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Distribution of ^{137}Cs, ^{238}Pu and $^{239+240}Pu$ in sediments of the southeastern Brazilian shelf–SW Atlantic margin

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Abstract

In this work levels of ^{137}Cs, ^{238}Pu and $^{239+240}Pu$ as well as activity ratios of anthrophogenic radionuclides in sediment samples from the southeastern Brazilian shelf are presented. Instrumental gamma spectrometry was used to determine ^{137}Cs and alpha spectrometry to determine ^{238}Pu and $^{239+240}Pu$ after a radiochemical procedure. The levels ranged from 0.30 to 1.79 Bq kg^{-1} for ^{137}Cs, from 15 to 150 mBq kg^{-1} for ^{238}Pu and, from 18 to 117 mBq kg^{-1} for $^{239+240}Pu$. There was a bathymetric differentiation in the radionuclides distribution. ^{137}Cs values were generally higher in the samples collected at water depths of less than 100 m. On the other hand, plutonium isotopes exhibit higher values at greater depths. The mean ratio of $^{239+240}Pu/^{137}Cs$ obtained was 0.112 ± 0.072 which is in agreement with the value reported for the Atlantic from atmospheric fallout of nuclear explosions in the past. The $^{238}Pu/^{230+240}Pu$ ratios varied widely (from 0.339 to 2.088) and showed the influence of the SNAP-9A accident in the ^{238}Pu levels for this area. The main goal of this work was to present unpublished anthropogenic radionuclide levels and activity ratios related to the contamination of the southeastern Brazilian shelf.
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1. Introduction

Radioactive pollution is as harmful as other forms of pollution. Thus the monitoring of anthropogenic radionuclides in the marine environment is of great importance, since the seas and oceans constitute the major repositories of this kind of element.

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Concentrations of anthropogenic radionuclides in the marine realm generally vary from region to region, according to the location and magnitude of the different sources of contamination. Radionuclides have been released into the environment from a multiplicity of sources, both planned and accidental (Calmet and Sjöeblom, 1992; Livingston and Povinec, 2000).

Radionuclides are powerful tracers for a variety of marine processes. Due to the relatively well-defined temporal and spatial characteristics of the introduction of radionuclides into the ocean, the knowledge of their input, transport and fate within the different marine compartments provides many insights into a large number of processes within the water column, and in both biological and sedimentary systems. The distinctly different geochemical behaviour of these radionuclides in the aqueous system makes this set of tracers valuable for investigating water circulation and sedimentary processes (Baskaran et al., 1996; Noureddine and Baggoura, 1997; Livingston and Povinec, 2000).

Among the radionuclides produced artificially and released into the marine environment ^{137}Cs and ^{239}Pu are of great importance. ^{137}Cs has a high fission yield and a half-life of 30 years, while ^{239}Pu has a longer half-life (24,000 years) and is extremely dangerous when released into the environment.

In the marine environment, these radionuclides may be retained by the sediments through fixation in suspended matter and sedimentation, direct precipitation of colloidal forms and the direct fixation by adsorption and deposition of organic matter which had previously incorporated the radionuclides (Ligero et al., 2004).

In the Southern Hemisphere, the main source of radioactive contamination for the South Atlantic is the fallout from past nuclear tests. The radionuclide inventories for this area are approximately 17.5 PBq of ^{137}Cs and 0.5 PBq of $^{239+240}$Pu (Aarkrog, 2003). Another main source of radioactive contamination was the accident with the SNAP-9A satellite, which contained a nuclear power generator onboard that released 0.6 PBq of ^{238}Pu into the atmosphere. Due to this accident, seawater samples from the Southern Hemisphere have shown enhanced ^{238}Pu/$^{239+240}$Pu activity, nearly an order of magnitude greater than that in ocean water from the Northern Hemisphere (Whitehead, 1988; Holm et al., 1991; Aarkrog, 2003). Detectable levels of artificial radionuclides have also been reported in the Antarctic region (Jia et al., 2000; Marzano et al., 2000; Desideri et al., 2003; Pourchet et al., 2003).

A research programme to establish the environmental radioactivity in marine samples from the southeastern Brazilian shelf has been carried out since 1993 to determine the levels of antrophogenic radionuclides. This area shows low levels of artificial radionuclides due to the atmospheric fallout, as previously reported in the literature (Cunha et al., 1993, 1999; Figueira et al., 1998, 2001, 2004; Saito et al., 2001a,b; Godoy et al., 2003). This work presents the levels of ^{137}Cs, ^{238}Pu and $^{239+240}$Pu and the activity ratios of manmade radionuclides in surface sediment samples collected in 1997 on the southeastern Brazilian shelf. The results obtained here are important for our knowledge of artificial radioactivity in the Southeastern Atlantic, an area for which the data on such parameters is scarce. In this sense our results may be taken as reference values for the monitoring of this region.

2. Study area

The study area corresponds to the southeastern Brazilian upper margin (Fig. 1), between the latitudes 28°40′S and 23°00′S, an area of approximately 320,000 km^2. The shelf width ranges from 73 to 231 km with an inclination of between 1:656 and 1:1333, the shelf break being located at between 120 and 180 m depth. As a rule, the inner shelf is covered with quartzose sandy sediments. From the 50-m isobath to the shelf break muddy sediments prevail. A few areas characterized by the occurrence of carbonate gravels are related to the presence of relict sediments (Mahiques et al., 2004). Most of the drainage systems run from the coastal range westwards into the interior, being part of the Parana-La Plata river basin; only small rivers draining granitic, gneissic and migmatitic terrains, run directly into the sea.

The southern sector of the study area presents allochthonous sediments, originating in the younger rocks of the Andean chain and reaching the shelf via

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