Marine Pollution Bulletin xxx (2016) xxx–xxx

Contents lists available at ScienceDirect

Marine Pollution Bulletin

journal homepage: www.elsevier.com/locate/marpolbul

Natural and man-made radionuclides in sediments of an inlet in Rio de Janeiro State, Brazil

Franciane Martins de Carvalho *, Dejanira da Costa Lauria, Fernando Carlos Araújo Ribeiro, Rafael Tonelli Fonseca, Sueli da Silva Peres, Nádia Soido Falcão Martins

Institute of Radiation Protection and Dosimetry (IRD), Av. Salvador Allende s/n — Barra da Tijuca, Rio de Janeiro — RJ, 22783-127, Brazil

ARTICLE INFO

Article history:
Received 29 November 2015
Received in revised form 21 March 2016
Accepted 27 March 2016
Available online xxxx

Keywords:
Radionuclides
Surface sediments
Geochemical mapping
Inlet
Brazil

ABSTRACT

The distribution of natural radionuclides ^{226}Ra, ^{228}Ra, ^{40}K and man-made radionuclides (^{54}Mn, ^{60}Co and ^{137} Cs) in the surface sediments of an inlet of Ribeira Bay were investigated. Sediment samples were collected and analyzed for radionuclides, organic matter, carbonate, sulfate, cationic exchange capacity and grain size composition. The natural radionuclide concentrations ranged from 4.4 to 45, from 10 to 93, from 66 to 1347 Bq·kg^{-1} dry weight for ^{226}Ra, ^{228}Ra and ^{40}K, respectively. Natural radionuclide concentrations tend to be higher in the silt fraction, which determines their pattern distributions. Only one sample presented measurable concentration for ^{137}Cs, while ^{54}Mn was detected in two samples and ^{60}Co in four sediment samples. Man-made radionuclides present a maximum value of dose external four times lower than the normal background and the potential risk due to the presence of man-made radionuclides in sediments is lower than the risk provided by the natural radionuclides.

1. Introduction

Radioactivity is ubiquitous in the earth crust. The naturally occurring radionuclides can be found in almost all environment media as rocks, soils, sediments, air, plants, surface water (river, lagoons and ocean) and groundwater and in the human beings themselves.

Although formation of water from oil and gas extraction can contribute to increase the radium isotopes concentrations, the major part of natural radionuclides present in marine environment is introduced by processes as erosion, weathering and mineral recycling of the terrestrial rocks (Dowdall and Lepland, 2012). On the other hand, man-made radionuclides have been introduced in the marine environment by anthropogenic sources, that include global fallout generated by atmospheric weapons test, the Chernobyl and Fukushima accidents, discharges from fuel reprocessing and nuclear power plants (UNSCEAR, 2000; Hu et al., 2010; Jha et al., 2012).

Humans are exposed to radiation through incorporation of radionuclides in food chain, and, as a result, an increase in the level of internal exposure occurs. On the other hand, the presences of radioactivity in water, soils and sediments are responsible for the external exposure of humans. Thus, surveys of concentration levels, distributions and geochemical behaviors of natural and man-made radionuclides are of great importance to human health concerns.

The knowledge of the concentrations and distribution of radionuclides in all materials is of great interest, since it provides essential information for the monitoring of environmental radioactivity and studies for assessing the environmental impact of nuclear facilities and nuclear accidents. Further, radionuclides are excellent tracers of environmental process. Due to the diversity of their geochemical behavior, they supply many insights about marine water circulation and sedimentary process (Noureddine and Baggoura, 1997; Livingston and Povinec, 2000).

From the health hazard standpoint, artificial radionuclides have largely become the focus of regulatory control. Among the artificial radionuclides, ^{137}Cs is the most abundant in the marine environment, since it was significantly provided by the fallout past nuclear tests. Although the magnitude of the fallout contamination in the Southern Hemisphere is not so high as the one of the Northern Hemisphere, several surveys pointed out the presence of artificial radionuclides in the South Atlantic Ocean, in particular, after the French nuclear tests in the South Pacific Ocean (Bruno et al., 1981; Cunha et al., 1993, 1995; Fiqueira et al., 1998; Holm et al., 1994; Livingston and Povinec, 2002; Pinõnes and Tominic, 1995; Santos, 1995; Godoy et al., 2003).

The liquid effluents from the Almirante Álvaro Alberto Power Plants discharge in the Piraquara de Fora Inlet present radionuclides produced of neutron activation of the reactor corrosion products. Besides ^{137}Cs ($T_{1/2}$ = 30.1 year), the main radionuclides found in the liquid effluents are ^{58}Co (half-life: 71. 3 days), ^{60}Co ($T_{1/2}$ = 5.2 years), ^{54}Mn ($T_{1/2}$ = 312.2 days), ^{51}Cr ($T_{1/2}$ = 27.8 days) and ^{59}Fe ($T_{1/2}$ = 45.1 days). Among them, ^{137}Cs, ^{134}Cs, ^{60}Co and ^{58}Co would be the highest contributor (99.6%) for the external dose due to human exposure to sediments (ELETRONUCLEAR, 2010a).

* Corresponding author.
E-mail address: francian@ird.gov.br (F.M. Carvalho).

http://dx.doi.org/10.1016/j.marpolbul.2016.03.059
0025-326X/

Fig. 1. Location of the CNAAA and the sediment sampling points.

Specifically, the objectives of the present study were to quantify and explain the spatial distribution of the natural radionuclides (^{226}Ra, ^{232}Th and ^{40}K) in surface sediments from Piraquara de Fora Inlet, providing a baseline of natural background for the area and to investigate the level of ^{137}Cs, ^{60}Co and ^{54}Mn, their possible origin, in order to assess the potential risk for human health due to their presence in the environment.

2. Study area

The Piraquara de Fora Inlet (PFI) is located in Ribeira Bay within the Ilha Grande Bay, Angra dos Reis (S23°00.840′; W44°26.493′) in the Rio de Janeiro State (Fig. 1). Geologically, the area is characterized by the presence of crystalline rocks as granites and migmatites, part of the Serra do Mar mountain chain, which reaches the shore in this region of coast. The liquid effluents from the Angra I and Angra II PWR power plants are discharged in this small inlet.

The PFI bottom is characterized by a small sedimentary plain and a narrow input of sediments. The sea floor topography is typical of inlets that are situated near an acclivity, on coastal areas and without river input. The water depths vary from 5 to 11 m. The shallow portion is located close to the borders and to the islands and outcrops and on the S/SE side, toward the deep end of the inlet, where the power plant water effluents are released. The slopes smooth in the major part of the inlet (<2°). Higher slopes are found in areas near the coast, following the coast contour (>20°) (ELETRONUCLEAR, 2005).

The local hydrodynamic circulation pattern is mostly governed by tidal wave spreading, which is marked by low-speed streams. However, due to the high-intensity flow of around 120 $m^3 \cdot s^{-1}$, the power plants liquid effluent discharge flux modifies the local hydrodynamic pattern by forming an extensive vortex and enhancing the streams near the release point, which move the fine sediment off the Saco de Piraquara de Fora region perimeter by hydrodynamic dragging. The water temperature varies from 23 °C to 31 °C. Local climate is subtropical with two seasons: dry season from July to December and rainy season from January to June. The annual mean precipitation surpasses values of 2000 mm.

Table 1
Properties of the Piraquara de Fora Inlet superficial sediments.

Parameter	Mean	Median	Standard deviation	Minimum	Maximum
Carbonate (%)	5.18	6.38	4.70	0.37	25.59
Organic matter (%)	1.86	1.80	0.86	0.63	5.06
CEC (mmol·kg^{-1})	134.9	144.0	27.8	12.0	157.0
Gravel (%)	1.8	0.3	3.1	<0.1	10.9
Sand (%)	43.5	34.9	30.9	4.5	93.0
Silt (%)	45.2	48.9	32.2	1.3	89.5
Clay (%)	9.5	7.9	10.4	<0.1	58.9

3. Material and methods

Fig. 1 shows a map of the area and the sampling point locations. Thirty-one surface bottom sediment samples (until 10 cm depth) were collect in a 250 m grid using a Van Veen Sampler. Afterwards, 3 to 4 kg of the samples were kept in polythene bags. At the site the water temperature and salinity was measured with a portable meter (HACH model HQ 40). Later in the laboratory, the sediment samples were split. Around 2 kg was separated to determine parameters such as grain size distribution, cationic exchange capacity, carbonate, sulfate and organic matter. The distribution of the grain sizes of the sediments was determined by sieve analysis (ABNT, 1995), cationic exchange capacity by replacement of sodium from the matrix and determination of displaced sodium by ICP-OES (USEPA, 1986). Sulfate by Turbidimetry Method (SM 4500E, 2011), Organic Matter by Heated-Persulfate Oxidation Method (SM 5310C, 2011) and total carbonate by Titrimetric Method (SM 4500C, 2011). The mineralogical analysis of the clay fraction was performed in two sediment samples (number 10 and 11) by X-ray diffraction (EMBRAPA, 1997).

Around 1 kg of the sediment samples was oven dried in a stove at 80 °C, for 48 h, at constant weight, being then smashed and sieved using 2 mm nylon sieves for further analysis. For gamma spectrometry analysis, aliquots of around 300 g of the sediments were transfered to polyethylene tubes, sealed and stored for at least 30 days in order to reach the radioactive equilibrium.

Then, the samples were analyzed by two Hyper Pure Germanium detectors (HPGe) and associated electronic devices (amplifier, HV source and MCA Analyzer) from Canberra, Inc. (USA), with 100% relative efficiency, for 60,000 s. The energy and efficiency calibrations were performed using standard sources supplied by the National Laboratory of Ionizing radiation (LNMRI/IRD). This laboratory was designated by the Brazilian authority for legal metrology — as the National Laboratory of Metrology of Ionizing Radiation, and therefore the LMNRI promotes and participates in inter-laboratory comparisons, in collaboration with International Bureau of Weights and Measures (BIPM), the Inter-American Metrology System (SIM) and its quality management system is regularly checked by international audit (IRD, 2016).

Please cite this article as: Carvalho, F.M., et al., Natural and man-made radionuclides in sediments of an inlet in Rio de Janeiro State, Brazil, Marine Pollution Bulletin (2016), http://dx.doi.org/10.1016/j.marpolbul.2016.03.059