Ocean and Coastal Research

ORIGINAL ARTICLE
Special Article Collection
Sousa, Carreira & Fonseca (eds), Subsidies for ecosystem-based management in an offshore oil and gas exploration area, Santos Basin, Brazil
http://doi.org/10.1590/2675-2824071.22059padlf
ISSN 2675-2824

Spatial trends in the distribution of natural radioisotopes in the bottom sediments of Santos Basin (Brazil)

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ABSTRACT

Activities related to the marine exploration of oil and gas reservoirs tend to cause a concentration of natural radionuclides in related materials. As such, knowledge regarding the distribution of radionuclides in sedimentary basins with current oil operations is essential for modeling radiological hazards and possible risks of site contamination. This study investigated the distribution of 226Ra and 228Ra, radioisotopes from the 232U and 232Th decay chains in bottom sediment of the Santos basin. Sediment samples were collected from sites in a sampling grid based on depth contours and were analyzed through high-resolution gamma spectrometry. A distribution model of the spatial variation of these isotopes, the first of its kind for the Basin, supported the interpretation of similar tendencies in their distribution. From studying the spatial trends of the mean levels of the isotopes with descriptive statistics and variance analysis, latitudinal and bathymetric differences in the content of radionuclides emerged. These differences are probably derived from the distinct sources of these elements, as ^{226}Ra originates in the deep ocean from the decay of parent ^{230}Th and from the patterns of sedimentation driven by open ocean circulation, while 228Ra is supplied by terrigenous materials transported by the northward-flowing Brazilian Coastal Current.

Descriptors: ^{226}RA, ^{228}RA, Gamma Spectrometry, Sources.

INTRODUCTION

Radioactive nuclei lose energy by radiation to become more energetically stable. This process, known as radioactive decay, results in nuclear transmutation and the emission of various ionizing particles. Since radioactive isotopes (a radioisotope) undergo the same natural processes as any other stable isotope, they are universally present on Earth. Their behavior follows the same environmental pathways as their stable counterparts (Atwood, 2013). Thus, the measurement of these radioisotopes, enabled by the advent and increasing availability of spectrometric techniques and mathematical approaches, supports their use as tracers of environmental processes (Poinssot, 2012).

Concerning marine processes, there is a multitude of published papers that used radioisotopes of various elements to examine age-depth relationships (Parry et al., 2013; Ferreira et al., 2014), source-to-sink dynamics (Zielinski, 2017), sediment fluxes (Eyrolle et al., 2012; Matisoff, 2014; Kappa et al., 2018), association with sedimentological features such as grain-size distribution (Ligero et al., 2001; Ferreira et al., 2020) and water circulation (Lu et al., 2014, Periáñez, 2020).

In recent years, research efforts in natural radioactivity have been devoted to the oil and gas

Submitted: 30-May-2022
Approved: 08-Nov-2022

Associate Editor: Renato Carreira

exploration industry. Extraction methods (e.g., hydraulic fracturing, enhanced oil recovery) lead to concentration of natural radioelements in oil and gas wastes, particularly ^{226}Ra, ^{228}Ra and their decay products (Fisher, 1998). These residues, enriched in radionuclides, are typically treated as TENORM (Technologically Enhance Naturally Occurring Radioactive Material) for occupational exposure management (Nabhani et al., 2017).

These isotopes reveal the ages of oil wastewater spills Lauer and Vengosh (2016) demonstrated, when previous information on background levels of radionuclides is already available. Dowdall and Lepland (2012) discovered increasing levels of Ra isotopes in sediment cores collected in the North Sea compared with adjacent sedimentary basins, indicating a possible influence of discharges related to oil and gas extraction activities. Dar and El Saman (2014) also detected radioactive Ra signatures that can be associated with nearby oil production and exploration fields in the bottom sediments of the Red Sea. In contrast, the levels of ^{226}Ra and ^{228}Ra measured in the sediments of the NW Persian Gulf could not be associated with the adjacent oil industrial facilities, but serve as a baseline to gauge for future monitoring (Uddin and Behbehani, 2018).

Therefore, knowledge concerning the levels and distribution of natural radioisotopes in ocean basins where petroleum reservoirs are explored is relevant, both before and during the activities of offshore platforms. This information is decisive for the evaluation of the need for the conception of site contamination assessments (Rahman et al., 2013), for the modeling of radiological hazards (Hilal et al., 2014) and for the proposal of waste disposal programs (Nabhani et al., 2016).

Since the end of the 1970s, when PETROBRAS (Petróleo Brasileiro S.A.) discovered the firsts oil and gas accumulations in the Santos Basin, there has been a continuous and increasing movement toward the exploration of those reservoirs (Bruhn et al., 2017). The Santos Basin is a southwest Atlantic marginal basin located between 23°S and 28°S (Figure 1). Its continental margin originated with the opening of the South Atlantic Ocean during the Early Cretaceous (Chang et al., 1992).

According to Faria et al. (2017), its current configuration as an ocean environment, set in a passive margin, resulted from the development of a shallow carbonate platform, continuously drowned since its formation. Most of its oil fields are in the sub-salt sequence (stratigraphically represented as the Guaratiba Group) (Moreira et al., 2007).

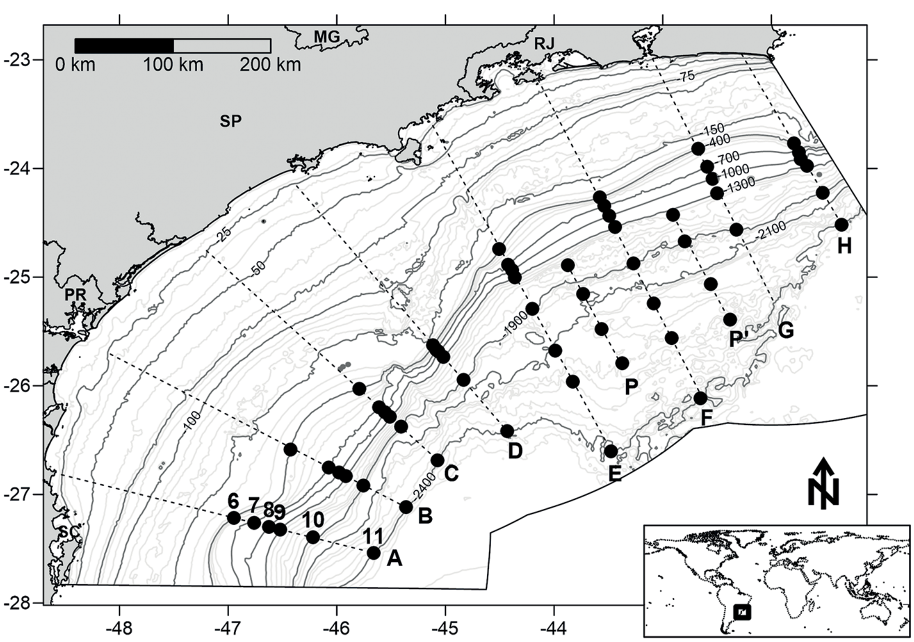

Figure 1. Santos Basin (South Atlantic Ocean) and isobathic lines. Survey transects and sampling sites.

The prospection and exploration of these fields are complex operations due to the thickness of the salt sequence (2,000-2,500 m thick) (Pereira and Macedo, 1990; Rodríguez et al., 2018) and high CO_2 contents in the reservoirs (Freitas et al., 2022).

It is considered a sediment-starved margin due to the lack of direct terrigenous sediment sources to the area from the adjacent continent (Mahiques et al., 2004), but with strong hydrodynamic drives on the distribution of sediment properties (Mahiques et al., 2004; Moller Jr. et al., 2008). Recent works stress the importance of reworking late Pleistocene sediments and inputs of terrigenous materials from the Río de La Plata drainage basin transported by the Brazilian Coastal Current (BCC) for its current sediment budget (Mahiques et al., 2008, Nagai et al., 2014, Ferreira et al., 2020).

This work aims to fill the knowledge gap regarding the levels of radionuclides from the ^{232}U and ^{232}Th decay chains in the sediments of the Santos Basin. The main objective of this study was the conception of a distribution model of ^{226}Ra and ^{228}Ra — radioisotopes representative of the main natural decay series — in bottom sediments between 400 and 2,400 m deep. This compartment of the South Atlantic Ocean has never been previously surveyed from a radiometric perspective. Thus, this purely radiometric survey of this basin is of great necessity to improve the current knowledge of its sediment sources, and for future radiological monitoring and environmental impact assessments. Moreover, graphical and statistical approaches were used to investigate the relationships between these radioisotopes.

METHODS

A sampling grid was established using the distance sampling method to reliably estimate the variables of interest in the study region. The survey design (represented in Figure 1) involved the sampling of bottom sediment (0-2 cm) in 59 sites defined based on the intersection of 10 line transects (described in Table 1) with isobathic lines. This sampling occurred. The sampling was executed during a scientific cruise in 2019 aboard R.V. Ocean Stalwart with a box-corer sampler (125 L) as part of the Santos Project (Santos Basin Environmental Characterization), coordinated by PETROBRAS/CENPES. On board, undisturbed samples were extracted from the collection instruments by inserting a stainless-steel tube in the sediment, transferring the sample into polyethylene bags, and freeze-drying until laboratory analyses.

The radioisotopes of interest were measured in a high-resolution gamma spectrometer (ORTEC, model GMX25190P, resolution of 1.97 keV for the 1332.35 keV ^{60}Co photopeak). The analytical method, described in details in Ferreira et al. (2014), consists of gamma counting of 10-15 g of macerated sediment samples, stored in air-sealed polyethylene containers, for 70,000 s. The production of the gamma spectra involved daily detector calibration with ^{60}Co and ^{137}Cs sources, background radiation detection, estimation of

Table 1. Description of the sampling transects.

Transect	Sample labels	Number of samples	Average latitude (°S)	Depth coverage (m)
A	A6-A11	6	27.34	400-2,400
B	B6-B11	6	26.83	400-2,400
C	C6-C11	6	26.30	400-2,400
D	D6-D11	6	25.85	400-2,400
E	E6-E11, P1-P2	8	25.39	400-2,400
P	P3-P6	4	25.33	1,720-2,135
F	F6-F11, P7-P8	8	24.92	400-2,400
P’	P9-P12	4	24.89	1,435-2,198
G	G6-G10	5	24.14	400-2,100
H	H6-H11	6	24.04	400-2,400

self-absorption, counting efficiency and minimum detectable activity (MDA), and statistical quality control (Supplementary Materials, Table S1).

The measured photopeaks correspond to the gamma-ray emission of two decay products: ^{214}Bi for ^{226}Ra (in the ^{238}U decay series) and ^{212}Pb for ^{228}Ra (in the ^{232}Th decay series). These peaks represent the activity of their parent within their respective decay chains if secular equilibrium is considered (achieved after one month of sample sealing). The quality control was evaluated through the analyses of the certified reference material IAEA-326. The precision was checked using relative standard deviation (RSD) and the accuracy using the relative error (RE). The quality control shows that the measured activities were close to the reported values with mean deviations and errors not exceeding 10% (Supplementary Materials, Table S1).

RESULTS

The analysis of the radiometric properties showed that ^{226}Ra level ranged from <MDA to 65.87 Bq kg^{-1}; ^{228}Ra from 7.74 to 41.28 Bq kg^{-1}. The mean activity of ^{226}Ra (21.55 Bq kg^{-1}, standard deviation = 11.90) was lower and statistically different than the mean for ^{228}Ra (24.64 Bq kg^{-1}, standard deviation = 7.47) as observed with the results of a two-sample t-test assuming different variances ($p = 0.034$, $\alpha = 5\%$). Only one of the samples (P1), collected in the São Paulo Plateau, showed ^{226}Ra activity below the MDA for that isotope. All the results for all analyses are documented in the Supplementary Materials (Table S2).

The Anderson-Darling test of normality of residuals was applied and its results for both ^{226}Ra ($S = 0.40$, $p = 0.36$) and ^{228}Ra ($S = 0.24$, $p = 0.75$) showed that their distribution could be considered statistically ($\alpha = 5\%$) normal. This result is essential as a premise for applying parametric statistic tests.

Figure 2 presents the spatial distribution of radioisotopes ^{226}Ra and ^{228}Ra in all bottom sediment samples. When observing the geographical distribution of the radioisotopes (Figure 2a and 2b), it is visible that the levels of ^{226}Ra are the highest in the southernmost region of the continental slope. These figures also show that the highest activities of ^{228}Ra can be found along the 400, 700 and 1,000 m isobaths.

DISCUSSION

The study of radionuclides in sediments is usually built on its relationships with sedimentological variables, given their known association in coastal and ocean sediments (Ligero et al., 2001; El-Reefy et al., 2014; Valan et al., 2017; Lin et al., 2020). Factors that explain this affinity are the availability of more adsorption sites in fine-grained sediments than in coarse-grained sediments, binding and fixation in organic matter, incorporation in suspended materials, and dilution effects caused by the presence of carbonates, among others (Ligero et al., 2001).

Previous surveys executed in the shelf area of Santos Basin found statistically significant correlations between natural radionuclides and mud content in surficial sediment samples (Nagai et al.,

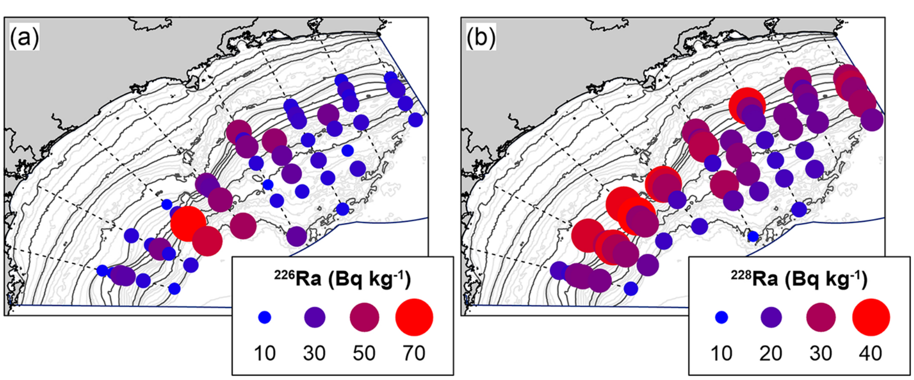

Figure 2. Spatial distribution of the levels of radioisotopes - ^{226}Ra (a) and ^{228}Ra (b) (in Bq kg^{-1}) in the bottom sediments of Santos Basin (400 - 2,400 m deep).

2014) and sediment cores (Ferreira et al., 2020). These relationships were all observed in the sediments of the studied region of the Santos Basin. When comparing the spatial patterns of the radionuclides with these studies, it can be noticed the same tendency of higher values in shallower sediments exists for both ^{228}Ra and mud.

The levels of ^{226}Ra were higher than those found on the adjacent continental shelf (Table 2), further corroborating the tendency of increasing ^{226}Ra levels toward higher depths (Figure 3). In addition, the range of levels is comparable to other regions where oil and gas reservoirs are explored, such as the Persian Gulf, the Barents Sea and

Table 2. Comparison of the levels of Ra isotopes in marine sediments (in Bq kg^{-1}) with other sites around the world.

			Activity (Bq kg^{-1})	
Region	**Reference**	**Site context**	**^{226}Ra**	**^{228}Ra**
Santos Basin	This study	Sediments preceding oil and gas exploration	5.45 – 65.87	7.74 – 41.28
	Ferreira et al. (2020)	Shelf sediments	4.16 – 44.63	ND1
North Sea	Dowdall & Lepland (2012)	Deep ocean samples	1.7 – 252.0	3.1 – 110.0
	Ahmad et al. (2021)	Sediments with oil and gas wastes	40 – 3200	ND1
Barents Sea	Yakovlev & Puchlov (2020)	Shelf sediments near oil and gas facilities	0.50 – 48.30	3.60 – 54.00
Persian Gulf	Patiris et al. (2016)	Shelf sediments preceding oil and gas exploration	3.9 – 20.5	4.3 – 21.2
	Alzahrani et al. (2022)	Shelf sediments near nuclear plants	3.7 – 7.4	2.6 – 4.2

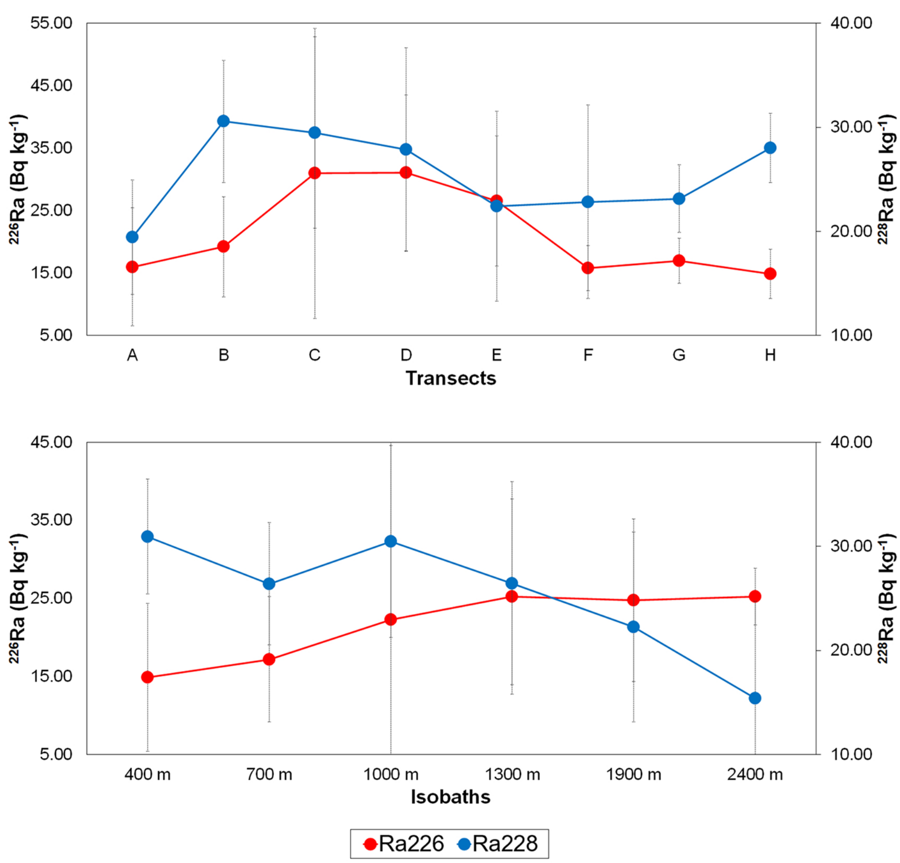

Figure 3. Trends on the mean levels of radioisotopes (in Bq kg^{-1}) in the bottom sediments of Santos Basin (400 - 2,400 m deep) between the (a) transects and (b) isobaths.

the North Sea (Table 2). However, they are much lower than deep ocean sediments and sediments contaminated by TENORM wastes from this industry (Table 2).

Spatial trends in the distribution of the radionuclides can be more clearly observed in Figure 3, with graphics depicting variations in the mean levels of both isotopes among the transects and isobathic lines. These graphics showed a remarkable change in the levels of radionuclides, i.e., an increase in ^{226}Ra and a decrease in ^{228}Ra toward offshore. They also denote a difference in the ^{228}Ra content between the samples from the southernmost transects (B-D) and those from the northernmost ones (transects E-H). This behavior was also observed for ^{226}Ra between south (represented by transects C-E) and north (F-H) compartments.

One-way analysis of variance (ANOVA) was used to verify if these differences in the distribution of radionuclides are statistically significant. After applying the Levene test to assess and validate the homoscedasticity in the data residuals, ANOVA showed a statistically significant (α = 5%) difference in the means between the transects for ^{226}Ra and between the isobaths for ^{228}Ra. Two approaches were presented here: one is visual, checking visual trends and using descriptive statistics (Figure 3, with maximum and means), and another is studying differences between the means (Table 3). This statistical analysis showed that, even though spatial trends are visible in the vertical graphics, these other trends (^{226}Ra with isobaths and ^{228}Ra with transects) are not statistically significant. Following these remarks, there is a need to ponder the possible sources of these isotopes and patterns of sedimentation driven by water circulation resulting in the distribution of radioisotopes in the Santos Basin.

Besides radioactive decay, the possible sources of ^{228}Ra to the Atlantic Ocean are submarine groundwater discharges (SGD) and freshwater inputs (Moore et al., 2008; Kwon et al., 2014), represented in the region by the southern Brazilian lagoonal systems and the Río de La Plata drainage basin. Differently from ^{228}Ra, ^{226}Ra in the oceans originates from the decay of its parent ^{230}Th and diffusion from deep-sea sediments, as less than 10% of ^{226}Ra inventory is supplied to the oceans by river inputs (Xu et al., 2022). Reports for the Atlantic Ocean (Broecker et al., 1976; Le Roy et al., 2018) showed the enrichment of this isotope in deep water sediments.

The contrasting provenances of these isotopes, combined with diverse sedimentary processes, must be behind the scene viewed in the distribution models and statistical analysis. ^{226}Ra presents higher activities in the deepest sediments, whereas ^{228}Ra is mainly introduced from terrigenous sources, thus richer in shallower areas, closer to the outer shelf, and transported, distributed and reworked by the local hydrodynamical agents. Meanwhile, from a latitudinal perspective, the southernmost zone is characterized by higher Ra content and diluted carbonate levels due to the contribution of terrigenous sediments from the Rio de La Plata mouth (Ferreira et al., 2020). These properties were observed in the samples from transects B-D, especially concerning ^{228}Ra, transported to this region by the BCC. Finally, these results independently validate the knowledge that

Table 3. One-way analysis of variance (ANOVA) (α = 5%) of the levels of radioisotopes between the transects and isobaths groups. Bold numbers denote results with *p*-value lower than the level of significance.

Radioisotope	Scenario	Test	Statistic result	*p*-value
^{226}Ra	Comparison of means between transects	Levene	W = 2.15	0.06
		One-way ANOVA	**F = 2.27**	**0.04**
	Comparison of means between isobaths	Levene	W = 0.75	0.59
		One-way ANOVA	F = 1.06	0.40
^{228}Ra	Comparison of means between transects	Levene	W = 0.92	0.51
		One-way ANOVA	F = 1,71	0,13
	Comparison of means between isobaths	Levene	W = 0.73	0.61
		One-way ANOVA	**F = 6.39**	**< 0.01**

there are latitudinal and bathymetric controls on the distribution of sediment properties in the area (Mahiques et al., 2004; Nagai et al., 2014).

CONCLUSION

This study presented the distribution model of ^{226}Ra and ^{228}Ra in the seafloor sediments of the Santos Basin between 400 and 2,400 m deep. Spatial trends in the geographical dispersion of these variables were observed, based on vertical graphics and statistical analyses. These trends were related to the latitudinal and bathymetric differences in the distribution of both radionuclides. The southernmost region, known to be dominated by fine-grained sediments with lower carbonate content, is richer in ^{226}Ra. At the same time, the continental slope, closer to the shelf break, presents the highest levels of ^{228}Ra.

These geographical changes are probably due to the contrasting sources for these elements, as ^{226}Ra originates in the deep ocean from the decay of parent ^{230}Th, and ^{228}Ra is supplied by terrigenous materials transported by the northward-flowing Brazilian Coastal Current. This interpretation further emphasizes the modern deposition of terrigenous materials in a passive margin lacking present drainage basins flowing to it. Its natural radioactivity levels are within the range of other studies in the region and other open ocean areas and are fully explained by natural processes.

Given the relevance of the Santos Basin to the oil and gas industry, the modeling of natural radioactivity of this region is imperative for simulations of radiological hazards and TENORM disposal. This study provided the first spatial distribution model in sediments of the continental slope of the Santos Basin. With a purely radiometric approach, it was possible to discern trends related to the complex hydrodynamics of the region. However, further studies are still needed to confirm these hypotheses and the sources of the radionuclides.

ACKNOWLEDGMENTS

The authors thank PETROBRAS for the opportunity to the analysis of the sediment samples from the Santos Project (Santos Basin Environmental Characterization) and ANP (*Agência Nacional de Petróleo, Gás Natural e Biocombustíveis*) for the investment of RDI (Research, Development and Innovation) resources. M.M.M. acknowledges CNPq for research grant no. 300962/2018-5.

AUTHOR CONTRIBUTIONS

P.A.L.F.: Conceptualization; Investigation; Methodology; Writing – original draft; Writing – review & editing;

R.C.L.F.: Conceptualization; Resources; Supervision; Validation; Writing – review & editing;

M.M.M.: Data Curation; Formal Analysis; Validation; Writing – original draft; Writing – review & editing;

S.H.M.S: Supervision; Funding Acquisition; Project Administration; Writing – review & editing.

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