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Journal of Environmental Radioactivity 81 (2005) 63–87
JOURNAL OF ENVIRONMENTAL RADIOACTIVITY
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^{90}Sr, ^{137}Cs and $^{239,240}Pu$ concentration surface water time series in the Pacific and Indian Oceans – WOMARS results

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Received 14 February 2004; received in revised form 24 November 2004; accepted 13 December 2004

Abstract

Under an IAEA's Co-ordinated Research Project "Worldwide Marine Radioactivity Studies (WOMARS)" ^{90}Sr, ^{137}Cs and $^{239,240}Pu$ concentration surface water time series in the Pacific and Indian Oceans have been investigated. The Pacific and Indian Oceans were divided

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doi:10.1016/j.jenvrad.2004.12.003

into 17 latitudinal boxes according to ocean circulation, global fallout patterns and the location of nuclear weapons test sites. The present levels and time trends in radionuclide concentrations in surface water for each box were studied and the corresponding effective half-lives were estimated. For the year 2000, the estimated average ^{90}Sr, ^{137}Cs and ^{239,240}Pu concentrations in surface waters of the Pacific and Indian Oceans varied from 0.1 to 1.5 mBq/L, 0.1 to 2.8 mBq/L, and 0.1 to 5.2 μBq/L, respectively. The mean effective half-lives for ^{90}Sr and ^{137}Cs in surface water were 12 ± 1 years for the North, 20 ± 1 years for the South and 21 ± 2 years for the Equatorial Pacific. For ^{239,240}Pu the corresponding mean effective half-lives were 7 ± 1 years for the North, 12 ± 4 years for the South and 10 ± 2 years for the Equatorial Pacific. For the Indian Ocean the mean effective half-lives of ^{137}Cs and ^{239,240}Pu were 21 ± 2 years and 9 ± 1 years, respectively. There is evidence that fallout removal rates before 1970 were faster than those observed during recent decades. The estimated surface water concentrations of ^{90}Sr, ^{137}Cs and ^{239,240}Pu in latitudinal belts of the Pacific and Indian Oceans for the year 2000 may be used as the average levels so that any new contribution from nuclear facilities, nuclear weapons test sites, radioactive waste dumping sites and from possible nuclear accidents can be identified.

Keywords: Radionuclides; Strontium-90; Caesium-137; Plutonium-239,240; Surface water; Time series; Pacific Ocean; Indian Ocean; Southern Ocean

1. Introduction

The origin of the majority of anthropogenic radionuclides in the Pacific and Indian Oceans can be traced to global fallout from nuclear weapons tests carried out in the 1950s and 1960s. The input function is relatively simple (Nakano and Povinec, 2003), as the main introduction of anthropogenic radionuclides to Pacific and Indian Ocean waters started in 1963 after series of large atmospheric nuclear weapons tests carried out at Novaya Zemlya in 1961–1962. However, local fallout originating from tests carried out at the Marshall Islands mainly in the 1950s has also contributed significantly to the inventory of anthropogenic radionuclides in the southern North–West Pacific Ocean. Other sources have introduced relatively smaller amounts of radionuclides into the Pacific and Indian Oceans (Livingston and Povinec, 2002). As the ocean is a dynamic system, radionuclides introduced to surface waters by wet and dry deposition did not stay in steady-stay conditions, but due to currents and processes in the water column, they have been transported to different regions, as well as to bottom waters and sediments.

One of the main interests in studying the behaviour of radionuclides in the ocean derives from the fact that radionuclides are powerful tracers providing basic insights into a variety of oceanic processes. For example, plutonium as a particle-reactive radioelement, 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 water column, and in biological and sedimentary systems.

As a contribution to these investigations, the International Atomic Energy Agency's Marine Environment Laboratory (IAEA-MEL) in Monaco has in collaboration with several marine institutes, completed a Co-ordinated Research Project (CRP) on "Worldwide Marine Radioactivity Studies (WOMARS)". The objectives of the CRP were: (i) to identify the major sources of anthropogenic radionuclides in the world ocean; (ii) to develop present knowledge of the distributions of key radionuclides (^{90}Sr, ^{137}Cs and Pu isotopes) in water and sediment of the world ocean; and (iii) to study the evolution of radionuclide concentrations in water with time using quality controlled data (e.g. from the GEOSECS (Geochemical Ocean Sections Study) programme of the mid-1970s (Bowen et al., 1980), 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 bottom sediment, and thus predict the radiological impact to be addressed. Another objective was to encourage and support marine radioactivity studies in IAEA Member States by providing methodological and analytical quality control assistance (IAEA, 2001).

Three anthropogenic radionuclides – ^{90}Sr, ^{137}Cs and ^{239,240}Pu – were chosen as the most radiologically important and typical of beta, beta/gamma and alpha emitters, respectively. They are also the most abundant anthropogenic radionuclides in the marine environment and can lead to the highest radiation doses to humans and marine biota. In this paper, surface water time series of ^{90}Sr, ^{137}Cs and ^{239,240}Pu, and their effective half-lives in the Pacific and Indian Oceans are presented and discussed. The spatial distribution of these radionuclides in the Pacific and Indian Oceans has already been discussed by Povinec et al. (2004) and therefore it is not included in the present paper.

2. Methods

2.1. Radionuclide database

Radionuclide data are relatively abundant in the Pacific but there are very limited data for the Indian Ocean. The IAEA (2001) recently completed a bibliography review covering the concentrations, distributions and behaviour of ^{90}Sr, ^{137}Cs and ^{239,240}Pu in water, sediment and biota in the world ocean. The GEOSECS programme carried out in 1973 and 1974 provided the first comprehensive data set on the lateral and vertical distributions of ^{90}Sr, ^{137}Cs and ^{239,240}Pu in the Pacific Ocean proper (Bowen et al., 1980). Japanese institutes have been active in investigating these radionuclides, especially in North-Western (NW) Pacific water (e.g., Nagaya and Nakamura, 1970, 1976, 1984, 1987, 1992, 1993; Aoyama and Hirose, 1995; Miyake et al., 1988; Nakanishi et al., 1990; Hirose et al., 1992, 1999; Hirose, 1997; Ikeuchi et al., 1999; Aoyama et al., 2001a,b). The nuclear weapons testing sites in the Marshall Islands were monitored extensively by the US (e.g., Noshkin and Wong, 1980; Noshkin et al., 1983; Robison and Noshkin, 1999), and the impact of the tests carried out in French Polynesia were assessed by France and IAEA (e.g. Bourlat et al., 1991, 1996; IAEA, 1998; Povinec et al., 1999).

The data evaluated in this paper were extracted from the Global Marine Radioactivity Database (GLOMARD) developed at IAEA-MEL (IAEA, 2000; Povinec et al., 2004). The main data sets were supplied by the following Japanese institutions: the Japan Chemical Analysis Center (JCAC), the Marine Safety Agency (MSA) and the Fisheries Agency (FA). Data from the joint Japanese–Korean–Russian expeditions in 1994 (STA, 1995) and 1995 (STA, 1997) were also used in the evaluations. Further data were obtained from a literature survey covering the western and central N Pacific (Nagaya and Nakamura, 1984, 1987, 1993; Aoyama and Hirose, 1995; Miyake et al., 1955; Nakanishi et al., 1990; Hirose et al., 1992; Yang et al., 1986; Livingston, 1986; Livingston and Noshkin; 1999) as well as the eastern Equatorial Pacific (Cochran, 1985). More recent data were obtained from expeditions organized by IAEA-MEL in the framework of the WOMARS project (Povinec et al., 2003a), or carried out with IAEA-MEL participation (Mulsow et al., 2003; Povinec et al., 2003b).

The authors noticed that from the 1950s to the 1970s the decrease in radionuclide concentrations in surface water was much faster than after the 1970s. Therefore, only data from 1971 onwards were evaluated, although all other available data are shown in figures presented in this paper. Water samples collected down to 50 m below the sea surface have been considered in this study as surface samples. The radionuclide data set for the Pacific and Indian Oceans will be available soon on the IAEA website (http://maris.iaea.org).

2.2. Radionuclide surface water time series and effective half-lives

In general, concentrations of ^{90}Sr, ^{137}Cs and ^{239,240}Pu in surface water appear to decrease exponentially with time. Therefore, the temporal variations of their concentrations in surface water may be described by a flux model (e.g., Hirose et al., 1992)

$$dC/dt = -kC,$$

where C is the concentration of a given radionuclide at time t, and k is the flux coefficient. Thus, the slope of the ln C and t of the best fit in property plots may be considered as an effective half-life of a given radionuclide in surface water. The flux coefficient, k, is a combination of the radioactive decay constant (λ) and of the removal coefficient. The removal of a given radionuclide from surface water other than by radioactive decay (e.g. scavenging, etc.) depends on radioelement chemistry and the involvement of radionuclides in biogeochemical processes occurring in the sea. The concentrations of ^{90}Sr, ^{137}Cs and ^{239,240}Pu in a given ocean region are determined by the horizontal and vertical movements of water masses in the ocean, particle formation processes, and radioactive decay. ^{90}Sr and ^{137}Cs are mainly present in soluble form and their concentrations peak in subsurface or surface water, and decrease with depth (Nakano and Povinec, 2003; Hirose and Aoyama, 2003). However, ^{239,240}Pu is highly particle-reactive, therefore, it is more tied to the particle formation. It sinks with particles and regenerates in deeper waters as a result of the

biological decomposition of the particles (Hirose, 1997). Pu concentration peaks presently at ca. 700 m depth and it is expected that this peak will descend over time (Livingston et al., 2001).

Pelagic sediment/water distribution coefficients, (the ratio of the concentration of an element in pelagic clays to that in deep-ocean water, K_d values) of ^{90}Sr, ^{137}Cs, and ^{239,240}Pu are 200, 2000, and 1×10^5, respectively (IAEA, 2004). The concentration factors (the ratio of the quantity of an element in biological material (e.g. phytoplankton) to that in seawater, CF values) of ^{90}Sr, ^{137}Cs, and ^{239,240}Pu are 10, 100, 2000, respectively (IAEA, 2004). Due to the conservative nature of Sr and Cs, their distribution should be primarily related to mixing processes in the oceans (as in the case of tritium and ^{14}C). On the contrary Pu will be impacted by scavenging processes, more similar to some degree to Th, although the forms of Pu remaining in the ocean surface seem more conservative than first expected.

3. Results and discussion

3.1. Pacific Ocean

For studying the temporal variations of radionuclides in the Pacific Ocean (as well as in the Indian Ocean) the spatial distribution of radionuclides in surface waters of the studied areas should be close to uniform, sources of radionuclides in the areas should be well established, as well as their main oceanographic features, important for surface and vertical transport of water masses in the region.

In order to study the distribution of radionuclides in surface water of the Pacific Ocean and its marginal seas, the Pacific has been divided into 14 different latitudinal belts (boxes) on the basis of known ocean current systems, the location of nuclear weapons test sites, a relatively uniform distribution of radionuclides, and the availability of recent data (IAEA, 2001). Boxes 1–12 are in the Pacific Ocean and Box 13 is in the Southern Ocean (Fig. 1). The Sea of Japan has been kept separate (Box 14) because of its marginal oceanographic specificity and importance in marine radioactivity studies.

Box 1 (north of 40°N) is the subarctic Pacific region, where the largest fallout of ^{137}Cs, ^{90}Sr and ^{239,240}Pu over the Pacific Ocean was observed in the late 1950s and 1960s (Hirose et al. 2001). UNSCEAR (2000) reported the largest ^{90}Sr deposition on the earth in the latitude belt of 40°–50°N. 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, respectively, which is a typical oligotrophic ocean current corresponding to the subtropical gyre. Radionuclide concentrations in these boxes have been influenced by nuclear weapons tests carried out mostly at Bikini and Enewetak Atolls (Livingston et al., 2001; Livingston and Povinec, 2002). The subtropical region of the western N Pacific (Box 4) was more affected by local fallout as compared to the subtropical region of the eastern N Pacific (Box 5). Boxes 6 and 7 (5°S–5°N) are downstream and upstream of the

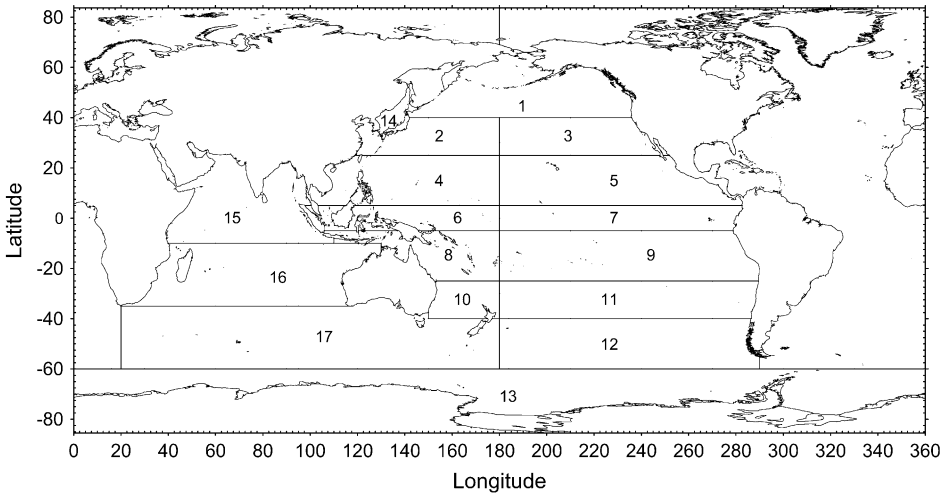

Fig. 1. Latitudinal boxes in the Pacific and Indian Oceans.

North Equatorial Current, respectively. The Equatorial Counter-Current forms a natural boundary between the tropical region (Boxes 4 and 5) and the equatorial region (Boxes 6 and 7). 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 the equatorial region (Boxes 6 and 7) and the tropical region (Boxes 8 and 9) approximately corresponds to the South Equatorial Counter-Current. Box 9 includes the French nuclear weapons test sites (Mururoa and Fangataufa Atolls). Box 10 (25°–40°S) corresponds to the Tasman Sea. Box 11 (25°–40°S) is the mid-latitude region of the South Pacific and includes the Southern Pacific Current. Box 12 (40°–60°S) corresponds to the Antarctic Circumpolar Current and it is connected to Box 17 in the Indian sector of the Southern Ocean. Box 13 (below 60°S) is in the Antarctic region and includes the polar front and the Continental Water Boundary.

Regression analysis of surface water ^{90}Sr, ^{137}Cs and ^{239,240}Pu concentrations developing with time has been used to estimate their average radionuclide concentrations in latitudinal boxes for the year 2000, as well as their effective half-lives. The estimated concentrations for the year 2000 were derived from the regression analysis of available measurement data in the time interval 1971–2000. In the N Pacific, the estimated concentrations of radionuclides for the year 2000 and the corresponding effective half-lives have been obtained with relatively good precision. However, if only a few data were available, then the average concentrations were estimated on the basis of recent measurements, in the case of ^{90}Sr and ^{137}Cs decay-corrected to January 1st, 2000.

^{90}Sr, ^{137}Cs and ^{239,240}Pu concentrations measured since 1950s in surface water of 13 regions of the Pacific Ocean are presented as a series of figures (Figs. 2–4). Concentrations in the 1950s and 1960s were mainly controlled by input processes

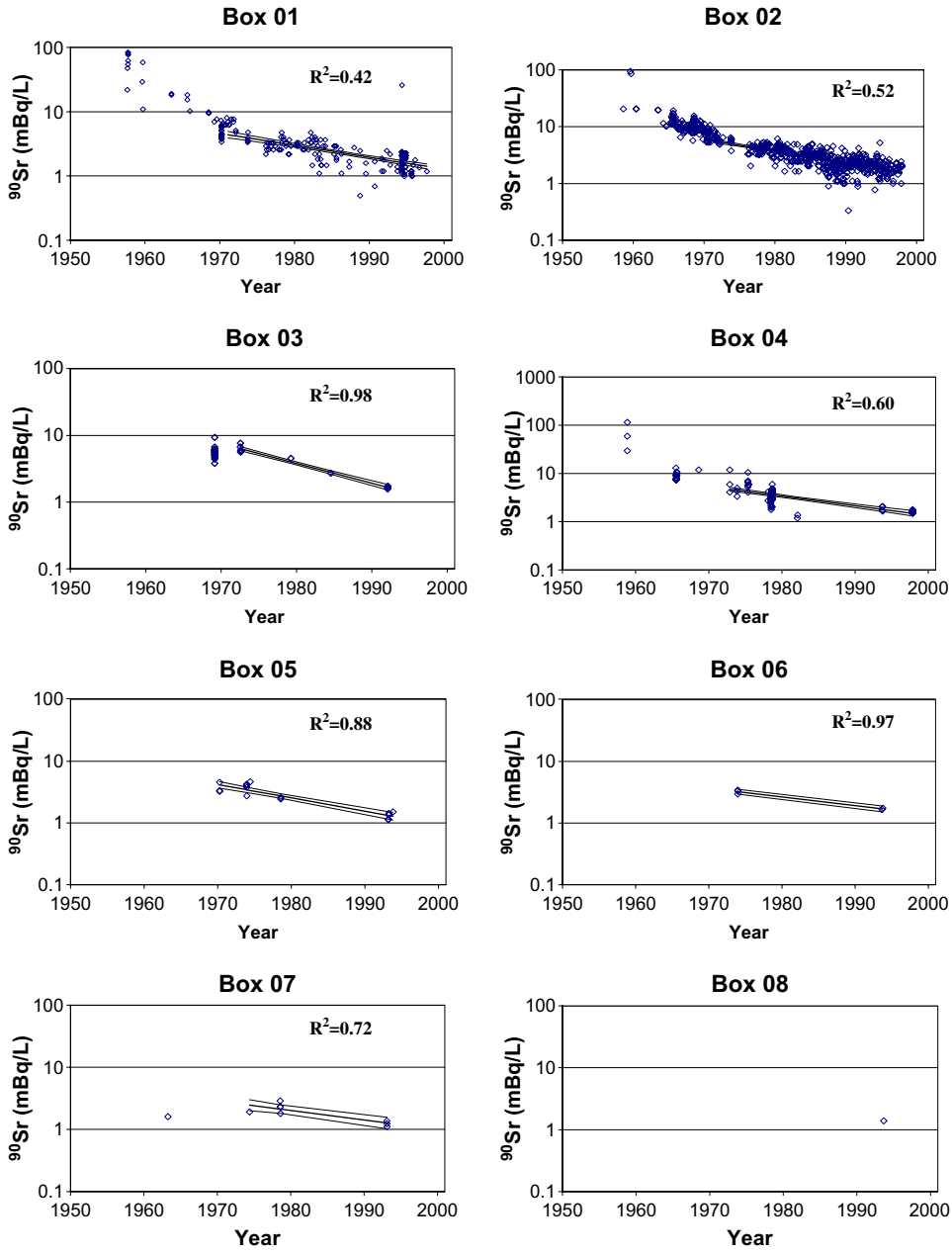

Fig. 2. ^{90}Sr in Pacific surface water (the thin lines represent in all figures the 95% confidence intervals; the correlation coefficients, R^2, are also given where possible).

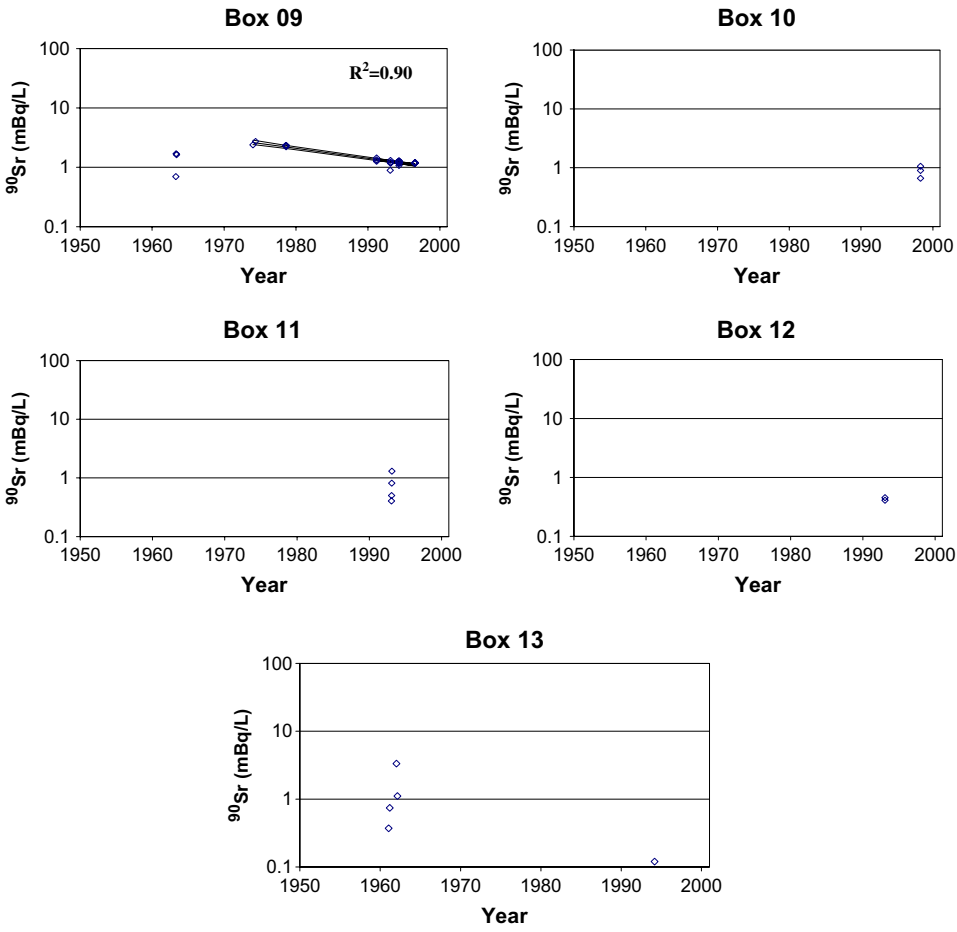

Fig. 2. (*continued*).

such as global and local fallout, and they were decreasing faster than in recent decades. However, starting from 1970s the radionuclide concentrations in surface water have been decreasing exponentially and they can be expressed in each box by a regression line. The 95% confidence intervals (shown in Figs. 2–4 as thin lines) vary greatly between boxes because of variations in data densities and radionuclide concentrations. These variations have also influenced the correlation coefficients (R^2 in Figs. 2–4), which vary between 0.02 and 0.98, showing no correlation between the data at one hand, or a strong correlation on the other hand, when homogeneous data sets have been available.

The average surface concentrations of ^{137}Cs, ^{90}Sr and ^{239,240}Pu calculated from the corresponding regression lines in each box of the Pacific for the year 2000 (or estimated from recent measurements if data sets have been very sparse) are presented

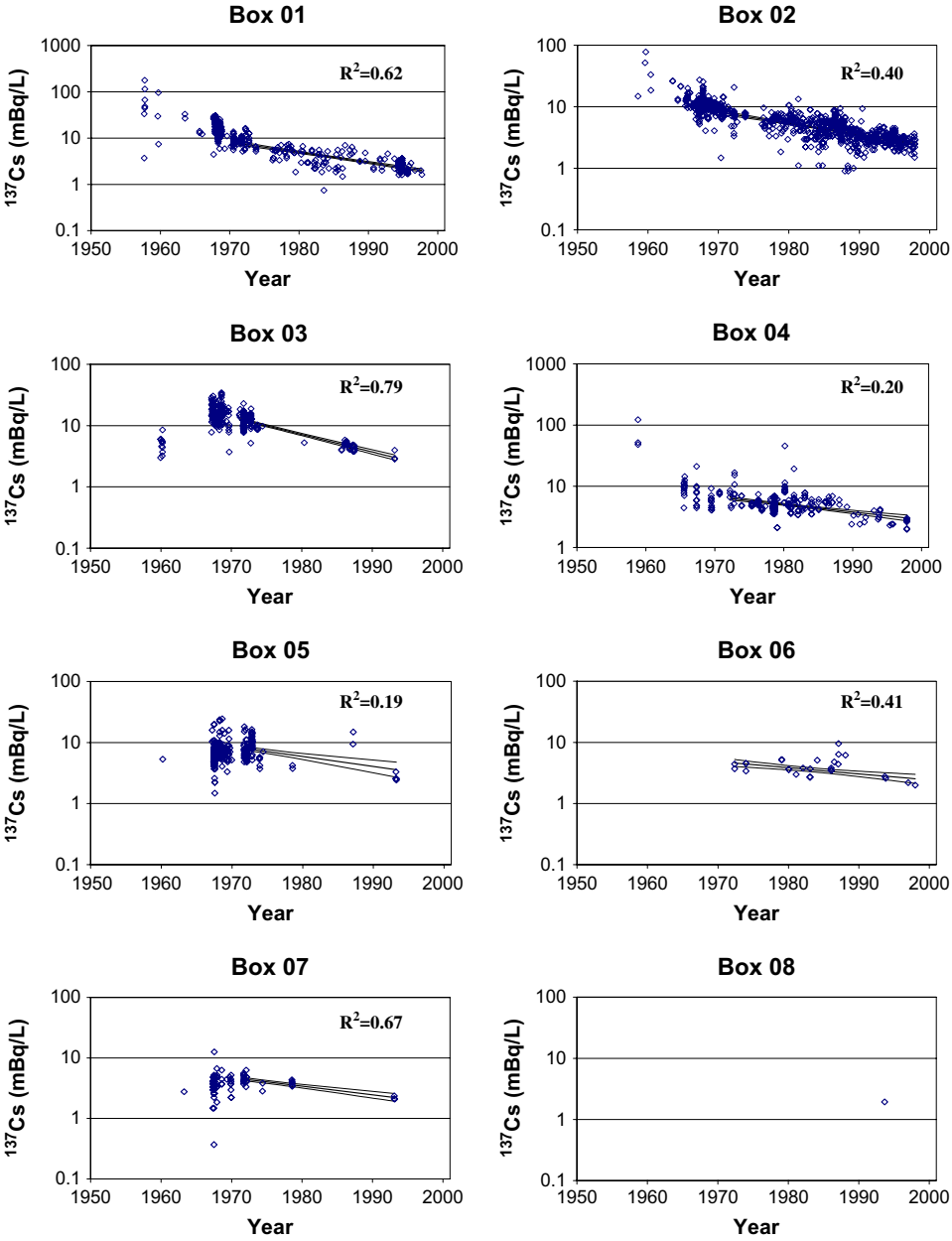

Fig. 3. ^{137}Cs in Pacific surface water.

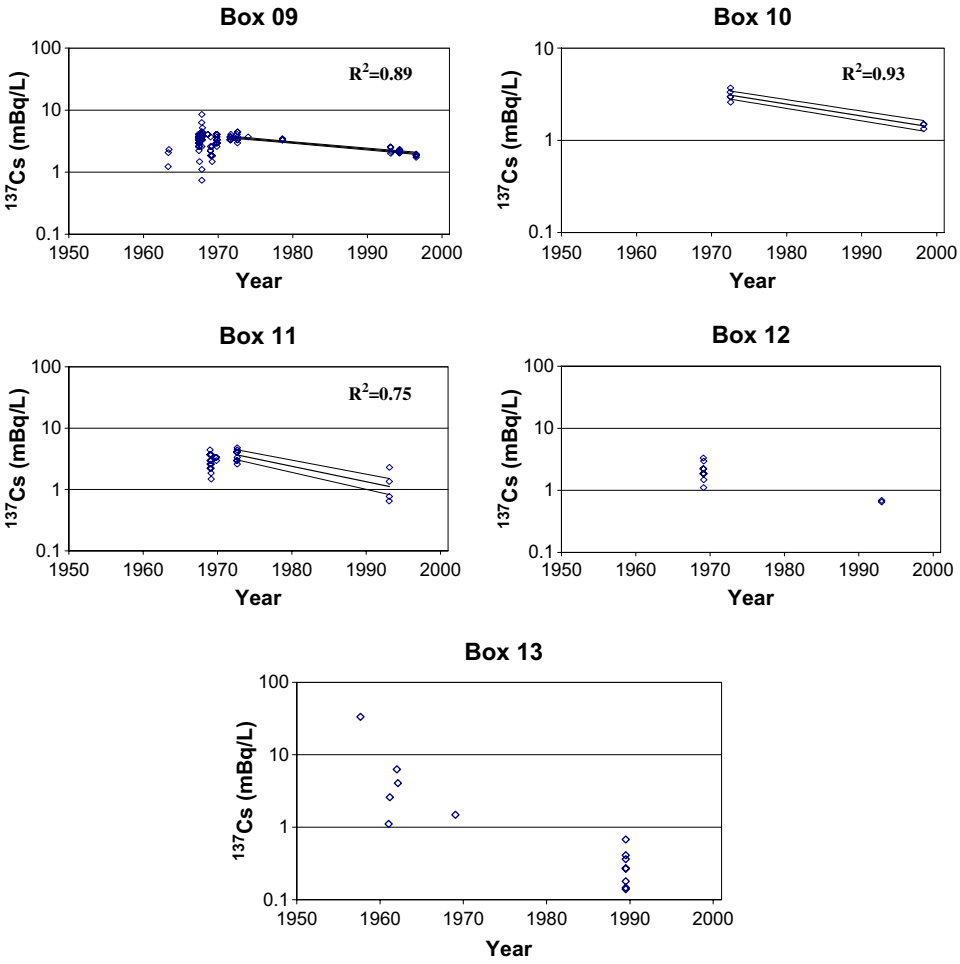

Fig. 3. (*continued*).

in Tables 1–3. The average ^{90}Sr concentrations in surface water are in the range of 0.4–1.5 mBq/L, except for Box 13 where the expected concentration is around 0.1 mBq/L (Table 1). Higher average surface ^{90}Sr concentrations (above 1.1 mBq/L) occur in the western N Pacific. The lowest values (below 0.4 mBq/L) appear in regions of high latitude in the South Pacific.

The calculated average ^{137}Cs concentrations in surface water for each box in 2000, together with the most recent data, are shown in Table 2. The average ^{137}Cs concentrations ranged from 0.6 to 2.8 mBq/L, except for Box 13 where the expected concentration is around 0.1 mBq/L. Similarly to ^{90}Sr, higher surface ^{137}Cs concentrations (above 2.3 mBq/L) occur in the western N Pacific. Lower values (below 1.3 mBq/L) appear in the South Pacific, and the lowest in the Antarctic region (0.1 mBq/L).

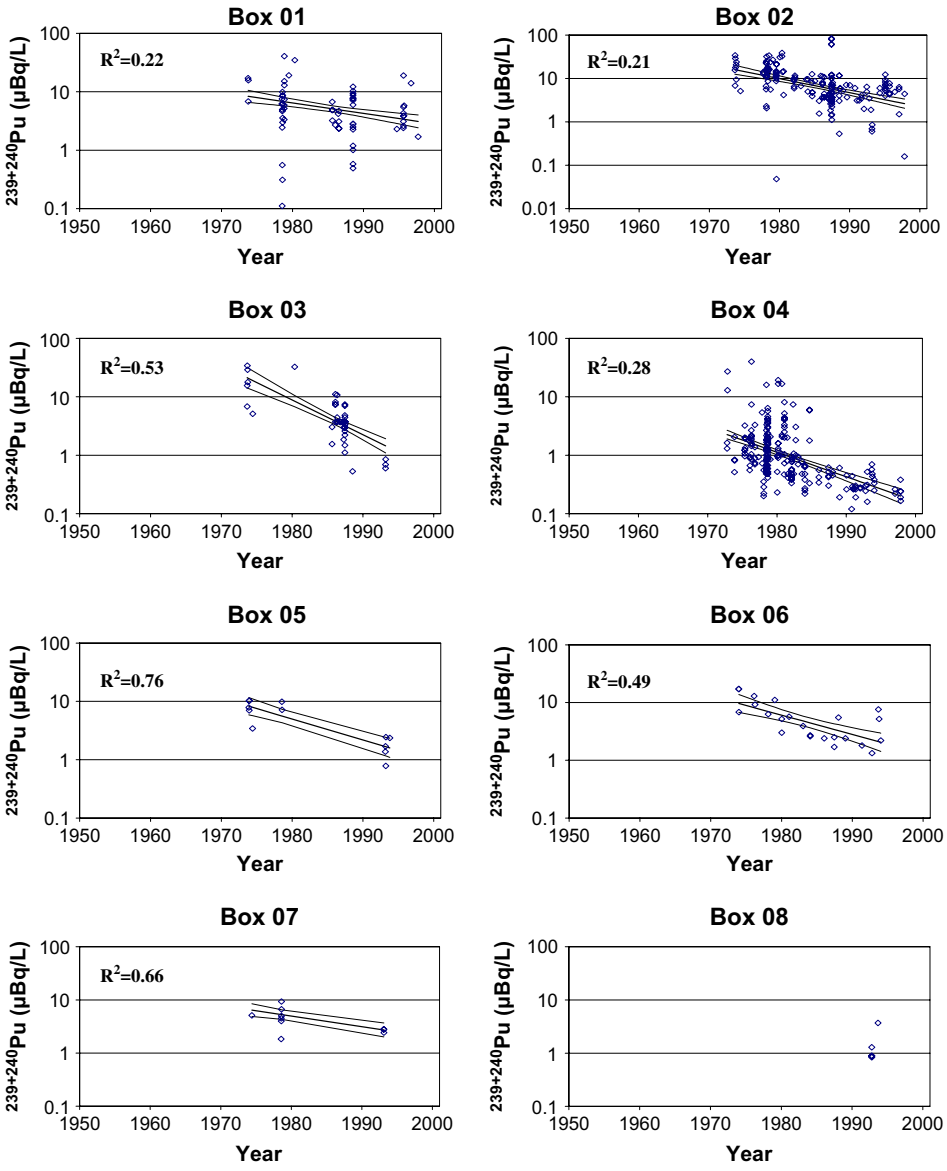

Fig. 4. ^{239,240}Pu in Pacific surface water.

The average ^{239,240}Pu concentrations in surface water in 2000 (Table 3) are estimated to be in the range of 0.5 (eastern N Pacific) to about 2.8 μBq/L in the subarctic Pacific (Box 1). There is only a small difference in surface ^{239,240}Pu concentrations between the mid-latitude regions of the Northern and Southern hemispheres. However, enhanced ^{239,240}Pu levels were observed in Box 4 affected by

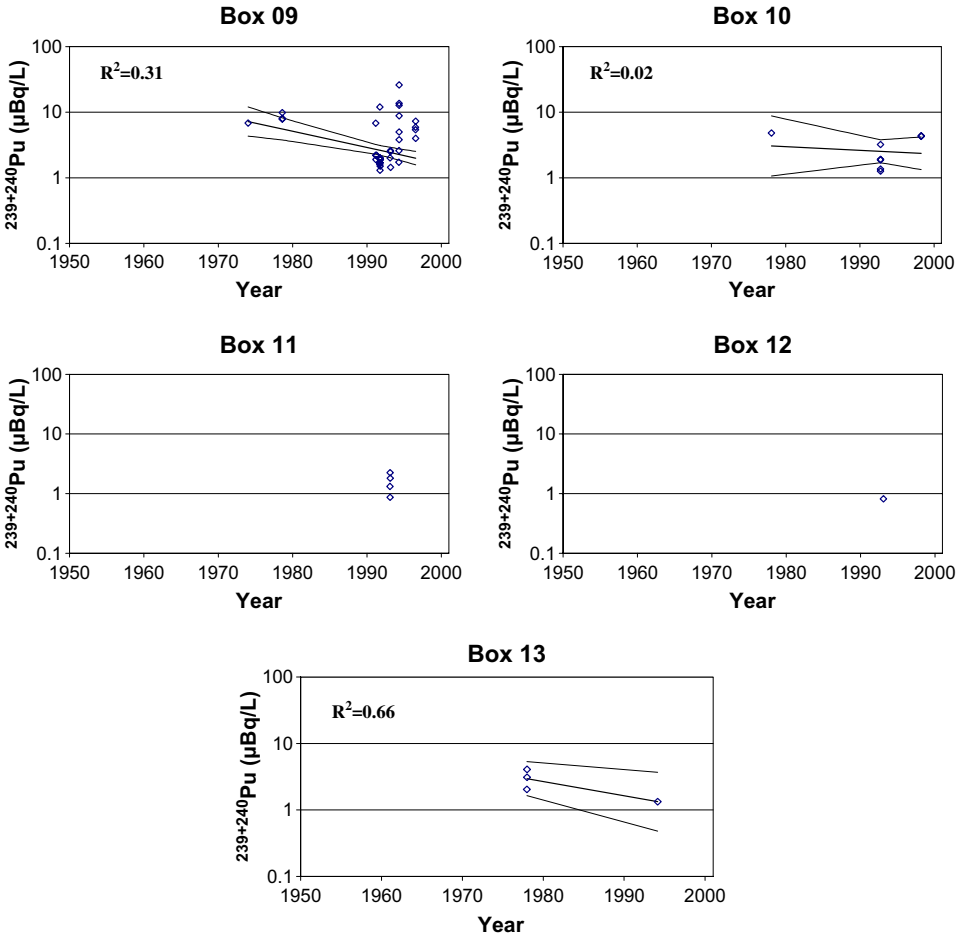

Fig. 4. (*continued*).

tests carried out at the Marshall Islands (Povinec et al., 2003a), as well as in the Tasman Sea, due to its low scavenging efficiency in low productivity waters (Povinec et al., 2003b).

In order to assess the environmental effects of ^{90}Sr, ^{137}Cs and ^{239,240}Pu, it is necessary to estimate their effective half-lives in each box. They can be calculated from the corresponding regression lines presented in Figs. 2–4. The results are summarized in Tables 1–3. Although in some cases the data sets were too small, the quoted 95% confidence intervals and the correlation coefficients (Figs. 2–4) gave reasonable estimates of the effective half-lives. The quoted weighted regression uncertainties are given as $\pm 1\sigma$, therefore one should keep this in mind when comparing results from different data sets. The effective half-life of ^{90}Sr in surface water ranged from 10 to 21 years. The shortest effective half-life (10.1 $\pm$ 0.5 years) is

Table 1
^{90}Sr effective half-lives and average concentrations in Pacific and Indian Ocean surface water for the year 2000

Box no.	Effective half-life[a] (year)	Est. concentration in 2000.01.01[a] (mBq/L)	Recent measurements		
			Averaged value[b] (mBq/L)	No. of data	Period
01	16.2 ± 1.5	1.3 ± 0.1	1.5 ± 0.3 (1.3)	3	1996–1997
02	14.4 ± 0.7	1.1 ± 0.1	1.7 ± 0.4 (1.6)	31	1996–1997
03	10.1 ± 0.5	1.0 ± 0.1	1.7 ± 0.1 (1.4)	3	1993
04	15.0 ± 1.4	1.2 ± 0.1	1.6 ± 0.1 (1.5)	13	1997
05	14.0 ± 1.4	0.9 ± 0.1	1.3 ± 0.2 (1.1)	5	1993
06	21.4 ± 2.0	1.5 ± 0.1	1.7 ± 0.1 (1.5)	2	1993
07	19.6 ± 5.0	1.0 ± 0.3	1.2 ±0.1 (1.1)	3	1993
08			1.4 ± 0.2 (1.2)	1	1993
09	17.9 ± 1.2	1.0 ± 0.1	1.2 ± 0.1 (1.1)	4	1996
10			0.9 ± 0.2 (0.8)	3	1998
11			0.8 ± 0.4 (0.6)	4	1993
12			0.4 ± 0.1 (0.4)	2	1993
13			0.1 ± 0.1 (0.1)	1	1994
14	14.1 ± 0.6	1.4 ± 0.1	1.8 ± 0.3 (1.6)	42	1996–1997
15			1.1 ± 0.2 (1.0)	15	1998
16			1.2 ± 0.2 (1.1)	7	1998–1999
17			0.7 ± 0.4 (0.7)	12	1998–1999

[a] Weighted regression uncertainties represent $\pm 1\sigma$.
[b] Decay-corrected values to January 1st, 2000 are given in brackets.

observed in the eastern N Pacific (Box 3). The longest effective half-lives, 21 ± 2 years and 20 ± 5 years, occur in the Equatorial Pacific, Boxes 6 and 7, respectively. The effective half-life of ^{137}Cs in surface water ranged from 11 to about 30 years. The shortest effective half-life (10.6 ± 0.5 years) is observed in the mid-latitude of the eastern N Pacific (Box 3). The longest effective half-life of ^{137}Cs is found in the Equatorial Pacific (30 ± 8 years for Box 6) and in the eastern subtropical South Pacific (28 ± 2 years for Box 9). The effective half-life of ^{239,240}Pu in surface water ranged from 5 to about 17 years. The shortest effective half-life (5.1 ± 1.7 years) is observed in the mid-latitude region of the eastern N Pacific (Box 3), as for ^{90}Sr and ^{137}Cs. The longest ^{239,240}Pu effective half-lives are found in the subarctic region (17 ± 4 years for Box 1) and in the Equatorial Pacific (15 ± 4 years for Box 7).

Observed effective half-lives of ^{90}Sr and ^{137}Cs are smaller almost in all boxes than the corresponding radioactive decay half-lives (28.15 and 30.15 years, respectively). In the case of ^{239,240}Pu the effect is even better visible, as the radioactive decay half-lives of ^{239}Pu and ^{240}Pu are much longer (24 110 and 6560 years, respectively). Therefore, the oceanic processes are important (even dominant in the case of ^{239,240}Pu) for removal of these radionuclides from the water column.

The obtained results suggest that in the mid-latitude region of the eastern N Pacific an enhanced removal of ^{90}Sr, ^{137}Cs and ^{239,240}Pu from surface water has been taking place. The surface water mass in this region has penetrated under the surface layer of the subtropical gyre. As a result, the ^{137}Cs subsurface maximum is formed in

Table 2
^{137}Cs effective half-lives and average concentrations in Pacific and Indian Ocean surface water for the year 2000

Box no.	Effective half-life[a] (year)	Est. concentration in 2000.01.01[a] (mBq/L)	Recent measurements		
			Averaged value[b] (mBq/L)	No. of data	Period
01	13.7 ± 0.8	1.7 ± 0.1	2.1 ± 0.5 (2.0)	4	1996–1997
02	16.5 ± 0.9	2.3 ± 0.1	2.6 ± 0.6 (2.4)	30	1996–1997
03	10.6 ± 0.5	1.9 ± 0.1	3.2 ± 0.6 (2.8)	3	1993
04	24.2 ± 3.1	2.8 ± 0.4	2.6 ± 0.4 (2.4)	7	1997
05	18.4 ± 4.2	2.7 ± 0.6	2.7 ± 0.4 (2.3)	4	1993
06	29.9 ± 7.7	2.4 ± 0.6	2.7 ± 0.1 (2.3)	2	1993
07	21.3 ± 3.0	1.7 ± 0.2	2.2 ± 0.1 (1.9)	3	1993
08			1.9 ± 0.4 (1.7)	1	1993
09	28.5 ± 1.6	1.8 ± 0.1	1.9 ± 0.1 (1.7)	4	1996
10	23.3 ± 2.6	1.3 ± 0.1	1.4 ± 0.1 (1.4)	3	1998
11	11.9 ± 2.0	0.7 ± 0.1	1.3 ± 0.8 (1.1)	4	1993
12			0.6 ± 0.3 (0.6)	2	1993
13			0.1 ± 0.1 (0.1)	1	1994
14	19.0 ± 0.9	2.5 ± 0.1	3.0 ± 0.5 (2.8)	44	1996–1997
15	20.2 ± 1.9	1.5 ± 0.1	1.6 ± 0.3 (1.6)	17	1998
16	26.3 ± 4.9	1.9 ± 0.4	2.2 ± 0.3 (2.1)	7	1998–1999
17	12.7 ± 7.0	0.7 ± 0.4	1.1 ± 0.6 (1.0)	12	1998–1999

[a] Weighted regression uncertainties represent $\pm 1\sigma$.
[b] Decay-corrected values to January 1st, 2000 are given in brackets.

the subtropical N Pacific (Bowen et al., 1980). The longest effective half-life of ^{239,240}Pu (except for the subarctic Pacific (Box 1) occurs in the eastern Equatorial Pacific (Box 7) due to the upwelling of subsurface water (with higher ^{239,240}Pu content) in the eastern equatorial region (Bowen et al., 1980), and thus maintaining there higher ^{239,240}Pu concentrations in comparison to other regions. The effective half-life of ^{239,240}Pu in each box is generally shorter than for ^{90}Sr and ^{137}Cs. Plutonium as a particle-reactive radionuclide in contrast to ^{137}Cs attaches to biogenic particles in surface water (Hirose and Sugimura, 1993), sinks with the particles, and regenerates in deeper water as a result of the remineralisation of the particles (Hirose, 1997).

When we group the ^{90}Sr data for the North, Equatorial and South Pacific (Table 4), the corresponding mean effective half-lives are 12 ± 1 years for the North, 18 ± 1 years for the South and 21 ± 2 years for the Equatorial Pacific. All Pacific data give the effective half-life of ^{90}Sr as 13 ± 1 years. The corresponding ^{137}Cs effective half-lives are 13 ± 1 years for the North, 22 ± 1 years for the South and 23 ± 3 for the Equatorial Pacific. The mean ^{137}Cs effective half-life for all the Pacific Ocean is 14 ± 1 years. The ^{239,240}Pu effective half-lives are 7 ± 1 years for the North, 12 ± 4 years for the South and 10 ± 2 years for the Equatorial Pacific. The mean ^{239,240}Pu effective half-life for all the Pacific Ocean is 7 ± 1 years.

Since the residence time of ^{90}Sr and ^{137}Cs in surface water of the S Pacific is longer than in the N Pacific, their concentrations will decrease more slowly with time in the

Table 3
^{239,240}Pu effective half-lives and average concentrations in Pacific and Indian Ocean surface water for the year 2000

Box no.	Effective half-life[a] (year)	Est. concentration in 2000.01.01[a] (μBq/L)	Recent measurements		
			Averaged value (μBq/L)	No. 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

[a] Weighted regression uncertainties represent $\pm 1\sigma$.

S Pacific than in the N Pacific. Therefore, after some time their concentrations in the S Pacific could be higher than in the N Pacific, without additional transport of these radionuclides from the N Pacific.

Recently Hirose and Aoyama (2003) investigated distribution of ^{137}Cs and ^{239,240}Pu in surface water of the Pacific Ocean using a similar WOMARS approach (IAEA, 2001). Although their HAM database for some of the boxes contains different data, their results are within uncertainties the same and conclusions similar to the present paper, which is based on the GLOMARD database. The present paper, however, also includes data on ^{90}Sr in the Pacific Ocean, and discusses results for all three radionuclides in the Sea of Japan and the Indian Ocean, as well.

Table 4
Mean effective half-lives of ^{90}Sr, ^{137}Cs and ^{239,240}Pu in Pacific Ocean surface water

Area	Boxes	Effective half-life[a] (year)		
		^{90}Sr	^{137}Cs	^{239,240}Pu
North Pacific Ocean	01–05	12 ± 1	13 ± 1	7 ± 1
Equatorial Pacific Ocean	06–07	21 ± 2	23 ± 3	10 ± 2
South Pacific Ocean	08–12	18 ± 1	22 ± 1	12 ± 4
All Pacific Ocean	01–12	13 ± 1	14 ± 1	7 ± 1

[a] Error weighted averaged values.

3.1.1. Sea of Japan

Box 14 (Sea of Japan) has the densest data sets of all the regions in the Pacific Ocean, largely due to Japanese researchers. ^{90}Sr and ^{137}Cs surface concentrations have been decreasing faster during the 1960s than from 1970s to the present, whereas surface ^{239,240}Pu concentrations show a slight decrease over the past two decades (Fig. 5). While the ^{90}Sr and ^{137}Cs data are well grouped around the estimated average values (except for higher ^{137}Cs levels observed after the Chernobyl accident in 1986), the Pu levels are much more scattered due to mixing processes in the water column. The average surface concentrations of ^{137}Cs, ^{90}Sr and ^{239,240}Pu in the Sea of Japan for the year 2000, estimated from the corresponding regression lines are 2.5 ± 0.1 mBq/L, 1.4 ± 0.1 mBq/L and 5.2 ± 0.9 μBq/L, respectively. The ^{137}Cs and ^{90}Sr levels are similar to the levels estimated for Boxes 2 and 4 in the western N Pacific, confirming that the horizontal transport of water masses by the Tsushima Warm Current (a branch of the Kuroshio Current) from the Pacific through the East China Sea over the sill of the Tsushima Strait into the Sea of Japan is responsible for the observed ^{137}Cs and ^{90}Sr concentrations in the Sea of Japan. However, the estimated average ^{239,240}Pu concentration in the Sea of Japan (around 5 μBq/L) is at least two times higher than the values estimated for any box in the Pacific. This may be explained by rapid cycling of ^{239,240}Pu between the surface and subsurface layers which is driven by cooling and strong winds during winter in the subarctic region of the Sea of Japan, transporting subsurface water with higher ^{239,240}Pu concentrations to the sea surface (Ito et al., 2003).

The mean effective half-lives of ^{137}Cs, ^{90}Sr and ^{239,240}Pu in surface water of the Sea of Japan are 19 ± 1, 14 ± 1, and 16 ± 3 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 effective half-life of ^{239,240}Pu (16 ± 3 years) seems to be very long in comparison with Box 4 (7 ± 1 years), however, it can be explained in a similar way as the high Pu levels observed in the Sea of Japan.

3.2. Indian Ocean

The Indian Ocean differs from the Pacific Ocean 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 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 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. During the South West Monsoon, the part of the SEC turning

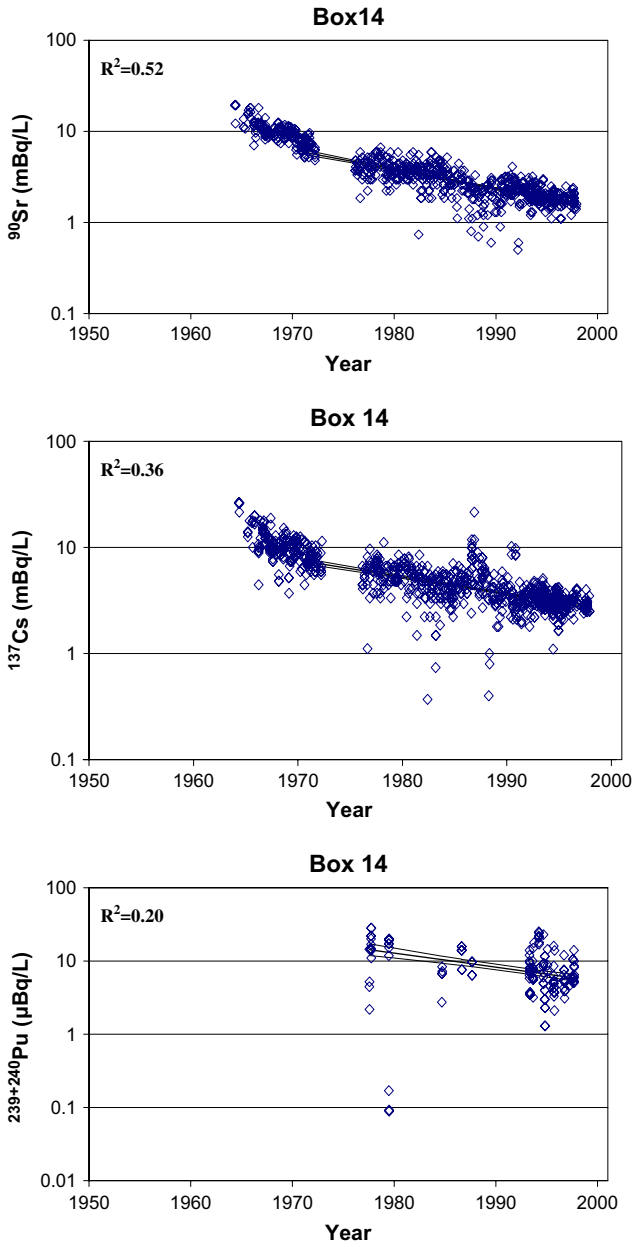

Fig. 5. ^{90}Sr, ^{137}Cs and ^{239,240}Pu time series in surface water of the Sea of Japan.

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. Complex interaction between water masses takes place at frontal regions in the Southern Ocean (south of 40°S) (Povinec et al., 2003b).

Following these oceanographic features, the Indian Ocean was divided into 3 boxes (Fig. 1). Box 15 covers the area affected by the monsoons, where seasonal currents flow. Box 16 is influenced by the South Equatorial Current. Box 17 is the zone defined by the Antarctic Circumpolar Current and is connected to Box 12 in the Pacific Ocean.

For the Indian Ocean only a few data on ^{90}Sr, ^{137}Cs and ^{239,240}Pu concentrations were reported. ^{137}Cs concentrations ranged between 1.6 and 2.3 mBq/L, while ^{90}Sr ranged from 1.1 to 1.5 mBq/L. Only 3 measurements were reported for ^{239,240}Pu prior to the year 2000 (around 1.4 μBq/L) (Miyake et al., 1988; Bourlat et al., 1996). The only comprehensive study was carried out in 1978 in the framework of the GEOSECS programme, which included several vertical profiles of tritium (and ^{14}C) in the water column (Broecker et al., 1986), however, no ^{90}Sr, ^{137}Cs and ^{239,240}Pu measurements were carried out. A latitudinal trend in tritium concentrations was observed, 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 (Broecker et al., 1986).

Recently in the framework of the WOMARS project new data were obtained for the Arabian Sea (Mulsow et al., 2003), as well as for the whole Indian Ocean (Povinec et al., 2003b). The ^{90}Sr, ^{137}Cs and ^{239,240}Pu concentrations (Tables 1–3) show the same latitudinal trend as the GEOSECS tritium: low concentrations of both ^{137}Cs and ^{90}Sr (1.1 ± 0.6 mBq/L and 0.7 ± 0.4 mBq/L, respectively – both values obtained from the recent measurements) characterize Box 17 (35°S –60°S). Proceeding north to Box 16 (10°S –35°S) higher concentrations of both radionuclides (2.2 ± 0.3 mBq/L for ^{137}Cs and 1.2 ± 0.2 mBq/L for ^{90}Sr) have been observed. Box 15 is again characterized with lower concentrations (1.6 ± 0.3 mBq/L for ^{137}Cs and 1.1 ± 0.2 mBq/L for ^{90}Sr). In the case of ^{239,240}Pu, the average levels in boxes are showing trends similar to conservative radionuclides, the highest concentrations were observed in Box 15 (3.0 ± 1.5 μBq/L) and the lowest in Box 17 (1.0 ± 0.5 μBq/L).

Although a latitudinal trend is still observed in the distribution of ^{90}Sr, ^{137}Cs and ^{239,240}Pu, it does not reflect the trend in the atmospheric input, as a re-distribution has taken place due to oceanic surface circulation. Higher radionuclide levels observed in Box 16 may be explained by the influx of Pacific waters through the Indonesian archipelago, and a system of equatorial currents that re-circulates surface water masses and maintains higher radionuclide concentrations in the region (Povinec et al., 2003b).

The ^{90}Sr, ^{137}Cs and ^{239,240}Pu surface water concentration time series for different latitudinal boxes in the Indian Ocean are presented in Figs. 6–8, and the estimated effective half-lives are given in Table 5. ^{90}Sr data were very sparse so any estimation of an effective half-life was not possible. ^{137}Cs data enabled to calculate its effective half-life which is around 20 years. The uncertainty in ^{239,240}Pu data was too high, except for Box 17, which gave an effective half-life of about 9 years. When the data are grouped together, the mean effective half-lives for ^{137}Cs and ^{239,240}Pu are 21 $\pm$ 2 years and 9 $\pm$ 1 years, respectively, in reasonable agreement with the Pacific results.

4. Conclusions

The Pacific and Indian Oceans were divided into 17 latitudinal boxes for which average concentrations of ^{90}Sr, ^{137}Cs and ^{239,240}Pu in surface water were estimated on the basis of time trends observed in radionuclide concentrations in surface water. The effective half-lives of the investigated radionuclides in surface water of latitudinal boxes were estimated as well. Generally, as expected, the differences between effective half-lives of ^{90}Sr and ^{137}Cs are within uncertainties, the same. It can be noticed that longer effective half-lives have been found in the Equatorial Pacific, as a result of specific circulation patterns there. The mean effective half-lives for ^{90}Sr and ^{137}Cs in surface water were 12 $\pm$ 1 years for the North, 20 $\pm$ 1 years for the South and 21 $\pm$ 2 years for the Equatorial Pacific. For ^{239,240}Pu the corresponding effective half-lives were 7 $\pm$ 1 years for the North, 12 $\pm$ 4 years for the South and 10 $\pm$ 2 years for the Equatorial Pacific. The grouped mean effective half-lives calculated for the Pacific Ocean are 13 $\pm$ 1 years for ^{90}Sr and ^{137}Cs, and 7 $\pm$ 1 years for ^{239,240}Pu. For the Indian Ocean, the mean effective half-lives are 21 $\pm$ 2 years for ^{90}Sr and ^{137}Cs, and 9 $\pm$ 1 years for ^{239,240}Pu.

The observed mean effective half-lives are much shorter that corresponding radioactive decay half-lives suggesting that oceanographic processes are important (even dominant in the case of ^{239,240}Pu) for removal of these radionuclides from the water column. The conservative nature of Sr and Cs in the water column is responsible for the fact that their distribution is primarily related to mixing in the oceans. On the contrary, Pu is heavily impacted by scavenging processes in the water column and therefore, as expected from other studies (e.g. Livingston et al., 2001), its mean effective half-life in surface water should be shorter than that for Sr and Cs. This study confirms on a global scale what has been learned earlier from regional studies, that because of different oceanic geochemistries (Sr and Cs vs. Pu) these radionuclides behave very differently in the water column (Livingston and Povinec, 2002).

There is evidence that the surface removal rates of Pu (and Cs and Sr as well) observed in the 1960s were higher than those observed during recent decades due to enhanced removal of particle-reactive forms initially followed by more conservative nature of all forms found in the ocean (Buesseler, 1997). Pu from local fallout in Pacific waters was removed quicker/deeper than Pu from global fallout (Buesseler, 1997; Livingston et al., 2001; Povinec et al, 2003a). Therefore, differences in source

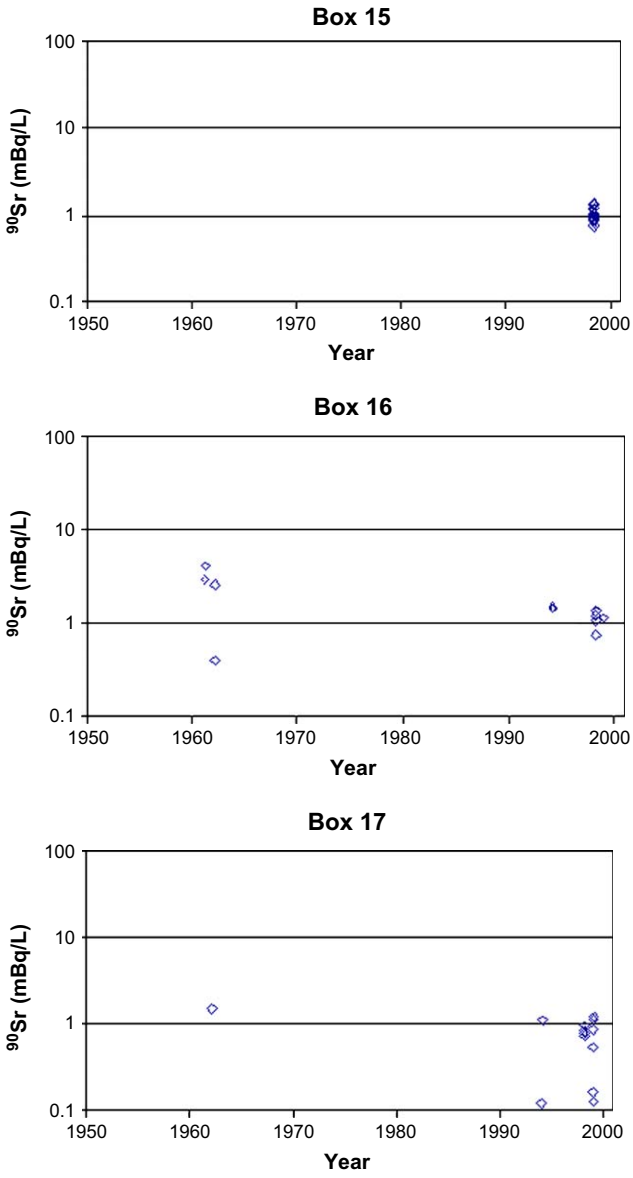

Fig. 6. ^{90}Sr time series in Indian Ocean surface water.

characteristics (i.e. global vs. local fallout) can have a significant effect on removal rates. Unfortunately, for most of the latitudinal boxes we have very limited data sets, which reflect the present status of investigation of the distribution of anthropogenic radionuclides in the world ocean.

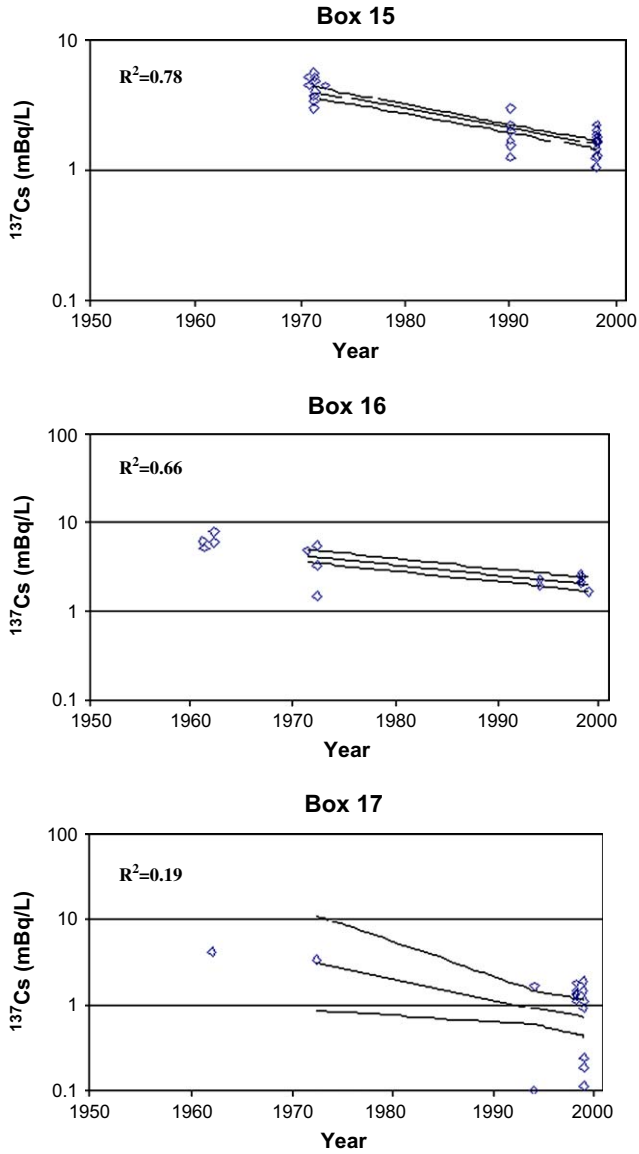

Fig. 7. ^{137}Cs time series in Indian Ocean surface water.

The results obtained in the framework of the WOMARS CRP provide the most complete available data set on levels of ^{90}Sr, ^{137}Cs and ^{239,240}Pu radionuclides in surface waters of the Pacific and Indian Oceans. The results can be used as the source on the average levels of anthropogenic radionuclides in the marine environment so that any further contributions from nuclear reprocessing plants, radioactive waste

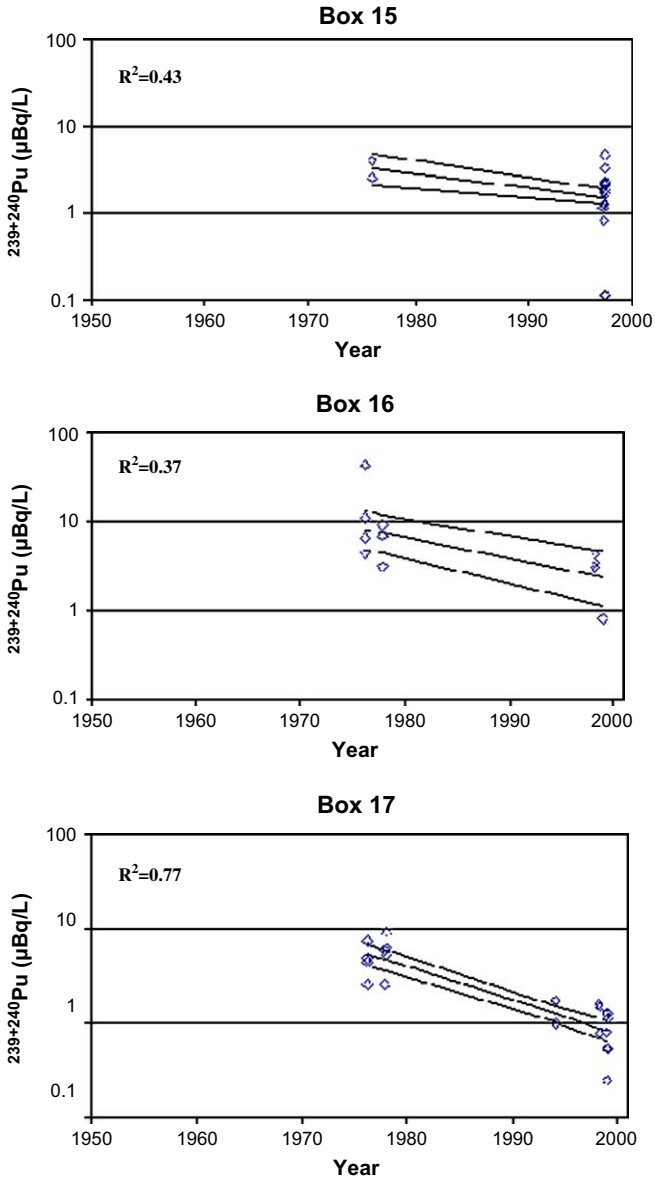

Fig. 8. ^{239,240}Pu time series in Indian Ocean surface water.

dumping sites, nuclear bomb test sites and possible nuclear accidents can be identified. As the fate of radionuclides for an assessment of adverse environmental or human health consequences needs to be well understood, the accumulated knowledge provides a critical basis for studying the impact of future potential releases of radionuclides into the marine environment.

Table 5
Mean effective half-lives of ^{137}Cs and $^{239,240}Pu$ in Indian Ocean surface water

Area	Boxes	Effective half-life[a] (year)	
		^{137}Cs	$^{239,240}Pu$
North Indian Ocean	15	20 ± 2	21 ± 8
South Indian Ocean	16 + 17	22 ± 4	9 ± 1
All Indian Ocean	15–17	21 ± 2	9 ± 1

[a] Error weighted averaged values.

Acknowledgements

This project would not have been possible without the generous support of the Government of Japan provided through the Science and Technology Agency. The support provided by the governments of France, India, Italy and the Russian Federation for IAEA-MEL's participation in international expeditions is highly acknowledged. Special thanks are due to Japanese institutions JCAC, MSA and FA for providing radionuclide data for the GLOMARD database. The authors would like to acknowledge colleagues who assisted in data collection and evaluation. The authors also thank three anonymous reviewers for their constructive comments. IAEA-MEL operates under a bilateral agreement between the IAEA and the Government of the Principality of Monaco.

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