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Radionuclides as Tracers of Water Fronts in the South Indian Ocean—ANTARES IV Results

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Anthropogenic ^{90}Sr, ^{239,240}Pu and ^{241}Am were used as tracers of water mass circulation in the Crozet Basin of the South Indian Ocean, represented by three main water fronts—Agulhas (AF), Subtropical (STF) and Subantarctic (SAF). Higher ^{90}Sr concentrations observed north of 43°S were due to the influence of AF and STF, which are associated with the south branch of the Subtropical gyre, which acts as a reservoir of radionuclides transported from the North to the South Indian Ocean. On the other hand, the region south of 43°S has been influenced by SAF, bringing to the Crozet Basin Antarctic waters with lower radionuclide concentrations. The ^{238}Pu/^{239,240}Pu activity ratios observed in water and zooplankton samples indicated that, even 35 years after the injection of ^{238}Pu to the Indian Ocean from the burn-up of the SNAP-9A satellite, the increased levels of ^{238}Pu in surface water and zooplankton are still well visible. The radionuclide concentrations in seawater and their availability to zooplankton are responsible for the observed ^{210}Po, ^{239,240}Pu and ^{241}Am levels in zooplankton.

Keywords:
· Anthropogenic radionuclides,
· seawater profile,
· zooplankton,
· ANTARES IV,
· Crozet Basin,
· South Indian Ocean.

1. Introduction

Natural and anthropogenic radionuclides have been used in many investigations as tools for tracing water masses in the open ocean (e.g. Bowen *et al*., 1980; Broecker and Peng, 1982; Livingston and Anderson, 1983; Broecker *et al*., 1986; Bayer and Schlosser, 1991; Livingston *et al*., 2001; Livingston and Povinec, 2002; Povinec *et al*., 2003a, b). Global fallout tracers extensively used in water circulation studies included ^{90}Sr (half-life 28.78 y) and ^{137}Cs (half-life 30.17 y). They are found mostly in a dissolved phase, they follow well the movement of water masses, and their removal from the water column is mainly due to their radioactive decay and diffusion (Livingston *et al*., 2001; Povinec *et al*., 2003a). Other useful anthropogenic tracers such as ^{238}Pu (half-life 87.74 y), ^{239}Pu (half-life 2.44×10^4 y), ^{240}Pu (half-life 6.58×10^3 y) and ^{241}Am (half-life 432 y) are particle-reactive and, due to their reactive nature, significant portions of these radionuclides in the world ocean have reached marine sediments (Bowen *et al*., 1980; Livingston and Anderson, 1983; Livingston *et al*., 2001; Lee *et al*., 2005).

The South Indian Ocean has been of interest to many research groups due to its key role in the regulation of CO_2 levels, biological productivity and biogenic fluxes. It represents a key region for the comprehension of the exchange of water masses between Antarctica and Equatorial regions, and it also plays important role in giving us a better understanding of oceanographic processes and the global climate (Samiento and Toggweiler, 1984; Taljaard and Van Loon, 1984; Michel *et al*., 1995; Gaillard, 1997; van Beusekom *et al*., 1997; DeMaster, 2002; Heinze, 2002; Ragueneau *et al*., 2002). However, only limited information on anthropogenic radionuclides in the South Indian Ocean has been available. Weiss and Roether (1980) and Broecker *et al*. (1986) studied the

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distribution of tritium in the Indian Ocean. A few results on ^{137}Cs, plutonium and americium distributions were published by Miyake *et al.* (1988). The absence of radionuclide sampling missions to the South Indian Ocean was mainly due to financial constraints as large volume water column sampling required for radionuclide analysis was very time consuming and therefore very costly. However, recent developments in underground counting (for ^{137}Cs) and mass spectrometry techniques (accelerator mass spectrometry or inductively coupled plasma mass spectrometry for Pu isotopes) have decreased the required sample size at least by a factor of ten, thus replacing large volume water samplers with frequently used Rosette systems (Povinec, 2005; Povinec *et al.*, 2008; Hirose *et al.*, 2009).

Due to these facts the South Indian Ocean was included in the recently completed international project Worldwide Marine Radioactivity Studies (WOMARS), coordinated by the International Atomic Energy Agency's Marine Environment Laboratories in Monaco (IAEA-MEL), and carried out in collaboration with several laboratories in Denmark, France, Germany, India, Italy, Japan, Korea (Republic of), New Zealand, Sweden, UK and USA (Povinec *et al.*, 2004, 2005). The aim of the project was to study the distribution and behaviour of anthropogenic radionuclides in the world ocean. Some of the results obtained for the North Indian Ocean have already been published (Povinec *et al.*, 2003b; Mulsow *et al.*, 2003; Bhushan *et al.*, 2003). This paper focuses on the results obtained from the ANTArctic RESearch (ANTARES) IV cruise describing the distribution of ^{90}Sr, ^{239,240}Pu and ^{241}Am in surface and deep waters, and in zooplankton in the South Indian Ocean. The paper deals mainly with radionuclides associated with water mass movement and the position of fronts.

2. Oceanographic Background

The Crozet Basin is well known for several frontal systems that meet there. The Agulhas Front (AF), Subantarctic Front (SAF), Subtropical Front (STF) and Polar Front (PF) represent narrow zones with sharp changes in temperature, salinity and oxygen content (Park *et al.*, 1991, 1993, 2002; Belkin and Gordon, 1996). As can be seen in Fig. 1, these frontal systems basically follow the complicated topography of the region. There is no information available on annual or seasonal variations of the fronts. The dominant physical control of the biogeochemical distribution of tracers in the South Indian Ocean is the banded structure of the Antarctic Circumpolar Current (ACC) (Gambéroni *et al.*, 1982; Pollard *et al.*, 2002). North of the ACC, there is a single narrow band (only about 200 km wide) of frontal zone along the shelf edge, where 80% of the ACC transport is concentrated (Park *et al.*, 1991). This frontal zone is formed by the confluence of the SAF and STF.

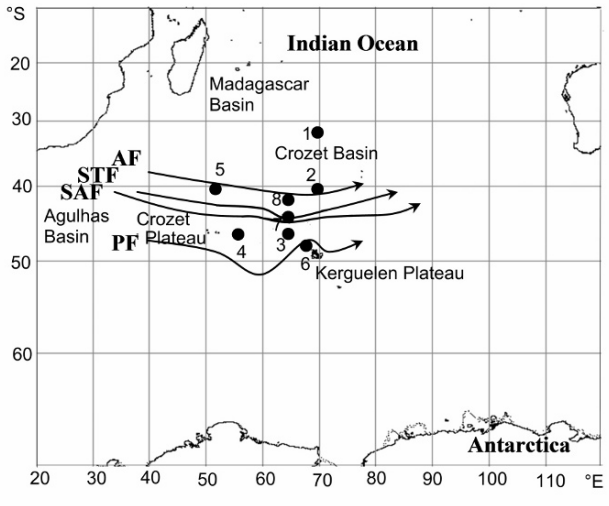

Fig. 1. Map of the study area for ANTARES IV cruise in the South Indian Ocean conducted aboard the R/V Marion Dufresne, showing the main water fronts flowing from the west to the east (AF—Agulhas Front, STF—Subtropical Front, and SAF—Subantarctic Front; modified after Park *et al.*, 1991, 1993), as well as seawater sampling stations. Zooplankton sampling was carried out at Stations 3, 7 and 8.

Sampling for the present study was carried out in the Crozet Basin (northwest of Kerguelen Plateau and east of Crozet Plateau) in the confluence of the AF, STF and SAF fronts (Coppola *et al.*, 2005, 2006). The most predominant current affecting the circulation in the Crozet Basin is the Agulhas Return Current (ARC), characterized by warm and saline water, extending eastward into the basin. Extending to east of 60°E, ARC re-circulates to the north, probably as part of an anticyclonic subtropical gyre (Park *et al.*, 1991). The STF is a boundary between the subtropical surface water and cooler, fresher subantarctic surface water (Pollard *et al.*, 2002). Temperature dominates the stratification north of the SAF, whereas to the south, salinity and temperature contribute about equally (Sievers and Nowlin, 1984; Nowlin and Klinck, 1986). The PF is not a part of the ACC main core, but is very close to the Kerguelen Plateau, since the island's climate and biogeography are subantarctic.

While the fronts represent the water mass structure on the surface of the Crozet Basin, the medium depth waters (300–1000 m) are influenced by Antarctic Intermediate Water (AAIW), the deep waters (1000–2500 m) by the North Atlantic Deep Water (NADW) and the North Indian Deep Water (NIDW), and the bottom waters by the Antarctic Bottom Water (AABW) (e.g. Park *et al.*, 2002).

It has therefore been great challenge to collect water samples in such a complex water transport region, and to use radionuclide tracers to investigate movement of water masses by frontal zones. This is the first time that radionuclide tracers have been used to study frontal systems in the South Indian Ocean.

3. Materials and Methods

3.1 Seawater samples

The ANTARES IV cruise was conducted in the South Indian Ocean (Fig. 1) in January–February, 1999, aboard the R/V Marion Dufresne (CNRS). Temperature and conductivity/salinity were measured using a commercial CTD mounted on the Rosette sampling system. Surface seawater was collected at eight stations in three main hydrological domains (AF, STF and SAF) between 33–48°S and 52–70°E by pumping from an average depth of 4 m. After filtering (0.45 μm mesh), onboard pre-concentration procedures were carried out to separate transuranics (MnO_2 co-precipitation), and ^{90}Sr by co-precipitation with oxalic acid. Transuranics were then purified at IAEA-MEL (using anion exchange resins and extraction chromatography), and electrodeposited on stainless steel disks for alpha-ray spectrometry (Lee *et al*., 2001). ^{90}Sr was determined by ^{90}Y in-growth followed by beta-ray counting (La Rosa *et al*., 2001). $^{18}O/^{16}O$ isotopic ratios were measured mass spectrometrically using the CO_2-H_2O equilibration procedure reported by Epstein and Mayeda (1953). The results are reported against the VSMOW (Vienna Standard Mean Ocean Water) as defined by Gonfiantini (1978) using conventional $\delta^{18}O$ notation.

3.2 Zooplankton samples

Zooplankton was collected at Stations 3, 7 and 8 during nights at depths above 200 m with a triple WP2 net (0.25 m^2 surface aperture and 200 μm mesh size). At some stations samples were collected during different nights to assure a better representation of collected zooplankton. Copepods were the dominant group in all net samples regardless of the area (Labat *et al*., 2002). The material was immediately frozen on board at –20°C. All zooplankton samples were freeze-dried at the laboratory and analysed following published procedures (Lee *et al*., 2001), so only a brief description will be given here. After dissolution of the sample by concentrated acids, ^{210}Po and transuranics were separated and purified using anion exchange resins and extraction chromatography. ^{210}Po was then electrodeposited on silver disks, and Pu and Am on stainless steel disks, and analysed by semiconductor alpha-ray spectrometry.

Radionuclide standards (Amersham) and IAEA reference materials (IAEA-381 Irish Sea water (Povinec *et al*., 2002; IAEA-414 mixed fish flesh from the Irish Sea and the North Sea (Pham *et al*., 2006)) for radionuclides in the marine environment were analysed simultaneously with the collected samples to assure data quality.

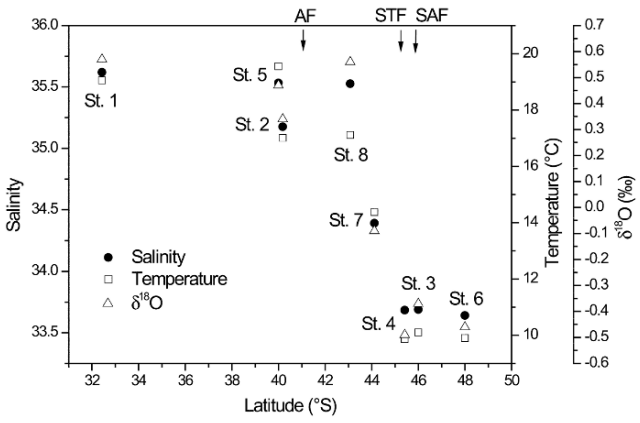

Fig. 2. Distribution of salinity, temperature and $\delta^{18}O$ as a function of latitude in South Indian Ocean surface water.

4. Results and Discussion

4.1 Radionuclides in seawater

The distribution of salinity, potential temperature and $\delta^{18}O$ as a function of latitude is shown in Fig. 2 (Table 1). We can group the stations visited into three groups: Stations 1, 2, 5 and 8 are located in the AF and north of STF, while Stations 3, 4 and 6 are located south of SAF. Station 7 lies between these two groups, located close to STF. Stations 1, 2, 5 and 8 are characterized by warmer (>17.00°C) and saltier (>35.2) waters. By contrast, Stations 3, 4 and 6, are characterized by cooler (<10.10°C) and less saline (<33.70) waters. Station 7 is characterized by a moderate temperature (14.37°C) and salinity (34.39). As a consequence, the fronts in the latitudinal bend of 40–48°S are acting as strong barriers, preventing mixing of water masses between them, so that the concentrations of radionuclides are well preserved in each of the frontal zones.

As shown in Fig. 3, the distribution of ^{90}Sr, ^{239,240}Pu and ^{241}Am concentrations in surface waters (Table 1) reveals a strong latitudinal variation defined by the water fronts. The surface ^{90}Sr concentrations are almost stable from 33°S to 43°S. These tendencies change remarkably from the south of 43°S, where the STF and SAF dominate, and where decreasing radionuclide levels were observed.

^{239,240}Pu shows increasing levels from 33° to 40°S, and then a decline down to 46°S; however, Station 6 situated at the most southerly site at 48°S shows almost a

Table 1. Concentrations of anthropogenic radionuclides in surface water of the South Indian Ocean.

Location	Station	Temperature (°C)	Salinity	$\delta^{18}O$ (‰)*	^{238}Pu (μBq l^{-1})	$^{239,240}Pu$ (μBq l^{-1})	^{241}Am (μBq l^{-1})	^{90}Sr (mBq l^{-1})	$^{238}Pu/^{239,240}Pu$	$^{241}Am/^{239,240}Pu$	$^{239,240}Pu/^{90}Sr$ $\times10^{-3}$
32°42.19′ S 70°00.00′ E	1	19.05	35.619	0.57	<0.14	0.78 ± 0.11	<0.30	1.15 ± 0.06	—	—	0.68 ± 0.10
40°17.98′ S 70°00.00′ E	2	17.01	35.177	0.34	0.19 ± 0.04	0.95 ± 0.07	0.53 ± 0.08	0.86 ± 0.05	0.20 ± 0.04	0.56 ± 0.10	1.10 ± 0.10
45°51.32′ S 55°00.00′ E	4	9.86	33.684	0.49	0.09 ± 0.03	0.49 ± 0.06	0.77 ± 0.14	0.13 ± 0.03	0.18 ± 0.06	1.6 ± 0.3	3.8 ± 1.0
40°00.00′ S 51°57.27′ E	5	19.55	35.533	0.47	0.10 ± 0.03	1.30 ± 0.10	0.45 ± 0.11	1.20 ± 0.07	0.08 ± 0.03	0.35 ± 0.09	1.08 ± 0.10
48°00.00′ S 68°38.50′ E	6	9.90	33.640	0.46	0.18 ± 0.05	1.40 ± 0.11	0.96 ± 0.08	0.07 ± 0.03	0.13 ± 0.04	0.69 ± 0.08	—
46°00.03′ S 63°03.58′ E	3	10.10	33.688	0.37	<0.19	0.28 ± 0.12	0.58 ± 0.08	0.16 ± 0.03	—	—	1.7 ± 0.8
44°11.45′ S 63°24.45′ E	7	14.37	34.392	0.09	<0.12	0.58 ± 0.05	0.52 ± 0.09	0.53 ± 0.03	—	0.90 ± 0.17	1.09 ± 0.11
43°08.03′ S 62°31.42′ E	8	17.11	35.526	0.56	0.10 ± 0.04	1.00 ± 0.10	0.30 ± 0.09	1.10 ± 0.10	0.10 ± 0.04	0.30 ± 0.10	0.91 ± 0.12

*Uncertainty at 1σ is ±0.1‰.

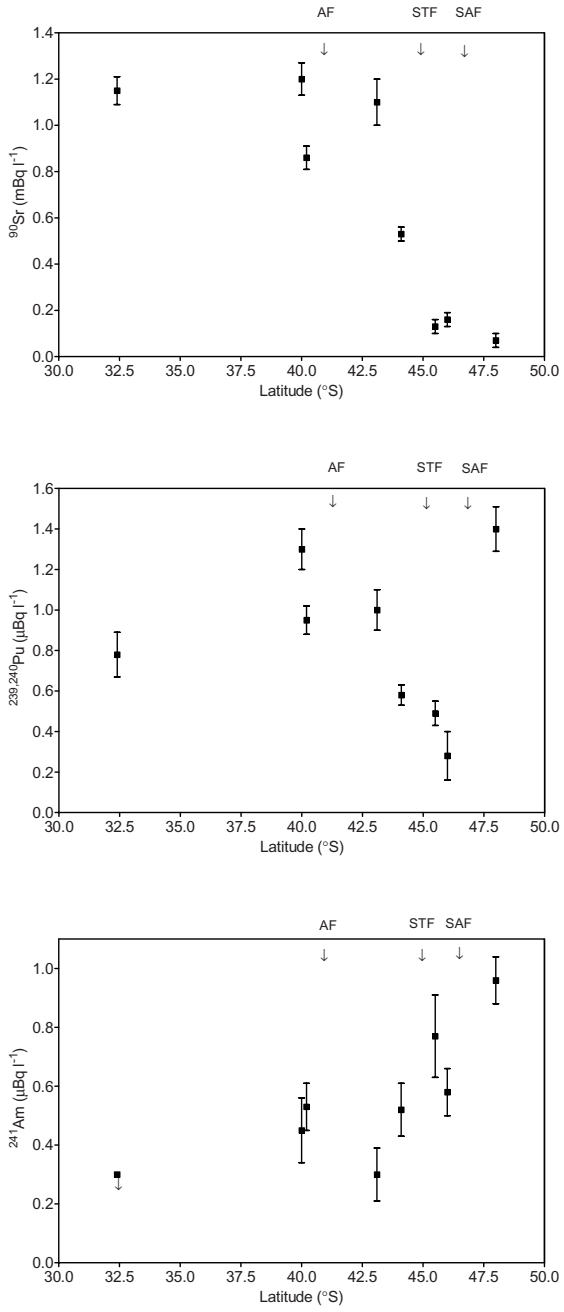

Fig. 3. Radionuclide concentrations in surface waters as a function of latitude (vertical lines represent the water fronts).

factor 5 higher ^{239,240}Pu concentration than Station 3, situated at 46°S. ^{241}Am does not fully copy the ^{239,240}Pu distribution (as would be expected, because of its similar particle reactive behaviour (Fowler *et al.*, 1983)), showing a peak at 40°S (Station 5), and a minimum at 43°S (Station 8). However, increasing levels were observed at Stations 7, 3 and 4 (in contradiction to the ^{239,240}Pu record), and at Station 6.

Most nuclear weapons tests with global radionuclide deposition were carried out in the Northern Hemisphere, and due to the specific stratosphere-troposphere mixing, the distinct latitudinal variations of the integrated global fallout density have been observed mainly in the Northern Hemisphere (UNSCEAR, 1993). Following the global deposition patterns, higher radionuclide concentrations are expected in the northern sampling areas (around 40°S), with values decreasing to the Equator and the South Pole.

Latitudinal variations in anthropogenic radionuclide concentrations related to global fallout deposition have been reported in several oceanic studies (e.g. Broecker *et al.*, 1986; Povinec *et al.*, 2003a, b). However, in the Southern Hemisphere the latitudinal effect of the integrated global fallout density is much weaker than in the Northern Hemisphere (Povinec *et al.*, 2003b; Mulsow *et al.*, 2003; Bhushan *et al.*, 2003).

Besides the global fallout patterns, there are several factors that could affect the distribution of radionuclides in surface waters of the South Indian Ocean, such as current systems, different water masses, primary productivity, etc. The circulation system of the equatorial currents and the monsoon-induced circulation has been found to play an important role in maintaining high radionuclide concentrations in the equatorial Indian surface waters (Povinec *et al.*, 2003b).

However, the ANTARES IV study area is distant from the equatorial region, and other circulation systems therefore have to be considered. We have already seen that the main force controlling the distribution of radionuclides in the Crozet Basin is the Sub-Antarctic frontal System (Fig. 1). Higher ^{90}Sr concentrations observed north of 43°S (Stations 1, 2, 5 and 8) are influenced by AF and STF, which are associated with the Subtropical gyre. The gyre accumulates radionuclides (with a period of around ten years; Y.-H. Park, personal communication) transported from the north-western Pacific Ocean via Indonesian Seas to the South Indian Ocean (Povinec *et al.*, 2003b, 2009). A similar observation was made in the framework of the SHOTS (Southern Hemisphere Ocean Tracer Studies) project, where higher radionuclide levels were found in surface waters of the South Indian Ocean along 20°S latitude, which is part of the northern branch of the Subtropical gyre (Povinec *et al.*, 2009). On the other hand, samples collected south of 43°S (Stations 3, 4, 6 and 7) have been affected by SAF, bringing to the Crozet Basin Antarctic waters with lower radionuclide concentrations.

The distribution of transuranics in surface waters of the Crozet Basin (Fig. 3) is different from that observed for ^{90}Sr. While transuranics are particle reactive elements, strontium is dissolved in the water column. Organic particles present in the seawater act as efficient scavengers of Pu and Am, which are later released in deep layers

Table 2. Natural (^{210}Po) and anthropogenic (^{238}Pu, ^{239,240}Pu and ^{241}Am) radionuclides in the mixed population of zooplankton (dry weight, dw) from the South Indian Ocean (sampling depth 0–200 m, net mesh 200 μm). At some stations several samples were collected during the cruise.

Station	^{210}Po (Bq kg^{-1} dw)	^{238}Pu (Bq kg^{-1} dw)	^{239,240}Pu (Bq kg^{-1} dw)	^{241}Am (Bq kg^{-1} dw)	^{238}Pu/^{239,240}Pu	^{241}Am/^{239,240}Pu
3	24.0 ± 0.3	—	0.018 ± 0.004	0.024 ± 0.006	—	1.33 ± 0.45
3	34.0 ± 0.3	—	—	—	—	—
3	89.0 ± 0.6	—	—	—	—	—
7	78.0 ± 0.6	—	0.023 ± 0.003	0.014 ± 0.003	—	0.61 ± 0.15
7	98.0 ± 0.6	—	0.025 ± 0.004	0.010 ± 0.002	—	0.40 ± 0.10
7	—	—	0.016 ± 0.005	—	—	—
7	—	—	0.013 ± 0.002	0.014 ± 0.003	—	1.08 ± 0.29
7	—	0.003 ± 0.001	0.022 ± 0.002	0.007 ± 0.002	0.14 ± 0.05	0.32 ± 0.10
8	130 ± 1	0.003 ± 0.001	0.019 ± 0.003	<0.002	0.16 ± 0.06	—
8	120 ± 1	—	—	—	—	—
8	—	—	0.024 ± 0.008	0.010 ± 0.004	—	0.42 ± 0.22
8	—	—	0.047 ± 0.018	0.019 ± 0.006	—	0.40 ± 0.20
8	—	—	0.019 ± 0.004	0.013 ± 0.004	—	0.68 ± 0.25
416*	—	0.080 ± 0.007	0.235 ± 0.014	0.018 ± 0.001	0.34 ± 0.04	0.077 ±0.015
417*	—	0.070 ± 0.007	0.267 ± 0.016	0.023 ± 0.002	0.26 ± 0.03	0.086 ± 0.009
418*	—	—	—	0.085 ± 0.015	—	—
419*	—	0.053 ± 0.013	0.220 ± 0.025		0.24 ± 0.06	—

*Mixed populations of zooplankton samples were collected during the cruise in the Arabian Sea in April 1998 in the framework of the WOMARS project (Mulsow *et al*., 2003).

during their decomposition (Bowen *et al*., 1980; Fowler *et al*., 1983; Livingston *et al*., 2001). On the other hand the distribution of strontium in the water column is affected mainly by physical processes (e.g. advection, radioactive decay; Povinec *et al*., 2003a). Therefore, primary productivity might be affecting concentrations of ^{239,240}Pu and ^{241}Am in surface waters.

However, the highest concentrations of ^{239,240}Pu and ^{241}Am observed at Station 6 (48°S), in contrast to the lowest ^{90}Sr levels observed at this station, should be associated with weather and topography. Station 6 is located at shallow water depth (100 m), and a severe storm which occurred during the sampling period (Coppola *et al*., 2005) might induce the surface seawater to mix with deeper water and/or resuspension of particles bearing ^{239,240}Pu from the bottom water and sediment. Therefore, if we exclude Station 6 from the ^{239,240}Pu record presented in Fig. 3, both ^{90}Sr and ^{239,240}Pu data shows a similar trend with latitude, thus indicating a dominant influence of the water fronts on the radionuclide concentrations.

The activity ratios of ^{239,240}Pu/^{90}Sr in surface water ranged in the interval $(0.7–3.8) \times 10^{-3}$, and they were significantly lower than the global fallout ratio (0.014). This may be due to a higher biological productivity in the region, followed by a greater scavenging of ^{239,240}Pu than ^{90}Sr from surface waters (Povinec *et al*., 2003b). Generally, the primary productivity in the South Indian Ocean is lower than in the Antarctic zone. (Labat *et al*., 2002; Mayzaud *et al*., 2002). Therefore, if the observed transuranics trends are related to productivity and global fallout deposition, the ^{241}Am record should follow that of ^{239,240}Pu, due to its similar behaviour. While the observed ^{239,240}Pu concentrations are about a factor of two lower than previous results observed by Miyake *et al*. (1988) in 1977–1978 for the eastern South Indian Ocean (effective half-life of ^{239,240}Pu is 9 ± 1 y; Povinec *et al*., 2005), for ^{241}Am they decreased only by ~30% in spite of the relative shorter effective half-life of ^{241}Am (~2 y; León Vintró *et al*., 1999). The main source of ^{241}Am in the region has been its in-growth from the decay of ^{241}Pu (half-life 14.4 y), after its delivery to the ocean by global fallout. Figure 3 also confirms, as expected, that Am is even more effectively scavenged from the surface water than Pu (and of course ^{90}Sr), resulting in a negative correlation between the ^{90}Sr and ^{241}Am levels. More information will be gathered when vertical profiles of ^{239,240}Pu and ^{241}Am in the water column of the South Indian Ocean become available.

The global fallout ratio of $^{241}Am/^{239,240}Pu$ is estimated to be ~0.4 in 1999. However, high yield US nuclear weapons tests carried in the fifties at the Marshall Islands (contributing about 30% of the total Pu inventory on the Ross Ice Shelf (Koide *et al*., 1982; Roos *et al*., 1994; Jia *et al*., 2000)), were characterized by a high $^{241}Pu/^{239,240}Pu$ ratio (~27, Koide and Goldberg, 1981; Koide *et al*., 1982; Jia *et al*., 2000), from which the $^{241}Am/^{239,240}Pu$ ratio estimated for 1999 should be ~0.7. The $^{241}Am/^{239,240}Pu$ activity ratios observed in surface waters in the Crozet Basin lie between 0.3 and 1.6, clearly indicating higher values for stations affected by SAF, carrying ^{241}Pu deposited in Antarctica by the Marshall Islands tests. These ratios are up to 8 times higher than those observed in the eastern South Indian Ocean during the Italica cruise in 1998, again supporting the hypothesis of the impact of Marshall Island tests on Pu and Am levels in the western South Indian Ocean.

A major injection of ^{238}Pu into the atmosphere occurred in April 1964 from the SNAP-9A accident, which occurred over the Mozambique Channel in the Indian Ocean. The total ^{238}Pu fallout over the Indian Ocean exceeded 2.5 times the fallout observed in the Northern Hemisphere (Perkins and Thomas, 1980). The surface ^{238}Pu concentrations in the South Indian Ocean ranged from the detection limit (<0.12 μBq l^{-1}) to 0.19 ± 0.04 μBq l^{-1} (Table 1), which are much lower (by a factor 2–4) than those observed during the Italica cruise in 1998 north of 40°S (Povinec *et al*., 2003b). The activity ratios of $^{238}Pu/^{239,240}Pu$ in surface water ranged from 0.08 ± 0.03 to 0.20 ± 0.04, and they were much higher than expected from global fallout (0.03). This result agrees well with previous observations (Povinec *et al*., 2003b), confirming the impact of the burn-up of the SNAP satellite on ^{238}Pu levels in the Indian Ocean surface waters, which can be still seen over 35 years after its injection.

4.2 Radionuclides in zooplankton

Concentrations of natural (^{210}Po) and anthropogenic radionuclides (^{238}Pu, $^{239,240}Pu$, and ^{241}Am) in a mixed population of zooplankton (Table 2) were determined in the three main fronts (AF, STF and SAF). ^{210}Po concentrations are largely different between stations, ranging from 24 to 130 Bq kg^{-1} dry weight (dw). Large variability in ^{210}Po levels (outside of analytical uncertainties) was observed for samples collected at the same sampling stations. By contrast, $^{239,240}Pu$ and ^{241}Am concentrations did not show such variabilities, the observed ranges lying between 0.01 and 0.05 Bq kg^{-1} dw for $^{239,240}Pu$ and from <0.002 to 0.02 Bq kg^{-1} dw for ^{241}Am.

To identify factors that may influence radionuclide concentrations in zooplankton, the observed values are plotted as a function of latitude (Fig. 4). The distribution of ^{210}Po clearly shows a latitudinal effect (coefficient of determination $R^2 = 0.79$). The zooplankton collected at AF and STF has more ^{210}Po than that collected at SAF. The latitudinal dependence is therefore driven by the position of the front, as ^{210}Po is generally uniformly distributed in the open ocean (Aarkrog *et al*., 1997).

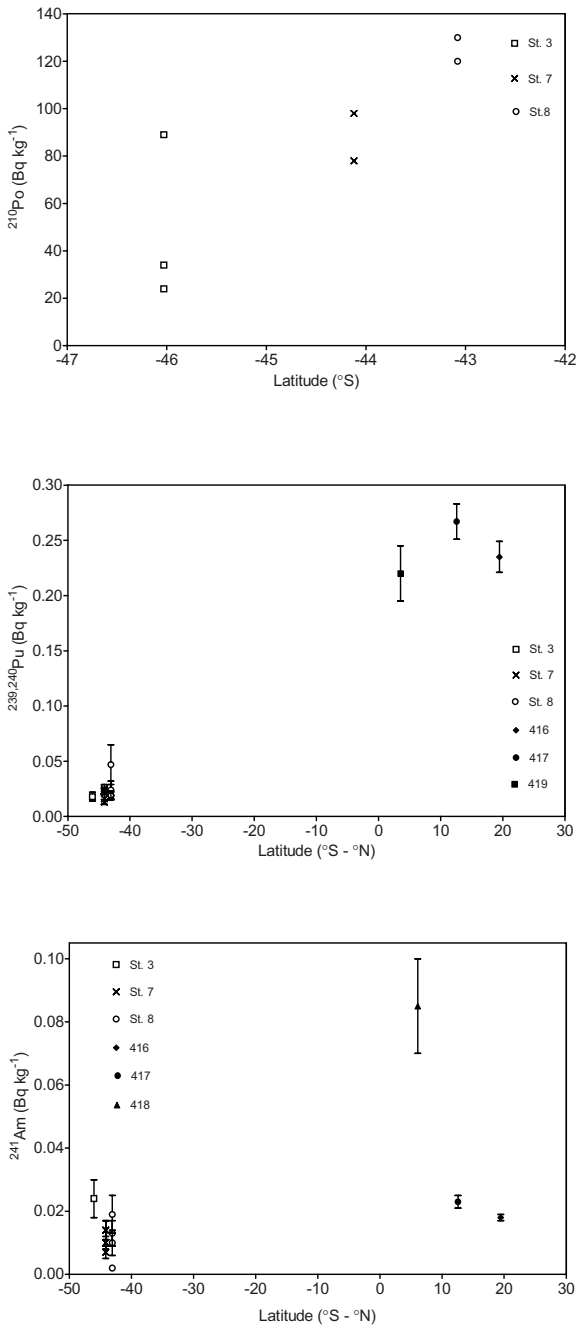

Fig. 4. Radionuclides in mixed population of zooplankton (sampling depth: 0–200 m; net mesh: 200 μm) as a function of a latitude in the South Indian Ocean (full circles) and in the Arabian Sea (Mulsow *et al*., 2003) (open circles).

^{239,240}Pu concentrations in zooplankton did not show a latitudinal dependence in the studied area; however, the differences in ^{239,240}Pu levels of seawater at stations where zooplankton was collected were not statistically significant (the zooplankton samples were collected at only three stations). Additional data from samples collected in the Arabian Sea (Mulsow *et al*., 2003), which are also plotted in Fig. 4, showed much higher levels, clearly associated with much higher concentrations of ^{239,240}Pu in surface waters (Livingston and Povinec, 2000; Povinec *et al*., 2003b).

Similarly, ^{241}Am concentrations in zooplankton collected in the Crozet Basin did not show any distinct trend with latitude, as the ^{241}Am levels in seawater at the sampling stations were statistically similar. Even when samples from the Arabian Sea (Mulsow *et al*., 2003) are included, the latitudinal dependence is not statistically significant.

The ratio of surface to volume of the collected zooplankton species (i.e. large vs. small species in the net) has been regarded as one possible factor controlling the concentrations of radionuclides in zooplankton (Cherry *et al*., 1987; Jeffrey *et al*., 1997; Hong *et al*., 2002). The species of zooplankton and their size collected at sampling stations and used in analyses were similar (Labat *et al*., 2002). Therefore, these parameters should not be critical factors affecting radionuclide concentrations in zooplankton. As a result, differences in the availability of elements for zooplankton (e.g. Po vs. Pu and Am), and differences in radionuclide concentrations in seawater, seem to be primary factors controlling the radionuclide levels in zooplankton.

^{238}Pu/^{239,240}Pu activity ratios in zooplankton samples varied between 0.14 and 0.34, clearly indicating the impact of the SNAP satellite (Livingston and Povinec, 2000) on the ^{238}Pu content of zooplankton. ^{241}Am/^{239,240}Pu ratios in the Crozet Basin zooplankton varied between 0.3 and 1.3, which is much higher than the ratios observed in the Arabian Sea zooplankton (0.08). This is mainly due to higher ^{241}Am seawater levels observed in the Crozet Basin than in the Arabian Sea (Povinec *et al*., 2003b).

5. Conclusions

The South Indian Ocean is a key region for the comprehension of the exchange of water masses between Antarctica and Equatorial regions, playing important role in affording us a better understanding of oceanographic processes and the global climate. This was the first time that radionuclides were measured in the Crozet Basin, where different water fronts meet in a relatively small area. The results obtained may be summarised as follows:

(i) Strong latitudinal variations of anthropogenic radionuclide concentrations have been found between 40–48°S in the South Indian Ocean, which are attributable to different water fronts present in the Crozet Basin. Higher ^{90}Sr concentrations observed north of 43°S were associated with the Subtropical gyre, which acts as a reservoir of radionuclides transported from the north-western Pacific Ocean via Indonesian seas to the North Indian Ocean and then to the South Indian Ocean. The subtropical region was under the influence of AF and STF, which are associated with the southern branch of the Subtropical gyre. On the other hand, the region south of 43°S has been affected by SAF, bringing to the Crozet Basin Antarctic waters with lower radionuclide concentrations.

(ii) Higher ^{238}Pu concentrations compared to ^{239,240}Pu observed in surface waters of the Southern Indian Ocean are associated with earlier fallout deposition from the SNAP satellite which burned over the Mozambique Channel. This signal is clearly visible in surface waters and zooplankton, even 35 years after ^{238}Pu injection into the Indian Ocean.

(iii) Differences in availability of elements for zooplankton (e.g. Po vs. Pu and Am), and differences in radionuclide concentrations in seawater, seem to be primary factors controlling the ^{210}Po, ^{239,240}Pu and ^{241}Am concentrations in zooplankton.

The observed distribution of radionuclides in the Crozet Basin may therefore be regarded as a useful complementary approach for tracing water masses in the ocean using anthropogenic radionuclides as tracers.

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