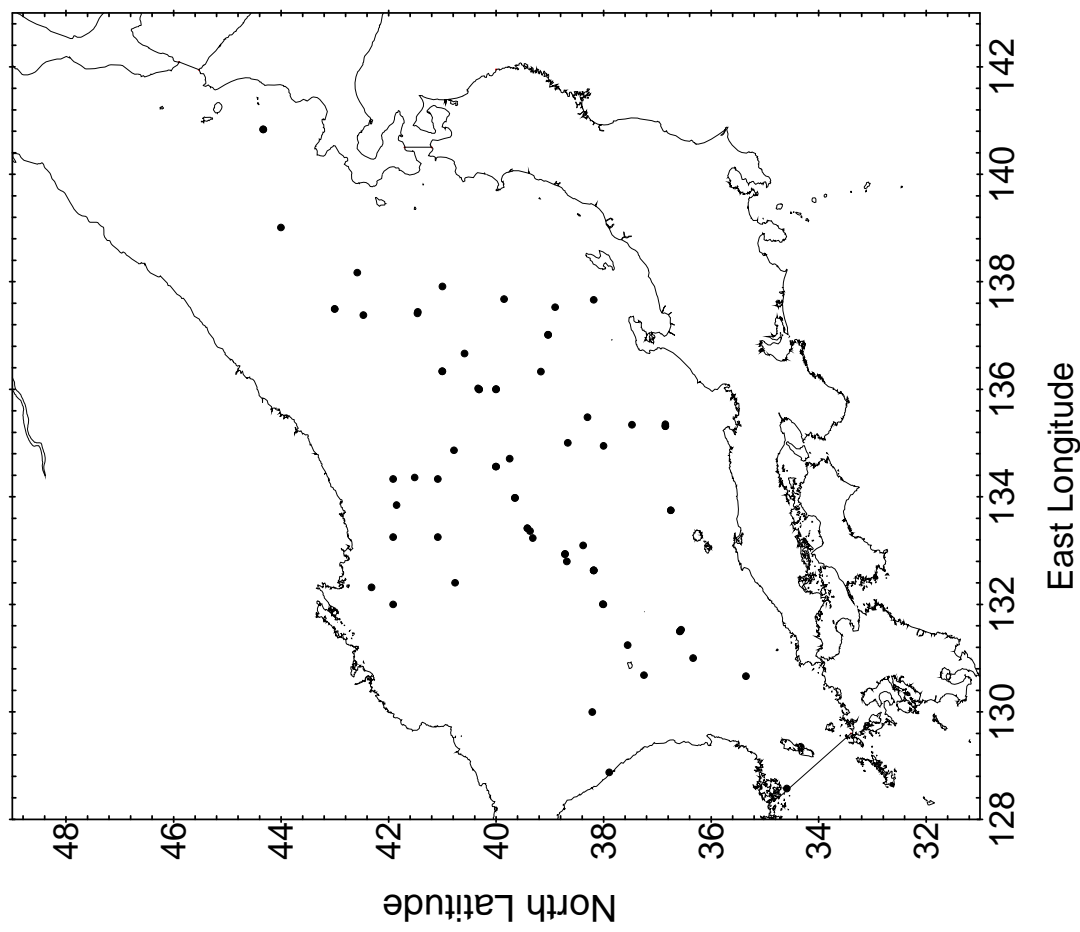
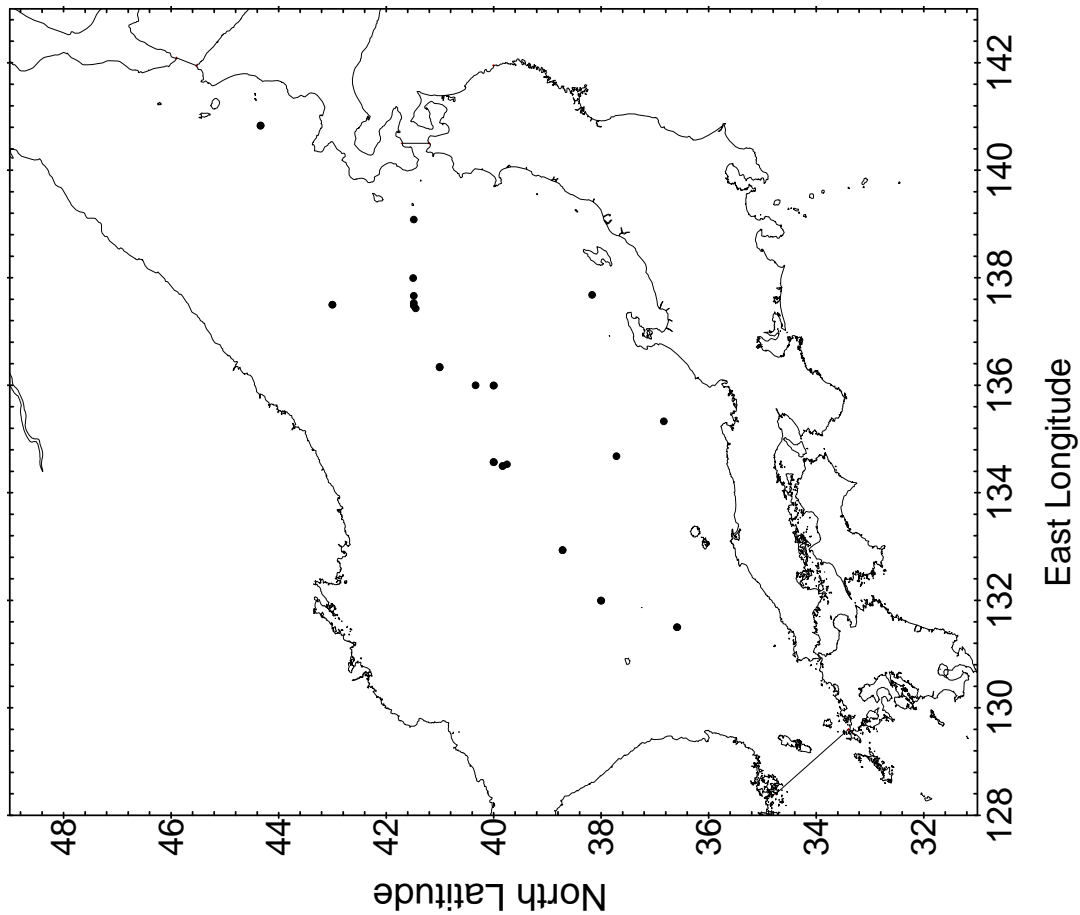


FIG. 68. $^{239,240}\text{Pu}$ sampling stations in the Japan Sea for the period 1991-1995 (left) and 1996-2000 (right).

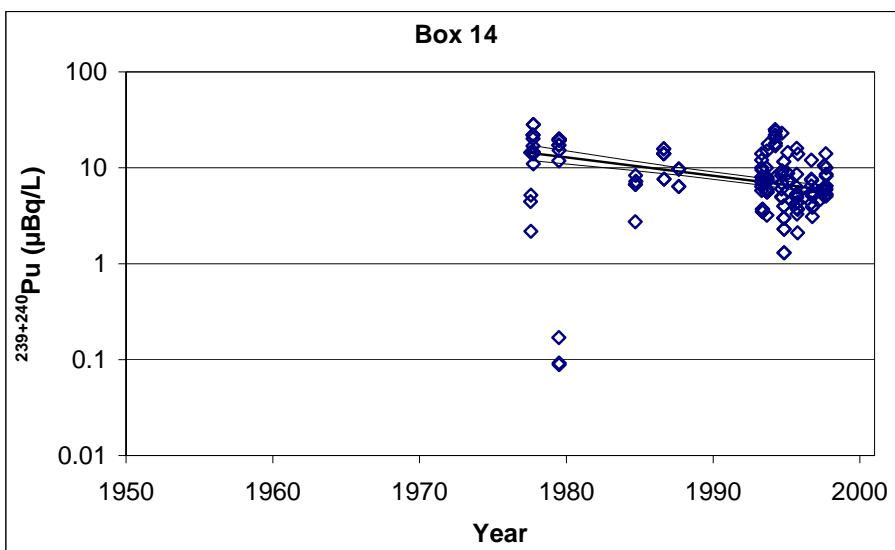
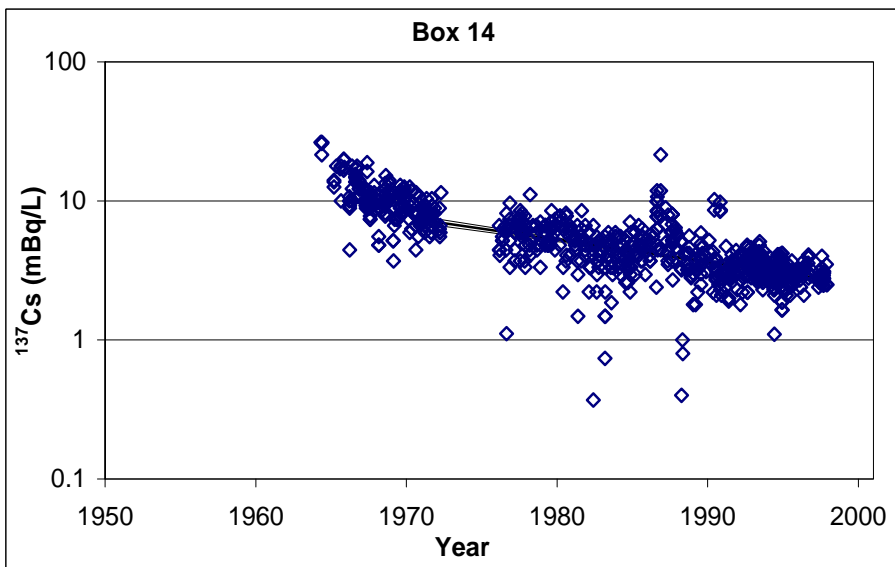
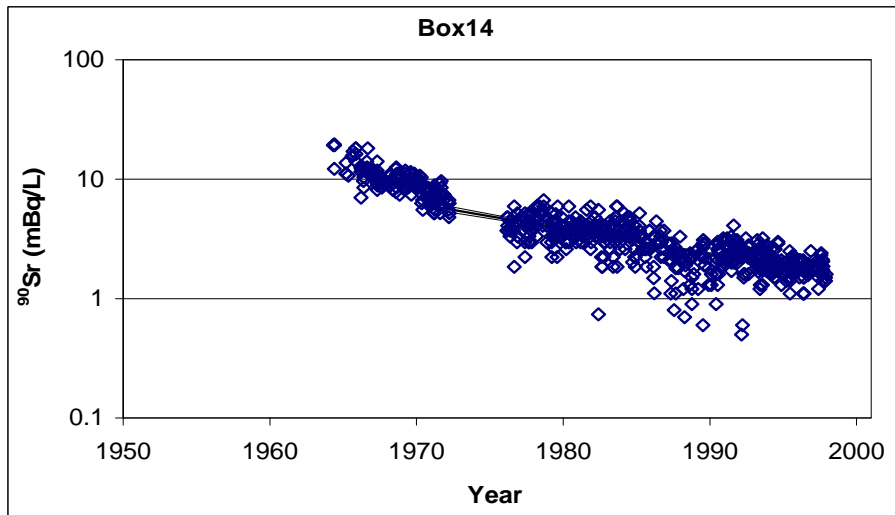


FIG. 69. ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ time series in surface water of the Sea of Japan.

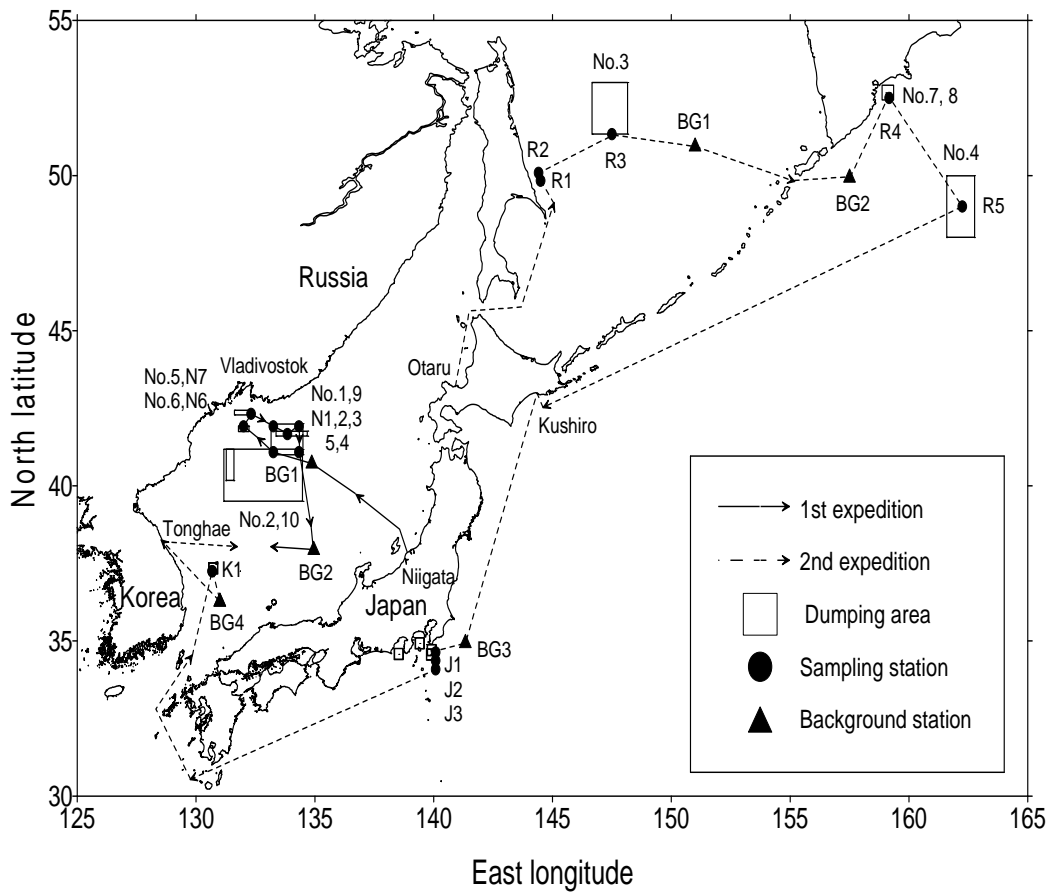


FIG. 70. Sampling stations in the Sea of Japan.

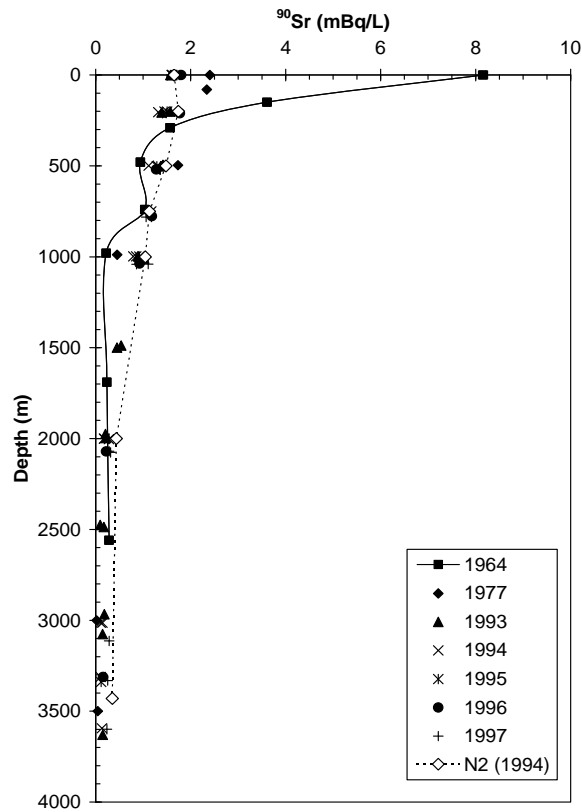


FIG. 71. ^{90}Sr vertical profiles in the Sea of Japan.

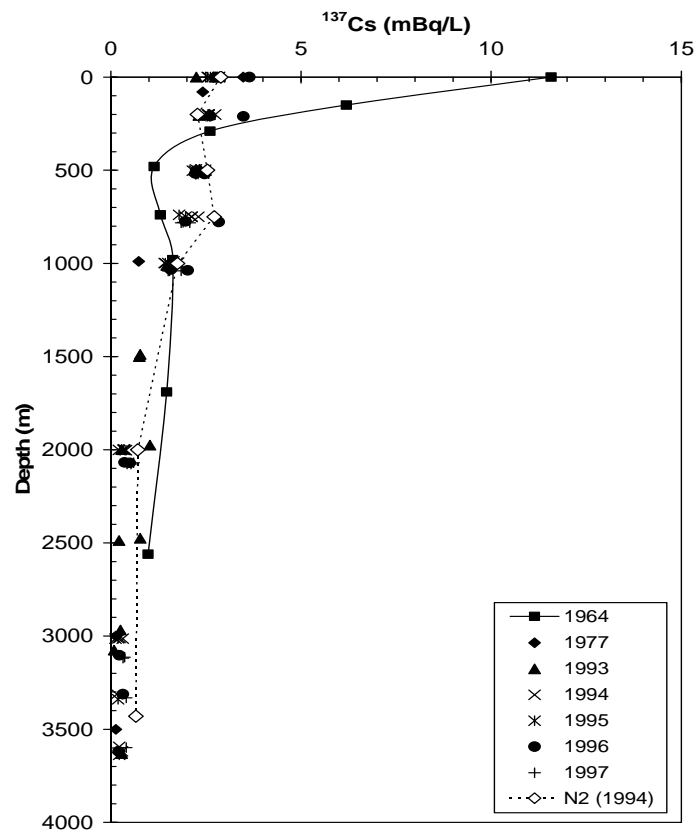


FIG. 72. ^{137}Cs vertical profiles in the Sea of Japan.

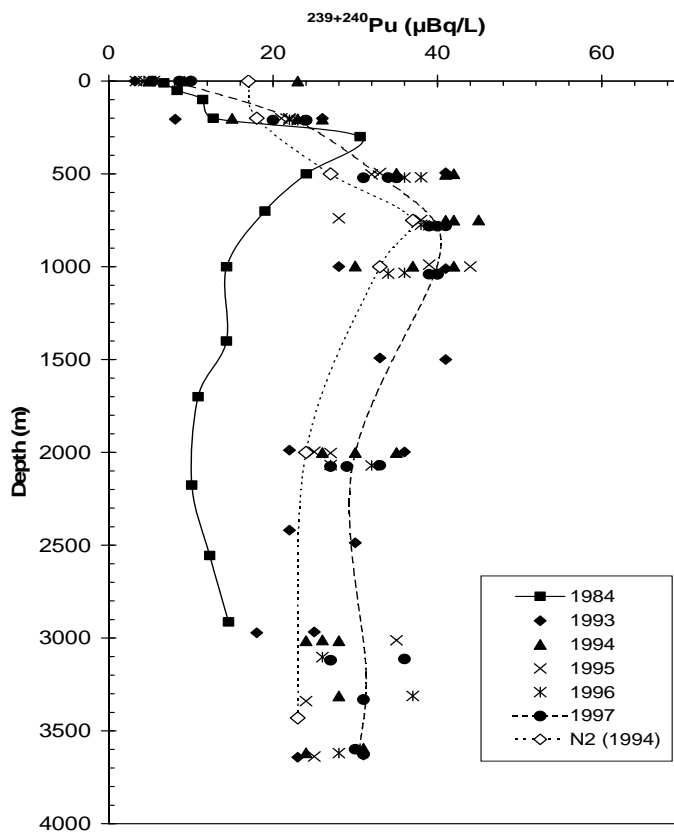


FIG. 73. $^{239,240}\text{Pu}$ vertical profiles in the Sea of Japan.

Unfiltered surface water in the East China Sea contained 2.5–3.0 mBq/kg ^{137}Cs , 1.9–2.8 mBq/kg ^{90}Sr and 3.8–6.3 Bq/kg $^{239,240}\text{Pu}$ in 1994 [107].

5. INDIAN AND SOUTHERN OCEANS

The Indian Ocean differs from the Atlantic and Pacific Oceans in its limited northward extent, to only 25°N. The southern boundary of the ocean is usually taken at the Subtropical Convergence at about 40°S. Its equatorial current system (Fig. 14) is strongly influenced by the seasonal variation in the winds north of the equator. From November to March these winds blow from the northeast (North East Monsoon), while from May to September they blow from the southwest (South West Monsoon). The South West Monsoon is a continuation across the equator of the South East Trade Winds that continue throughout the year. The change of wind direction north of the equator then results in a change of currents there. During the North East Monsoon season the North Equatorial Current (NEC) flows westward from 8°N to the equator; the Equatorial Counter-Current (ECC) flows eastward from the equator to 8°S and the South Equatorial Current (SEC) flows westward from 8°S to 15–20°S. During the South West Monsoon the flow north of the equator is reversed. This combines with the eastward ECC and the whole eastward flow from 15°N to 7°S becomes the Monsoon Current. The SEC continues to the west, south of 7°S but is stronger than during the North East Monsoon season.

During the North East Monsoon season, the SEC, when reaching the African coast, supplies both the ECC to its north and the Agulhas Current flowing south, close to the African shore. When it reaches the southern tip of Africa, the Current turns east into the Circumpolar Current. During the South West Monsoon, the part of the SEC turning north supplies the Somali Current up to the east coast of Africa. The SEC, the Somali Current and the Monsoon Current then comprise a strong wind-driven gyre in the Northern Indian Ocean. Strong upwelling occurs at this time along the Somali and Arabian coasts.

Along the eastern boundary of the Indian Ocean, off Western Australia, the Leeuwin Current flows poleward along the continental shelf break from about 22°S to 35°S. The Current then turns eastward. The Leeuwin Current is warm and of relatively low salinity, low dissolved oxygen and high phosphate content, and transports a significant amount of heat to the south.

Complex interaction between water masses takes place at frontal regions in the Southern Ocean (south of 40°S). Antarctic bottom water (AABW) has been shown to spread throughout the Southern and Northern hemispheres at 3000 to 4000 m water depth at frontal areas in mid-latitudes. Because of these oceanographic parameters, mid-latitudes in the Southern Ocean are high-nutrient low-chlorophyll zones (HNLC). The Southern Ocean region has also been documented as a major sink of CO_2 and is thus a highly productive area.

5.1. Radionuclides in surface water

Very few data have been reported to date on anthropogenic radionuclide concentrations in the Indian Ocean. The only comprehensive study was carried out in 1978 in the framework of the GEOSECS programme and included several vertical profiles of tritium in the water column [108].

There was a latitudinal trend of tritium concentrations, but, already in 1978, it did not reflect the fallout deposition in the area, but was influenced by water mass transport. Low

levels characterized the area below 40°S, while the highest concentrations were found between 30°S and 10°S. Towards the north, tritium concentration regularly decreased around the equator, to 10°N and levelled in the Arabian and Red Seas.

A few other data were obtained in [46] in the 1980s (^{137}Cs from 2.8 to 2.9 Bq/m³) and in [56] in 1994 when ^{137}Cs concentrations ranged between 1.6 and 2.3 Bq/m³, while ^{90}Sr ranged from 1.1 to 1.5 Bq/m³. Only 3 results were reported for plutonium isotopes. Again, the lowest levels characterized the area below 30°S.

The Indian Ocean together with the Southern Ocean was divided into 4 boxes (Fig. 15). Data for ^3H , ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ for the years 1991–1995 and 1996–2000 are plotted in Figs 74 to 77. There were only a few data for the first time interval, therefore a proper evaluation was not possible. For the second time period, thanks to the WOMARS project, more data have become available from cruises to the Arabian Sea, the Southern Ocean and the Indian Ocean transect. All investigated radionuclides show similar latitudinal trends with the smallest values in Box 13 (below 60°S) and the highest in the centre box between 10°S and 35°S (Box 16, Tables VI–VIII). The broken latitudinal dependence in radionuclide concentrations in surface water must be due to specific circulation patterns in the Indian Ocean.

In the framework of the WOMARS project, a sampling campaign was carried out in March–April 1998, along a track from New Zealand to the Mediterranean Sea (Fig. 78), to determine the present concentrations of anthropogenic radionuclides in surface water. The results for ^3H , ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ are shown in Figs 79–82 as a function of latitude. The latitudinal trend of radionuclide concentrations shows exactly the same trend as that of GEOSECS tritium: low concentrations of both ^{137}Cs and ^{90}Sr (1.1–1.8 Bq/m³ and 0.7–1 Bq/m³, respectively) characterize the Tasman Sea and the whole area south of Australia, at latitudes ranging between 40°S and 35°S. Proceeding northwest, all other stations in Box 16 show higher concentrations of both radionuclides (2.08–2.23 Bq/m³ for ^{137}Cs and 0.76–1.5 Bq/m³ for ^{90}Sr). Upwelling near the equator is probably responsible for the decrease in concentrations in the latitudinal band 10°S and 10°N.

The Arabian and Red Seas are characterized by similar radionuclide concentrations, in the range of 1.4–1.7 Bq/m³ for ^{137}Cs and 0.9–1.4 for ^{90}Sr . The mean ratio $^{137}\text{Cs}/^{90}\text{Sr}$ is 1.63 ± 0.19 (Fig. 83).

^3H data (Fig. 79) show the same latitudinal trend as the other radionuclides. The region south of Australia and the Arabian Sea have lower concentrations than other areas. The $^{137}\text{Cs}/^3\text{H}$ ratio is 0.019 ± 0.004 and the correlation coefficient is 0.74 ± 0.09 (Fig. 84). $^{239,240}\text{Pu}$ data in Fig. 82 show a similar latitudinal dependence to the other radionuclides.

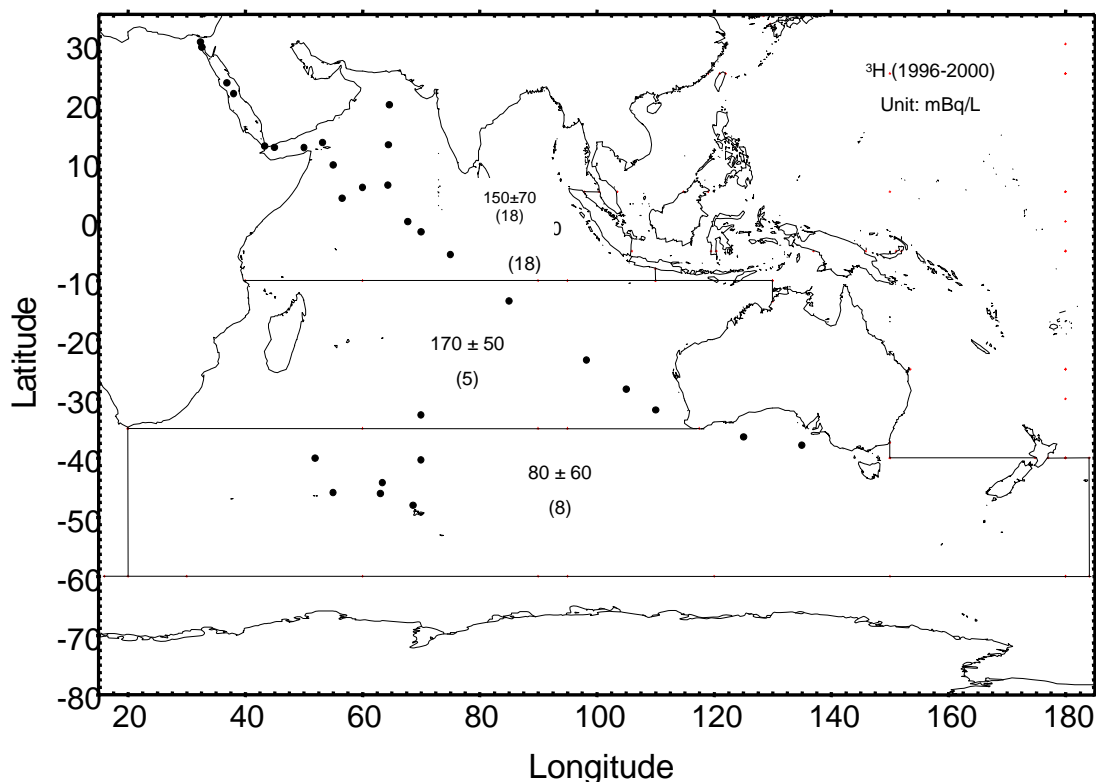


FIG. 74. ^3H in the Indian and Southern Oceans (no data for 1991–1995) (data corrected to 01.01.2000).

5.2. Surface water radionuclide time series

Surface water ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ concentration time series for boxes 15, 16 and 17 (Fig. 15) were evaluated using data stored in the GLOMARD database. Figs 85–87 show the data used for evaluation. The estimated effective half-lives of ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ in surface water for different latitudinal boxes are given in Tables VI–VIII. ^{90}Sr data were very sparse so any estimation of effective half-lives was not possible. The concentrations for the year 2000 were estimated from the few recent measurements available. ^{137}Cs data enabled to calculate the effective half-life which is around 20 years. The uncertainty in $^{239,240}\text{Pu}$ data was too high, except for Box 17 which gave an effective half-life of about 9 years.

When the data are grouped (Table XII), the effective half-lives for ^{137}Cs and $^{239,240}\text{Pu}$ are 20 ± 2 years and 9 ± 2 years, respectively.

5.3. Radionuclides in the water column

GEOSECS Stations 413 ($13^{\circ}21'\text{N}$; $53^{\circ}13'\text{E}$), 416 ($19^{\circ}45'\text{N}$; $64^{\circ}37'\text{E}$) and 417 ($3^{\circ}57'\text{N}$; $56^{\circ}30'\text{E}$) in the Arabian Sea (Fig. 88) were re-visited in 1998 to sample large volumes of water at different depths. While Station 413, east of the Gulf of Aden, is relatively shallow (bottom depth 2830 m), Station 416 in the north was 3210 m deep and Station 417 was the deepest (4114 m). ^{137}Cs data are presented in Figs 89–91. As there are no water profile data for ^{137}Cs from GEOSECS or other cruises, no direct comparison can be made for the Indian Ocean. However, it is possible to compare the ^{137}Cs profiles calculated from the measured GEOSECS ^3H profiles using the estimated $^{137}\text{Cs}/^3\text{H}$ ratio, as shown in Figs 89–91.

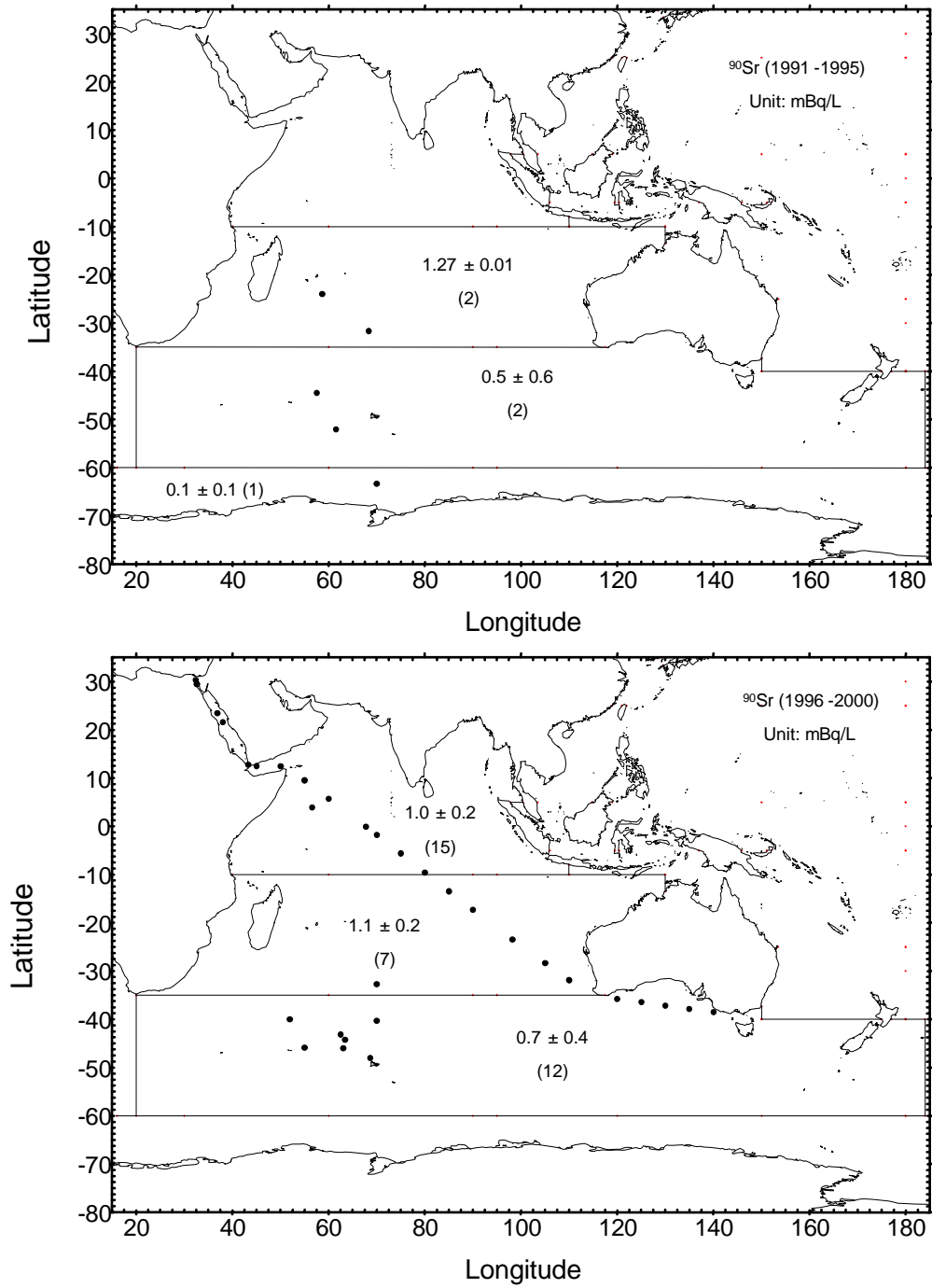


FIG. 75. ^{90}Sr in the Indian and Southern Oceans (data corrected to 01.01.2000).

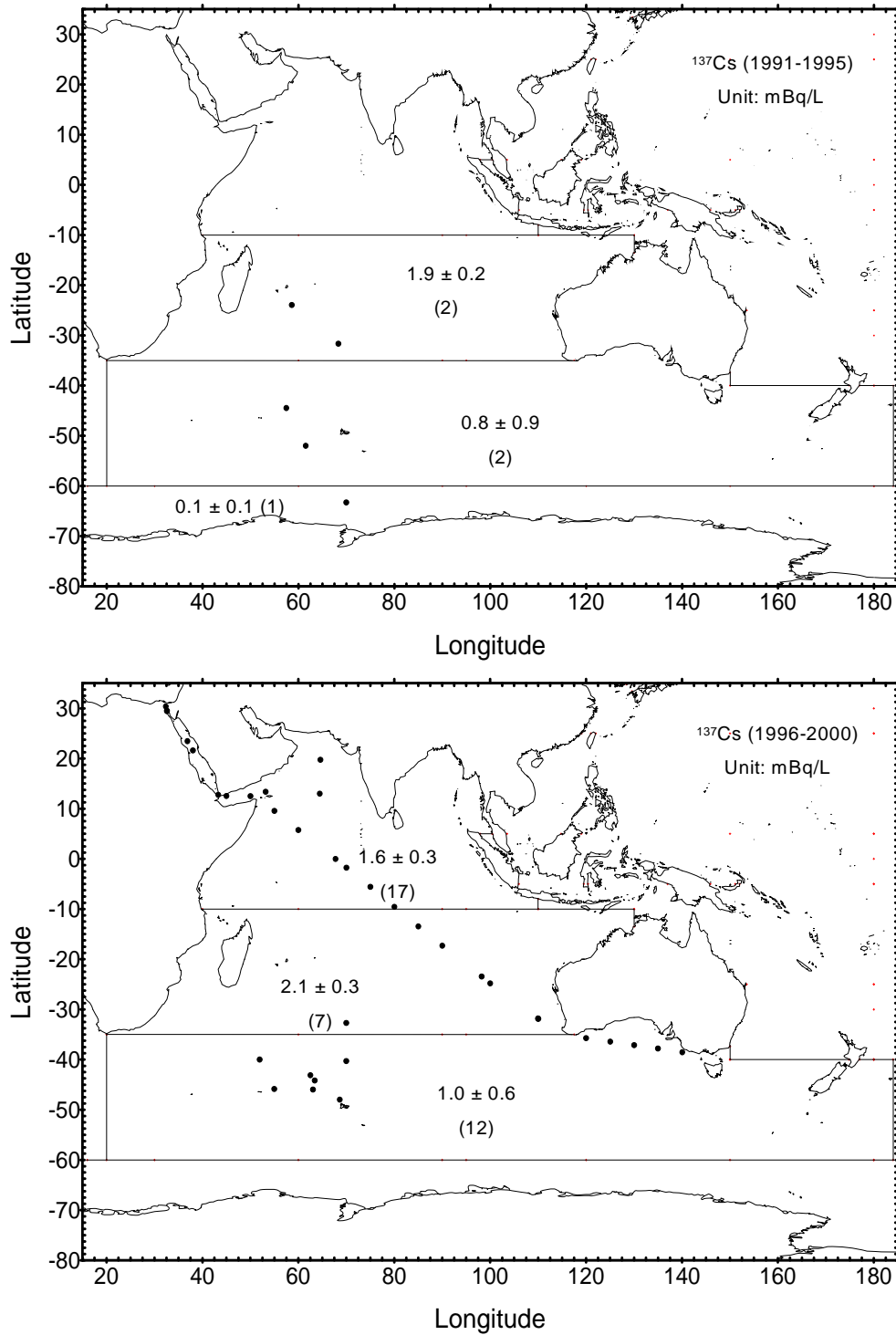


FIG. 76. ^{137}Cs in the Indian and Southern Oceans (data corrected to 01.01.2000).

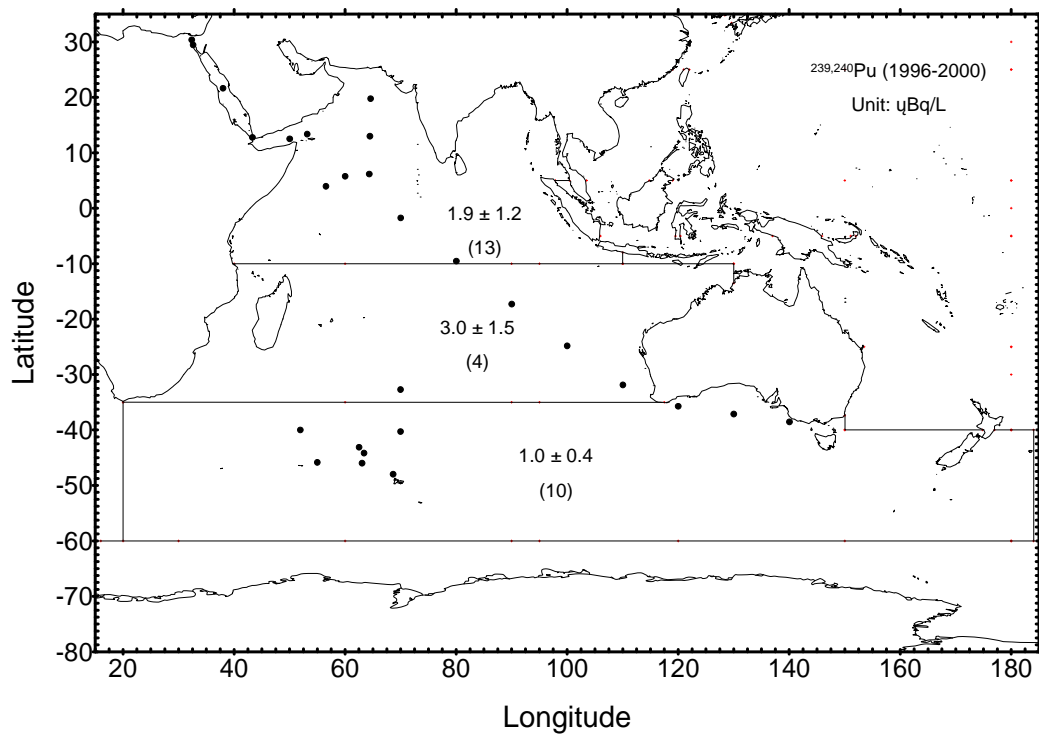
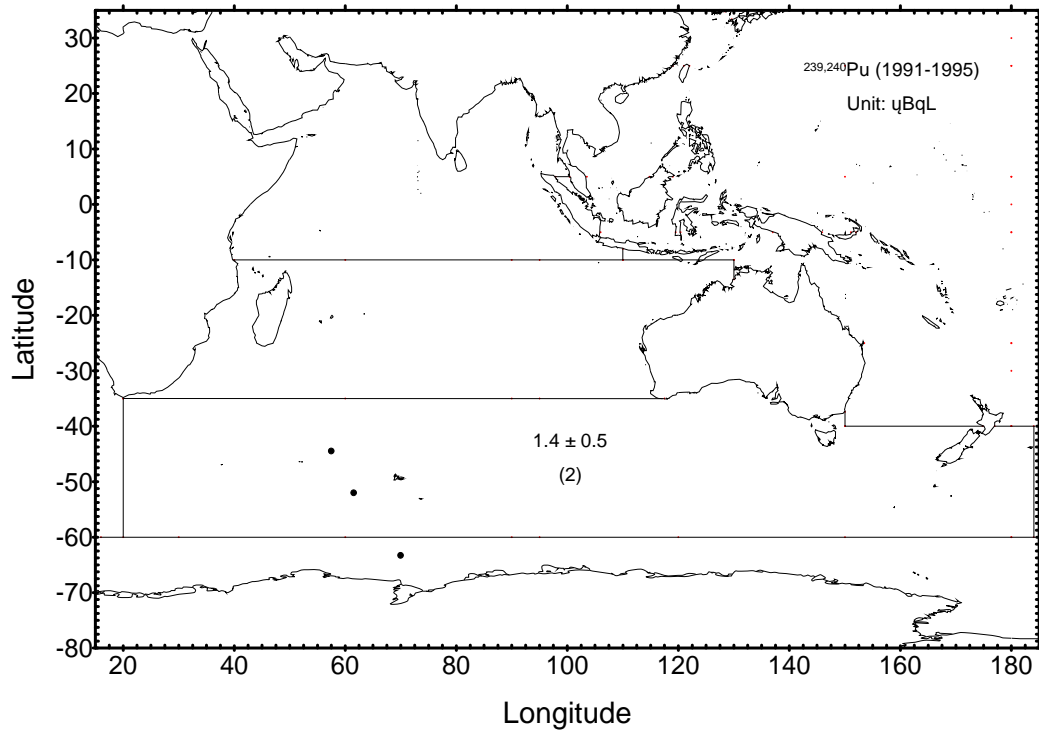


FIG. 77. $^{239,240}\text{Pu}$ in the Indian and Southern Oceans.

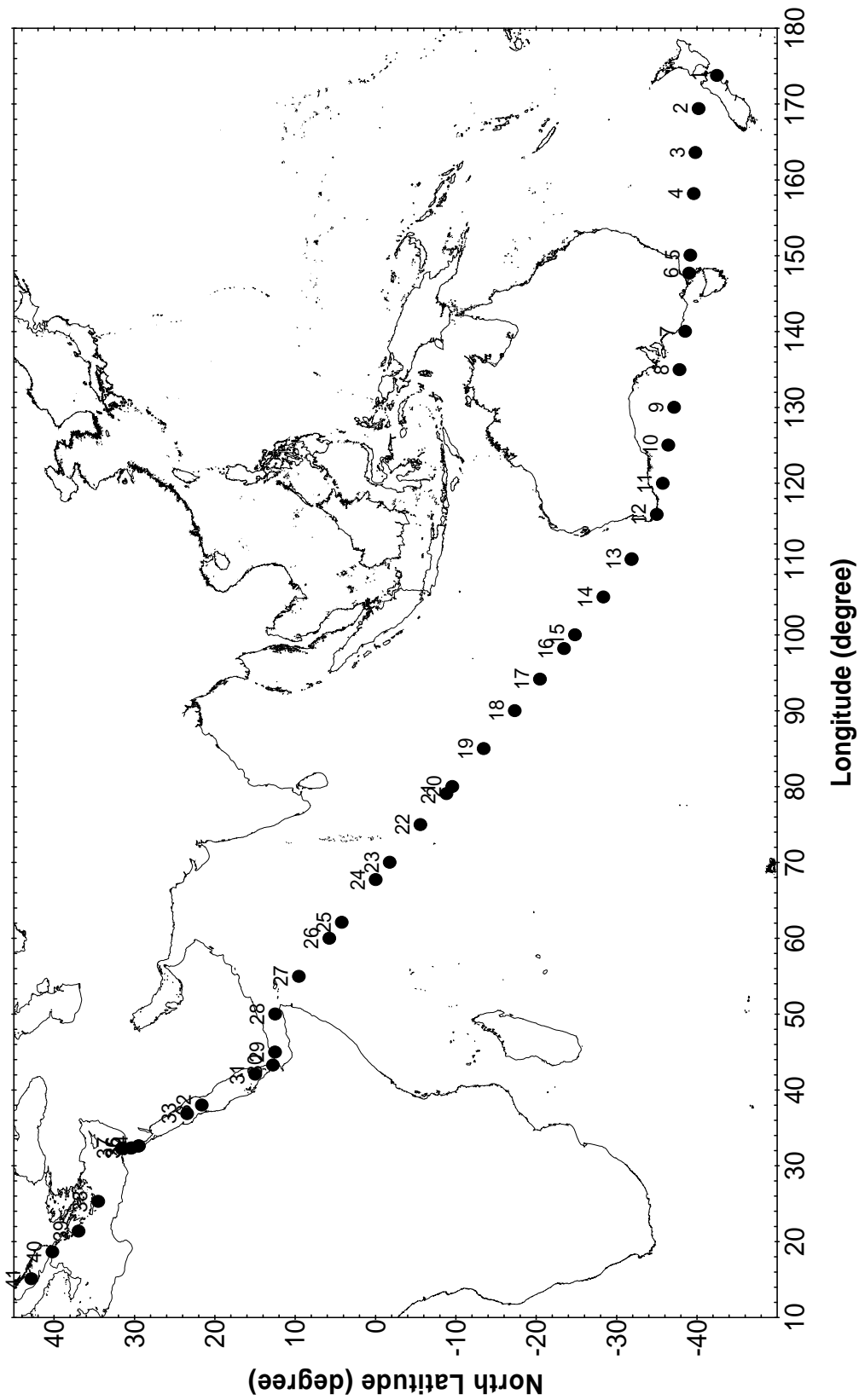


FIG. 78. Sampling stations of the Indian Ocean Transect Cruise, 1997.

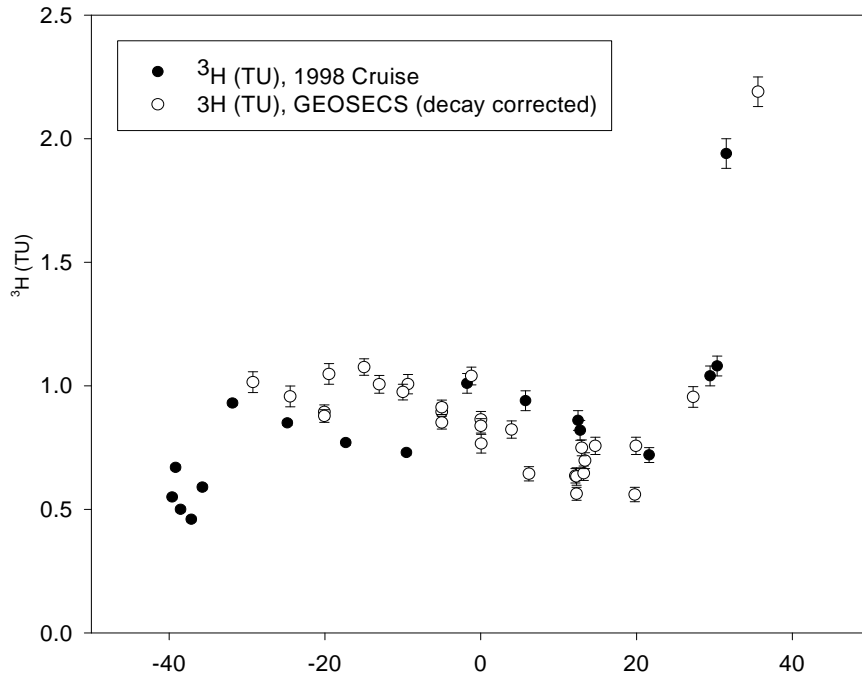


FIG. 79. ^3H concentrations in surface water of the Indian Ocean, the Red and Mediterranean Seas as a function of latitude.

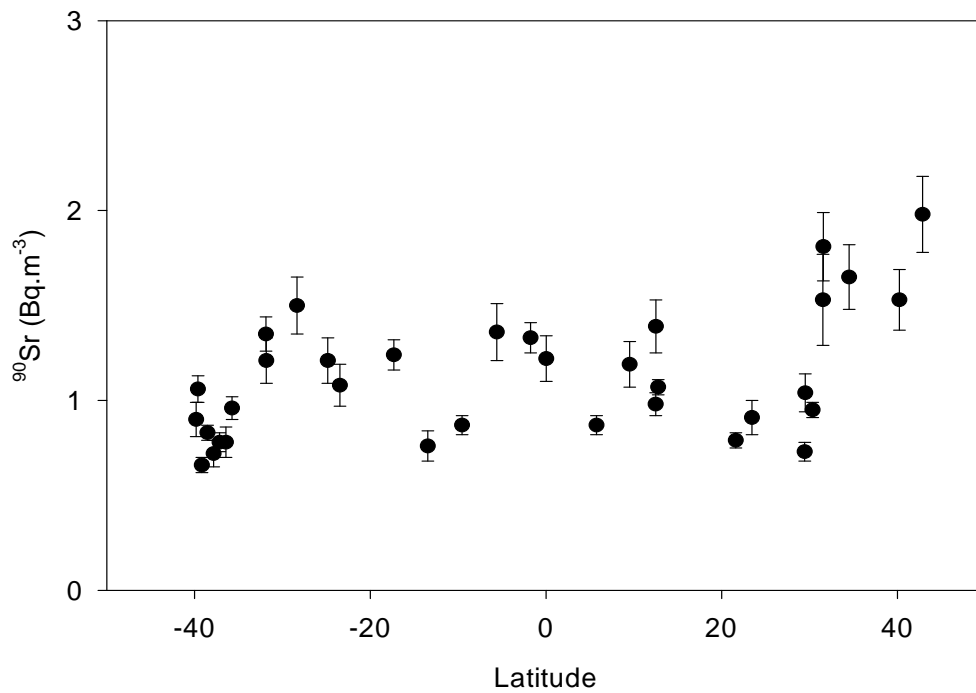


FIG. 80. ^{90}Sr concentrations in surface water of the Pacific and Indian Oceans, the Red and Mediterranean Seas as a function of latitude.

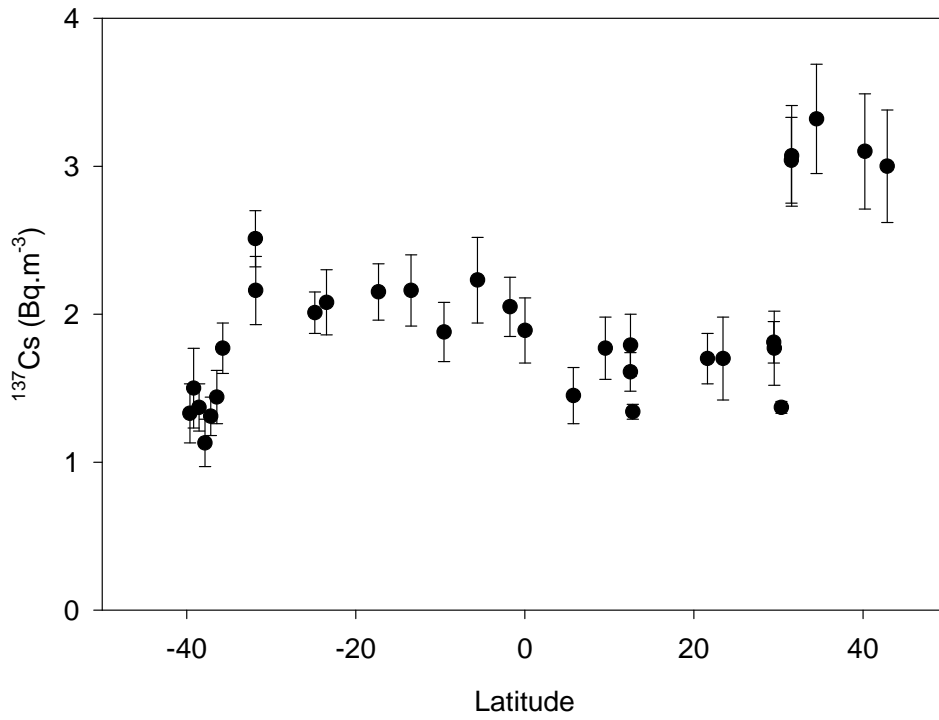


FIG. 81. ^{137}Cs concentrations in surface water of the Pacific and Indian Oceans, the Red and Mediterranean Seas as a function of latitude.

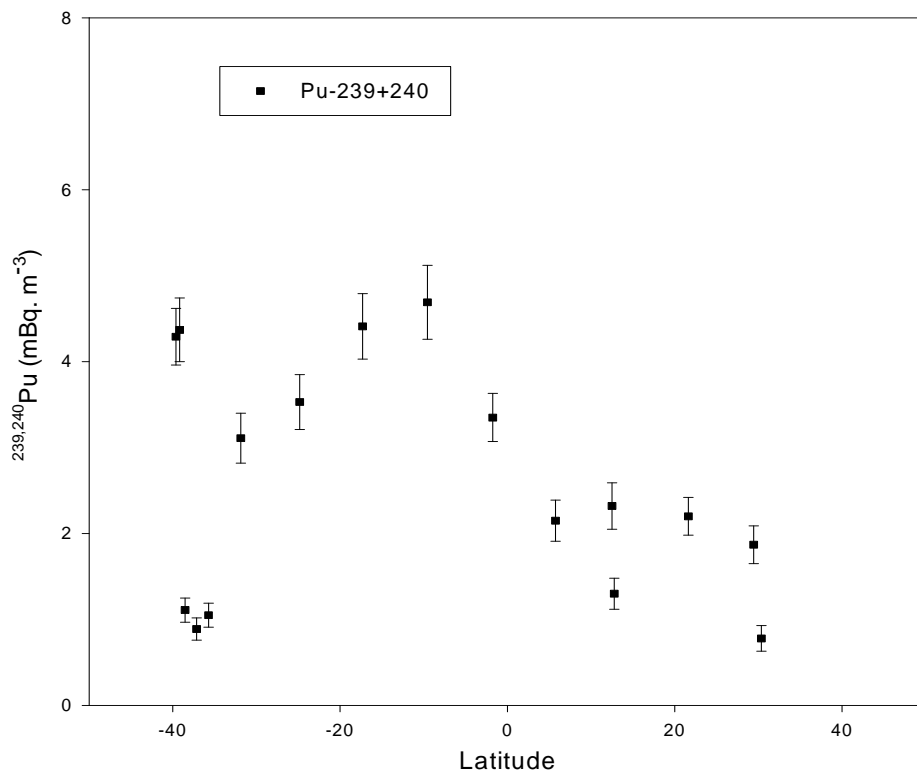


FIG. 82. $^{239,240}\text{Pu}$ concentrations in surface water of the Pacific and Indian Oceans, the Red and Mediterranean Seas as a function of latitude.

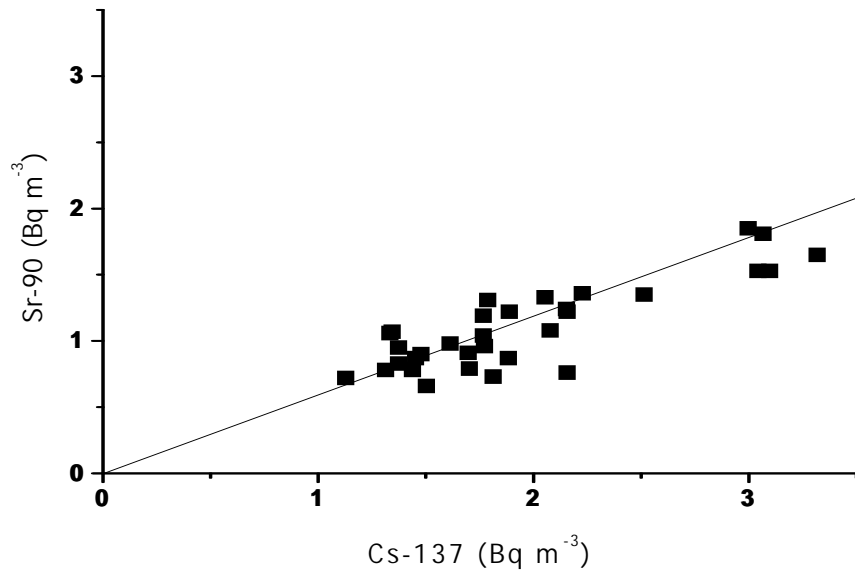


FIG. 83. Relationship between ¹³⁷Cs and ⁹⁰Sr in surface seawater.

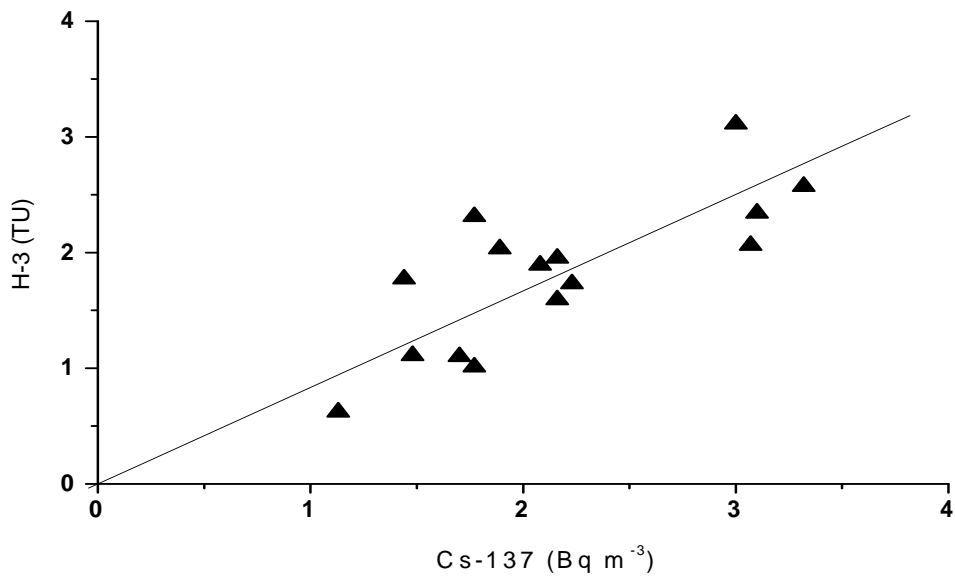


FIG. 84. Relationship between ¹³⁷Cs and ³H in surface seawater.

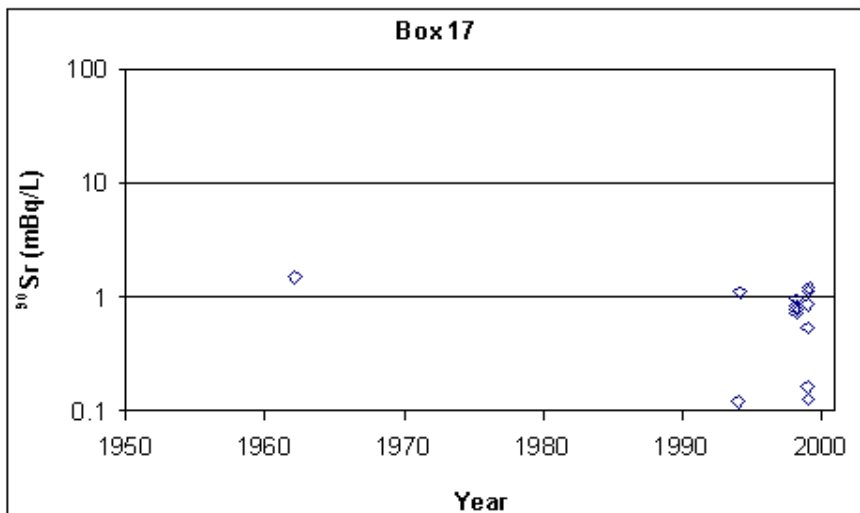
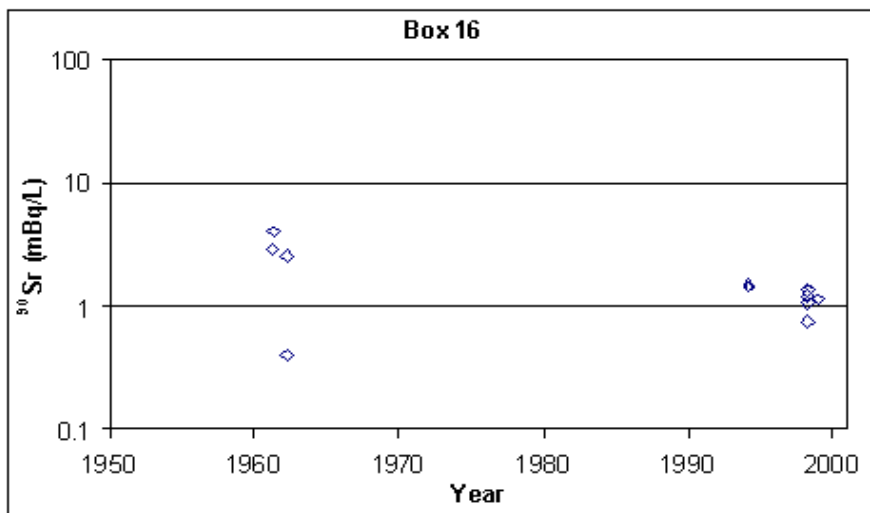
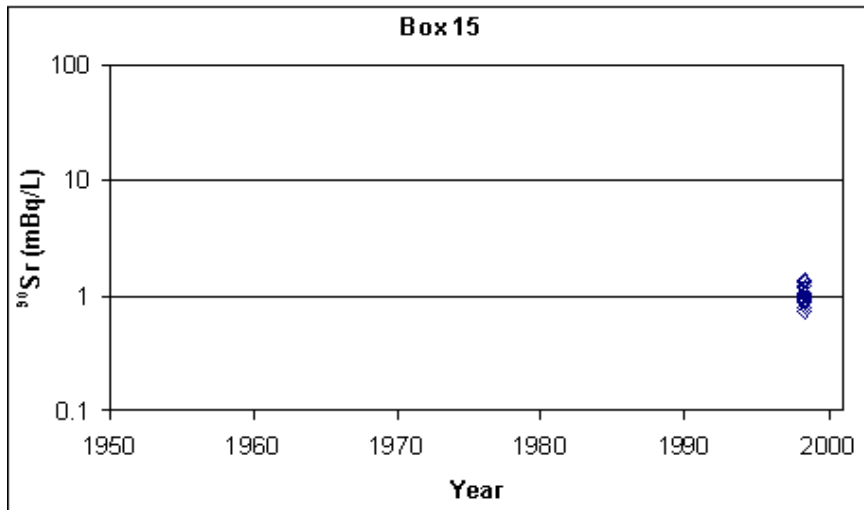


FIG. 85. ^{90}Sr time series in Indian Ocean surface water.

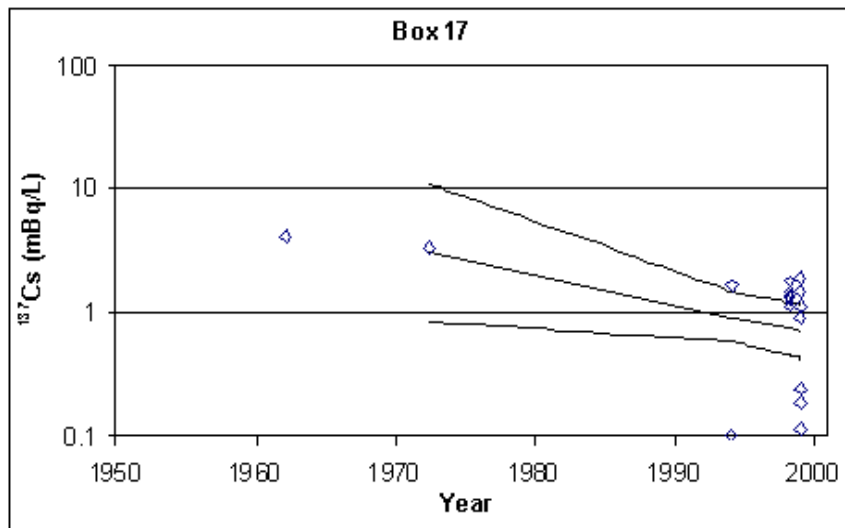
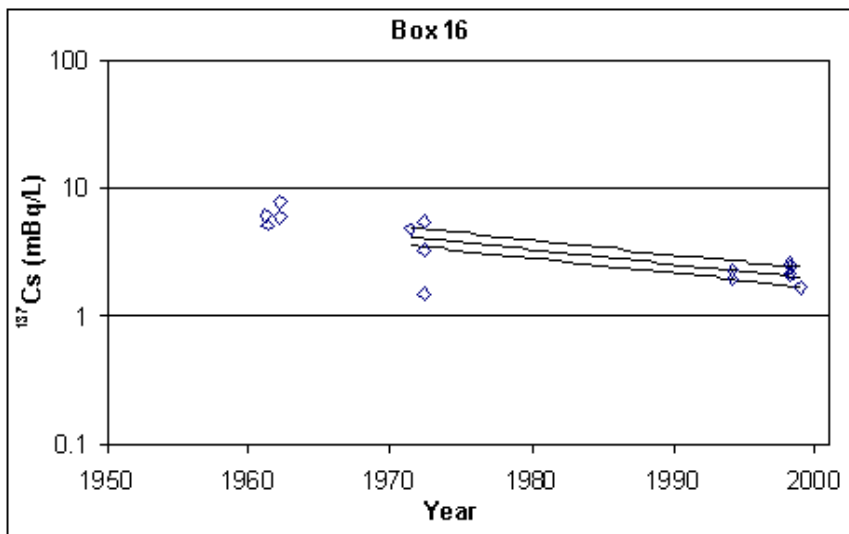
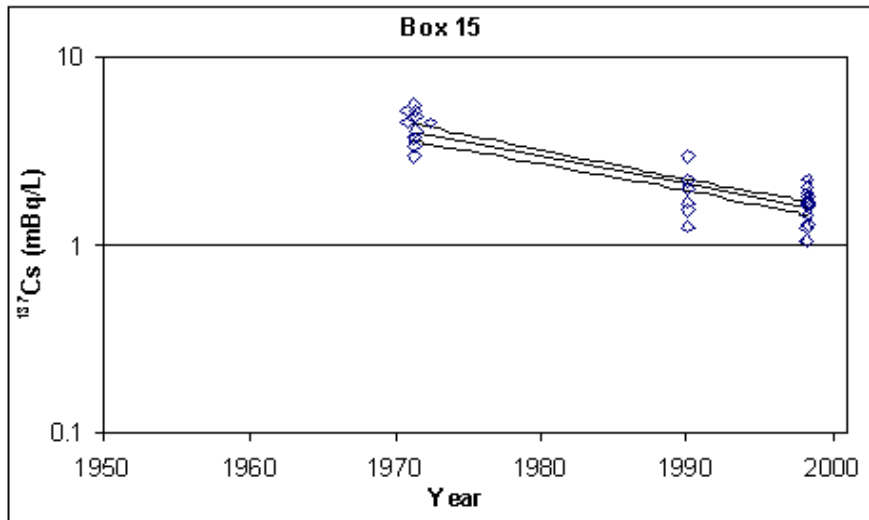


FIG. 86. ^{137}Cs time series in Indian Ocean surface water.

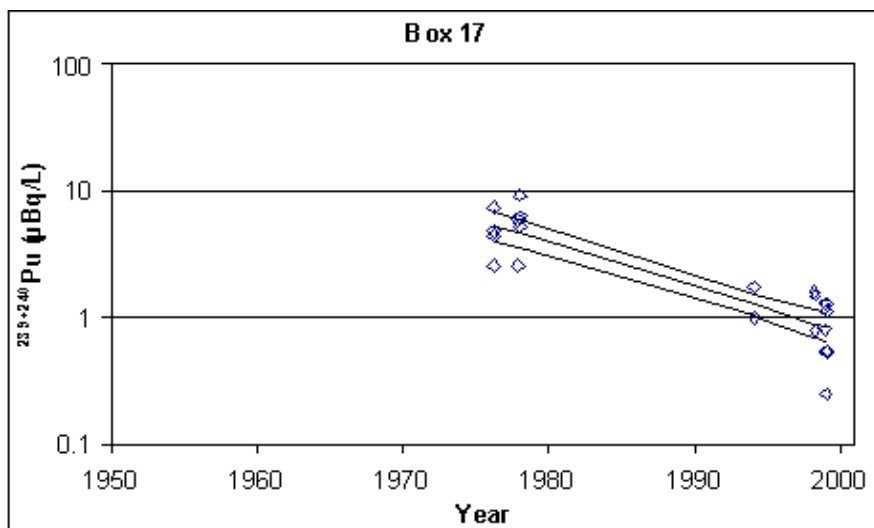
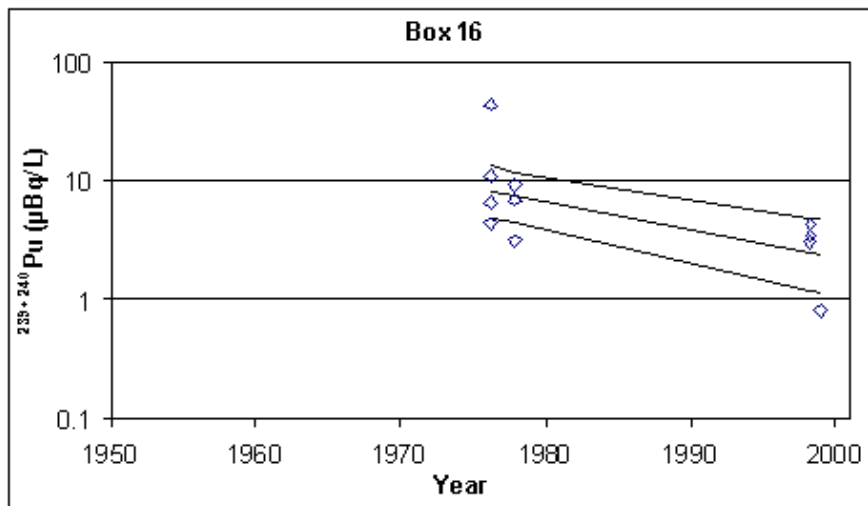
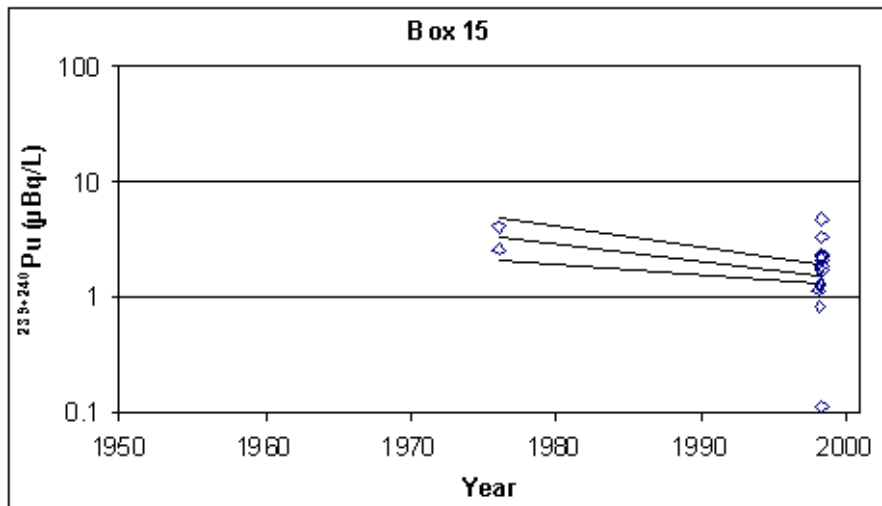


FIG. 87. $^{239,240}\text{Pu}$ time series in Indian Ocean surface water.

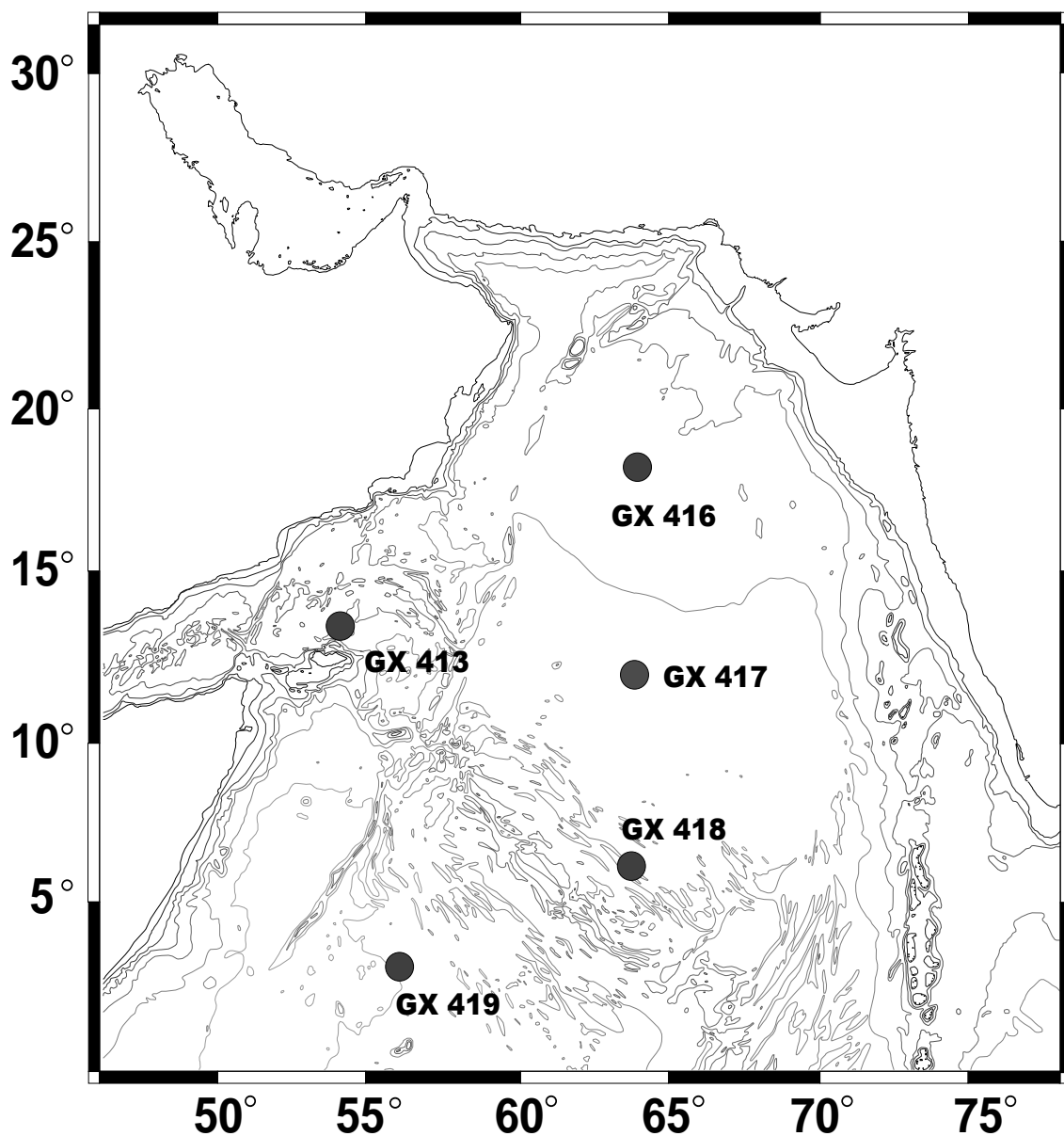


FIG. 88. GEOSECS stations in the Arabian Sea revisited in 1998.

TABLE XII. MEAN EFFECTIVE HALF-LIVES OF ^{137}Cs AND $^{239,240}\text{Pu}$ IN INDIAN OCEAN SURFACE WATER

Area	Boxes	Effective half-life $\pm 1\sigma^*$ (year)	
		^{137}Cs	$^{239,240}\text{Pu}$
North Indian Ocean	15	20.2 \pm 1.9	21.2 \pm 7.6
South Indian Ocean	16+17	21.9 \pm 6.3	8.8 \pm 1.0
All Indian Ocean	15-17	20.3 \pm 1.8	9.0 \pm 1.7

* Error weighted averaged values

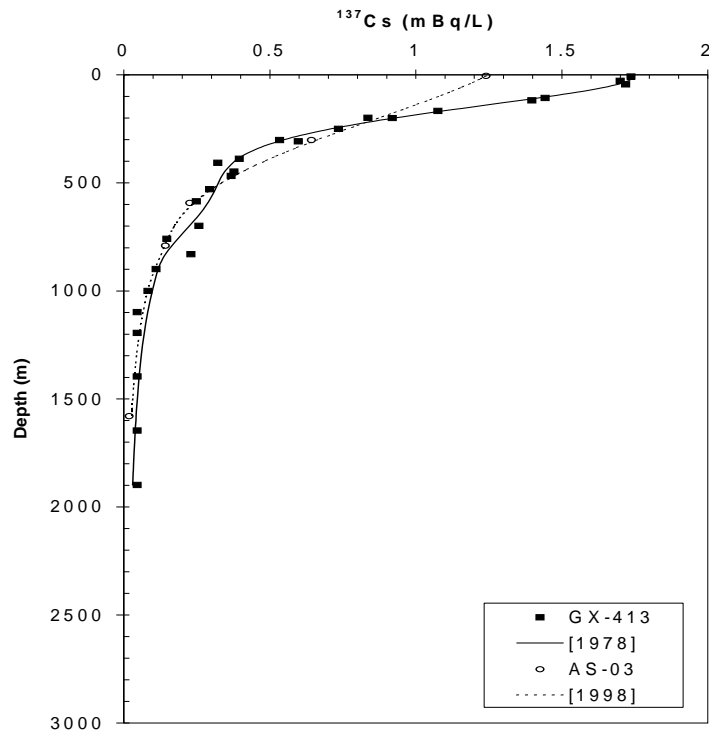


FIG. 89. Comparison of measured (dotted line) and calculated (continuous line), using GEOSECS ^3H data, ^{137}Cs profiles at GEOSECS Station 413.

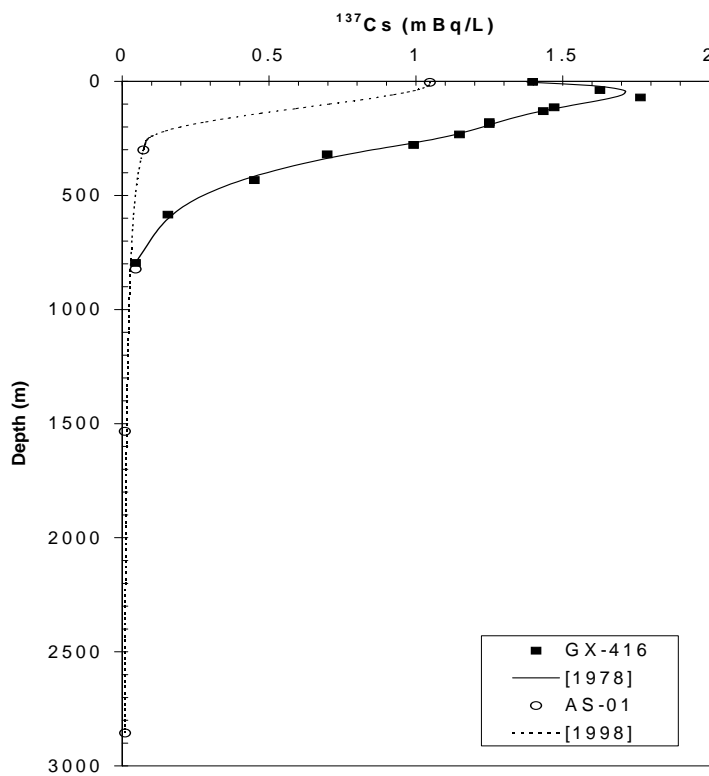


FIG. 90. Comparison of measured (dotted line) and calculated, (continuous line) using GEOSECS ^3H data, ^{137}Cs profiles at GEOSECS Station 416.

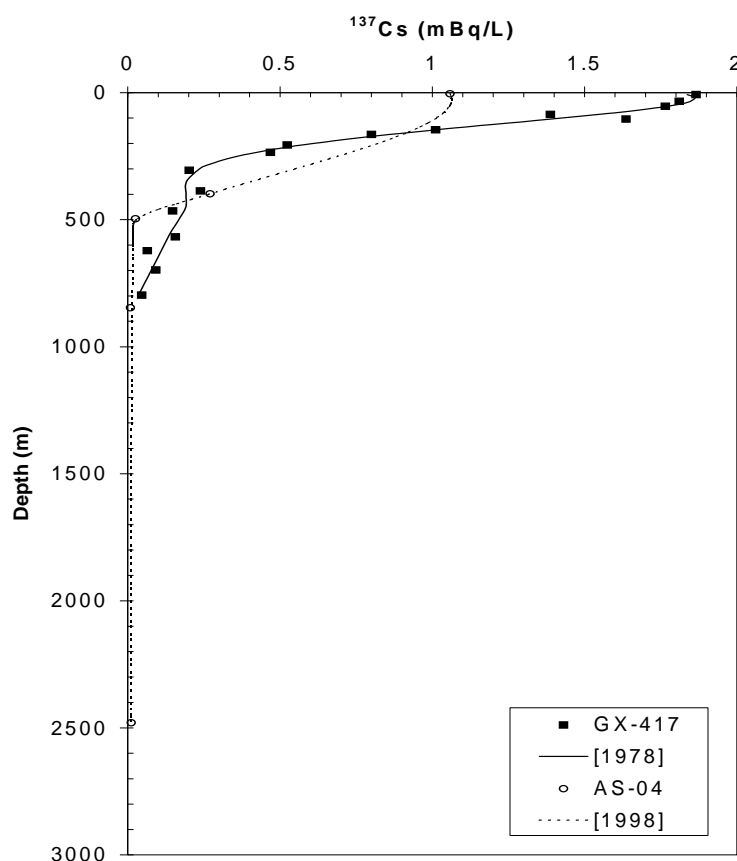


FIG. 91. Comparison of measured (dotted line) and calculated (continuous line), using GEOSECS ^3H data, ^{137}Cs profiles at GEOSECS Station 417.

6. ATLANTIC AND ARCTIC OCEANS

The radioactive contamination of the Atlantic and Arctic Oceans originates from three main sources:

- Global fallout, due to nuclear weapons testing in the atmosphere, which occurred from 1945 to 1980,
- Nuclear reprocessing in western Europe notably Sellafield in the UK and Cap de la Hague in France. The maximum discharges to the sea took place from 1970 to 1985,
- The Chernobyl accident in the Ukraine in 1986.

In contrast to the Pacific Ocean, the Atlantic has not received local fallout from nuclear weapons testing to any significant extent. Due to the inputs from reprocessing and Chernobyl, the NE Atlantic shows higher concentrations of, in particular, ^{137}Cs , ^{99}Tc and ^{129}I than any other part of the world ocean. Furthermore the variation in radionuclide concentrations is higher than seen elsewhere. The marginal seas of Europe show concentrations of e.g. ^{137}Cs and ^{99}Tc , which vary orders of magnitude within distances of a few hundred kilometres. It thus is difficult to give meaningful average concentrations for these radionuclides in marginal seas such as the North Sea, the Baltic Sea, the Black Sea and especially, the Irish Sea.

Intensive sampling of the NE Atlantic, including deep-water samples, was carried out in the area of the NEA/OECD dumpsites for low level radioactive wastes by the Bundesamt für

Seeschiffahrt und Hydrographie (BSH) (Germany) between 1983 and 1985 [109]. The levels detected are a consequence of global fallout from atmospheric weapons tests in the sixties. ^{137}Cs levels were between 2.8 and 4.5 Bq/m³ with an average concentration around 3.3 Bq/m³. The ^{90}Sr concentration was found to have an almost linear relation to ^{137}Cs values with the $^{137}\text{Cs}/^{90}\text{Sr}$ ratios close to the theoretical value of about 1.5 given in the UNSCEAR Report [110]. $^{239,240}\text{Pu}$ values were found to be relatively homogeneously distributed in the water column between 5 and 30 mBq/m³, whereas maximum values were found at about 1000 m depth. These observations were found to be similar to other oceans.

In order to collect more recent data on the background radionuclide levels in Atlantic water flushing through the North Sea, a cruise on board the German R/V "Deneb" was carried out in the spring of 1997 to the Mediterranean (Monaco). Samples of water taken at depths down to 500 m were analysed for ^{137}Cs , ^{90}Sr and transuranics. The average levels found in surface water of the eastern Atlantic and the Mediterranean are not statistically distinguishable (Table XIII).

TABLE XIII. AVERAGE ^{90}Sr AND ^{137}Cs LEVELS IN 1997 IN ATLANTIC AND MEDITERRANEAN SURFACE WATER (Bq/m³)

	^{137}Cs	^{90}Sr
Atlantic Ocean	2.33 ± 0.21 (24 values)	1.53 ± 0.21 (24 values)
Mediterranean Sea	2.63 ± 0.42 (25 values)	1.77 ± 0.31 (24 values)

The activity ratios of $^{137}\text{Cs}/^{90}\text{Sr}$ were 1.52 and 1.48, close to the expected fallout value. The distribution of transuranics in surface water of the Atlantic is less homogeneous. For $^{239,240}\text{Pu}$ values were between 2.7 and 8.9 mBq/m³, while concentrations in deeper water increased up to 17 mBq/m³. The activity ratios of $^{238}\text{Pu}/^{239,240}\text{Pu}$ are about 0.1, indicating slightly higher ratios than expected from global fallout. The few data for the French Mediterranean coast are above levels for the Atlantic, between 9.3 and 16.9 mBq/m³ for $^{239,240}\text{Pu}$. Water from 500 m depth with levels up to 28 mBq/m³ of $^{239,240}\text{Pu}$ possibly indicate the influence of the nuclear complex at Marcoule on the Rhone river. The concentrations of ^{241}Am are mostly below 3 mBq/m³ down to 500 m depth and there is no obvious difference between the data from the Atlantic and Mediterranean.

6.1. Surface water radionuclide time series in the Atlantic Ocean

The Atlantic and Arctic Oceans with their marginal seas have been divided into Boxes 18–30 (Fig. 92). The maps of the distributions and average concentrations for ^3H , ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ in Atlantic surface water for the period 1991–1995 and 1996–2000 are shown in Figs 93 to 96 and 97 to 100, respectively. Unfortunately, for the south Atlantic Ocean, the radionuclide data are very sparse, therefore a proper evaluation has not been possible.

Data for ^{137}Cs for the period 1991–1995 clearly show a latitudinal effect with mean concentrations from the south of 0.6 Bq/m³ (Box 30) to 3.5 Bq/m³ in the north (Box 18). The highest concentrations were however, observed in the Baltic Sea (mean value of about 60 Bq/m³) and the Black Sea (about 40 Bq/m³). The data for the period 1996–2000 (where available) show a similar trend.

The latitudinal effect is clearly seen for ^{90}Sr as well. Higher mean concentrations were observed in the Baltic Sea (about 10 Bq/m³), the English Channel (about 8 Bq/m³) and the

North Sea (about 5 Bq/m³) resulting from discharges from Sellafield and Cap de la Hague. In 1996–2000, the mean concentrations were about 10 Bq/m³ in the Baltic Sea, about 4 Bq/m³ in the English Channel, and about 4 Bq/m³ in the North Sea.

The effective half-lives of ⁹⁰Sr, ¹³⁷Cs and ^{239,240}Pu in Atlantic surface water were calculated using data stored in the GLOMARD database, mostly based on Risö reports [111], BSH and CEFAS (The Centre for Environment, Fisheries and Aquaculture Science, UK) data. All data used for the calculation of effective half-lives of ⁹⁰Sr, ¹³⁷Cs and ^{239,240}Pu in the different boxes dividing the Atlantic Ocean (except for Box 19, where no data were available) are shown in Figs 101–103 (for ⁹⁰Sr), 104–106 (for ¹³⁷Cs) and 107–109 (for ^{239,240}Pu). The calculated effective half-lives and predicted concentrations are given in Tables VI–VIII. The values for the different boxes are very widely scattered, so it is more useful to group data for the north, central and south Atlantic Ocean (Table XIV).

⁹⁰Sr and ¹³⁷Cs show similar half-lives of 16 ± 4 and 19 ± 4 years, respectively, for the north Atlantic, 24 ± 2 and 24 ± 6 years for the central Atlantic and 17 ± 9 and 20 ± 6 years for the south Atlantic Ocean. The total of all Atlantic data is 22 ± 2 and 20 ± 3 years, for ⁹⁰Sr and ¹³⁷Cs, respectively. The ^{239,240}Pu data are close, giving a value for all the Atlantic Ocean of 9 ± 1 year.

Data for all oceans (Table XV) gave similar effective half-lives for ⁹⁰Sr and ¹³⁷Cs, 19 ± 3 and 20 ± 2 years, respectively. As expected, the half-life is similar for both radionuclides, therefore, one can accept a mean value of 19 ± 2 years. The ^{239,240}Pu effective half-life in surface water is 9 ± 1 year. The corresponding residence times of ⁹⁰Sr (¹³⁷Cs) and ^{239,240}Pu in surface water are 27 ± 2 and 13 ± 1 years, respectively.

In 1988–1989 the SWEDARP expedition collected surface sea water in the Atlantic Ocean between 73°N and 72°S [112]. The samples were analysed for ¹³⁷Cs and ^{239,240}Pu and were compared with the GEOSECS data from the expedition in 1972–1973. The effective half-life of ¹³⁷Cs was determined to be 23 years (or longer), corresponding to a mean residence time of 33 years. The effective half-life of ^{239,240}Pu was determined to be 7–8 years.

TABLE XIV. MEAN EFFECTIVE HALF-LIVES OF ⁹⁰Sr, ¹³⁷Cs AND ^{239,240}Pu IN ATLANTIC OCEAN SURFACE WATER

Area	Boxes	Effective half-life ± 1σ [*] (year)		
		⁹⁰ Sr	¹³⁷ Cs	^{239,240} Pu
North Atlantic	25+28	15.6 ± 4.3	18.6 ± 4.2	9.0 ± 0.6
Central Atlantic	29	24.2 ± 2.1	23.7 ± 5.7	9.8 ± 2.9
South Atlantic	30	17.3 ± 8.8	20.2 ± 6.3	10.7 ± 4.9
All Atlantic	25–30	22.3 ± 2.4	20.4 ± 3.0	9.1 ± 0.5

* Error weighted averaged values

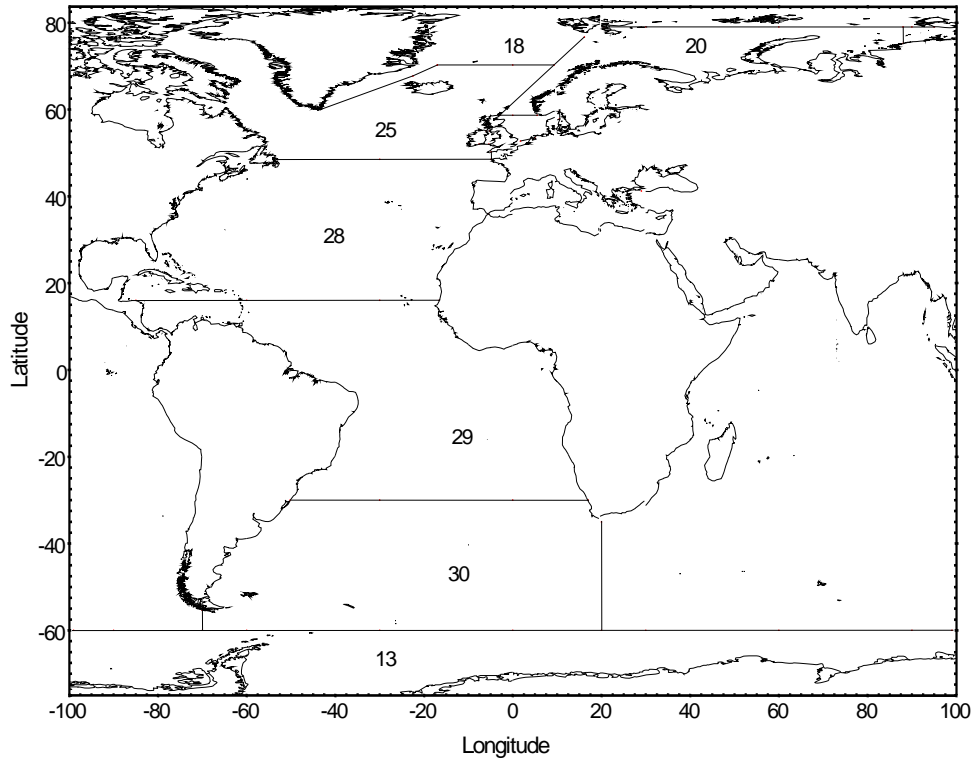


FIG. 92. Latitudinal boxes in the Atlantic Ocean.

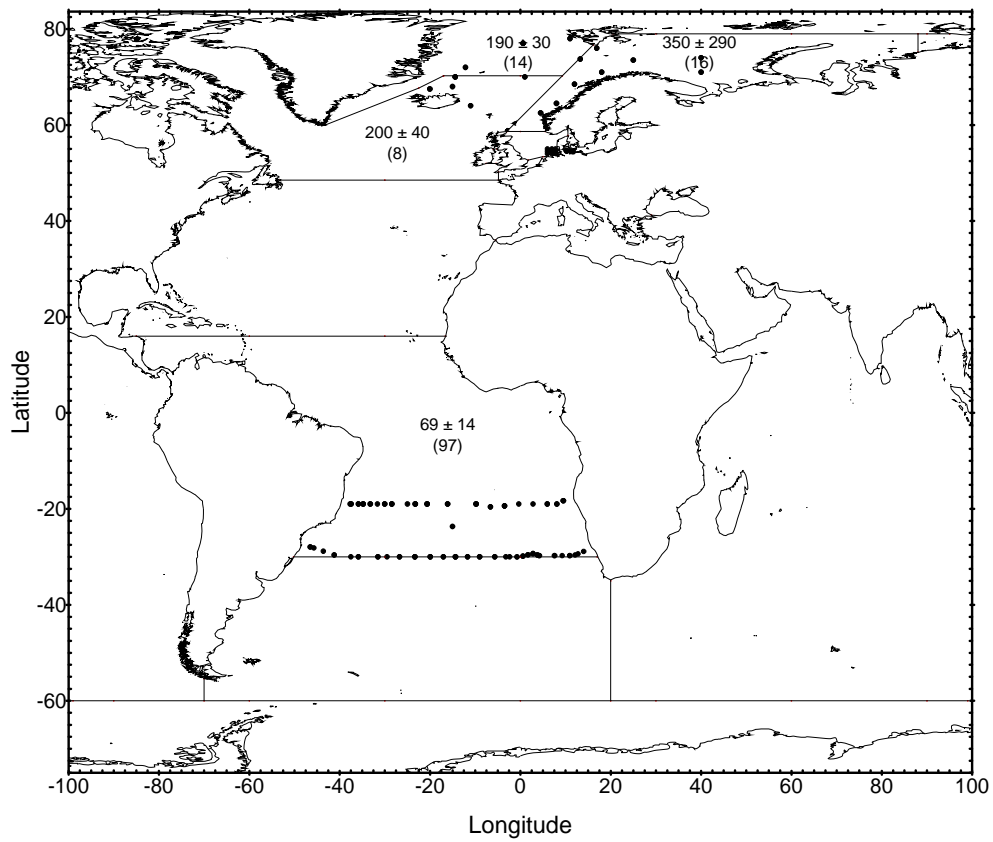


FIG. 93. ^3H in Atlantic surface water (1991–1995), decay-corrected to 01.01.2000, in mBq/L.

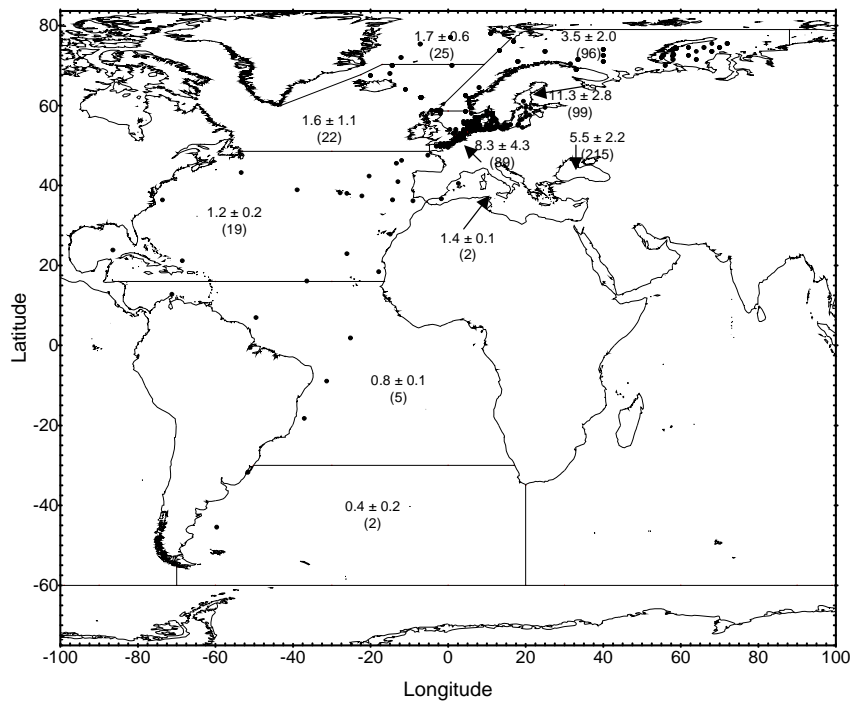


FIG. 94. ^{90}Sr in Atlantic surface water (1991–1995), decay-corrected to 01.01.2000, in mBq/L.

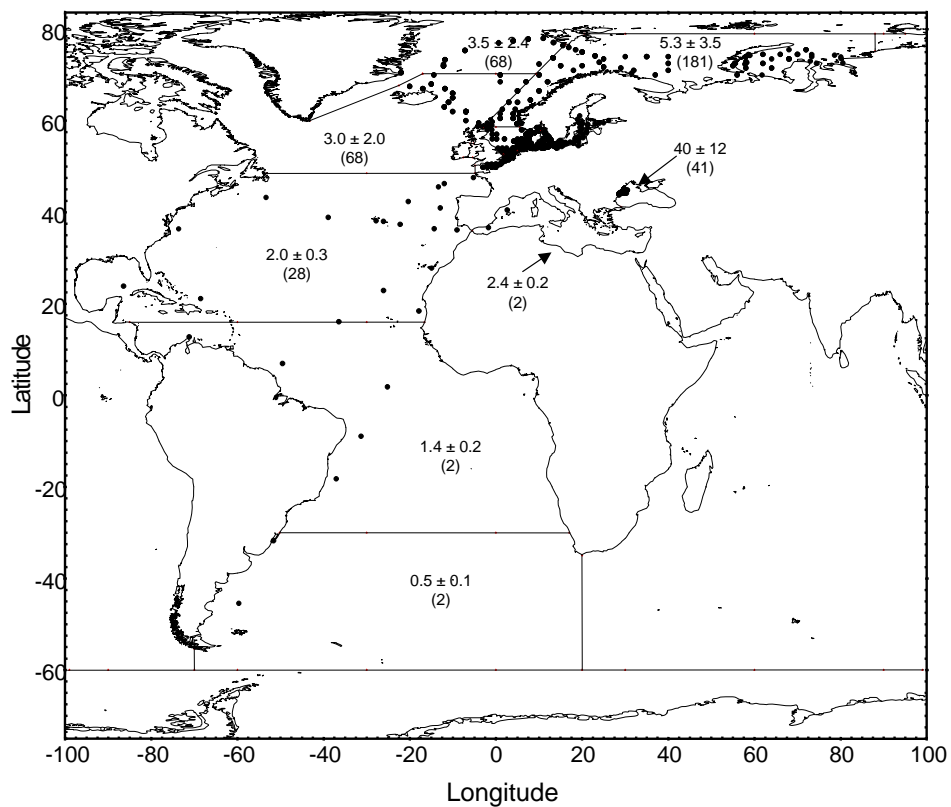


FIG. 95. ^{137}Cs in Atlantic surface water (1991–1995), decay-corrected to 01.01.2000, in mBq/L.

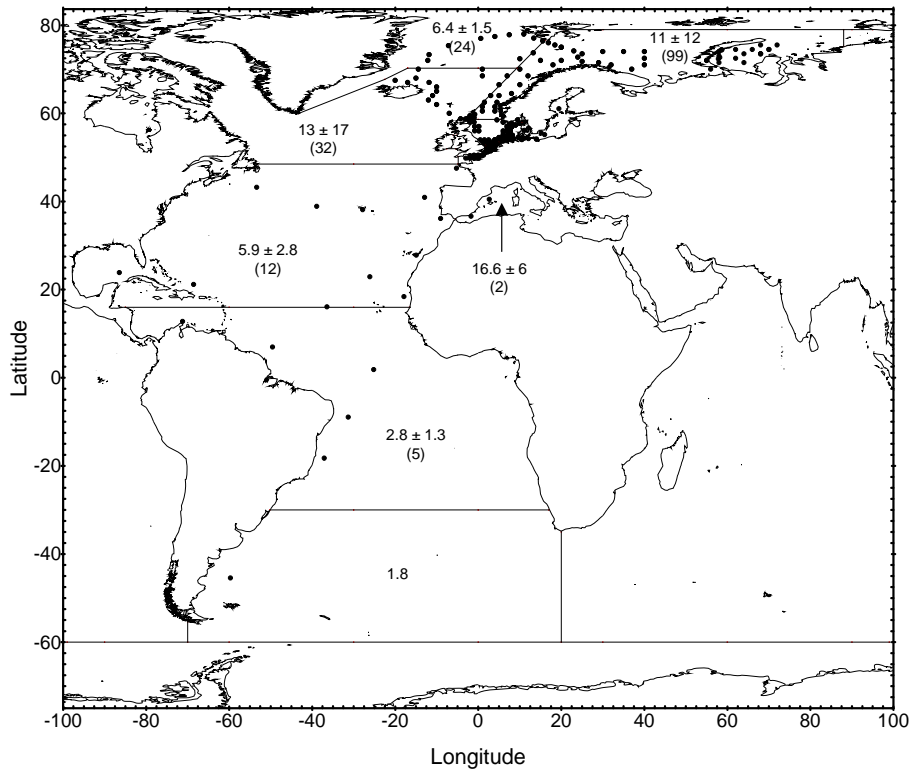


FIG. 96. $^{239,240}\text{Pu}$ in Atlantic surface waters (1991–1995), in $\mu\text{Bq/L}$.

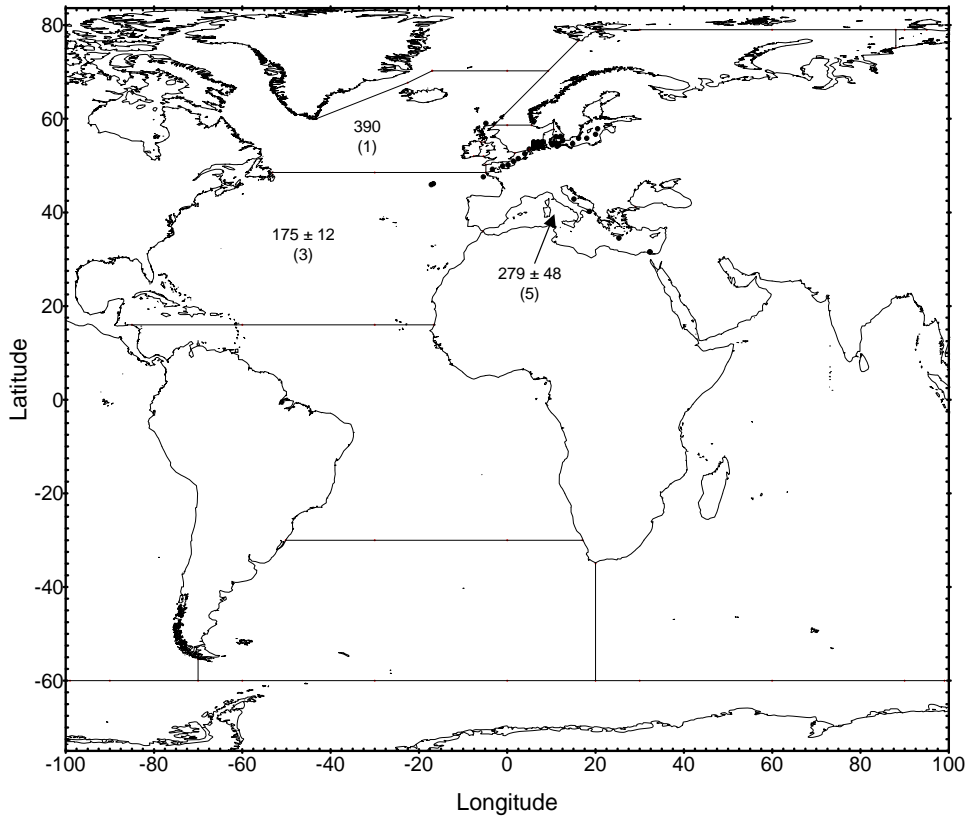


FIG. 97. ^3H in Atlantic surface water (1996–2000), decay-corrected to 01.01.2000, in mBq/L .

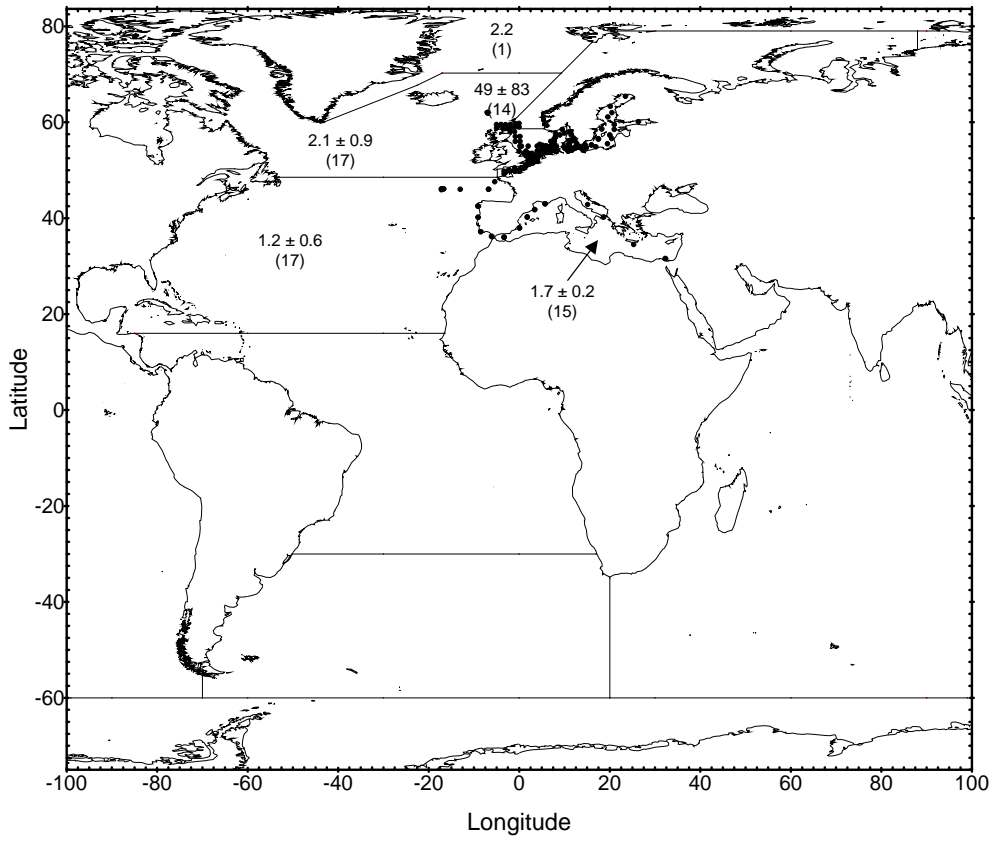


FIG. 98. ^{90}Sr in Atlantic surface water (1996–2000), decay-corrected to 01.01.2000, in mBq/L.

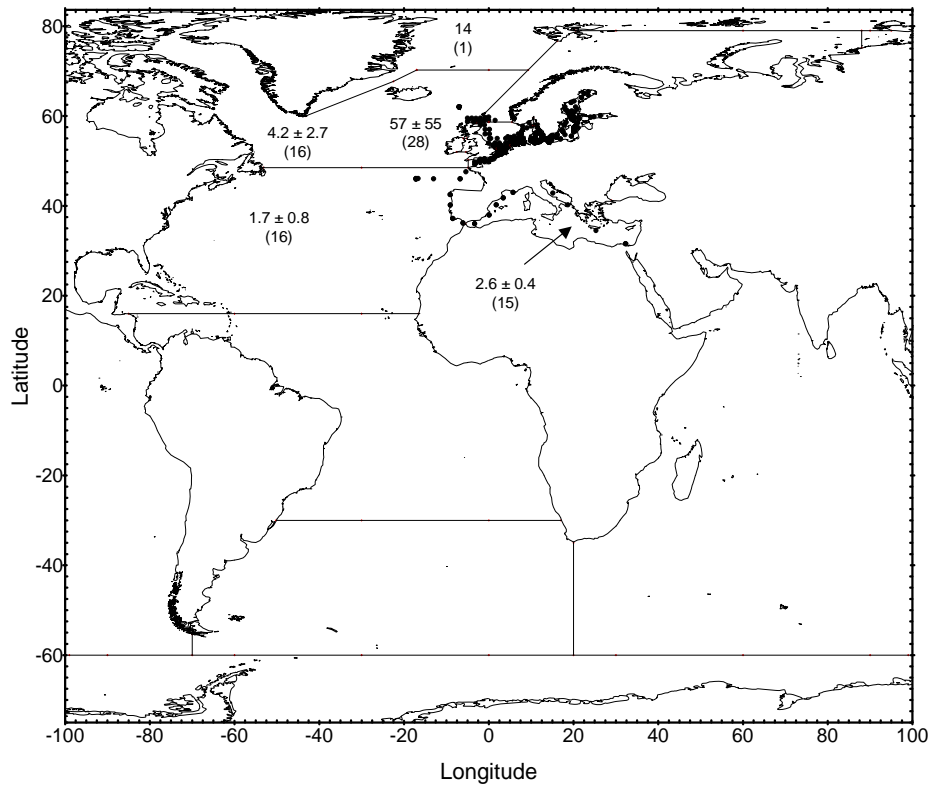


FIG. 99. ^{137}Cs in Atlantic surface waters (1996–2000), decay-corrected to 01.01.2000, in mBq/L.

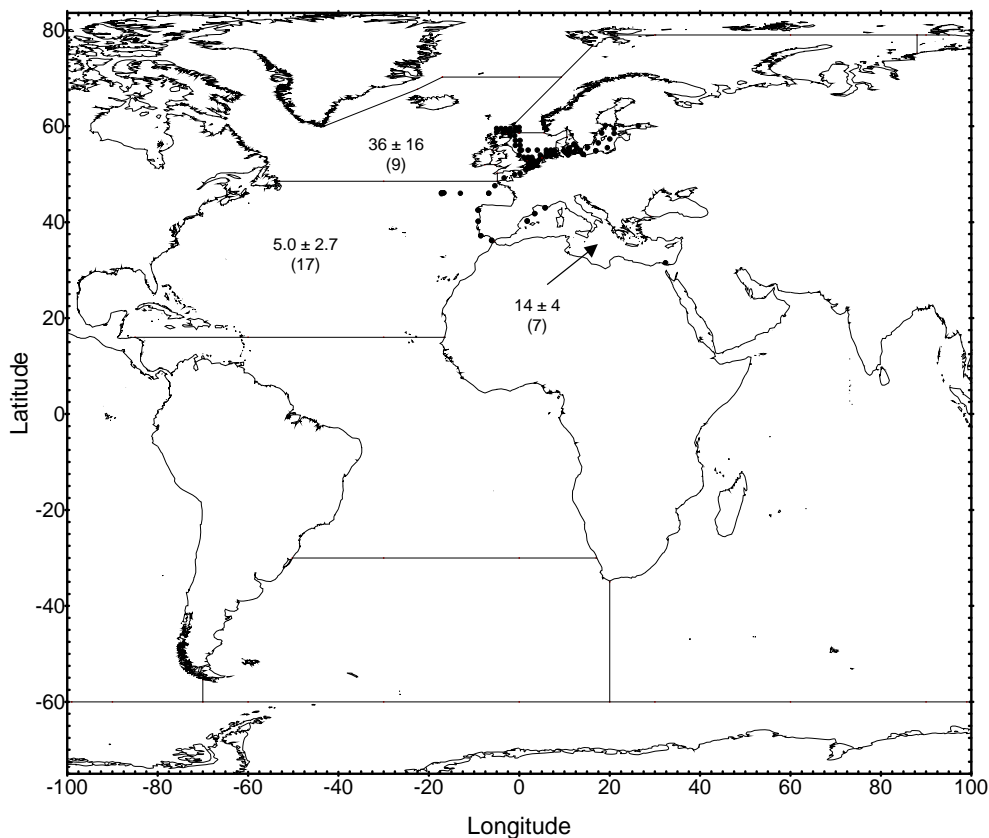


FIG. 100. $^{239,240}\text{Pu}$ in Atlantic surface waters (1996–2000), in $\mu\text{Bq/L}$.

TABLE XV. MEAN EFFECTIVE HALF-LIVES OF ^{90}Sr , ^{137}Cs AND $^{239,240}\text{Pu}$ IN SURFACE WATER OF THE WORLD OCEAN

Area	Boxes	Effective half-life $\pm 1\sigma^*$ (year)		
		^{90}Sr	^{137}Cs	$^{239,240}\text{Pu}$
North Pacific	01-05	12.4 \pm 1.1	12.7 \pm 1.4	6.7 \pm 0.9
Equatorial Pacific	06+07	21.1 \pm 1.9	28.6 \pm 3.0	10.2 \pm 2.4
South Pacific	08-12	17.9 \pm 1.0	22.2 \pm 5.2	12.1 \pm 3.6
All Pacific	01-12	15.9 \pm 2.4	15.9 \pm 4.3	7.4 \pm 1.1
North Indian	15		20.2 \pm 1.9	21.2 \pm 7.6
South Indian	16+17		21.9 \pm 6.3	8.8 \pm 1.0
All Indian	15-17		20.3 \pm 1.8	9.0 \pm 1.7
North Atlantic	25+28	15.6 \pm 4.3	18.6 \pm 4.2	9.0 \pm 0.6
Central Atlantic	29	24.2 \pm 2.1	23.7 \pm 5.7	9.8 \pm 2.9
South Atlantic	30	17.3 \pm 8.8	20.2 \pm 6.3	10.7 \pm 4.9
All Atlantic	25-30	22.3 \pm 2.4	20.4 \pm 3.0	9.1 \pm 0.5
All Oceans		19.1 \pm 3.2	19.8 \pm 1.5	8.8 \pm 1.0

* Error weighted averaged values

6.2. Water column radionuclide time series in the Atlantic Ocean

Sampling stations for the evaluation of ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ water profiles time series in the Atlantic Ocean are shown in Fig 110. Corresponding ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ water profiles in the north Atlantic Ocean (north of Iceland) are shown in Figs 111, 112 and 113, respectively. Data for 1972 have been extracted from the GEOSECS project and for 1985 and 1995 from the BSH database. Data are fairly widely scattered for ^{90}Sr , however, data for 1985 (except for two outliers) are lower than those for 1972. The results from 1995 are in between the 1972 and 1985 values. Also, the 1985 values below 1000 m are much higher than for 1972. Higher 1995 values may be due to discharges from Cap de la Hague in the eighties.

^{137}Cs profiles clearly show increasing concentrations from 1972, to 1985 and 1995, reflecting ^{137}Cs releases from Sellafield. According to Dahlgard [113] the expected transit time from Sellafield to the investigated sites is around 10 years, which appears in good agreement with maximum ^{137}Cs releases in the mid-seventies. However, the further increase in 1995 is difficult to explain.

$^{239,240}\text{Pu}$ data show a remarkable decrease from a peak value of around 58 mBq/m^3 in 1972 to about 13 mBq/m^3 in 1995.

Another comparison of water profiles was made for ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ obtained from the GEOSECS programme (1973) and the "Walther Herwig II" cruise of August–September 1996 organized by Federal Research Centre for Fisheries (Germany) in which IAEA-MEL participated. Although the positions of the stations are not the same (see Fig. 110), the ^{90}Sr data presented in Fig. 114 show a decrease in surface water concentrations from 2 to 1 Bq/m^3 and an increase below 750 m water depth in 1996 data. ^{137}Cs data show similar behaviour (Fig. 115).

The $^{137}\text{Cs}/^{90}\text{Sr}$ activity ratios from the 1996 data show values of around 1.5, except for layers between 1500–1400 m, where ratios of up to 2 were observed. $^{239,240}\text{Pu}$ profiles shown in Fig. 116 clearly show the displacement of the maximum from around 600 m, observed in 1973, down to 750–1500 m in 1996. Unfortunately, there are not enough GEOSECS data to make a more precise comparison. The remarkable $^{239,240}\text{Pu}$ peak, as well as the ^{90}Sr , ^{137}Cs , ^3H and ^{14}C peaks observed in deep-water layers are due to specific transport processes in the NE Atlantic [114].

6.3. Inventories of ^{90}Sr and ^{137}Cs in the NE Atlantic

From 1983 to 1995 BSH collected and analysed water column samples from the NE Atlantic. Table XVI summarizes the results of ^{90}Sr and ^{137}Cs inventory calculations based on the data supplied by them. It appears that the ^{137}Cs observations are in reasonable agreement with the known inputs to the Atlantic Ocean, but the ^{90}Sr observations are about double the expected values. The explanation may be that the number of ^{90}Sr determinations is relatively small. But the discrepancy may also be due to the fact that the samples may not be representative of the western and central parts of the N Atlantic.

The inventory of ^{137}Cs in ocean sediment is in the order of $0.1\text{--}0.2 \text{ kBq/m}^2$ i.e. about 5% of that present in the water column. Hence, sediment does not influence the inventory estimates significantly. The reasonable agreement between observations and inputs may be less favourable for ^{137}Cs if one considers that the NE Atlantic contains both reprocessing and Chernobyl ^{137}Cs . The inventories of ^{90}Sr and ^{137}Cs determined from measurements of water column samples collected in the NE Atlantic are up to twice the known inputs to the N Atlantic Ocean. The discrepancy is higher for ^{90}Sr than ^{137}Cs .

6.4. Radionuclides in northern European seas

The northern European seas have received input of anthropogenic radionuclides from different sources, mainly :

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

In addition to these sources, nuclear power stations, fuel production facilities, nuclear research facilities, and dumping of low level radioactive waste in the deep NE Atlantic Ocean may contribute to the contamination of the immediate environment. The discharges or releases from these sources are not detectable in the open marine environment and their impact on the local environment is monitored regularly by the competent authorities.

TABLE XVI. WATER COLUMN INVENTORIES OF ^{90}Sr AND ^{137}Cs IN THE NE ATLANTIC IN THE YEAR 2000

Year	BoxNo.	Columns	Year 2000 mean (kBq/m ²)	1 SD	N Atlantic latitude	Area (10 ¹² m ²)	Inventory (PBq)	Atlantic input (PBq)	INV/INP
^{137}Cs									
1985	25	5	3.16	0.64					
1995	25	4	3.26	0.51					
Mean	25		3.21	0.07	70°-60° N	4.560	14.6	16.8	0.87
1985	18	9	4.67	1.66					
1995	18	6	4.84	0.67					
Mean			4.76	0.12	80°-70° N	3.369	16.0	13.1	1.22
1983	28	7	3.05	0.21					
1984	28	10	3.34	0.27					
1985	28	3	3.29	0.33					
Mean			3.23	0.16	60°-30° N	20.842	67.3	48.3	1.39
					90°-30° N	28.771	97.9	78.2	1.25
^{90}Sr									
1985	25	2	2.01	0.43	70°-60° N	4.560	9.2	4.5	2.04
1985	18	3	2.58	1.53	80°-60° N	3.369	8.7	2.2	3.95
1983	28	1	2.62						
1984	28	5	2.07	0.24					
Mean			2.35	0.39	60°-30° N	20.842	48.9	24.2	2.02
					90°-30° N	28.771	66.7	30.9	2.16

NOTE: The inputs from reprocessing and Chernobyl were assumed by the year 2000 to have been equally divided between the three Atlantic boxes. Any input to the Arctic Ocean was neglected.

In the Arctic Ocean various additional potential sources exist, such as: dumped radioactive waste in the Kara and Barents Seas [12, 115]; discharges from nuclear activities on the Kola peninsula; the sunken nuclear submarine "Komsomolets"; plutonium contamination at the Thule, Greenland, accident site.

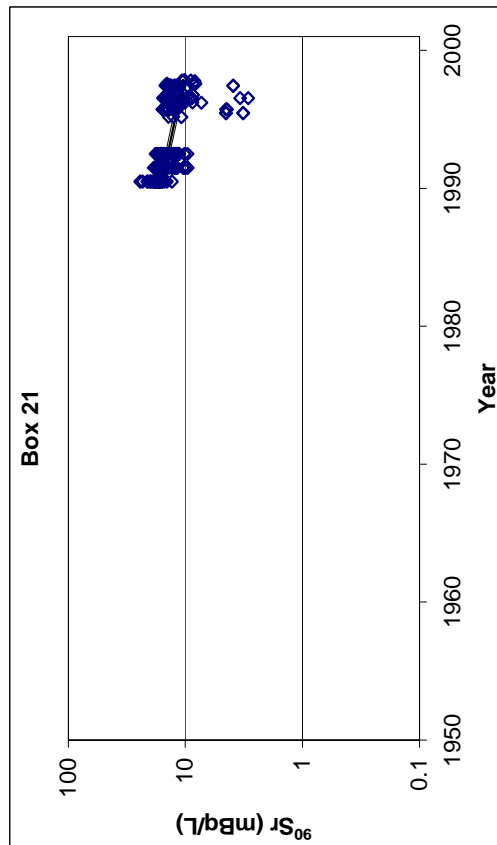
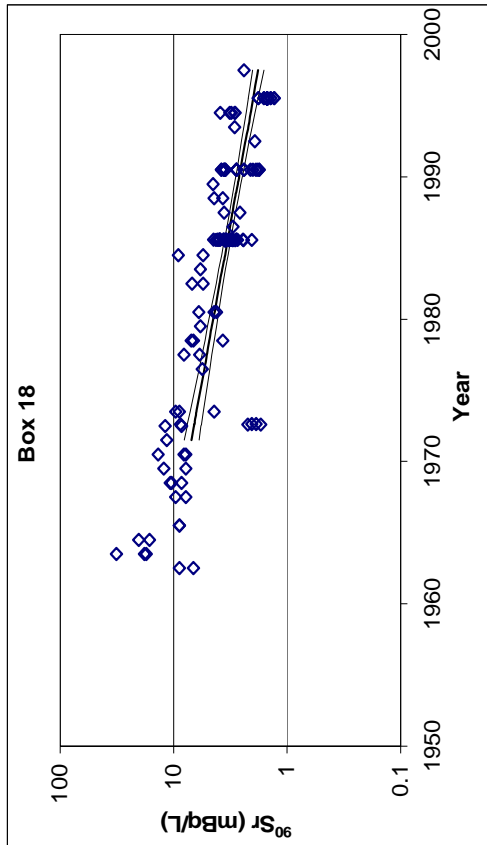
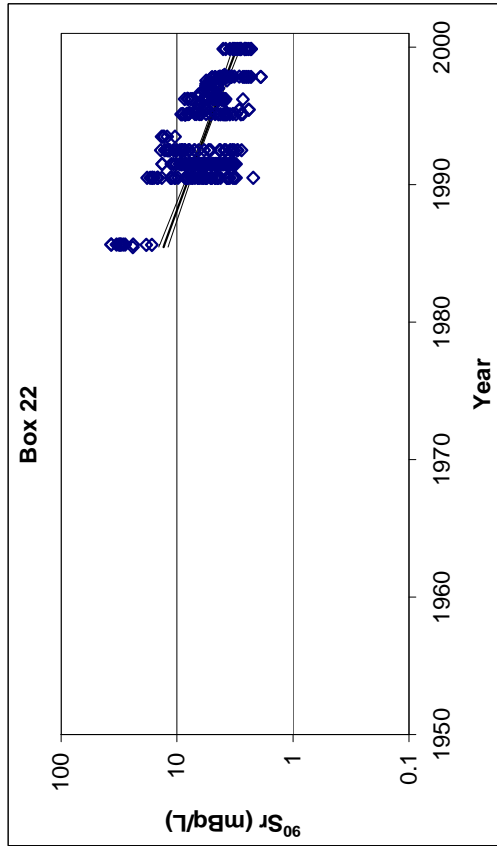
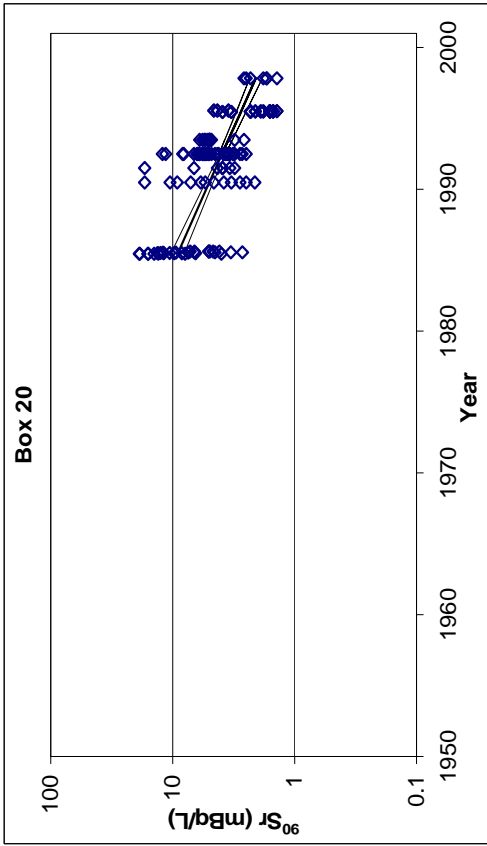


FIG. 101. ^{90}Sr surface water time series in the Atlantic Ocean.

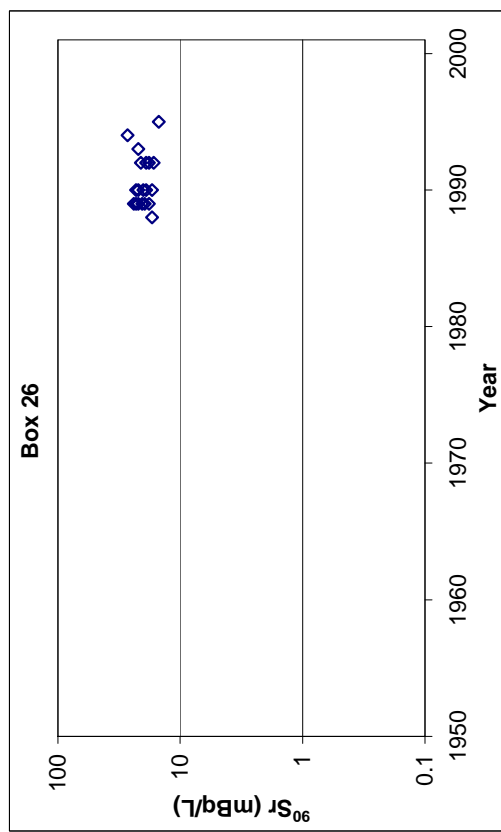
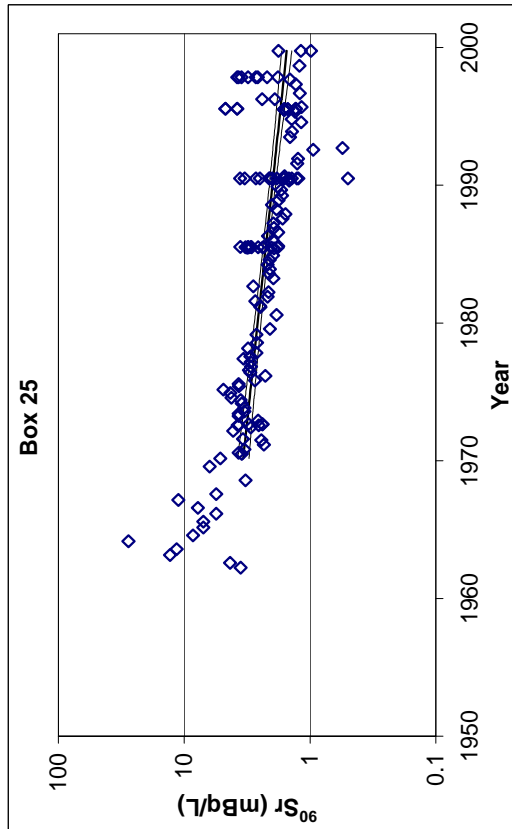
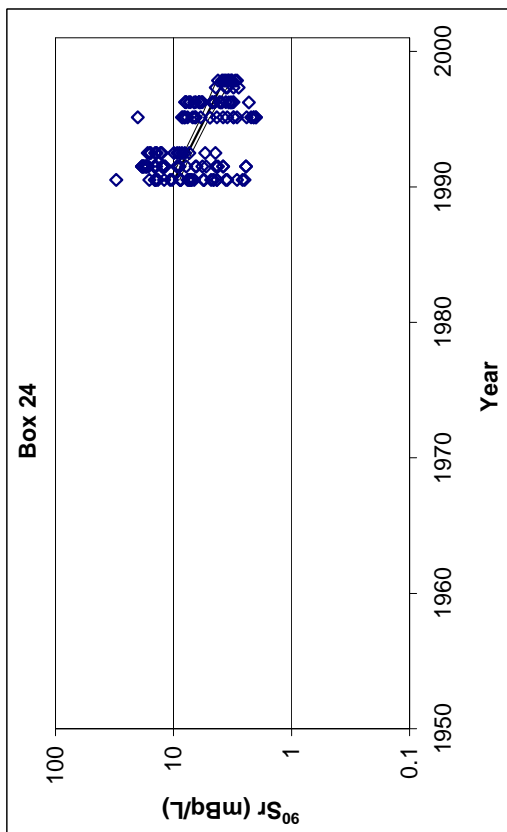
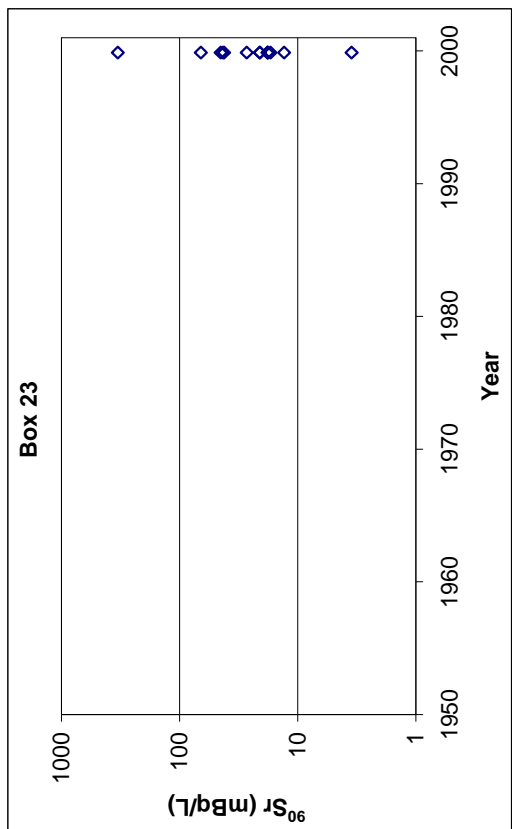


FIG. 102. ^{90}Sr surface water time series in the Atlantic Ocean.

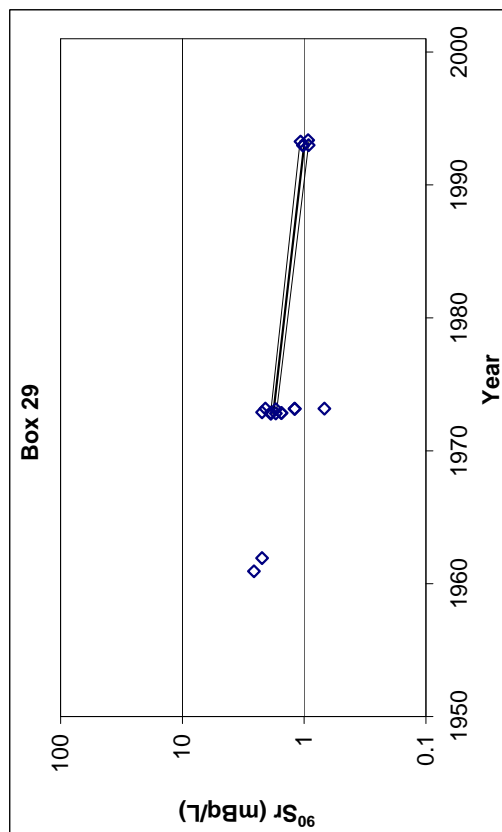
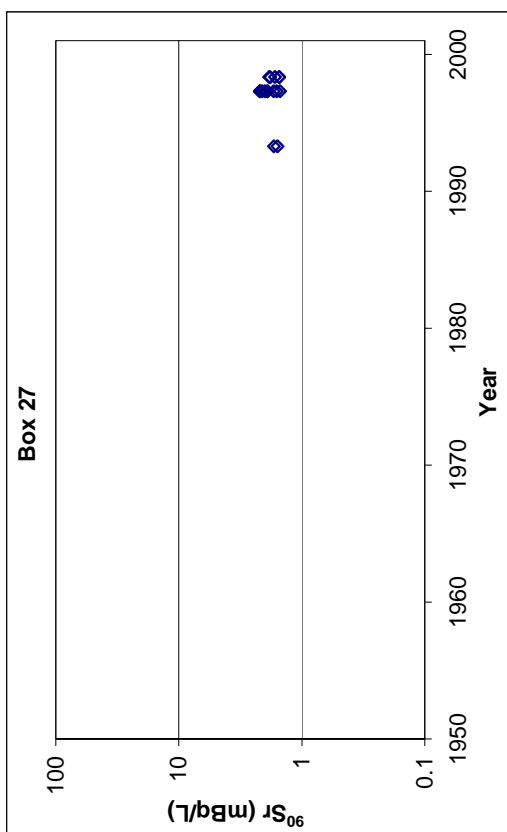
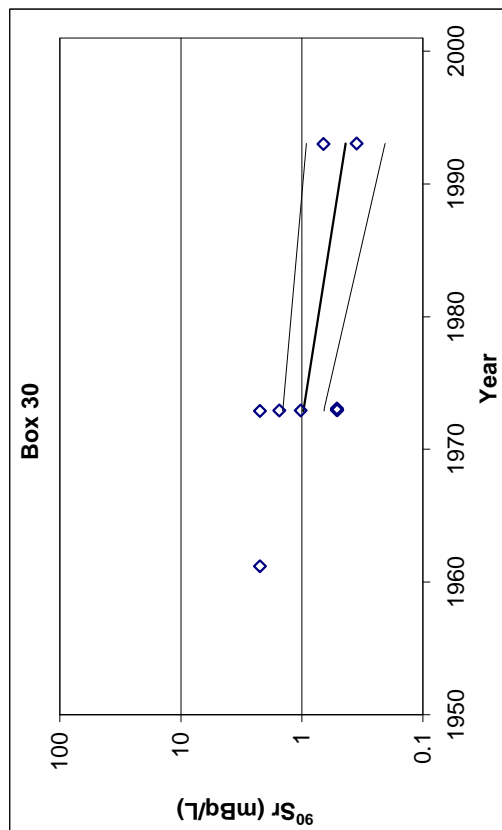
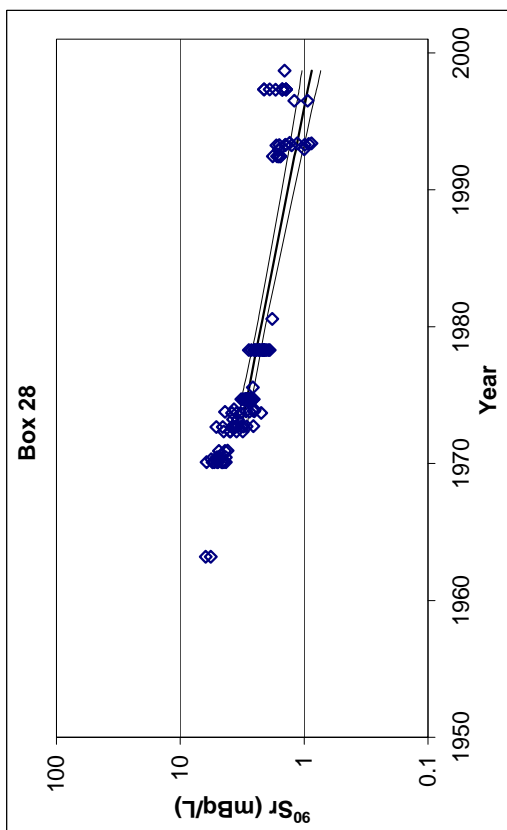


Fig. 103. ^{90}Sr surface water time series in the Atlantic Ocean.

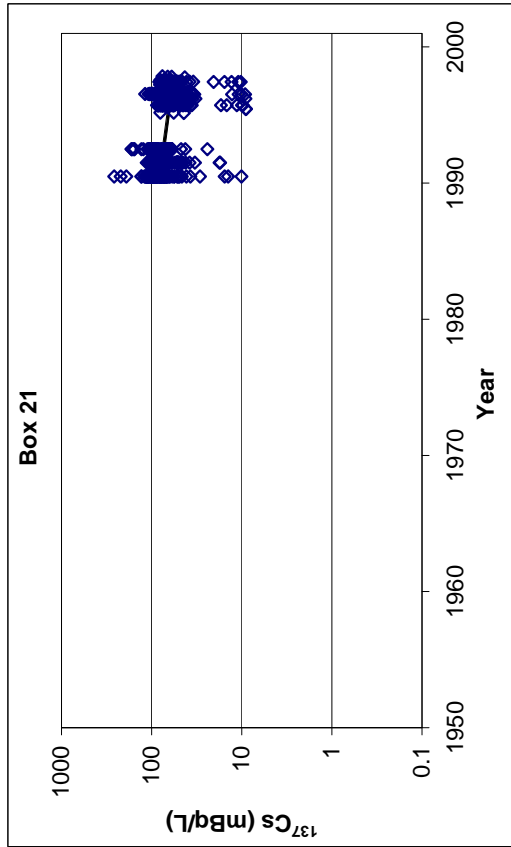
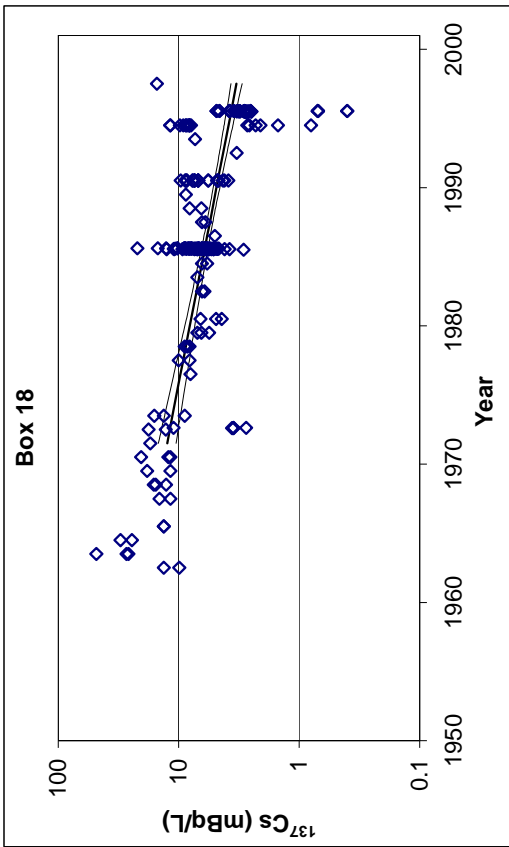
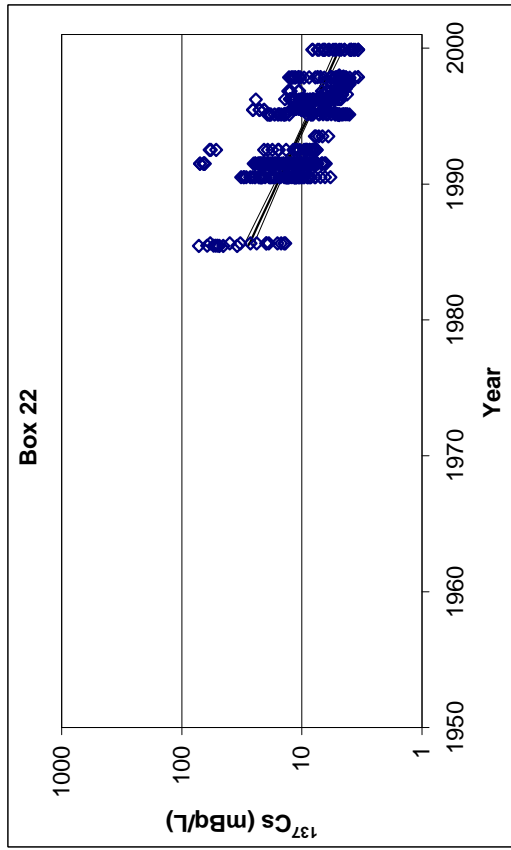
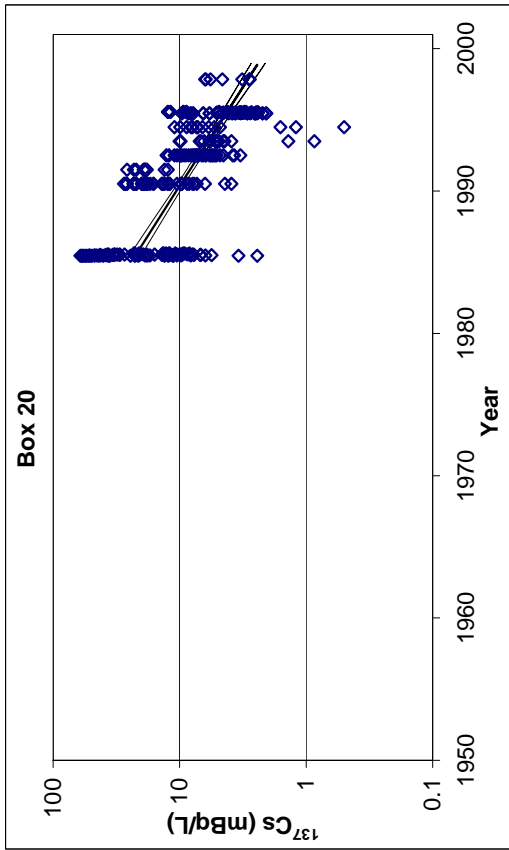


FIG. 104. ^{137}Cs surface water time series in the Atlantic Ocean.

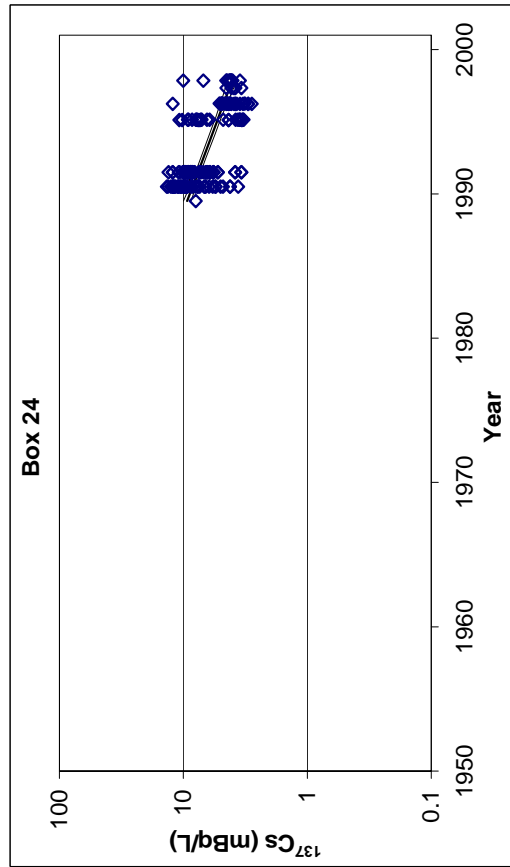
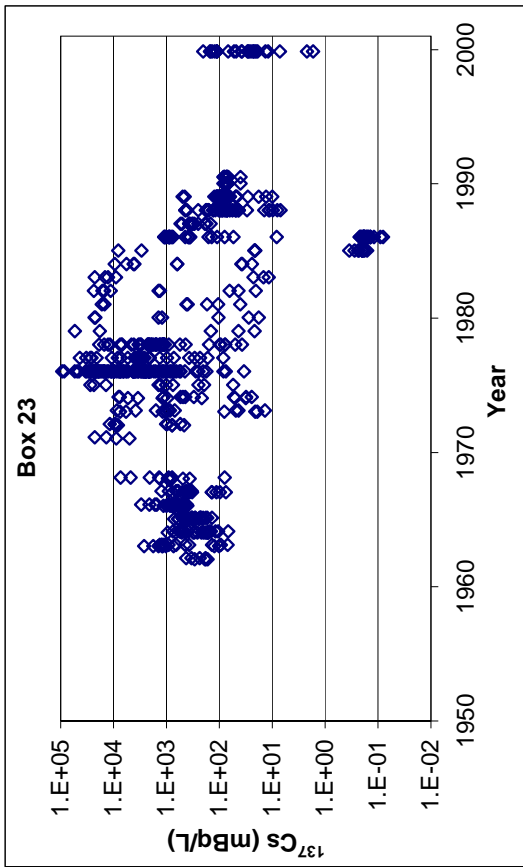
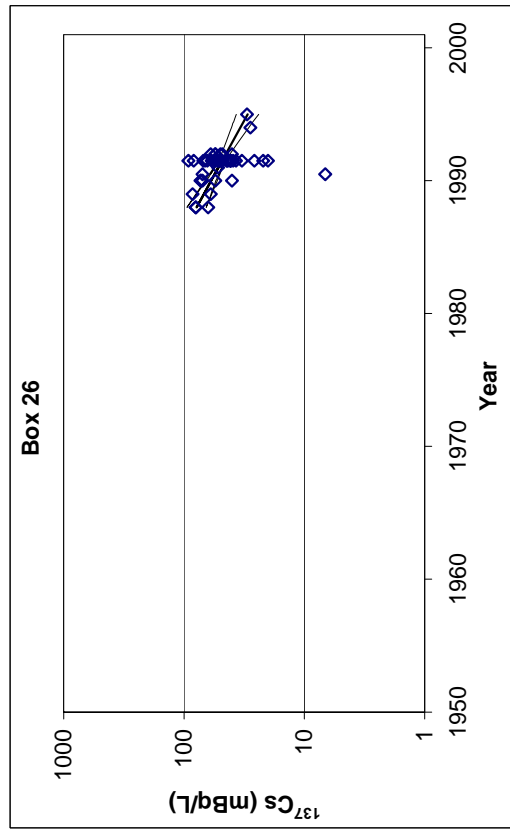
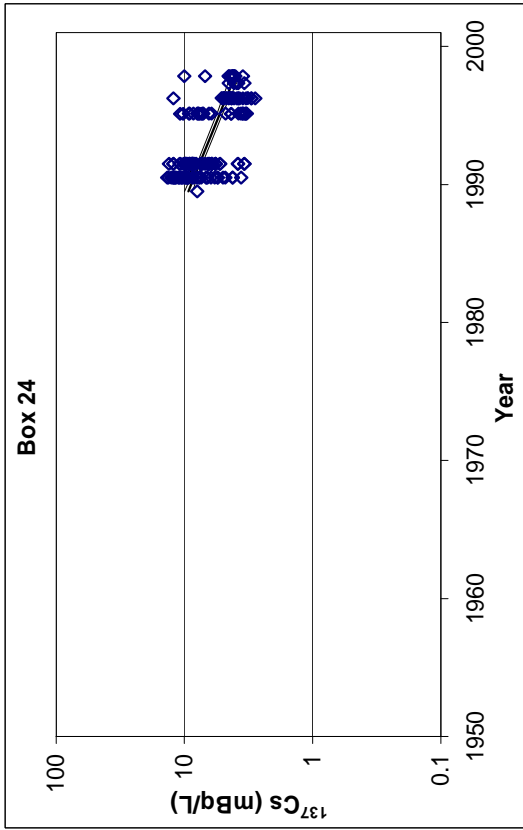


FIG. 105. ^{137}Cs surface water time series in the Atlantic Ocean.

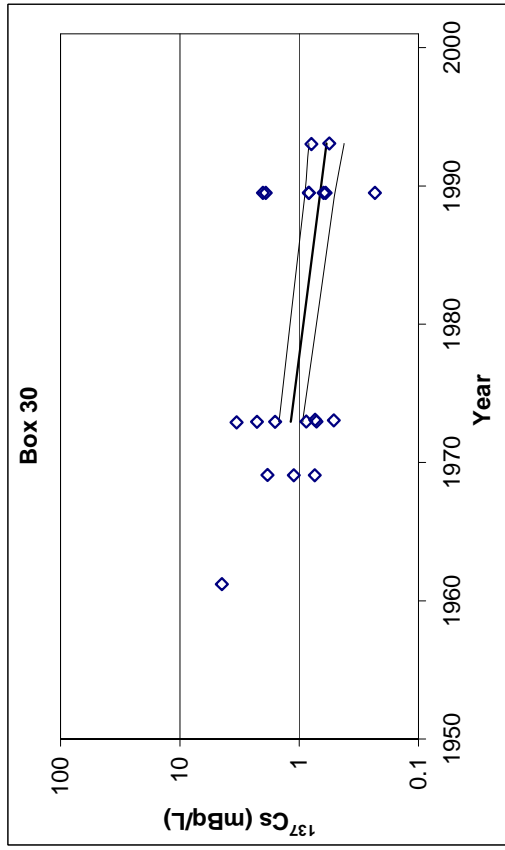
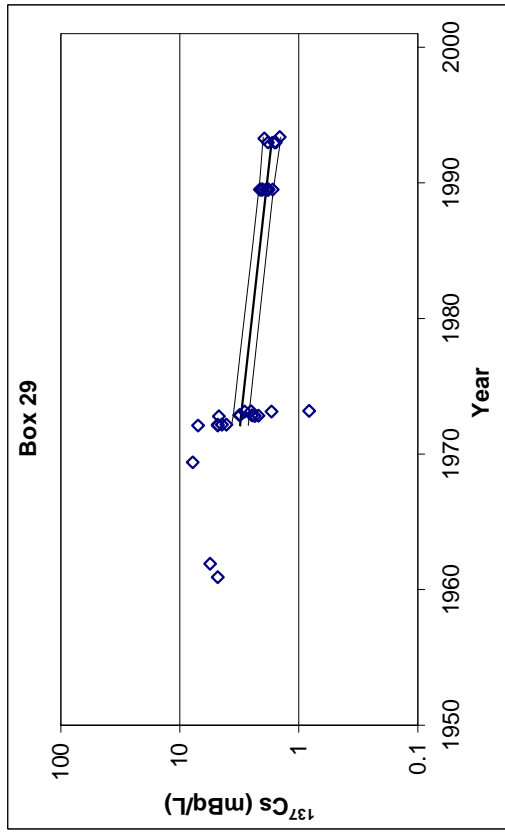
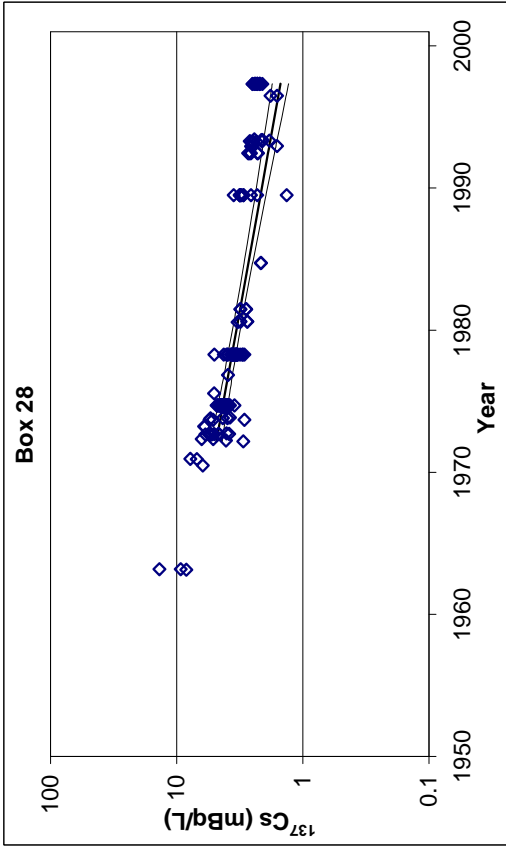
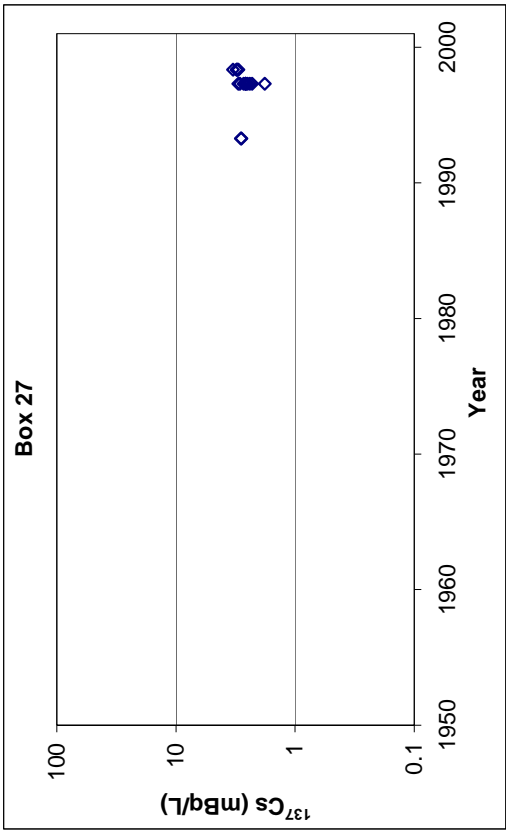


FIG. 106. ^{137}Cs surface water time series in the Atlantic Ocean.

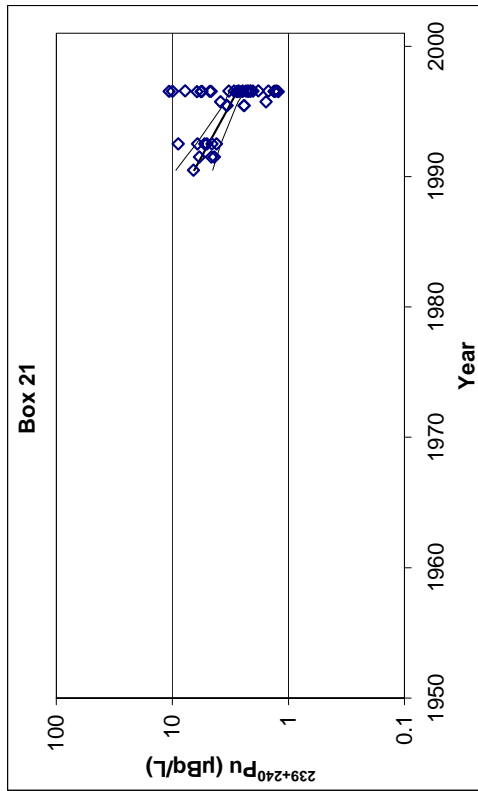
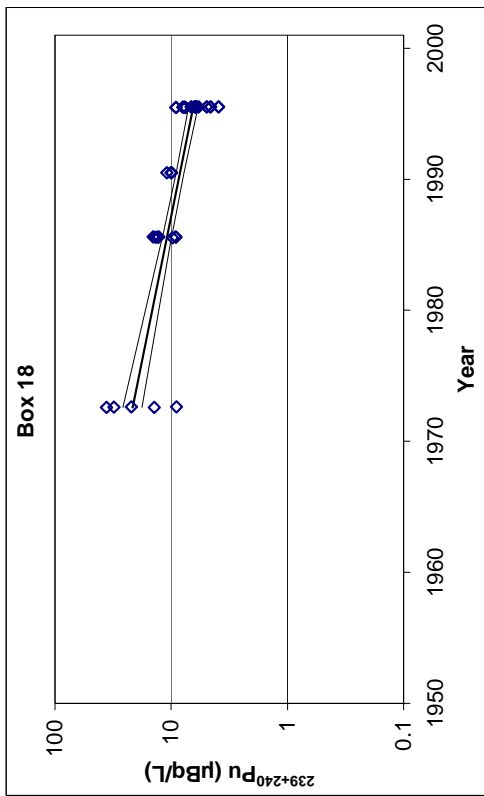
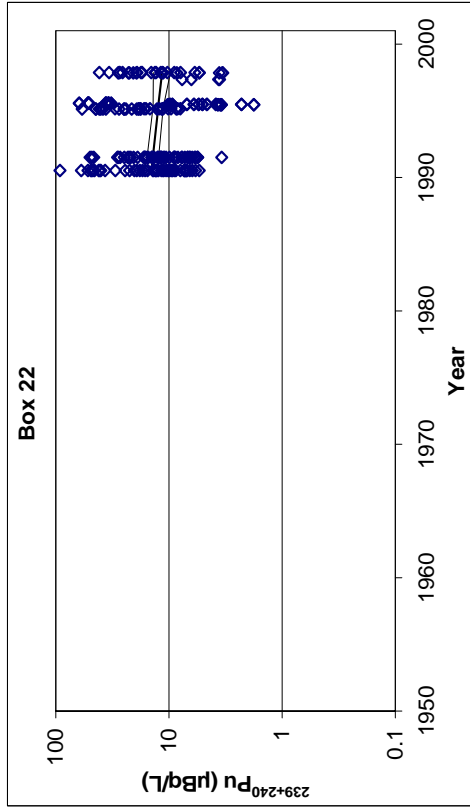
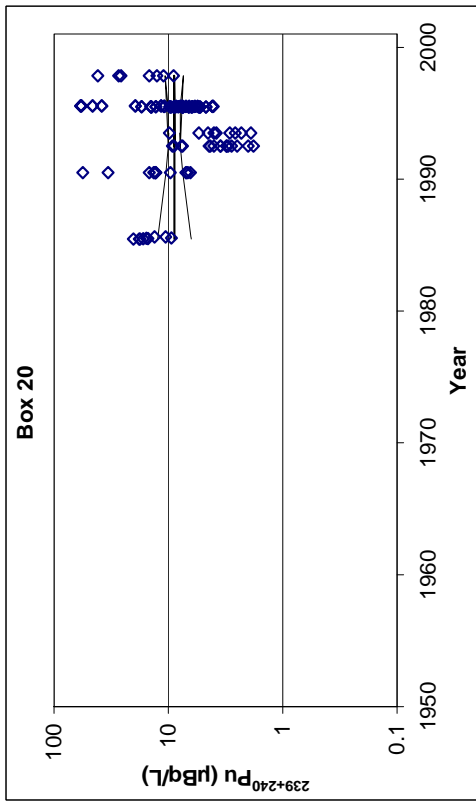


FIG. 107. $^{239,240}\text{Pu}$ surface water time series in the Atlantic Ocean.

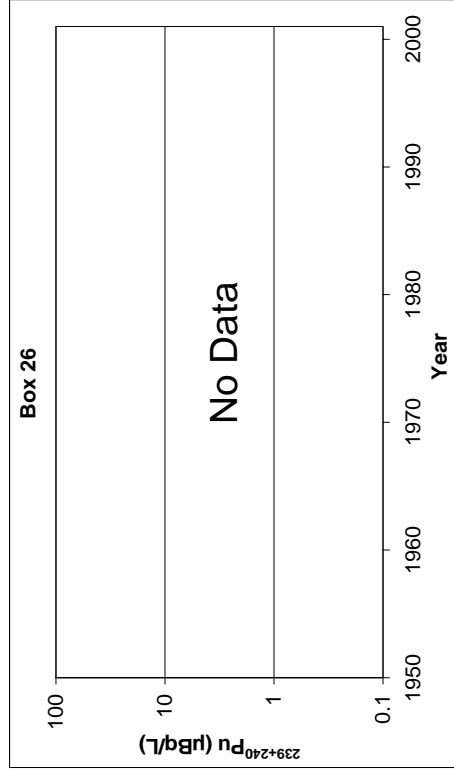
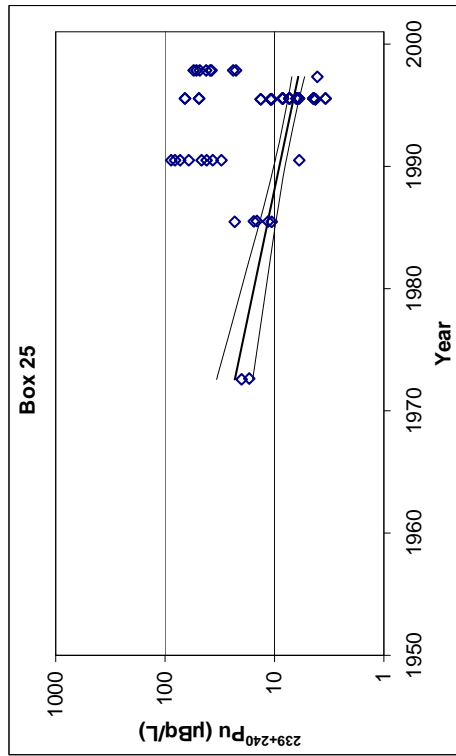
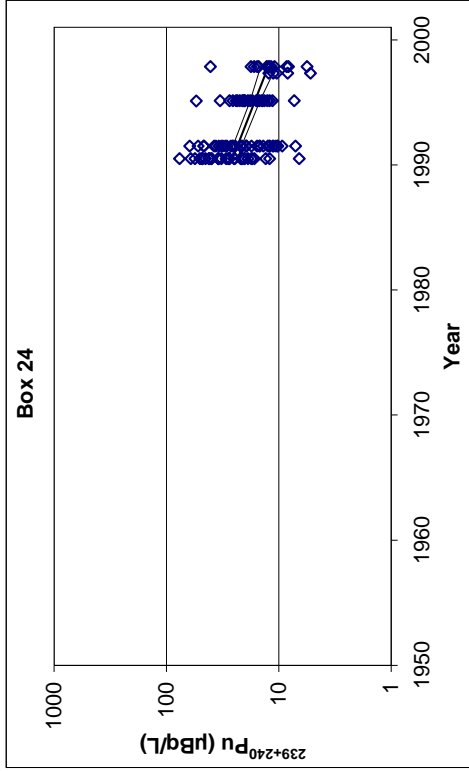
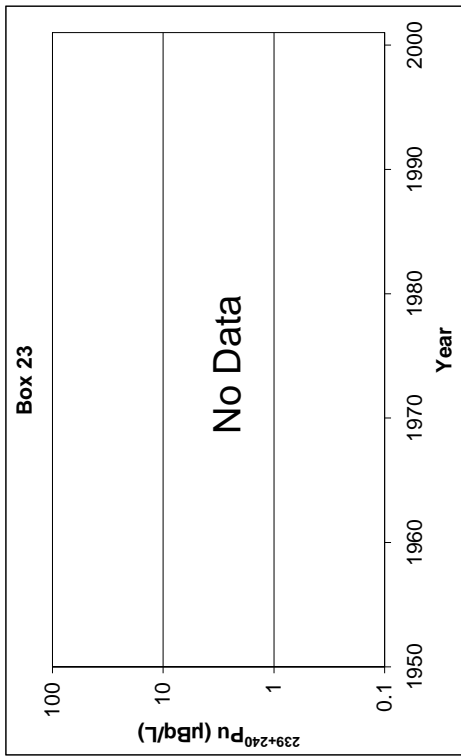


FIG. 108. $^{239,240}\text{Pu}$ surface water time series in the Atlantic Ocean.

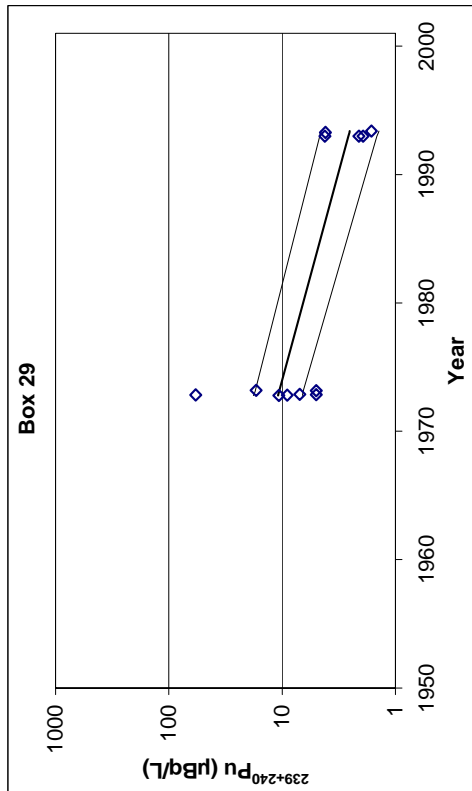
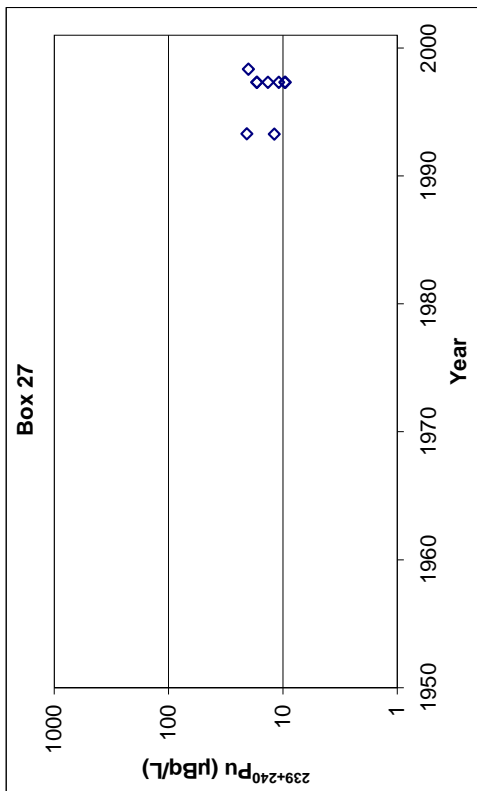
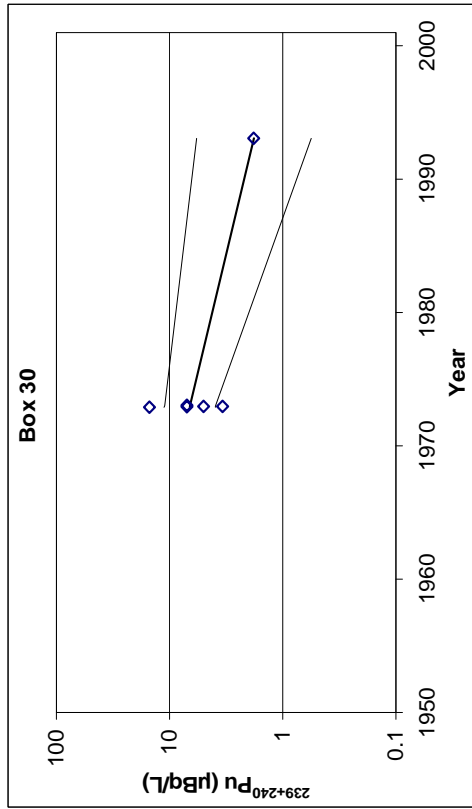
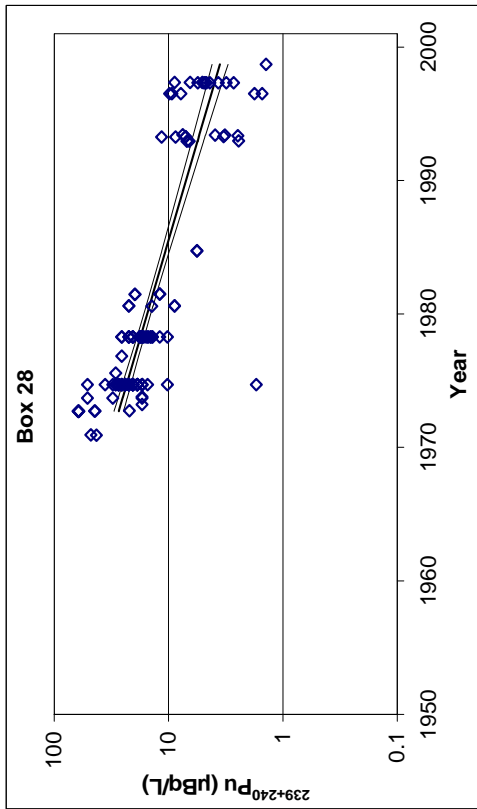


FIG. 109. $^{239,240}\text{Pu}$ surface water time series in the Atlantic Ocean.

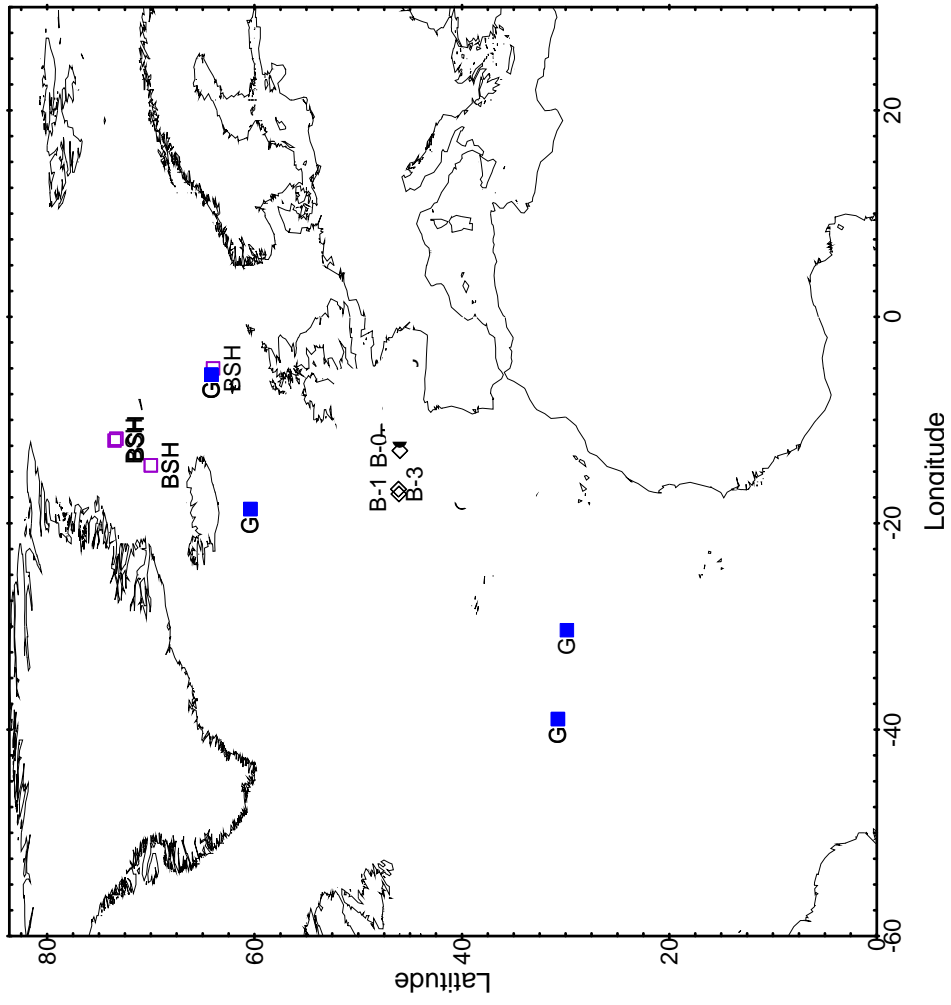


FIG. 110. Vertical profile stations in the North Atlantic (G-GEOSECS sites, BSH-BSH sites, B-IAEA sites).

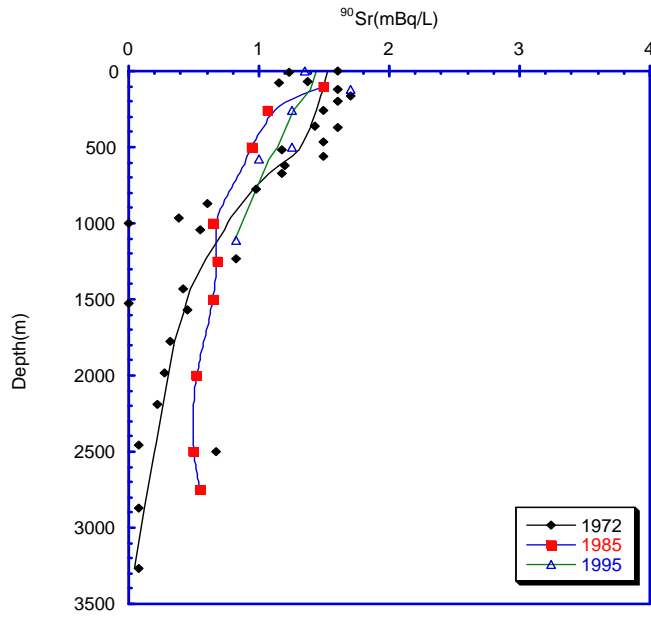


FIG. 111. ^{90}Sr water profiles time series in the North Atlantic (north of 60°N).

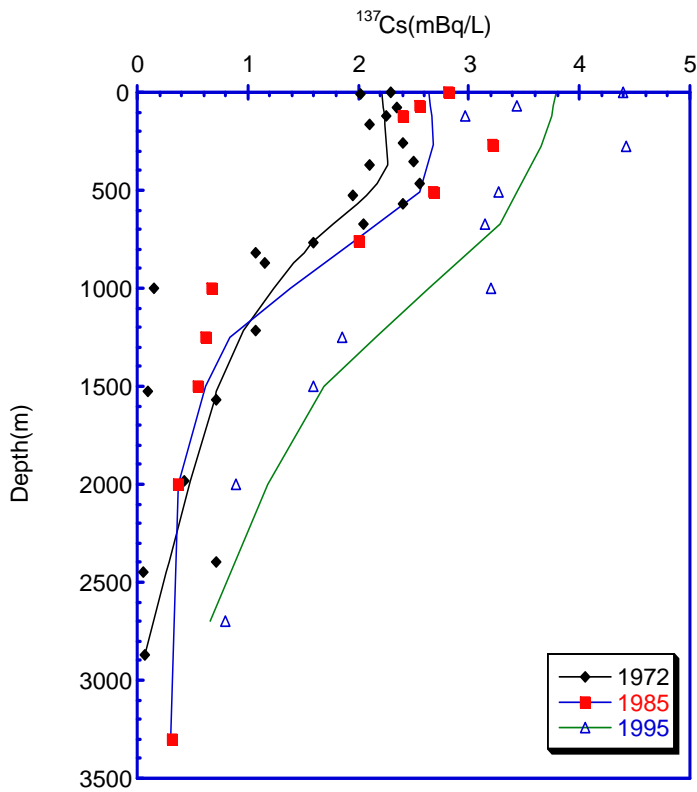


FIG. 112. ^{137}Cs water profile time series in the North Atlantic (north of 60°N).

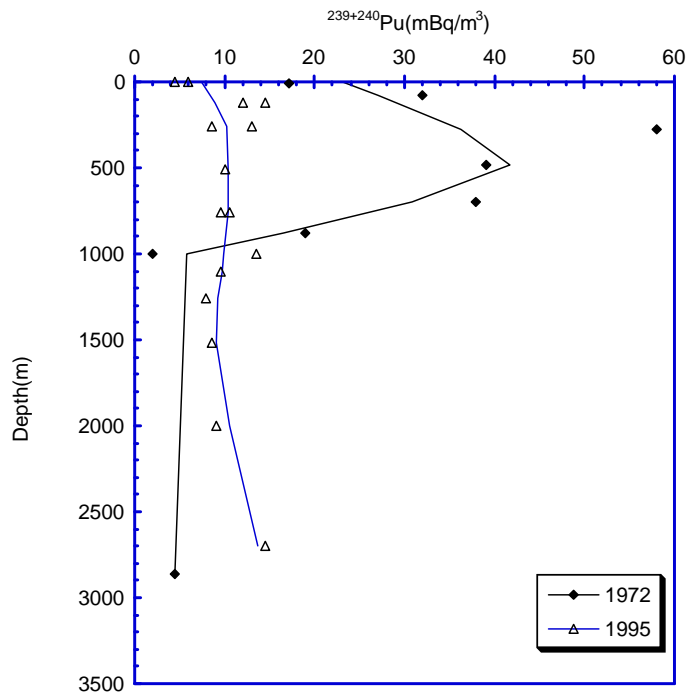


FIG. 113. $^{239,240}\text{Pu}$ water profile time series in the North Atlantic (north of 60°N).

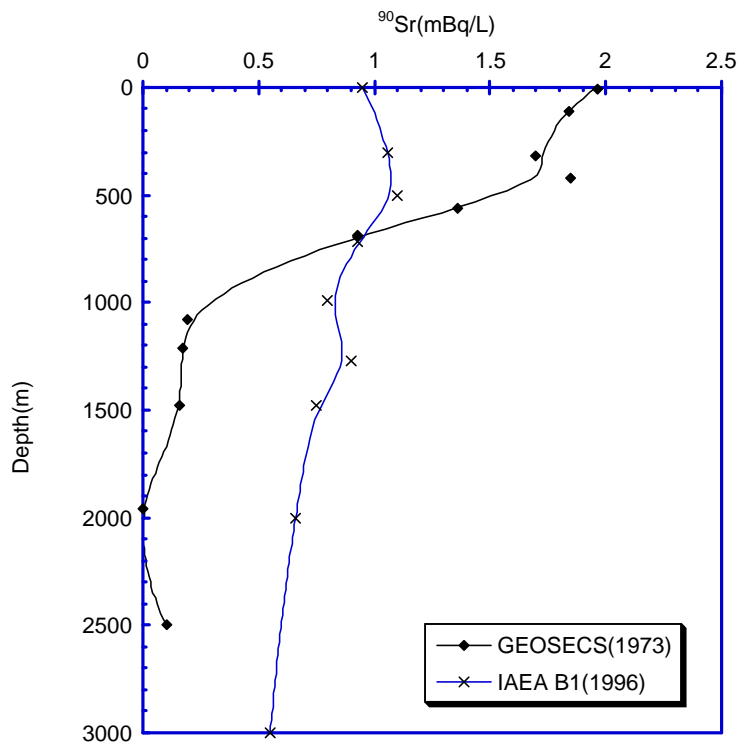


FIG. 114. ^{90}Sr water profile time series in the North Atlantic (south of 50°N).

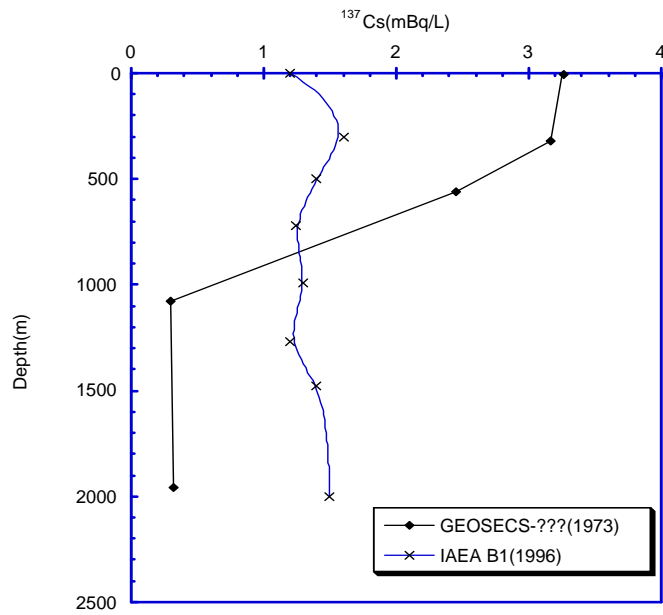


FIG. 115. ^{137}Cs water profile time series in the North Atlantic (south of 50°N).

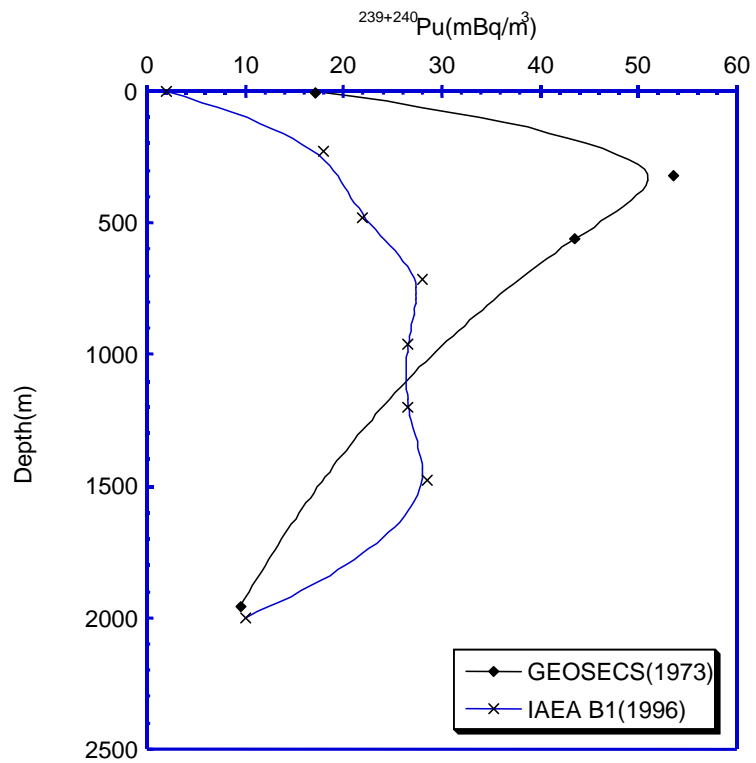


FIG. 116. $^{239,240}\text{Pu}$ water profile time series in the North Atlantic (south of 50°N).

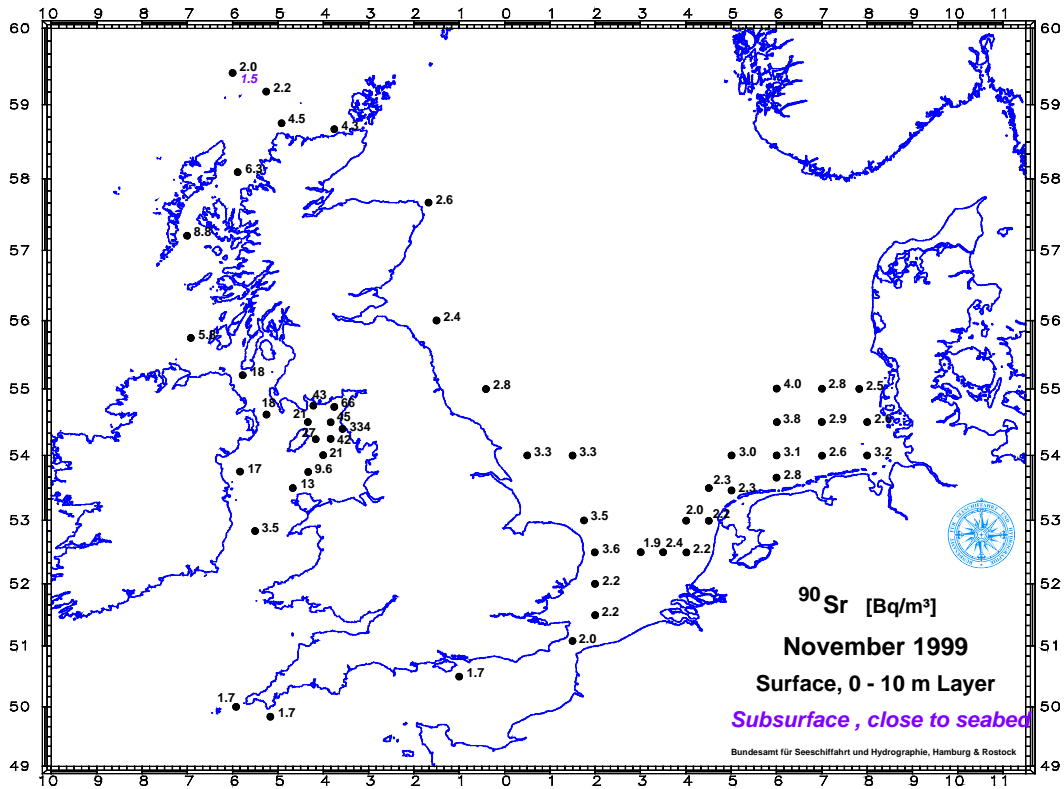


FIG. 117. ^{90}Sr distribution in water of the Irish and North Seas, including the English Channel in November, 1999. The relative uncertainty for the data is less than 5%.

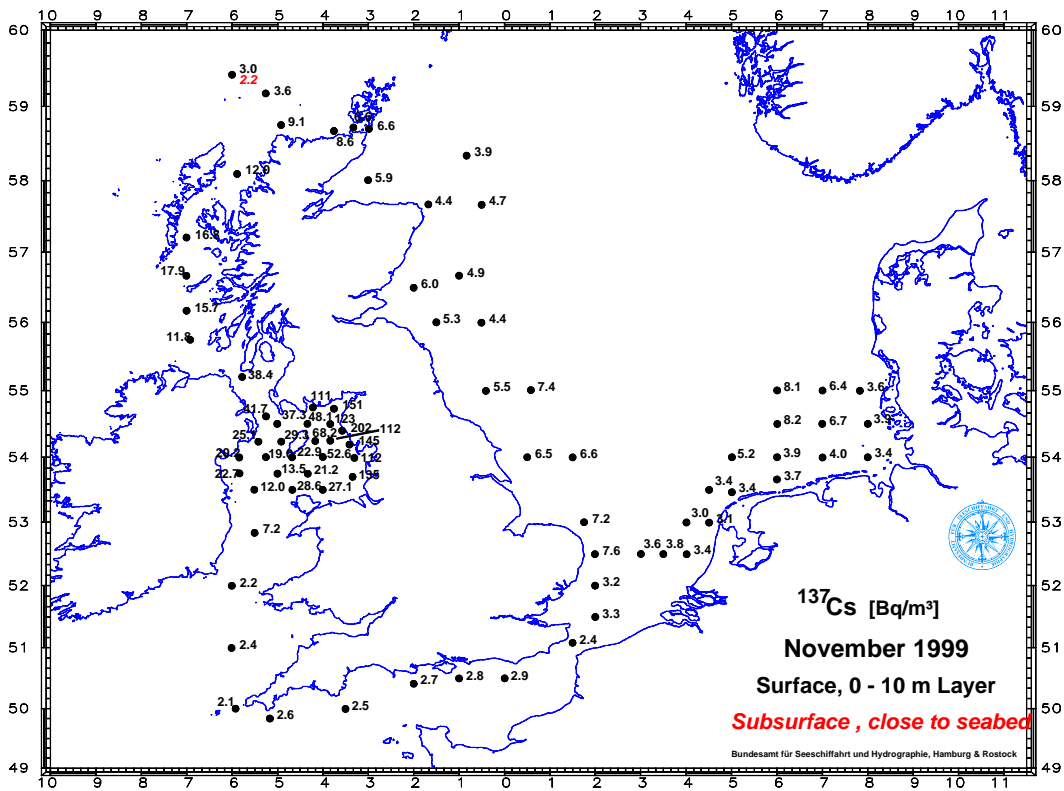


FIG. 118. ^{137}Cs distribution in water of the Irish and North Seas, including the English Channel, in November 1999. The relative uncertainty for the data is less than 5%.

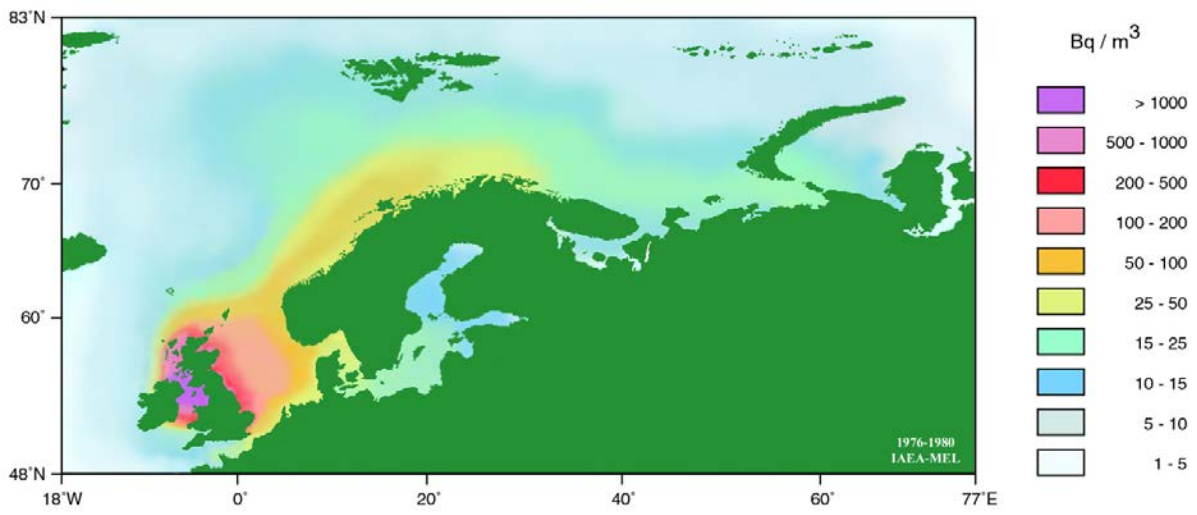


FIG. 119. ^{137}Cs in surface water of European seas (1976–1980).

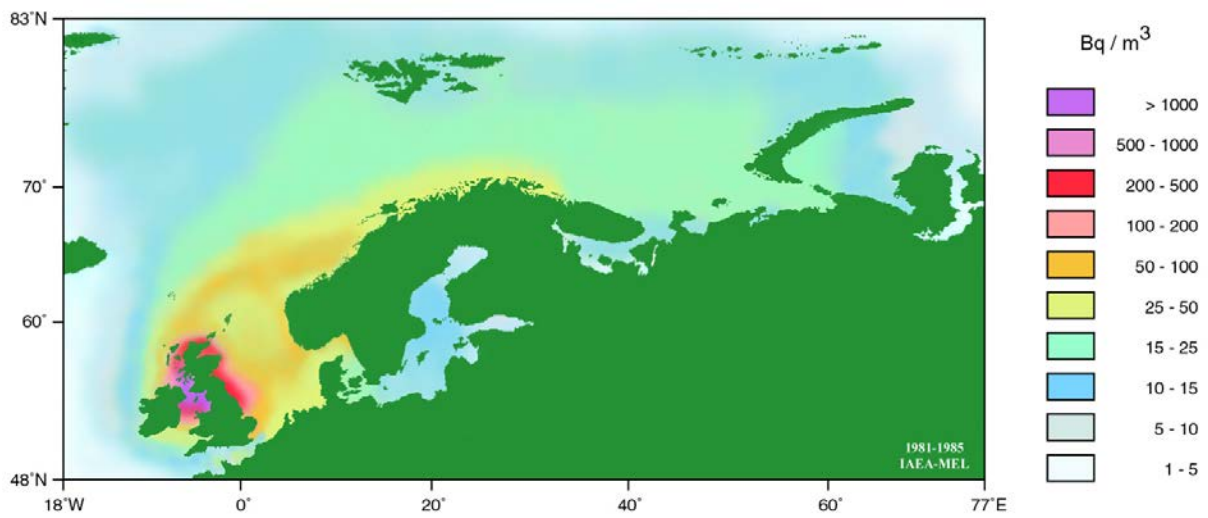


FIG. 120. ^{137}Cs in surface water of European seas (1981–1985).

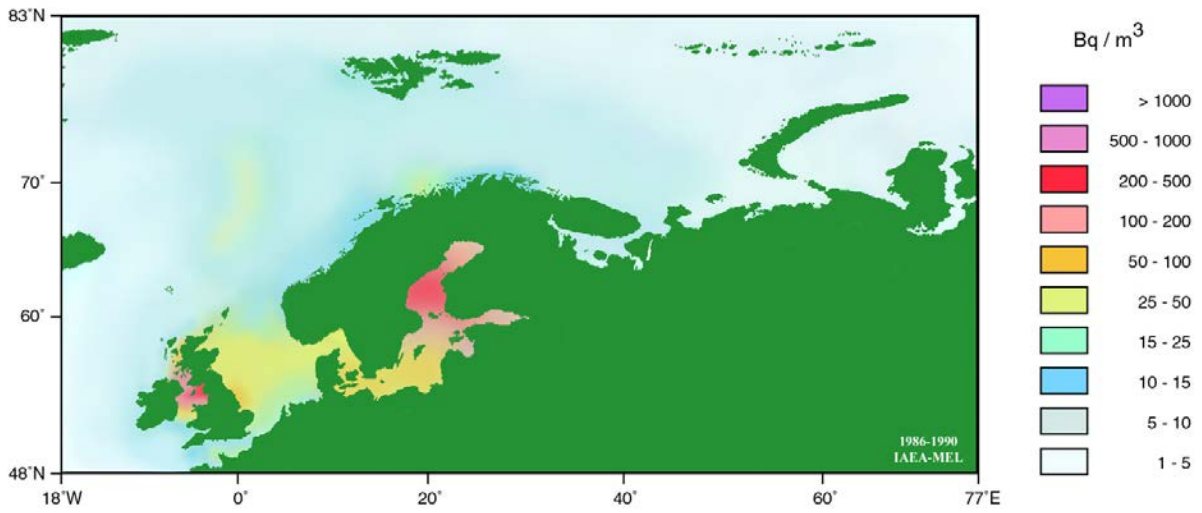


FIG. 121 ¹³⁷Cs in surface water of European seas (1986–1990).

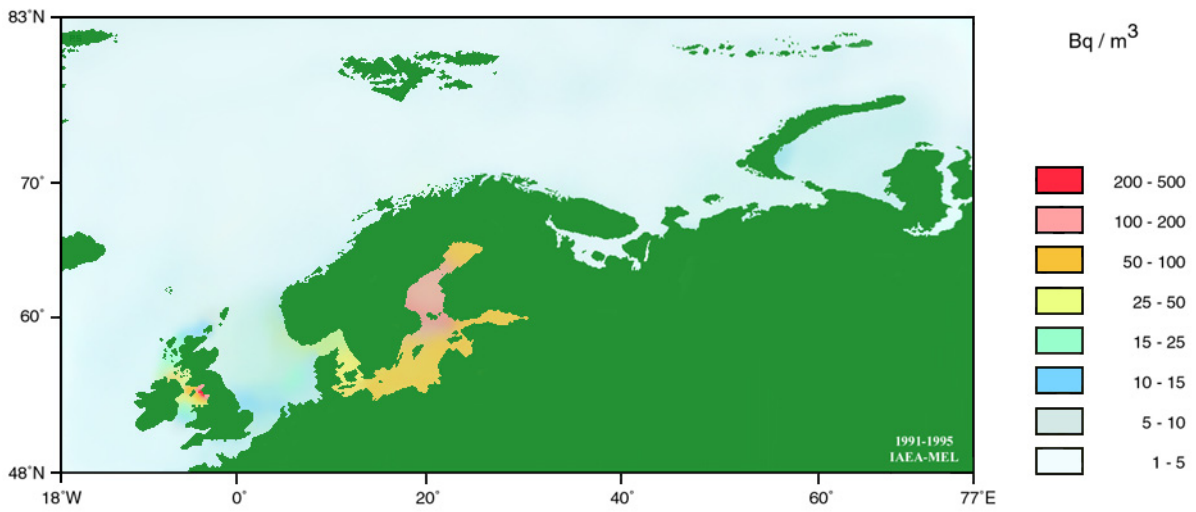


FIG. 122. ¹³⁷Cs in surface water of European seas (1991–1995).

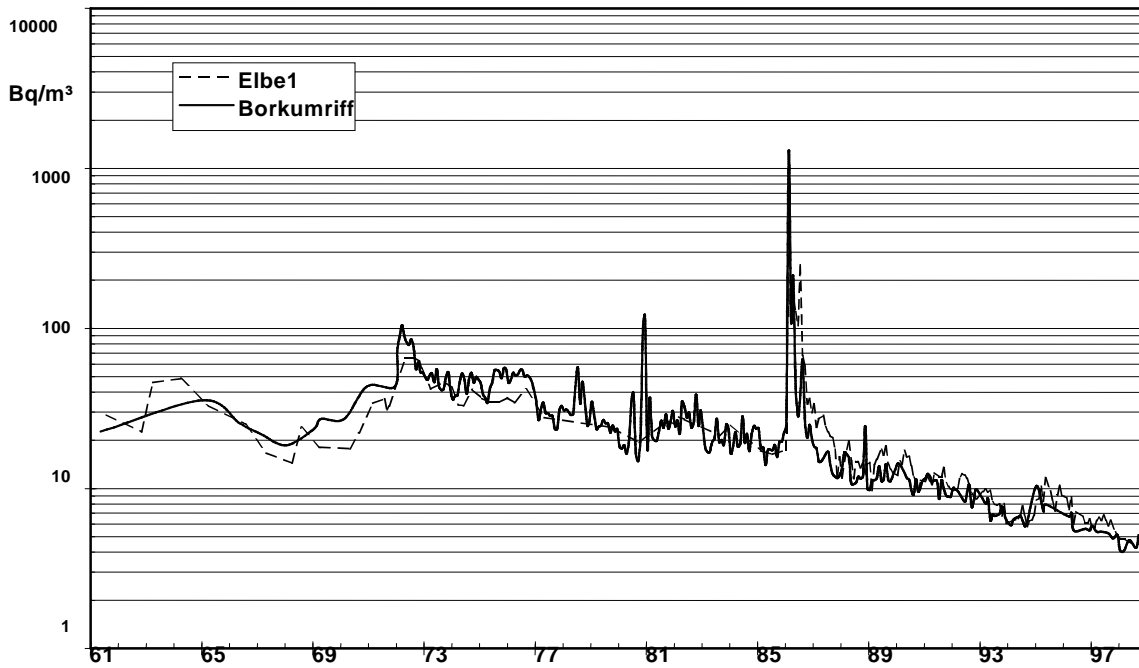


FIG. 123. Temporal trend of ^{137}Cs concentrations at two locations in the inner German Bight. This area is influenced by La Hague discharges.

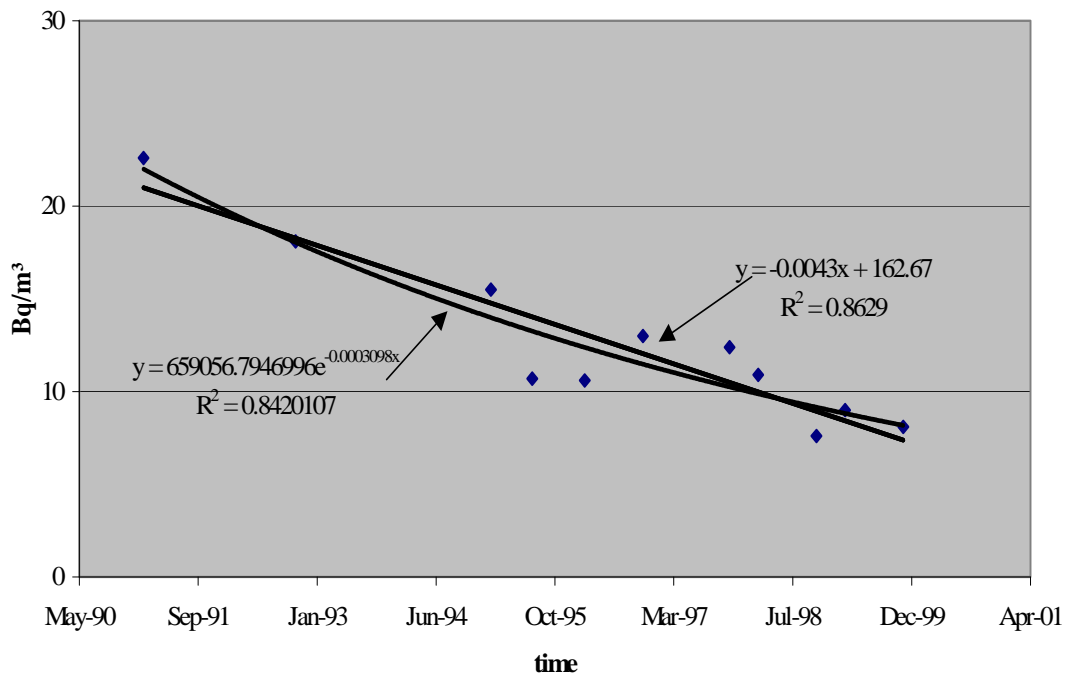


FIG. 124. Temporal trend of ^{137}Cs concentrations in the outer German Bight (55°N , 6°E). This location is influenced by Sellafield discharges.

Several cruises have recently been organized to northern European seas mainly by Germany and the UK [116–119].

6.4.1. *The Irish Sea*

The Irish Sea (Box 23) was recently monitored for ^{90}Sr , ^{137}Cs and ^{99}Tc in the autumn of 1999 by BSH on a cruise by the R/V "Gauss" in co-operation with British and Irish scientists. The distribution of the activity concentrations are given in Figs 117 and 118 for ^{90}Sr and ^{137}Cs , respectively.

The different sources of ^{90}Sr and ^{137}Cs in the Irish Sea can be identified by the activity ratio $^{137}\text{Cs}/^{90}\text{Sr}$. It becomes evident that the only position, where the typical discharge ratio as given by BNFL [120] is found is directly at the discharge area, where $^{137}\text{Cs}/^{90}\text{Sr} = 0.6$. In other areas this ratio is significantly different, proving that resuspension of sediment is the dominant source of ^{137}Cs at present in the Irish Sea.

As the distribution of ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ (Tables VI–VIII) in the Irish Sea has been controlled by discharges, effective half-lives were not calculated. The average ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ concentrations in the Irish Sea in the year 2000 have been estimated to be 50 Bq/m^3 , 60 Bq/m^3 and 500 mBq/m^3 , respectively.

^{137}Cs concentrations in surface water grouped for the periods 1976–80, 1981–85, 1986–90 and 1991–95 are shown in Figs 119, 120, 121 and 122, respectively, clearly indicating transport of ^{137}Cs from Sellafield reprocessing plant.

6.4.2. *The North Sea*

The North Sea (Box 22) receives contamination from the reprocessing plants at Sellafield and Cap de la Hague by the prevailing residual currents from the Irish Sea and the English Channel. Consequently, the concentrations of ^{137}Cs , ^{90}Sr and other radionuclides follow with delay the temporal trends of the discharges. The maximum discharge of ^{137}Cs and other β -emitting nuclides at Sellafield occurred in 1975; the highest concentrations of ^{137}Cs were measured around 1979 in the North Sea, with values of about 500 Bq/m^3 . After 1975, the discharges decreased steadily as mentioned in Chapter 2.2. Due to significantly decreasing discharges in recent years, the concentrations of ^{137}Cs , ^{90}Sr and transuranics are presently only slightly above the global fallout levels for North Atlantic surface water. Remobilization of ^{137}Cs from highly contaminated sediment in the Irish Sea is now the dominant source also for water contamination of the North Sea [121]. Levels of ^{137}Cs in November 1997 were between 3.5 Bq/m^3 in the southeastern North Sea and 12.4 Bq/m^3 in the central southern North Sea. The concentrations of ^{90}Sr were between 2.3 and 4.2 Bq/m^3 in the south, with lower levels in the north.

The general downward trend over recent years of ^{90}Sr and ^{137}Cs can be seen at several locations in the North Sea. This is due to both the physical decay of the global fallout level, decreasing discharges from the reprocessing plants at Sellafield and Cap de la Hague, as well as transport of these radionuclides to the north. The trend at two positions in the German Bight is shown in Fig. 123, whereas the outer German Bight (55°N , 6°E) reflects more the contamination from Sellafield about three years after the release into the Irish Sea (Fig. 124).

The Sellafield signal can also be detected in the North Sea for transuranic elements; the highest levels were found near the Scottish coast with $^{239,240}\text{Pu}$ concentrations of up to 55 mBq/m^3 in Pentland Firth in 1997. The concentrations decrease along the British east coast to

the southern central North Sea, where levels of less than 20 mBq/m³ are found. In the German Bight the values are even less than 10 mBq/m³. The activity ratio of ²³⁸Pu/^{239,240}Pu in the southern North Sea indicates a source of plutonium other than the Cap de la Hague plant, where the ratios are sometimes even higher than unity. In general, the activity ratios in the whole area indicate that the reprocessing plants are the dominant source of plutonium.

The ³H, ⁹⁰Sr, ¹³⁷Cs and ^{239,240}Pu surface water time series are shown in Figs 101, 104 and 107, respectively.

Combining all available data for ⁹⁰Sr, ¹³⁷Cs and ^{239,240}Pu, the effective half-lives could be calculated (keeping in mind that they could have been affected by releases from reprocessing plants and the Chernobyl accident (Tables VI–VII)). The effective half-lives for ⁹⁰Sr and ¹³⁷Cs are 6.9 ± 0.4 and 5.7 ± 0.3 years, respectively. The ^{239,240}Pu value is not sufficiently precise (24 ± 17 years). The average ⁹⁰Sr, ¹³⁷Cs and ^{239,240}Pu concentrations, estimated from recent measurements, are for 2000: 4, 4.5 Bq/m³ and 15 mBq/m³, respectively.

6.4.3. *The Baltic Sea*

The accident at Chernobyl deposited large quantities of radionuclides over the northern part of the Baltic Sea (Box 21). The levels of ¹³⁷Cs in the Baltic Sea (Fig. 125; [122]) were dominated by this fallout from 1986 onwards [5]. Before 1986, ¹³⁷Cs was due to global fallout and the inflow of North Sea water through the Danish Straits. Since 1986, a great proportion of ¹³⁷Cs was deposited in the fine grained sediment of the Baltic Sea. The inventory in the water phase has decreased by about a factor of 2 since then. The concentrations of ⁹⁰Sr have remained fairly unchanged over the years and are in the range between 10 and 15 Bq/m³ in surface water of the Baltic Sea. The main source of this radionuclide is global fallout, whereas the activity ratio of ¹³⁷Cs/⁹⁰Sr is not typical due to certain inputs from rivers. This mechanism is almost negligible for ¹³⁷Cs.

Due to the permanent halocline in the Baltic Sea there still exists a significant difference in the concentrations of ¹³⁷Cs in surface water and in the deeper layers below the halocline. This is true in particular for the Belt Sea (western Baltic) where the inflow from the North Sea with lower levels occurs below the halocline and the outflow of low salinity water at the surface carries higher contamination levels. In addition, Chernobyl fallout was very unevenly distributed. However, mixing of water masses has led to a more homogeneous distribution in different parts of the Baltic over years. The temporal trend in ⁹⁰Sr and ¹³⁷Cs concentrations at one station in the western Baltic Sea is shown in Fig. 126.

The ⁹⁰Sr, ¹³⁷Cs and ^{239,240}Pu surface water time series are given in Figs 101, 104 and 107, respectively. The calculated effective half-lives for ⁹⁰Sr, ¹³⁷Cs and ^{239,240}Pu in the Baltic Sea are 12.0 ± 1.7, 14.1 ± 1.8 and 4.9 ± 1.4 years, respectively. The average concentrations of ⁹⁰Sr, ¹³⁷Cs and ^{239,240}Pu in surface water estimated from recent measurements for the year 2000 are around 10 Bq/m³, 60 Bq/m³ and about 3 mBq/m³, respectively (Tables VI–VIII).

6.4.4. *The Arctic Seas*

The Arctic Seas (the Norwegian, Barents and Kara Seas) were recently investigated in detail in order to study potential transport of radioactive contaminants from nuclear dumpsites in the Kara and Barents Seas into the NE Atlantic [12, 115]. The levels of ¹³⁷Cs were

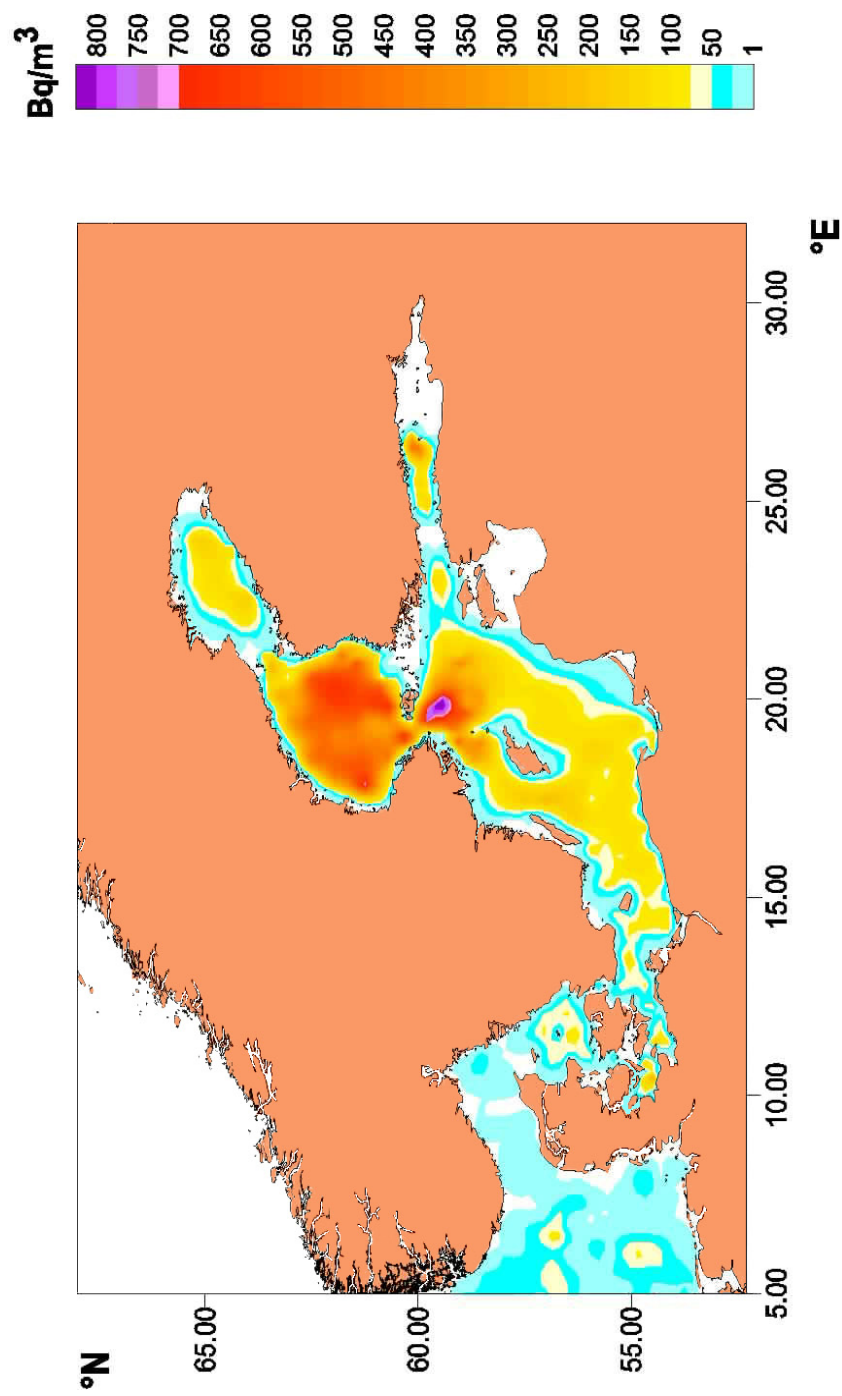


FIG. 125. ^{137}Cs contours of Baltic Sea surface waters after the Chernobyl accident (for the period 1986–1988) as extracted from the GLOMARD database.

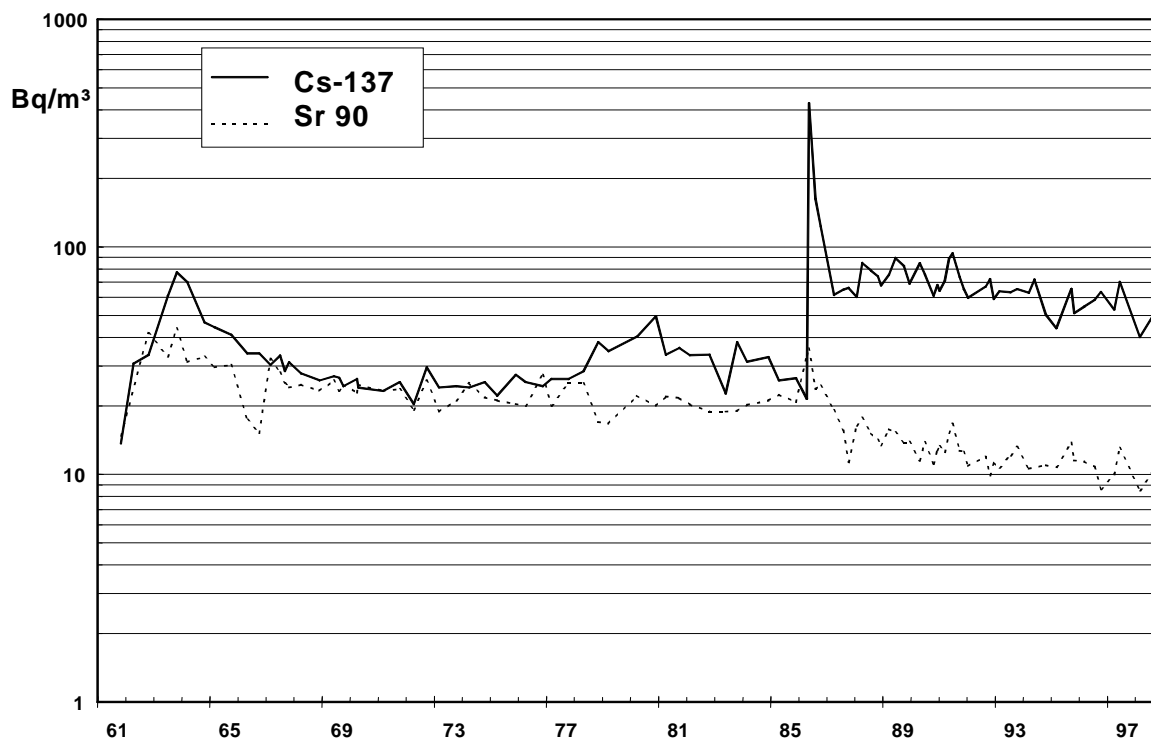


FIG. 126. Temporal trends of ^{137}Cs and ^{90}Sr in the western Baltic Sea.

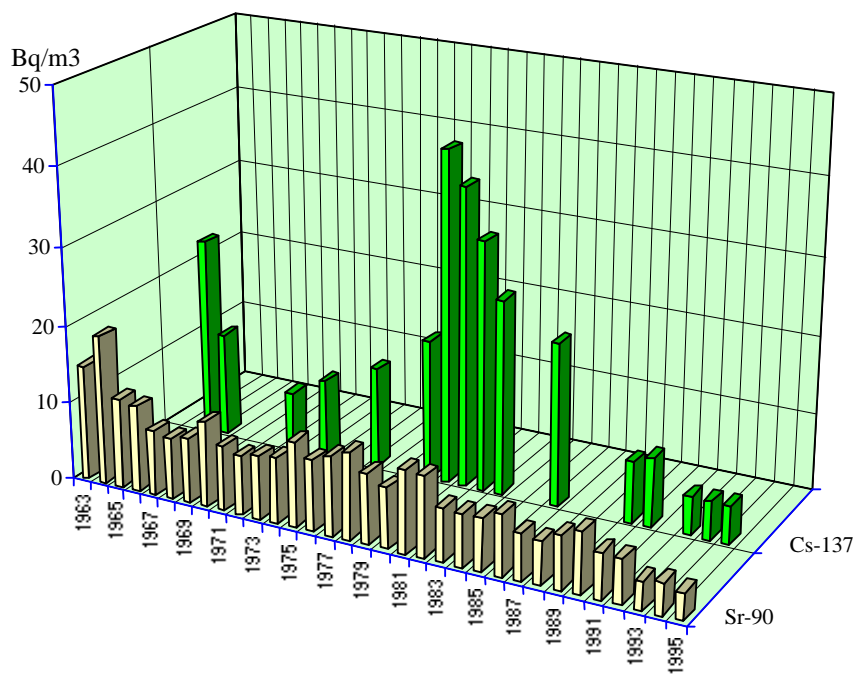


FIG. 127. ^{90}Sr and ^{137}Cs in Barents Sea surface water as extracted from the GLOMARD database.

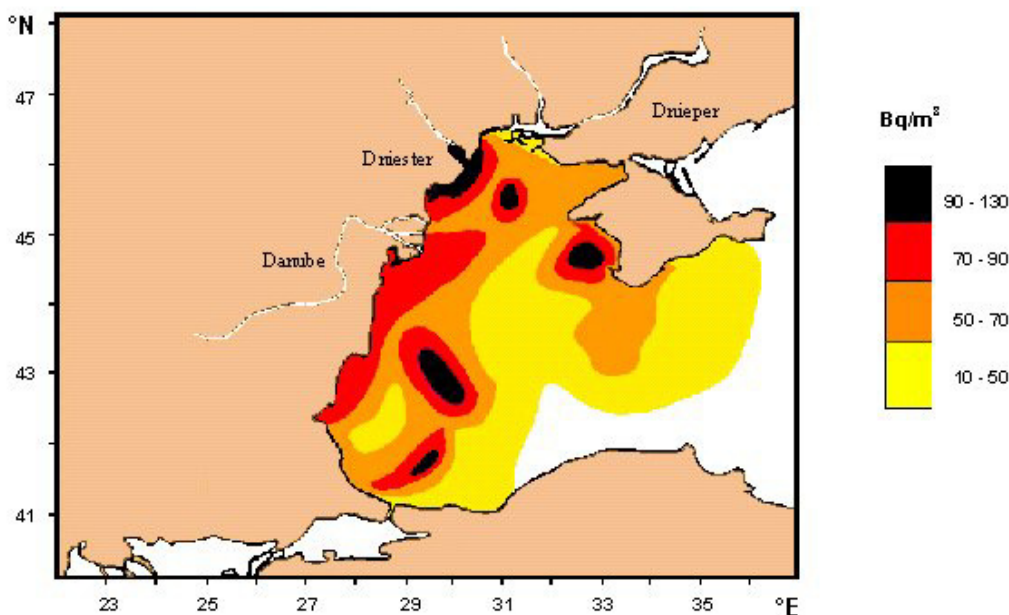


FIG. 128. ^{137}Cs contours of western Black Sea surface water after the Chernobyl accident in 1988, as extracted from the GLOMARD database.

significantly lower than in 1985 due to lower levels in the North Sea as a consequence of decreased discharges at Sellafield. It was also found that at deeper water layers, Sellafield-derived transuranics identified by their activity ratios are transferred into the Norwegian Sea [123].

In Arctic waters there is no indication of long range contamination from nuclear wastes and other radioactive objects dumped by the former USSR. Samples taken in its vicinity during 1992 and 1995 indicate that there is also no long range contamination from the sunken nuclear submarine "Komsomolets" in the northern Norwegian Sea. Except for global fallout, the main source of radionuclides in the Arctic Seas remains the contributions from nuclear reprocessing plants. Fig. 127 confirms this statement showing the time evolution of ^{90}Sr and ^{137}Cs activities in Barents Sea surface water.

The distribution maps and average ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ concentrations for the Arctic Seas are given in Figs 94–96 for 1991–1995 and 98–100 for 1996–2000, respectively. The corresponding time series (Box 20) are shown in Figs 101, 104 and 107. The calculated effective half-lives for ^{90}Sr , ^{137}Cs in the Barents Sea are 5.8 ± 0.5 and 4.2 ± 0.2 years, respectively. The average ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ concentrations estimated from recent measurements are for the year 2000 around 2 and 3 Bq/m^3 , and 20 mBq/m^3 , respectively (Tables VI–VIII).

6.5. Radionuclides in the Mediterranean Sea

The Mediterranean Sea is an enclosed, deep basin that communicates with the Atlantic Ocean through the narrow, shallow Strait of Gibraltar. Here a surface flux of Atlantic, low salinity water (36–36.5‰) enters the Mediterranean Sea, while a subsurface water mass, characterized by salinity higher than 37.5‰ leaves the basin. The entire Mediterranean is a concentration basin, with evaporation exceeding precipitation and runoff. Due to the increase in salinity and consequent density instabilities, deep and intermediate waters are formed by vertical convection at some key sites, as discussed below.

The general circulation of the basin is complex and can be summarized as follows: the major phenomenon influencing the surface circulation is the Atlantic water entering via the Strait of Gibraltar. After some meandering in the Alboran Sea, this water mass proceeds eastward, along the north African coast. After crossing the Sicilian Sill, the Atlantic stream occupies the southern part of the Ionian Sea, becoming an intensified mid-Mediterranean jet. In the area between Crete and Rhodes, extensive vertical convection gives rise to the formation of the saltier and denser Levantine Intermediate Water (LIW) that stabilizes at 200–600 m depth, moves westward and passes over the Sicilian Sill into the western basin. After following a rather complicated path throughout the basin, the LIW finally flows, mixed with some Western Mediterranean Deep Water, out of the Mediterranean, crossing the Strait of Gibraltar at an average depth of more than 150 m.

The deepest Mediterranean water is mainly formed in areas where prolonged periods of cold winds sharply increase the evaporation rates and produce, at the surface, a dense, cold, well oxygenated water mass that sinks rapidly, reaches the bottom and spreads all over the basin.

The Gulf of Lions, the Liguro-Provençal Basin, the Adriatic and the Cretan Seas have been identified as the most important sites for deep-water formation. More recently, important changes have been registered in deep-water formation and circulation in the Eastern Basin, with deep and intermediate water masses also being formed in the Aegean Sea.

The major source of anthropogenic radionuclides to the Mediterranean Sea is fallout from atmospheric weapons testing in the early sixties (12 PBq of ^{137}Cs and 0.19 PBq of $^{239,240}\text{Pu}$) [124–126]. Fallout from the Chernobyl accident contributed significantly to the total input of caesium isotopes. It is estimated that 3 to 5 PBq of ^{137}Cs were deposited after the accident, mainly in the eastern and northern parts of the basin. An additional input of Chernobyl radionuclides came through the Black Sea (0.3 PBq of ^{137}Cs) [127]. Figure 128 documents the ^{137}Cs levels in the western Black Sea in 1988 as obtained from the GLOMARD database [128]. The input deriving from the nuclear industry is very small compared to other sources. The Marcoule (France) reprocessing plant is authorized to discharge low level effluents into the Rhone river which flows into the NW Mediterranean. The discharges up to 1995 amount to a total of 30.2 TBq of ^{137}Cs and 0.26 TBq of plutonium isotopes.

6.5.1. Surface water radionuclide time series

No time series stations have been established in the Mediterranean Sea for anthropogenic radionuclide concentrations in open waters. However, Fukai et al. [129] showed that before the Chernobyl accident there were no significant differences in surface ^{137}Cs concentrations in the different basins of the Mediterranean Sea. In 1977, the mean values ranged from $4.4 \pm 0.7 \text{ Bq/m}^3$ in the SW Mediterranean and in the Tyrrhenian Sea, to $4.8 \pm 1.8 \text{ Bq/m}^3$ in the NW Mediterranean, Adriatic and Ionian Seas. Slightly lower values were found only in the SE Mediterranean basin ($4.2 \pm 0.3 \text{ Bq/m}^3$).

Also for plutonium, the few studies conducted over the whole of the Mediterranean, did not show significant differences in the levels measured in open surface water of the different basins [130], while there were some noticeable differences between the open sea and coastal areas [131].

It is thus reasonable to calculate the time trend of anthropogenic radionuclide concentrations in surface sea water using average annual values obtained in different areas of the Mediterranean Sea.

The time trend of ^{137}Cs concentrations in surface sea water is shown in Fig. 129. An exponential decrease is apparent in the period 1970–1985. In 1986, Chernobyl fallout deeply modified ^{137}Cs distribution in the Mediterranean, producing a significant increase in the levels of the northern and eastern basins. ^{137}Cs concentrations were back to pre-Chernobyl values over the entire Mediterranean Sea in 1990.

Figure 130 shows the plutonium time trend over the period 1970–1997. As for caesium, there is a clear exponential decrease in concentrations. The calculated effective half-life for $^{239,240}\text{Pu}$ is 13 ± 2 years and the corresponding mean residence time is about 19 years. These data are in reasonable agreement with the residence times of 12 years [132] and 20–30 years [106] calculated by other methods. As expected, these parameters are slightly lower than those derived for caesium, as plutonium is removed from the surface layer not only by physical processes, but also by association with sinking organic and inorganic particles.

Four main data sets were analysed to estimate recent radionuclide concentrations: the first obtained by Papucci and Delfanti for the eastern Mediterranean in 1995–97 [133], the second by BSH for the western Mediterranean in 1997 [136], the third for the Strait of Gibraltar in 1997, and the fourth in the framework of the WOMARS project (ITALICA cruise) for the eastern Mediterranean in 1998 [135].

^{137}Cs concentrations range from 2.5 Bq/m^3 in the low salinity Atlantic water inflow to an average value of 3.1 Bq/m^3 in the eastern Mediterranean. For ^{90}Sr there are no significant differences between the two basins ($1.8 \pm 0.2 \text{ Bq/m}^3$). For plutonium, data are available only for the western Mediterranean. The mean concentrations of anthropogenic radionuclides in Mediterranean surface water are given in Table XVII.

$^{239,240}\text{Pu}$ concentration (around 12 mBq/m^3) for the reference date of January 1st, 2000 was estimated using the effective half-life of 11 years obtained from the analysis of the time trend evolution of its concentration in the period 1970–1994. The average ^{137}Cs concentration (around 2.6 Bq/m^3) was estimated using the recent data, because ^{137}Cs effective half-life could not be determined for the period following the Chernobyl accident. The same approach was adopted for ^{90}Sr (the average concentration is around 1.7 Bq/m^3).

TABLE XVII. MEAN CONCENTRATION OF ANTHROPOGENIC RADIONUCLIDES IN SURFACE WATER OF THE MEDITERRANEAN SEA

Radionuclide	Year	Area	Mean concentration (Bq/m^3)	St. dev.	No. of data	Decay corrected for 2000 (Bq/m^3)
^{137}Cs	1995	E. Med.	3.4	0.5	5	3
^{137}Cs	1997	W. Med., Gibraltar	2.5	0.2	13	2.4
^{137}Cs	1998	E. Med.	3.1	0.1	4	3
^{90}Sr	1997–98	All Med.	1.8	0.2	6	1.7
$^{239,240}\text{Pu}$	1997	W. Med.	0.012	0.003	10	0.012
^{238}Pu	1997	W. Med.	0.002	0.001	10	0.002
^{241}Am	1997	W. Med.	0.0014	0.0001	3	0.001

6.5.2. Recent data on ^{137}Cs and $^{239,240}\text{Pu}$ in the water column of the Mediterranean Sea

Only a few data are available for the period 1995–2000 on radionuclide vertical profiles in the Mediterranean Sea. The two main data sets published to date concern the eastern Mediterranean and the Strait of Gibraltar.

Eastern Mediterranean

The eastern Mediterranean has been studied in detail, also in relation to the so-called "deep water transient", the change in circulation of the deep and intermediate water that made the basin of interest in the 1990s [150]. A first analysis of radionuclide data was published by Papucci and Delfanti [133], who discussed the present distribution in relation to water circulation and to the input of Chernobyl radionuclides. The data were collected in 1995 and 1996–97. The surface concentration of ^{137}Cs ranged between 3.3 and 4.0 Bq/m³. The vertical profiles showed relative maxima in recent intermediate and deep water, also characterized by an increase in dissolved oxygen concentration. Figure 131 shows ^{137}Cs vertical profiles at 4 stations in the Ionian Sea and southeastern Mediterranean in 1995–96.

Strait of Gibraltar

The Strait of Gibraltar is a narrow and shallow strait that separates the Mediterranean Sea from the Atlantic Ocean. Vertical profiles of ^{137}Cs and $^{239,240}\text{Pu}$ were determined here in order to estimate radionuclide fluxes between the Atlantic Ocean and the Mediterranean Sea [134]. In this area, low salinity (S=36.0-36.5‰) water enters the Mediterranean Sea in the surface layer, at a flux of 0.72 Sv, while an almost equal amount (0.68 Sv) of saltier (S>37.5‰) and denser water leaves the basin [151]. Radionuclide concentrations were determined in the two different water masses near the Gibraltar Sill. The results are shown in Table XVIII.

TABLE XVIII. CONCENTRATIONS OF ANTHROPOGENIC RADIONUCLIDES AT THE STRAIT OF GIBRALTAR (MEAN VALUES, JUNE AND SEPT. '97)

Radionuclide	AW inflow (S 36.0-36.5‰)	MW outflow (S > 37.5‰)
$^{239,240}\text{Pu}$	10.0 ± 2.5 mBq/m ³	21.8 ± 2.3 mBq/m ³
^{137}Cs	2.6 ± 0.3 Bq/m ³	2.4 ± 0.3 Bq/m ³

There are no significant differences in ^{137}Cs concentrations in the waters leaving and entering the Mediterranean Sea. As a consequence, the input flux of ^{137}Cs to the Mediterranean Sea is balanced by the output to the Atlantic Ocean. The present situation is markedly different from that reported for the past, when surface concentrations were much higher than in deep water. The net annual input of ^{137}Cs to the Mediterranean appears to be markedly reduced compared to 1988 (120 TBq/y) [125]. In contrast, the levels of $^{239,240}\text{Pu}$ are considerably different in the water entering and leaving the Mediterranean Sea: about 10 mBq/m³ in the Atlantic inflow and 22 mBq/m³ in the Mediterranean outflow. The net annual output of $^{239,240}\text{Pu}$ from the Mediterranean to the Atlantic is presently 0.34 TBq/y, about one third of the value estimated in 1988. This is due to the vertical transport of plutonium and the consequent decrease of its concentration at intermediate depths.

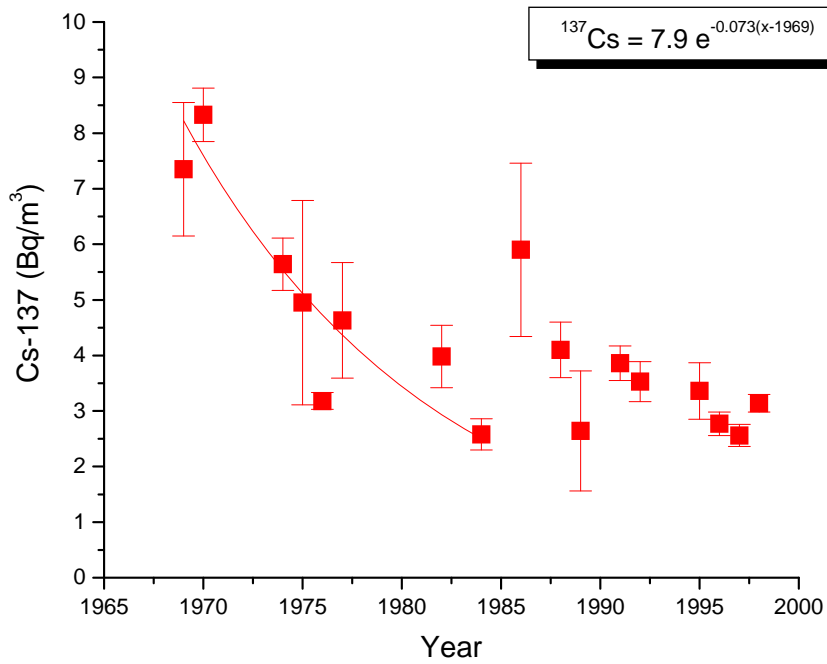


FIG. 129. Time trend of ¹³⁷Cs concentrations in surface water of the Mediterranean Sea, 1969–1998. Data from [129, 131, 133, 136–140].

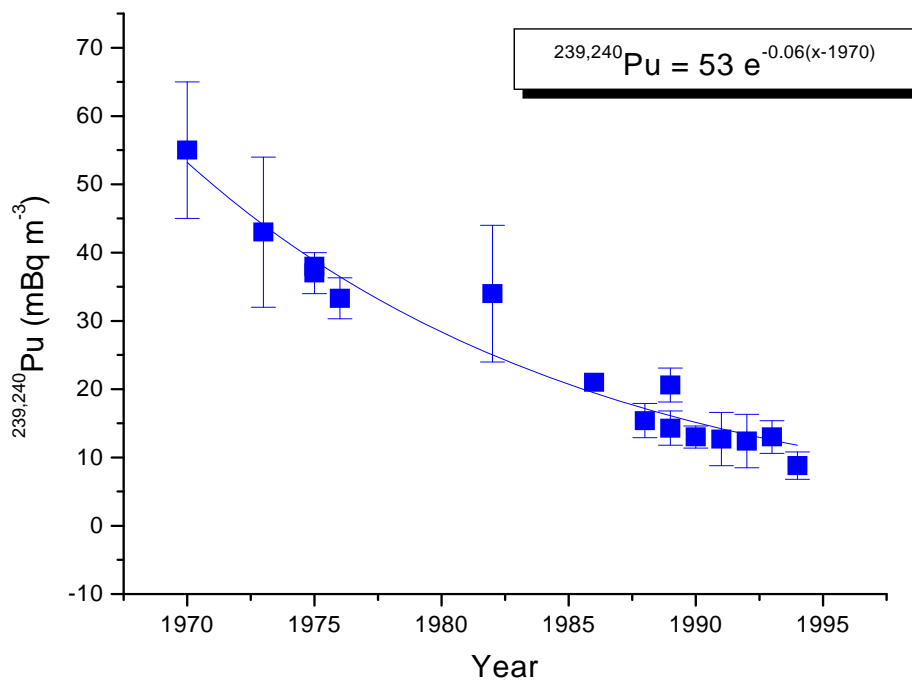


FIG. 130. Time trend of ^{239,240}Pu in surface seawater of the Mediterranean Sea, 1970–1997. Data from: [106, 131, 141–149].

The most recent vertical profiles of $^{239,240}\text{Pu}$ in the deep Mediterranean Sea [149] were measured in the Alboran Sea and the Algerian Basin in 1994. They show increasing concentrations from surface to a depth of 300–600 m and almost constant values from this depth to the bottom. In the vertical profiles measured in the 1970s and 1980s, maximum plutonium concentrations, found between 100–400 m depth, were higher by a factor of two than the present levels.

6.5.3. *Water column radionuclide time series*

The time trend of the vertical profiles of ^{137}Cs in the water column of the Mediterranean Sea has been analysed, based on published data [129, 131, 136, 140].

The average vertical profiles of ^{137}Cs for the period 1970–1982 (decay-corrected to 1994) and 1992–94 are compared in Fig. 132. While the ^{137}Cs concentration has not significantly changed in the depth interval 0–400 m, there has been a clear increase in the deeper layers. The inventory corresponding to a water depth of 2700 m was about 3 kBq/m² in the period 1970–82 and increased to about 6 kBq/m² in 1994, reflecting vertical transport to the deep sea by diffusion and convection processes and, to a lesser extent, additional input from the Chernobyl accident.

The time trend of plutonium vertical profiles has recently been analysed in detail [106]. They have shown that deep-water column (0–2000 m) inventories have not substantially changed over the period 1976–1990, but the inventory at a given depth in the deeper layers has increased considerably, demonstrating the slow downward movement of plutonium. The increase at depth is almost completely balanced by the decrease in the upper mixed layer (0–200 m). As an example, Fig. 133 shows the vertical profiles of $^{239,240}\text{Pu}$ in the NW Mediterranean in 1976 and 1990 [106, 130].

6.5.4. *Radionuclide inventories in the water column of the Mediterranean Sea*

^{137}Cs inventories in the water column have been calculated from the vertical profiles determined in the period 1995–97 in the eastern Mediterranean by Papucci and Delfanti [133]. The inventories have been calculated by simply integrating the concentration profiles, assuming a linear variation in concentration between sampling depths. The inventories range between 4.2 and 9.8 kBq/m², for water depths from 2000 to 4000 m.

These new data were combined with those obtained in the western Mediterranean in the period 1991–94 [140, 152] and plotted versus depth. All inventories were decay-corrected to the reference date of 2000. A linear increase in inventories with depth is evident (Fig. 134).

Plutonium and americium inventories in the Mediterranean Sea have been discussed in detail in two recent papers [106, 149].

Leon Vinturò et al. [149] discussed the relationship between water depth and plutonium inventories, using data obtained in the period 1990–94 (Fig. 135). The figure has been updated, adding a small number of data obtained in the Mediterranean in 1997 [134]. Based on the conclusions of Fowler et al. [106], who show that plutonium inventories in the water column of the Mediterranean Sea have not significantly changed in the depth interval 0–2000 m over the period 1976–1990, one can assume that these inventories are representative also for the period covered by the WOMARS programme (1995–2000).

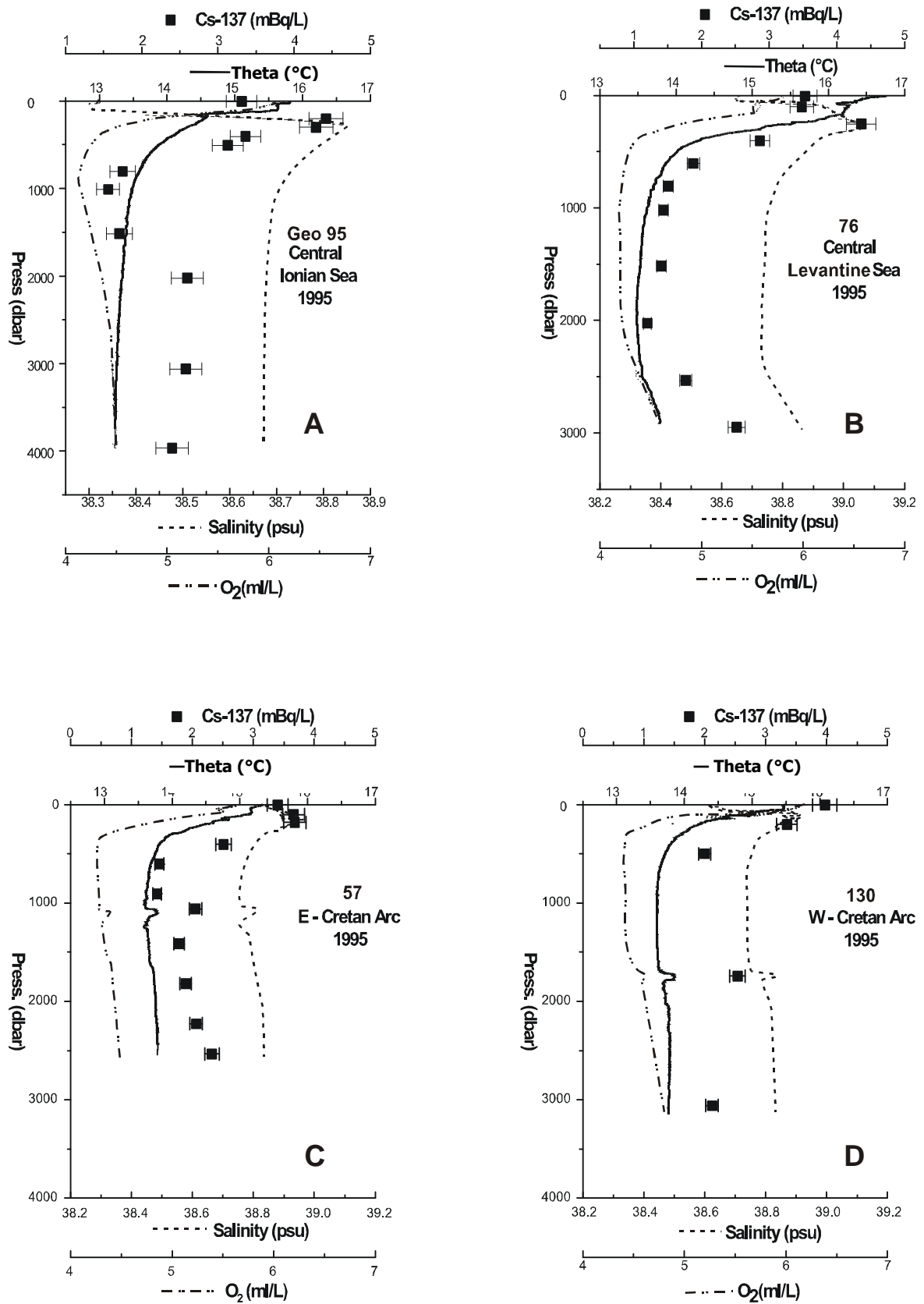


FIG. 131. Salinity, temperature, dissolved O_2 and ^{137}Cs vertical profiles in the eastern Mediterranean, 1995.

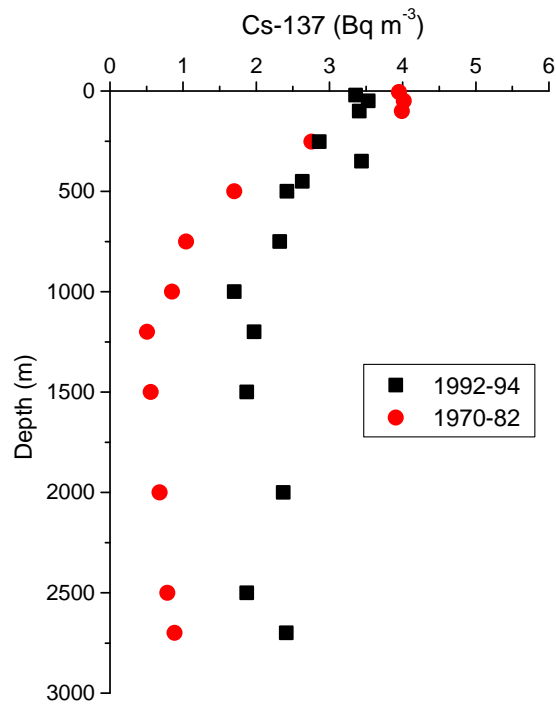


FIG. 132. Mean vertical profiles of ^{137}Cs in the water column of the Mediterranean Sea in 1970–82 (decay-corrected to 1994) and in 1992–94.

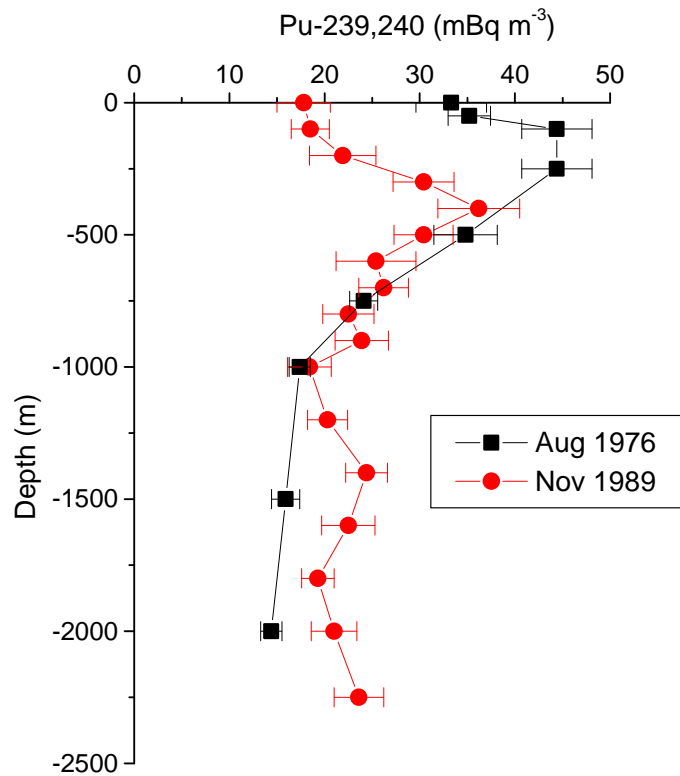


Fig. 133. Vertical profiles of $^{239,240}\text{Pu}$ in the Northwestern Mediterranean in 1976 and 1989 (data from [106, 130]).

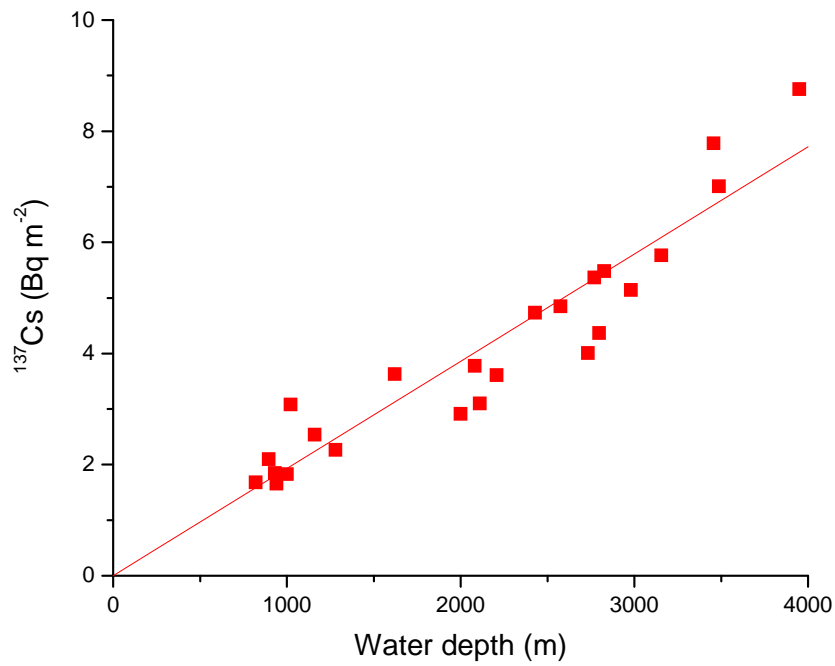


FIG. 134. Relationship between ¹³⁷Cs inventory and water depth (data from [133, 139, 140, 152] decay-corrected to 2000).

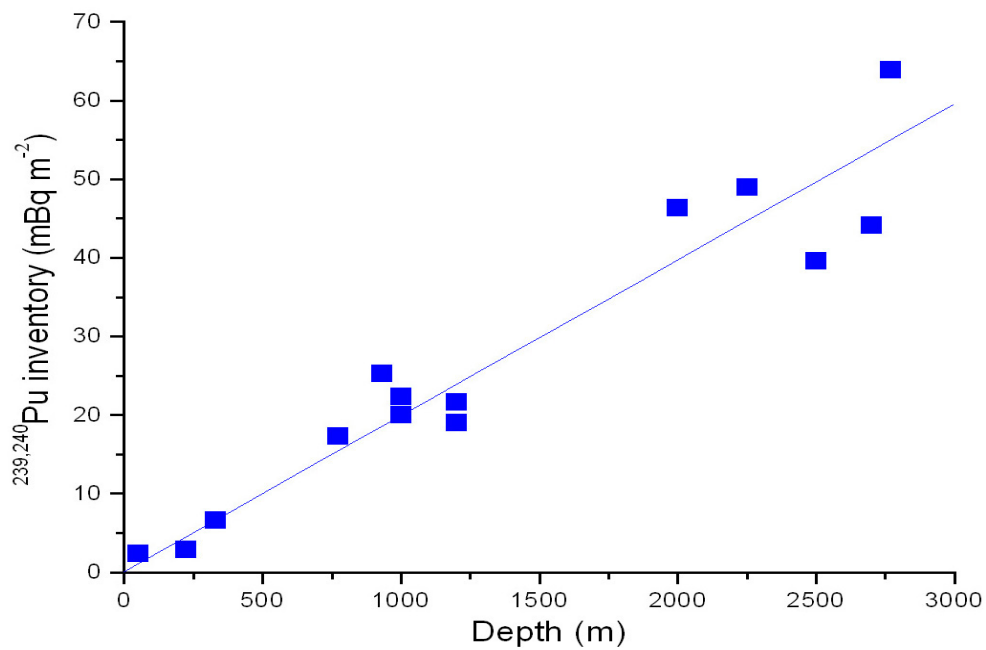


FIG. 135. Relationship between plutonium inventories and water depth in the Mediterranean Sea (1990–1994), data from [134, 147–149].

6.5.5. Radionuclide inventories in the deep Mediterranean Sea

Radionuclide concentrations and inventories are highly variable in the Mediterranean Sea, highest on the continental shelf and near river mouths and lowest in deep-sea environments.

Only a few studies have been conducted on radionuclides in deep-sea sediment. Livingston et al. [153] determined the inventories of ^{137}Cs and $^{239,240}\text{Pu}$ in sediment cores collected in the Western Mediterranean and Ionian Sea in 1975. ^{137}Cs inventories ranged from 90 to 260 Bq/m² and $^{239,240}\text{Pu}$ inventories from 2 to 7 Bq/m².

Studies conducted in the 1990s in the western Mediterranean [140, 154] indicate inventories very similar to those reported in 1975: 230 Bq/m² for ^{137}Cs and 3 Bq/m² for $^{239,240}\text{Pu}$.

7. CONCLUSIONS

The aim of the WOMARS CRP was to:

- (i) Identify the major sources of anthropogenic radionuclides in the world's oceans,
- (ii) Develop the knowledge of the distribution of key radionuclides (^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$) in the water column and in sediment,
- (iii) Study the development with time of radionuclide concentrations in water.

Three anthropogenic radionuclides, ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ were chosen as the most important and radiologically typical of each class of marine radioactivity. They are the most abundant anthropogenic radionuclides present in the marine environment and could lead to the highest radiation doses to humans and marine biota.

The results confirm that the dominant source of anthropogenic radionuclides in the marine environment is global fallout. The total ^{137}Cs input from global fallout was estimated to be 311 PBq for the Pacific Ocean, 201 PBq for the Atlantic Ocean, 84 PBq for the Indian Ocean and 7.4 PBq for the Arctic Ocean. For comparison, about 40 PBq of ^{137}Cs was released to the marine environment from Sellafield and Cap de la Hague reprocessing plants. The Chernobyl accident contributed about 16 PBq of ^{137}Cs into seas, mainly the Baltic and Black Seas where the present average concentrations of ^{137}Cs in surface water were estimated to be about 60 and 25 Bq/m³, respectively, while the worldwide average concentration due to global fallout was about 2 Bq/m³.

Figure 136 and Tables XIX–XXI show the best estimates of averaged ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ concentrations in the world's oceans and seas adjusted to the year 2000. As expected, the highest concentrations were observed in European Seas and the lowest in the Southern hemisphere, especially in the Antarctic Ocean.

For the purposes of this study, the world ocean was divided into latitudinal belts, for which average radionuclide concentrations were estimated. Further, where available, time trends in radionuclide concentrations in surface water were studied and mean residence times of radionuclides in these areas as well as in the world ocean were estimated. The results confirm similar residence times for ^{90}Sr and ^{137}Cs in surface water, varying from 11 to 30 years and from 5 to 15 years for $^{239,240}\text{Pu}$.

The mean residence times of radionuclides in surface water are listed in Table XXII.

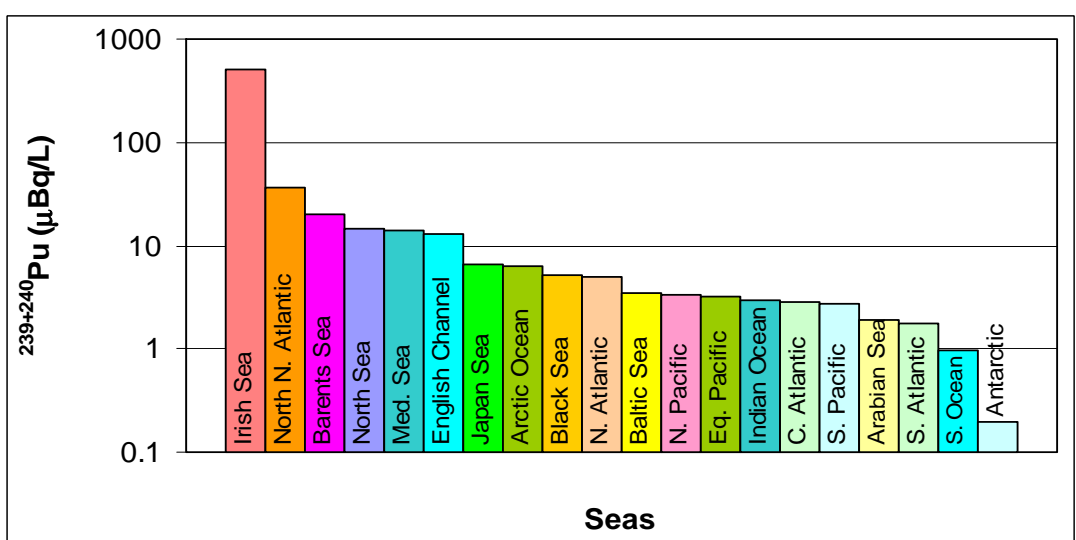
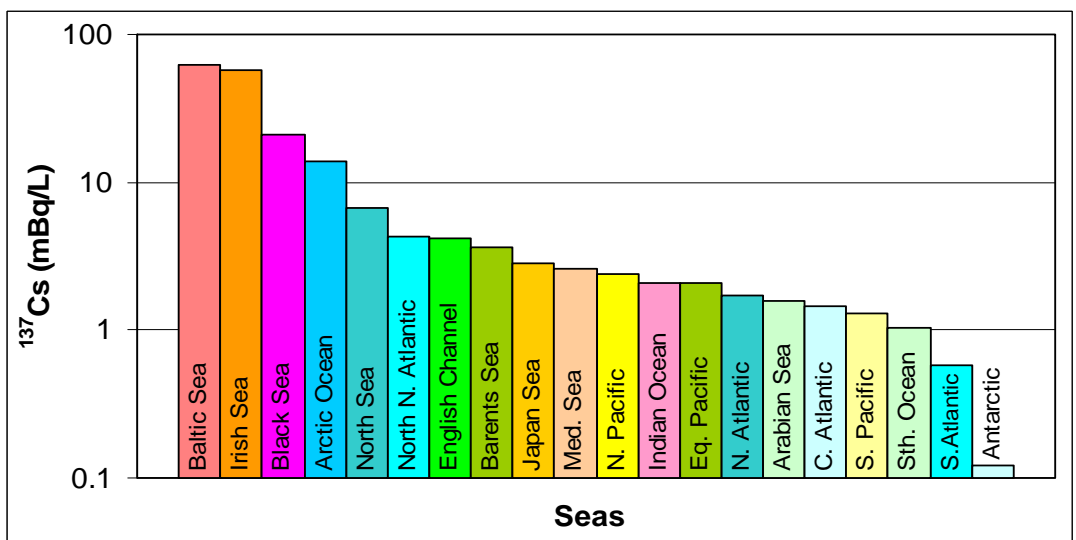
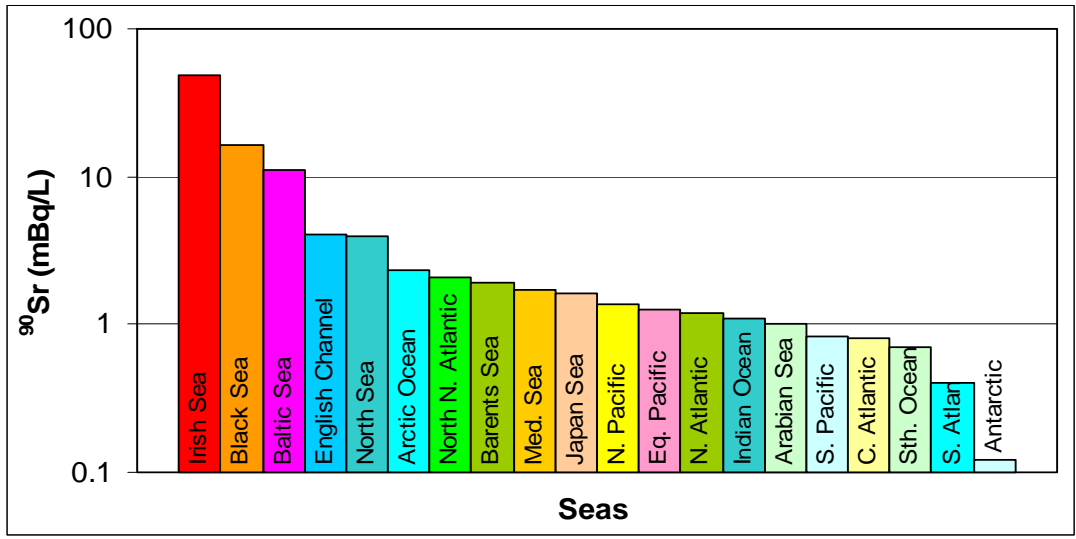


FIG. 136. ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ in the world's oceans and seas (adjusted to 01.01.2000).

TABLE XIX. ^{90}Sr IN SURFACE WATER OF THE WORLD'S OCEANS AND SEAS

Area	Box No.	Number of data	Averaged value on 01.01.2000 (mBq/L)	
			Box	Area $\pm 1\sigma$
North Pacific	1	3	1.3	1.4 ± 0.2
	2	31	1.6	
	3	3	1.4	
	4	13	1.5	
	5	5	1.1	
Equatorial Pacific	6	2	1.5	1.3 ± 0.3
	7	3	1.1	
South Pacific	8	1	1.2	0.8 ± 0.3
	9	4	1.1	
	10	3	0.8	
	11	4	0.6	
	12	2	0.4	
	13	1	0.1	
Sea of Japan	14	42	1.6	1.6 ± 0.3
Arabian Sea	15	15	1.0	1.0 ± 0.2
Indian Ocean	16	7	1.1	1.1 ± 0.2
Southern Ocean	17	12	0.7	0.7 ± 0.4
Arctic	18	1	2.3	2.3
	19			
Barents Sea	20	7	1.9	1.9 ± 0.4
Baltic Sea	21	71	11.1	11.1 ± 2.9
North Sea	22	118	4.0	4.0 ± 1.2
Irish Sea	23	14	49	49 ± 83
English Channel	24	43	4.1	4.1 ± 1.4
North N. Atlantic	25	17	2.1	2.1 ± 0.9
Black Sea	26	15	17	17 ± 6
Mediterranean Sea	27	15	1.7	1.7 ± 0.2
North Atlantic	28	17	1.2	1.2 ± 0.6
Central Atlantic	29	5	0.8	0.8 ± 0.1
South Atlantic	30	2	0.4	0.4 ± 0.2

Normal Font: Standard deviation for area-averaged value

Italic Font: Standard deviation for box-averaged value

TABLE XX. ¹³⁷Cs IN SURFACE WATER OF THE WORLD'S OCEANS AND SEAS

Area	Box No.	Number of data	Averaged value on 01.01.2000 (mBq/L)	
			Box	Area $\pm 1\sigma$
North Pacific	1	4	2.0	2.4 ± 0.3
	2	30	2.4	
	3	3	2.8	
	4	7	2.4	
	5	4	2.3	
Equatorial Pacific	6	2	2.3	2.1 ± 0.3
	7	3	1.9	
South Pacific	8	1	1.7	1.3 ± 0.5
	9	4	1.7	
	10	3	1.4	
	11	4	1.1	
	12	2	0.6	
Antarctic	13	1	0.1	0.1
Sea of Japan	14	44	2.8	2.8 ± 0.5
Arabian Sea	15	17	1.6	1.6 ± 0.3
Indian Ocean	16	7	2.1	2.1 ± 0.3
Southern Ocean	17	12	1.0	1.0 ± 0.6
Arctic Ocean	18	1	14	14
	19			
Barents Sea	20	9	3.6	3.6 ± 2.0
Baltic Sea	21	214	61	61 ± 19
North Sea	22	144	6.7	6.7 ± 2.9
Irish Sea	23	28	57	57 ± 55
English Channel	24	44	4.2	4.2 ± 1.5
North N. Atlantic	25	15	4.2	4.2 ± 2.7
Black Sea	26	20	25	25 ± 3
Mediterranean Sea	27	18	2.6	2.6 ± 0.4
North Atlantic	28	16	1.7	1.7 ± 0.8
Central Atlantic	29	5	1.4	1.4 ± 0.2
South Atlantic	30	2	0.6	0.6 ± 0.1

Normal Font: Standard deviation for area-averaged value

Italic Font: Standard deviation for box-averaged value

TABLE XXI. ^{239,240}Pu IN SURFACE WATER OF THE WORLD'S OCEANS AND SEAS

Area	Box No.	Number of data	Averaged value on 01.01.2000 (μBq/L)	
			Box	Area ± 1σ
North Pacific	1	2	7.9	3.3 ± 2.8
	2	6	3.8	
	3	3	0.7	
	4	7	2.4	
	5	2	1.7	
Equatorial Pacific	6	5	3.6	3.1 ± 0.7
	7	3	2.7	
South Pacific	8	5	1.5	2.8 ± 2.1
	9	4	5.7	
	10	2	4.3	
	11	4	1.6	
	12	1	0.8	
Antarctic	13	1	1.3	1.3 ± <u>0.7</u>
Sea of Japan	14	30	6.6	6.6 ± 2.5
Arabian Sea	15	13	1.9	1.9 ± 1.2
Indian Ocean	16	4	3.0	3.0 ± 1.5
Southern Ocean	17	10	1.0	1.0 ± 0.5
Arctic	18	24	6.4	6.4 ± 1.5
	19			
Barents Sea	20	7	20	20 ± 12
Baltic Sea	21	29	3.4	3.4 ± 2.5
North Sea	22	33	15	15 ± 10
Irish Sea	23	30	500	500 ± 400
English Channel	24	18	13	13 ± 8
North N. Atlantic	25	9	36	36 ± 16
Black Sea	26	5	5.3	5.3 ± 2.3
Mediterranean Sea	27	7	14	14 ± 4
N. Atlantic	28	17	5.0	5 ± 3
C. Atlantic	29	5	2.8	2.8 ± 1.3
S. Atlantic	30	1	1.8	1.8 ± <u>0.6</u>

Normal Font: Standard deviation for area-averaged value

Italic Font: Standard deviation for box-averaged value

Italic + Underline: Analytical error only

TABLE XXII. MEAN RESIDENCE TIME OF ^{90}Sr , ^{137}Cs AND $^{239,240}\text{Pu}$ IN SURFACE WATER

Area	Boxes	Mean residence time $\pm 1\sigma^*$ (year)		
		^{90}Sr	^{137}Cs	$^{239,240}\text{Pu}$
North Pacific	1-5	17.9 ± 1.6	18.4 ± 2.0	9.7 ± 1.3
Equatorial Pacific	6+7	30.5 ± 2.7	41.3 ± 4.4	14.7 ± 3.4
South Pacific	8-12	25.8 ± 1.4	32.0 ± 7.5	17.5 ± 5.2
All Pacific	1-12	23.0 ± 3.5	22.9 ± 6.2	10.7 ± 1.6
North Indian	15		29.2 ± 2.7	30.6 ± 11.0
South Indian	16+17		31.6 ± 9.2	12.7 ± 1.5
All Indian	15-17		29.4 ± 2.6	13.0 ± 2.4
North Atlantic	25+28	22.6 ± 6.2	26.9 ± 6.0	13.0 ± 0.8
Central Atlantic	29	34.9 ± 3.0	34.3 ± 8.2	14.1 ± 4.2
South Atlantic	30	25.0 ± 12.7	29.2 ± 9.1	15.4 ± 7.1
All Atlantic	25-30	32.2 ± 3.5	29.4 ± 4.3	13.1 ± 0.8
All Oceans		27.5 ± 4.6	28.6 ± 2.1	12.7 ± 1.4

* Error weighted averaged values

Generally, as expected, the differences between the effective residence times of ^{90}Sr and ^{137}Cs are, within uncertainties, the same. Longer residence times can be seen in both the central Pacific and the central Atlantic Oceans. The mean residence time calculated for the world ocean is 28 ± 3 years for ^{90}Sr and ^{137}Cs , and 13 ± 1 year for $^{239,240}\text{Pu}$.

Changes in radionuclide concentrations in water profiles with time in the N Atlantic and Pacific Oceans were also studied. A clear decrease of radionuclide concentrations in surface water was observed due to transport of radionuclides to medium water depths.

The results obtained in the framework of the WOMARS CRP provide the most complete data set available to Member States on levels of anthropogenic radionuclides in the marine environment. The results will be used as the international reference source on the average levels of anthropogenic radionuclides in the marine environment, so that any further contributions from nuclear reprocessing plants, radioactive waste dumping sites, nuclear bomb test sites and possible nuclear accidents can be identified.

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Further, the main data sets used in the project were kindly provided by the participating institutions: The Federal Maritime and Hydrographic Agency (Germany), Risø National Laboratory (Denmark), Japan Atomic Energy Research Institute, Marine Research Laboratory, Mutsu Establishment (Japan), Japan Marine Science Foundation, Mutsu Marine Laboratory (Japan), Korean Ocean Research Institute (Republic of Korea), Lawrence Livermore National Laboratory (USA), Woods Hole Oceanographic Institute (USA) and many other institutes (e.g. The Centre for Environment, Fisheries and Aquaculture Science, (UK), The Institute of Radioprotection and Nuclear Safety (France)) as well as researchers.

It is a great pleasure to acknowledge the institutes and researchers who contributed to this project by organizing oceanographic expeditions in which IAEA-MEL staff participated, for example, the Joint Japanese-Korean-Russian expert groups, 1994 and 1995; the Federal Fisheries Research Centre (Germany), 1996, 1998 and 2000; the Physical Research Laboratory (India), 1998; the Marine Environment Research Centre, La Spezia (Italy), 1998 and the Centre National de Recherche Scientifique (France), 1999.

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ANNEX I. TRANSFER COEFFICIENTS FOR ^{90}Sr AND ^{137}Cs FOR DOSE CALCULATIONS

I-1. Introduction:

In the MARDOS report [3] an attempt was made to calculate the collective effective doses from ingestion of marine products contaminated by ^{137}Cs . In this Annex, the most recent information on input of ^{90}Sr and ^{137}Cs to the world ocean has been used. Transfer factors between input and collective effective dose have been calculated for the terrestrial and the marine environment. Table I shows the calculation of the ingestion doses from marine foods (fish and shellfish, i.e. crustaceans and molluscs).

I-2. Input:

It has been assumed that all input of ^{137}Cs (and ^{90}Sr) from reprocessing and the Chernobyl accident occurred in the North Atlantic and Arctic Oceans and that these parts of the world ocean still contain this activity (or the rest after decay). Most of the activity is probably still present in the 90-60°N latitude band rather than in the 90-30°N band. Hence, one may have underestimated the radiological impact from these two sources, as the fish catch is high in the 90-60°N belt.

I-3. Volume:

The mixed layer of the ocean is assumed to be presented by the upper 100 m water layer. Hence, the volume of the mixed layer is obtained by multiplying the area (given in m^2) by 100.

I-4. Catch:

The fishing areas used in the FAO statistics do not strictly follow the division used here. Therefore, it has been necessary to make some adjustments. The catch in areas 71 and 77 in the Pacific have been totalled and divided by two; one half has been allocated to 30-0°N and the other to 0-30°S. In the Indian Ocean the three areas: 51, 57 and 58 have been added together and the total catch has been divided between the latitude bands 30-0°N, 0-30°S and 30-90°S, proportional to the area of these latitude bands for the Indian Ocean (i.e. 16%, 36% and 48%, respectively).

The catch data are the FAO's 1990 data used in the MARDOS report [3]. The doses may be overestimated because the catch has increased over the years, whereas the concentrations for ^{90}Sr and ^{137}Cs have decreased.

I-5. Concentrations in fish and shellfish:

The ^{137}Cs concentrations in fish and shellfish were obtained by multiplying the water concentration by 100 and 30, respectively [3].

I-6. Time integral:

It has been assumed that the mean residence time for ^{137}Cs (and ^{90}Sr) in the mixed layer of the ocean is 25 years. Time integrals were thus calculated by multiplying the concentrations by 25. If the mean residence times of ^{90}Sr and ^{137}Cs are shorter in the N

Atlantic Ocean as indicated by observations in the Faroes and Greenland, the doses from the North Atlantic are overestimated by approximately one third.

I-7. Dose calculation:

The collective effective dose was calculated by multiplying the sum of the ^{137}Cs inventories in the fish and shellfish catch by $1.25 \cdot 10^{-8}$ Sv/Bq.

Table II shows the calculation of transfer coefficients.

The transfer coefficient (manSv/PBq) for ^{90}Sr in global fallout was calculated from the transfer coefficient for ^{137}Cs taking into account that the dose factor for ^{90}Sr (ingestion) is $2.8/1.3 = 2.15$ times higher than that of ^{137}Cs , whereas the marine concentration factor (consumption weighted) is 44 times higher for ^{137}Cs than for ^{90}Sr (fish and shellfish) [3]. Hence, the manSv/PBq transfer coefficient for ^{90}Sr becomes $(2.15/44) \cdot 15 = 0.73$. For global fallout the terrestrial dose consists for ^{137}Cs of 64% external radiation and 36% ingestion. For ^{90}Sr , 92% comes from ingestion and 8% from inhalation.

I-8. Chernobyl

In the UNSCEAR report [2], exposures and effects of the Chernobyl accident are reported. This Annex deals preliminarily with the situation in the most contaminated areas of the FSU, i.e. areas that have received more than $37 \text{ kBq } ^{137}\text{Cs m}^{-2}$ from the accident. The total collective effective doses to the population in these areas are 40300 manSv from external exposure and 20400 manSv from ingestion (thyroid doses are not included). The ingestion dose comes essentially from ^{137}Cs , ^{134}Cs and ^{90}Sr . UNSCEAR [2] estimates that the ^{90}Sr contribution to the effective dose from internal exposure does not exceed 5–10%, i.e. ~ 1500 manSv. The releases from the Chernobyl accident were ~ 10 PBq ^{90}Sr , ~ 54 PBq ^{134}Cs and ~ 85 PBq ^{137}Cs . The amount of ^{137}Cs deposited in the contaminated areas ($>37 \text{ kBq } ^{137}\text{Cs m}^{-2}$) of the FSU was 29 PBq, the amounts of ^{134}Cs and ^{90}Sr were 18 PBq and 10 PBq, respectively. The transfer coefficient from input to collective dose for ^{90}Sr is calculated to be $1500/10 = 150$ manSv/PBq for the Chernobyl accident. In order to calculate the corresponding transfer coefficient for ^{137}Cs one has to correct for the ^{134}Cs contribution to the effective doses. In the case of external exposure the ratio between the collective doses from the same inputs of ^{137}Cs and ^{134}Cs is 5.372 and for ingestion the ratio is 1.44. Hence, the ratio between the external ^{137}Cs and ^{134}Cs Chernobyl dose becomes $(29/18) \cdot 5.372 = 8.6549$, and for the ingestion dose the ratio becomes $(29/18) \cdot 1.44 = 2.32$. The external collective effective dose for ^{137}Cs then becomes

$$40300 \frac{8.6549}{9.6549} = 36125 \text{ manSv and the corresponding dose from ingestion:}$$

$$(20400-1500) \frac{2.32}{3.32} = 13207 \text{ manSv.}$$

The total dose from the 29 PBq ^{137}Cs becomes 49333 manSv and the transfer coefficient for ^{137}Cs is calculated to be $\frac{49333}{29} = 1700$ manSv/PBq. The total terrestrial deposition of ^{137}Cs

was $85-16 = 69$ PBq and if one assumes the same transfer factor valid for the remaining part of the terrestrial environment as found for the FSU, the collective effective dose from 69 PBq ^{137}Cs is calculated to $69 \cdot 1700 = 117400$ manSv. This figure is about 3 times less than earlier estimates.

The collective effective dose from Chernobyl released to the marine environment was calculated by applying a weighted transfer coefficient: $0.75 \cdot 98 + 0.25 \cdot 15 = 77$ manSv/PBq ^{137}Cs . The assumption was that 75% of the debris went to coastal water and 25% to the open sea. For coastal water the transfer coefficient (98 manSv/PBq ^{137}Cs) used for reprocessing discharges was applied and for the open sea the transfer coefficient applied for global fallout was used.

I-9. Reprocessing

UNSCEAR has calculated the transfer coefficient for releases of ^{90}Sr and ^{137}Cs into coastal water to be 4.7 manSv/PBq ^{90}Sr and 98 manSv/PBq ^{137}Cs . These figures have been used together with the release data of the two radionuclides from Sellafield and Cap de la Hague in the calculation of the collective effective doses from water-borne discharges from nuclear reprocessing.

I-10. Comparison between transfer coefficients

If all sources are considered, Table II shows that the terrestrial pathway for ^{137}Cs (ingestion and external radiation) has a $6928/22 \approx 300$ times higher transfer coefficient than the marine pathway. For ^{90}Sr , the ratio between the two pathways is even higher namely $2634/0.80 \approx 3000$. If one considers global fallout and ingestion only, the ratio for ^{137}Cs becomes $\frac{8100 \cdot 0.36}{15} \approx 200$ and for ^{90}Sr it is $\frac{2740 \cdot 0.92}{0.73} \approx 3000$. Discharges in coastal seas, as has been seen from nuclear reprocessing, show a higher marine transfer coefficient than global fallout.

TABLE I. INGESTION DOSES FROM MARINE FOODS

	Atlantic and Arctic Oceans				Pacific and Indian Oceans				World		
	90-30°N	30-0°N	0-30°S	30-90°S	90 N-90°S	90-30°N	0-30°S	30-9°S		90N-90°S	
Input source											
Global fallout	115.4	49.3	16.5	27.3	208.5	116.7	126.6	66	85.5	394.8	603.3
Reprocessing	40				40						40
Chernobyl	16				16						16
Total	171.4	49.3	16.5	27.3	264.5	116.7	126.6	66	85.5	394.8	659.3
Annual catch											
Fish (t/year)	1117555	503861	1466179	146617	191465	471845	463481	5705171	479905	1985749	9004020
	7	0		9	25	5	0		9	5	
Shellfish (t/year)	2394607	732627	511030	511031	414929	531600	707820	855427	641662	7520910	1670205
					5	1					
Mixed ocean layer											
Area	37.28	23.493	19.145	26.604	106.522	26.725	67.147	78.807	81.909	254.588	361.11
Volume	3.728	2.3493	1.9145	2.6604	10.6522	2.6725	6.7147	7.8807	8.1909	25.4588	36.111
Conc. in water	45.97639	20.9849	8.618438	10.2616	85.8414	43.6669	18.8541	8.374890	10.4384	81.33443	167.1759
	5	74	2	15	22	78	56	6	13	8	
Time integral in water	1149.409	524.624	215.4609	256.540	2146.03	1091.67	471.353	209.3722	260.960	2033.361	4179.397
	9	36	6	37	56	45	9	6	33		
Conc. in fish	4597.639	2098.49	861.8438	1026.16	8584.14	4366.69	1885.41	837.4890	1043.84	8133.443	16717.59
	5	74	2	15	22	78	56	6	13	8	
Conc. in shellfish	1379.291	629.549	258.5531	307.848	2575.24	1310.00	565.624	251.2467	313.152	2440.033	5015.276
	8	23	5	44	27	94	67	2	4	1	
Time integral in fish	114940.9	52462.4	21546.09	25654.0	214603.	109167.	47135.3	20937.22	26096.0	203336.1	417939.7
	9	36	6	37	56	45	9	6	33		
Time integral in shellfish	34482.29	15738.7	6463.828	7696.21	64381.0	32750.2	14140.6	6281.167	7828.81	61000.82	125381.9
	6	31	7	11	67	34	17	9		9	

Inventory in fish catch	TBq	1.284529	0.26433	0.031590	0.03761	1.61807	0.51510	0.21846	0.119450	0.12523	0.978252	2.596323
		6	78	4	34	11	17	36	5	64	1	1
Inventory in shell fish catch	TBq	0.082571	0.01153	0.003303	0.00393	0.10133	0.17410	0.01000	0.005373	0.00502	0.194505	0.29584
		5	06	2	3	84	03	9	1	34	8	42
Dose calculation												
Intake by fish consumption	TBq	0.449585	0.09251	0.011056	0.01316	0.56632	0.18028	0.07646	0.041807	0.04383	0.342388	9087131
		3	82	7	47	49	56	23	7	27	2	2
Intake by shellfish consumption	TBq	0.041285	0.00576	0.001651	0.00196	0.05066	0.08705	0.00500	0.002686	0.00251	0.097252	1479221
		8	53	6	65	92	01	45	5	17	9	9
Collective effective dose	ManSv	6136	1229	159	189	7712	3342	1018	556	579	5496	13208

TABLE II. CALCULATION OF COLLECTIVE EFFECTIVE DOSES FOR MARINE AND TERRESTRIAL PATHWAYS
(WORLD POPULATION: $5.6 \cdot 10^9$)

Source	Pathway	Release in PBq		manSv/PBq		(manSv)*10 ³		References
		Sr-90	Cs-137	Sr-90	Cs-137	Sr-90	Cs-137	
Global fallout	marine	377	603	0.73	15	0.28	9.1	a) UNSCEAR [2]
	terrestrial	235	316	2740	8100	643.9	2550.0	c) WOMARS
Nuclear reprocessing	marine	6.6	39.7	4.7	98	0.03	3.9	a
	terrestrial	0	0	0	0	0	0	(this work)
Chernobyl accident	marine	0	16	3.7	77	0	1.2	c
	terrestrial	10	69	150	1700	1.50	117.3	a,c
Total	marine	383.6	658.7	0.80	22	0.31	14.2	
	terrestrial	245	385	2634	6928	645.40	2667.3	
	Sum	629	1043.7	1027	2569	646	2681.5	

ANNEX II. LIST OF PUBLICATIONS PUBLISHED BY THE IAEA-MEL/RML STAFF ON WORLDWIDE MARINE RADIOACTIVITY STUDIES

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