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Radium isotope (^{223}Ra, ^{224}Ra, ^{226}Ra and ^{228}Ra) distribution near Brazil's largest port, Paranaguá Bay, Brazil

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ABSTRACT

This work investigates the ^{223}Ra, ^{224}Ra, ^{226}Ra and ^{228}Ra isotope distribution in river, estuarine waters and sediments of the Paranaguá Estuarine Complex (PEC). The stratification of the Ra isotopes along water columns indicate differing natural sources. In sediments, the radium isotope activities was inversely proportional to the particle size. The highest concentrations of ^{223}Ra, ^{224}Ra, ^{226}Ra and ^{228}Ra in the water column were found in the bottom more saline waters and towards the inner of the estuary. These relatively high concentrations towards the bottom of the estuary may be attributed to the influence of tidally driven groundwater source and desorption from particles at the maximum turbidity zone. The apparent river water ages from the radium isotope ratios, ^{223}Ra/^{224}Ra and ^{223}Ra/^{228}Ra, indicate that the principal rivers that flow into the estuary have residence times from between 6 and 11 days.

Recently, there is a growing analytical applicability of measuring natural uranium and thorium decay series radioisotopes (de Oliveira et al., 2006; Huang et al., 2015; Lu et al., 2013). The outcome of such studies has contributed to a better understanding of the geological processes occurring on the Earth's crust, including sedimentation dynamics and water mass transport processes (Burnett et al., 2008; Sanders et al., 2011; Santos et al., 2008). However, to implement radionuclide tracer mass balance calculations baseline studies are needed.

Within the natural occurring ^{238}U, ^{235}U and ^{232}Th decay series are the radium isotopes (^{223}Ra, ^{224}Ra, ^{226}Ra and ^{228}Ra). The short lived radium isotopes, ^{223}Ra ($T_{1/2}$ 11.4 days), ^{224}Ra ($T_{1/2}$ 3.66 days) are continuously regenerated by the decay of their parent Th adsorbed on the sediment surface, or on aquifer particulate materials. Thus, Th isotopes continuously provide isotopes Ra short half -lives to salt water. In contrast, radioisotopes of long half-lives, ^{226}Ra (half-life 1600 years) and ^{228}Ra (half-life of 5.75 years) require a considerable time for regeneration. The differences in production rates lead to differences in the fluxes of each of these radionuclides to the coastal environment. The objective of this study was to examine the spatial distribution of the naturally occurring isotopes, ^{223}Ra, ^{224}Ra, ^{226}Ra and ^{228}Ra, in surface water and sediments along the Paranaguá Estuarine Complex (PEC).

The PEC is located in the southern coast of Brazil, 25°16′ and 25°34′ S and 48°17′ and 48°42′ W, occupying a total area of 612 km^2 (Fig. 1). The

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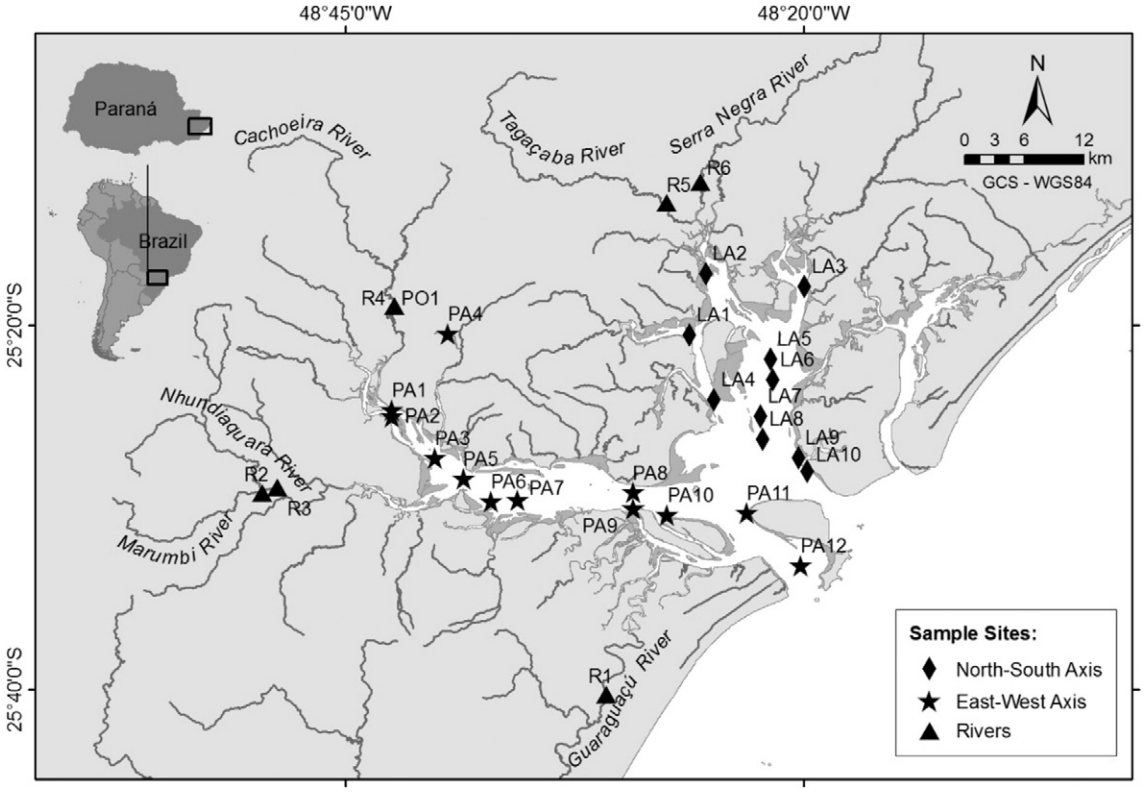

Fig. 1. Study sites along the Paranaguá Estuarine Complex, Southern Brazil.

PEC is comprised of two main drainage basins, the east-west axis is formed by Paranaguá and Antonina bays with an average depth of 5.4 m, and the north-south axis - with an average depth of 2.7 m covers the Laranjeiras Bay (Lana et al., 2001). The spring tide in PEC ranges from 1.7 m to 2.7 m (Marone et al., 2005). The rivers Cachoeira and Nhundiaquara are the major tributaries to the PEC (Lana et al., 2001) of the east-west axis of the PEC - present average flows of 21.13 and 15.88 $m^3\ s^{-1}$, respectively (Lana et al., 2001; Marone et al., 2005).

The sampling campaign for this work was carried out from 18 to 21 March 2013, covering a total of 28 sampling stations (Fig. 1). The sampling points were distributed in transects along the main salinity gradient of each axis, during low tide. Salinity was measured in the field by a refractometer (ATAGO) and CTD (JFE Compact), as well as the turbidity, temperature and depth. Samples were taken from Guaraguaçu Rivers (R1), Sacred (R2), Nhundiaquara (R3), Cachoeira (R4), Tagaçaba (R5) and Serra Negra (R6). A sample from an artesian well near the Rio Cachoeira was also sampled (Fig. 1).

To measure the Ra isotopes activities in water, approximately 60 L of water were pumped, then filtered in situ (average flow rate of 1 L min^{-1}), by means of acrylic fibers impregnated with manganese dioxide (Moore, 2008; Moore and Arnold, 1996). At each sampling station, near 500 g of sediments from rivers and PEC were collected with a Petit Ponar. The ^{224}Ra and ^{223}Ra activities in surface and bottom water samples of the rivers and estuary were determined by a delayed coincidence system (RaDeCC sys) (Moore and Arnold, 1996). The ^{224}Ra, ^{228}Ra and ^{226}Ra water and sedimentary measurements were performed by high-resolution gamma spectrometry, after being sealed for over 21 days before measuring (Smoak et al., 2012). The ^{226}Ra and ^{228}Ra activity concentrations were then determined through their daughters ^{214}Pb and ^{214}Bi, and ^{228}Ac, respectively (Godoy et al., 1994; Moore, 1983, 1984).

The depths along the estuary ranged from 1.2 to 11.9 m, from the inner to the mouth of the estuary. Water temperature showed little variation, ranging between 21 and 25 °C along the sampling stations. The salinity fluctuated significantly along estuary, ranging from 0.04 ppt at the innermost point (PA1) to 29.15 ppt towards the mouth of the estuary (PA12) (Fig. 2). The salinity was generally higher along the bottom (Fig. 2), likely triggered by denser water currents at the beginning of the flood tide. The highest turbidity in surface water, 54.71 FTU, was detected at the point LA6, while lowest turbidity was found towards the mouth of the estuary (PA12) 2.55 FTU (Fig. 3).

The sediment radium isotope concentrations showed a general decrease from the rivers to the mouth of the estuary (Figs. 4, 5 and 6). In general, the highest and lowest activities were found in Nhundiaquara (R3) and Guaraguaçu (R2) Rivers, respectively. It should be noted that the headwaters of these rivers originate from the Serra do Mar Mountain range, except for the Guaraguaçu River, which originates from springs on the coastal plain and is the river with lowest sediment radium isotope concentrations. In the sediments, it is apparent that the radium isotope concentrations are a function of grain size as the ^{224}Ra, ^{226}Ra and ^{228}Ra activities were inversely proportional to the sediment particle size and driven by the Serra do Mar. geology (Lamuar et al., 2004). Even though there are not many studies in which to compare our findings, the ^{226}Ra results in this work are similar to what was found in the surface sediments in earlier studies (Sanders et al., 2006; Sanders et al., 2012).

It is proposed that the diffusion potential of Ra isotopes is enhanced in fine sediments (Stieglitz et al., 2013). However the water concentrations did not follow a specific pattern that could be attributed to grain size. Indeed, the distribution of the ^{223}Ra, ^{224}Ra, ^{226}Ra and ^{228}Ra are likely driven by a range of sources (Carroll et al., 1993; Stieglitz et al., 2013). This is because estuarine systems present multiple potential secondary activity sources (end members), such as rivers, tidal channels, mangroves, coastal waters and groundwater discharge (Santos et al., 2010; Smoak et al., 2012; Stieglitz et al., 2013). Although activity distributions in sediment and water surface did not follow the same patterns, the concurrent influence of diffusion in the sediment-water interface on Ra isotopes activities in the water column cannot be discarded, as the aqueous matrix is also affected by tidal factors, wind and river discharge

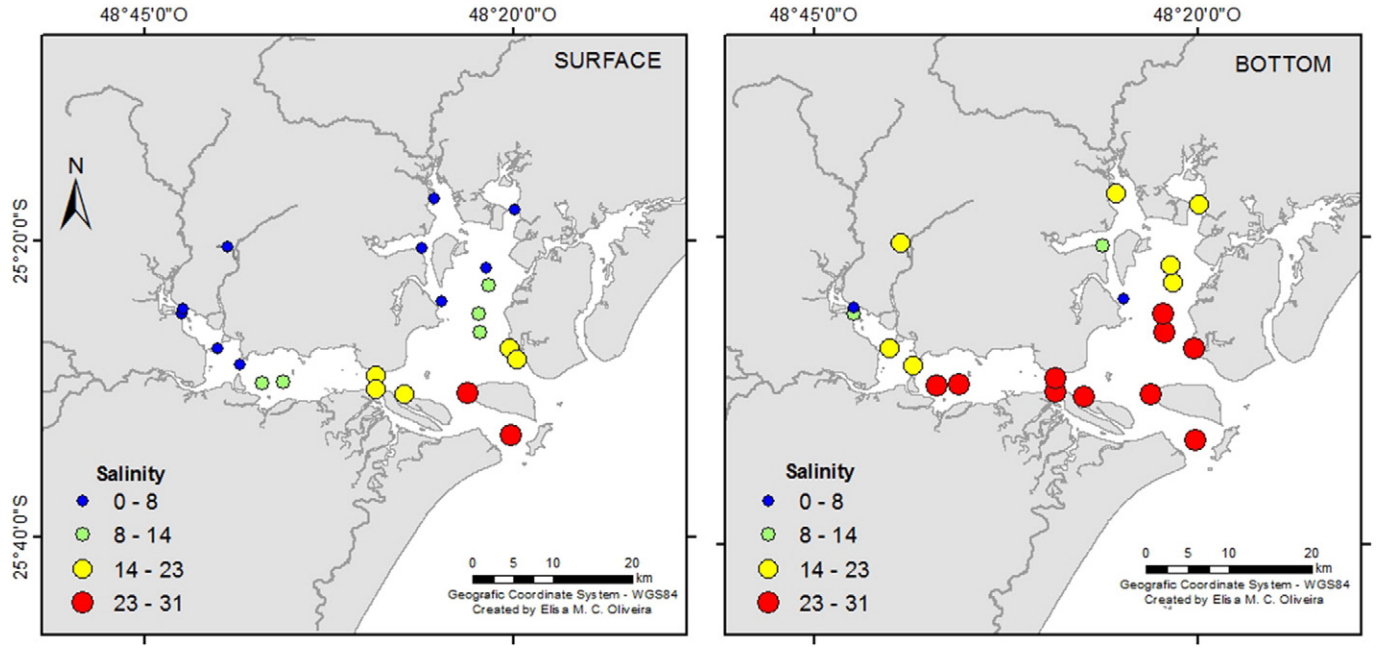

Fig. 2. Salinity distribution along the Paranaguá Estuarine Complex.

which in turn depend on climatic conditions. The Ra activities are noticeably higher in the east-west axis relative to conservative mixing between the observed endmembers. The highest activities may be related to the particulate suspended matter driven by turbidity (Fig. 2), and associated to the maximum turbidity zone (ZMT) (Noernberg, 2006).

High Ra activities may be related to particulate matter yielded by lateral river discharge, as well as alternative sources such as nearby mangroves and submarine groundwater discharge (Santos et al., 2009, 2012). According to Gleeson et al. (2013), an increase of natural isotopes by a tidal creek during ebb tide can be explained by the contributions from recirculating water in sediments (^{223}Ra and ^{224}Ra) and bioturbation by crabs (^{226}Ra) (Stieglitz et al., 2013). Indeed, the peak ^{223}Ra activity around the estuary mouth (LA10) can be related to the influence of the fluvial discharge arisen from the Island of Peças and its mangroves, or even to the occurrence of local groundwater sources (Santos et al., 2010).

The ^{224}Ra activities in water samples from rivers discharging into PEC surpassed those from estuary surface waters. In contrast, the long lived Ra isotopes (^{228}Ra and ^{226}Ra) highest concentrations were measured in the Nhundiaquara River (R03) and the Serra Negra River, respectively (Fig. 1). The discharge from Cachoeira and Nhundiaquara Rivers are important because they accounted for 82% of the total freshwater inflow to the Bay (Lana et al., 2001). Furthermore, the higher radium concentrations in the Cachoeira and Nhundiaquara Rivers may be related to the lower residence times. The river residence times were estimated from the apparent ages, which were calculated using the ratio of radium isotopes tracers (Table 2) (Dulaiova and Burnett, 2008). In these estimates, the main source of uncertainty lies in the variability

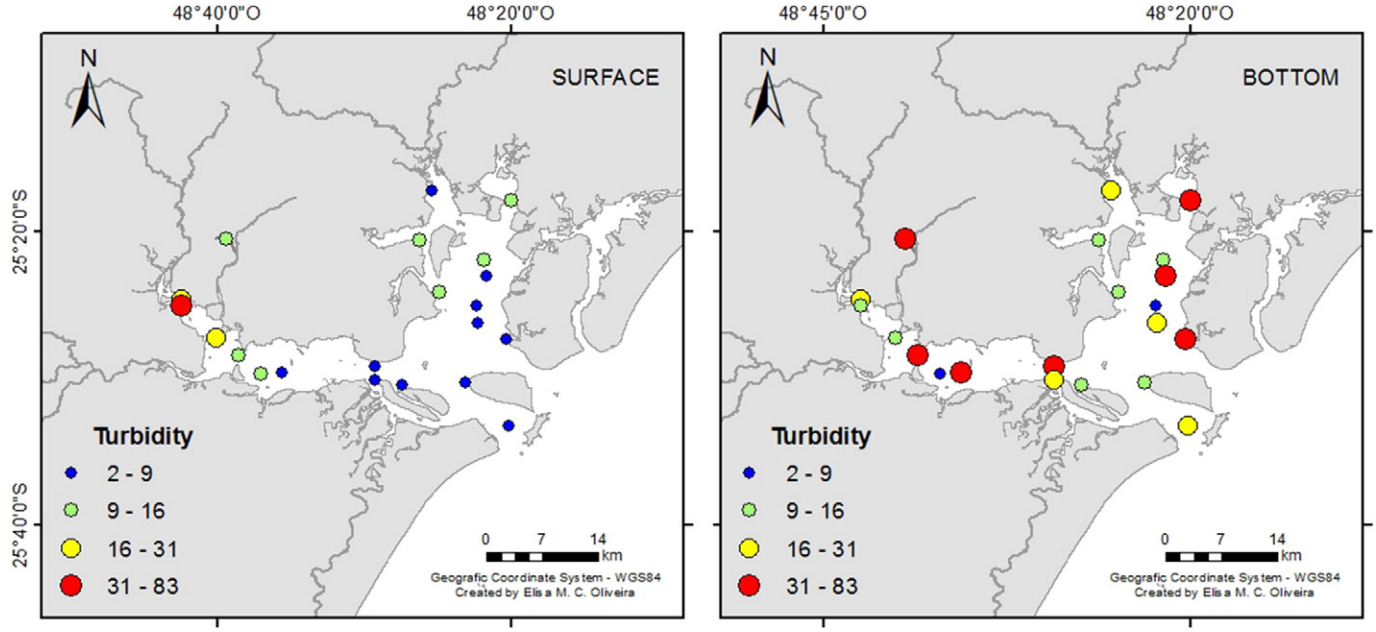

Fig. 3. Turbidity distribution along the Paranaguá Estuarine Complex.

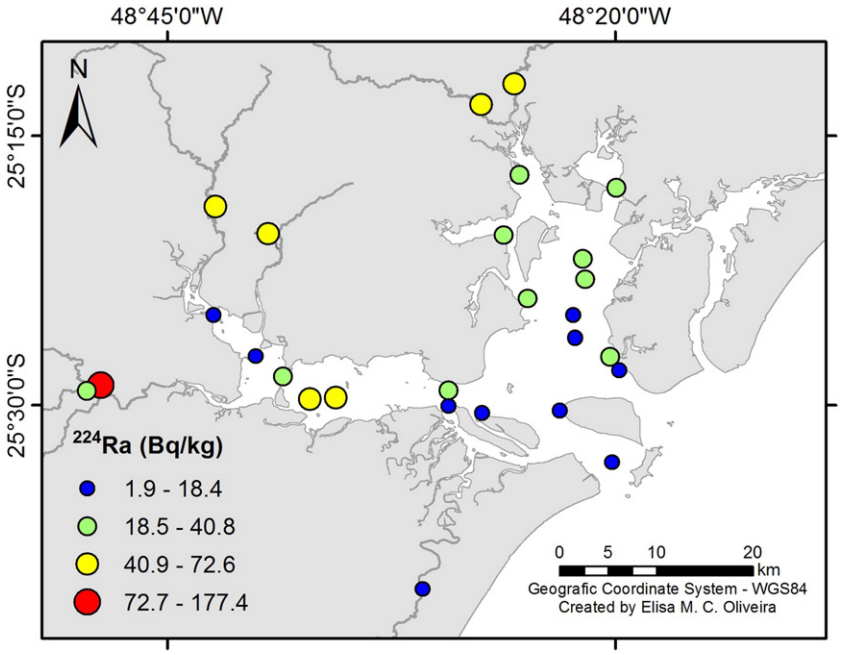

Fig. 4. Radium-224 distribution in the sediments along the Paranaguá Estuarine Complex.

of the ratio of end-member, associated with several factors and physical-chemical and geochemical processes (Gonneea et al., 2008; Tomasky-Holmes et al., 2013).

Using the highest measured activity ratio (AR) in low to mid-salinity waters to assign the initial ratio (Burnett et al., 2008; Moore and Krest, 2004), we estimated apparent ages based on $^{224}Ra/^{223}Ra$ (Xu et al., 2013):

$$T_1 = [\ln(AR_{obs}/AR_i)]/\left(\lambda^{223} - \lambda^{224}\right)$$

and on $^{223}Ra/^{228}Ra$ (Tomasky-Holmes et al., 2013):

$$T_2 = [\ln(AR_{obs}/AR_i)]/\lambda^{223}$$

where T is the age of the radio water or the time elapsed since the short half-life isotopes were added to the system; AR_{obs} is the ratio of the $^{224}Ra/^{223}Ra$ activities (T_1) or $^{223}Ra/^{228}Ra$ (T_2) at a certain distance from the innermost part of the system; AR_i is the highest measured ratio in the salinity gradient (the one with minimum decay); λ^{223} and λ^{224} are the decay constants of 0.189/day and 0.0608/day, respectively. The calculated ages are given in Table 4. Through this model, the water in the East-West axis takes 6 days (T1) to 10 days (T2) to run a distance of 48 km, from the portion innermost to the mouth of the system. In comparison, Marone et al. (2005) estimated an average of 3.5 days for this axis, based on tidal prism estimation.

Radium enrichment in the PEC was calculated by subtracting the average ^{228}Ra and ^{226}Ra ocean end member concentrations, 9.6 and 7.9 dpm 100 L^{-1} (Smoak et al., 2012), from the average values within

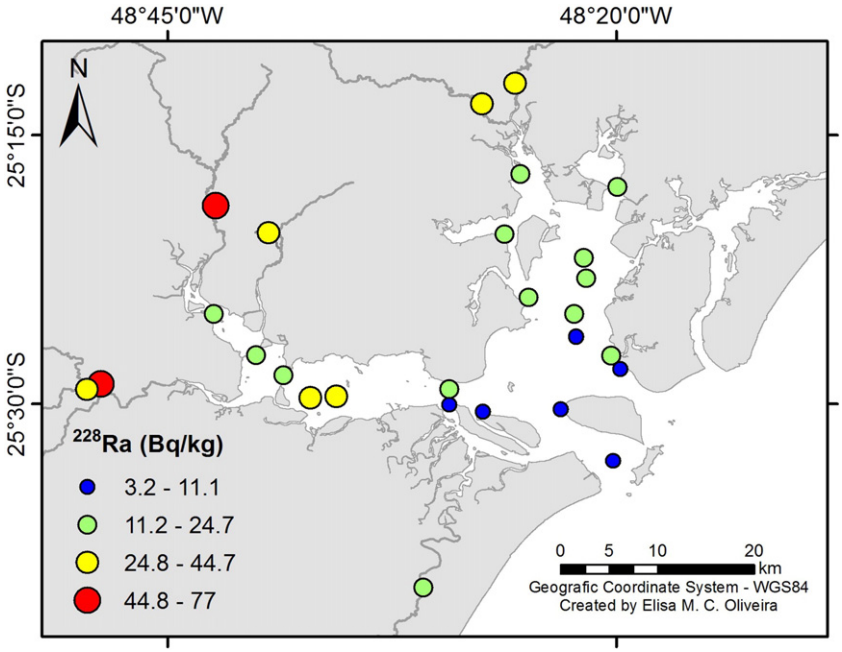

Fig. 5. Radium-228 distribution in the sediments along the Paranaguá Estuarine Complex.

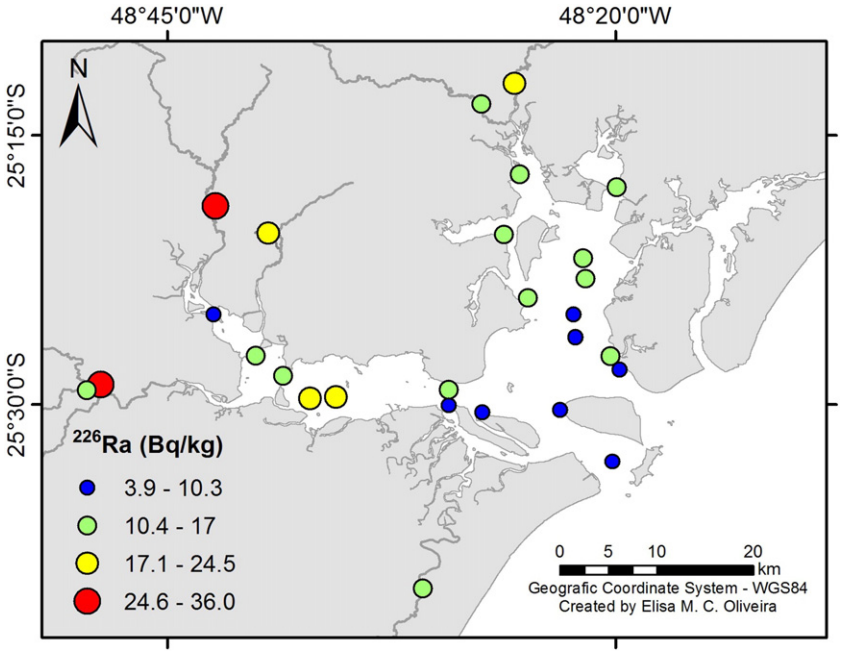

Fig. 6. Radium-226 distribution in the sediments along the Paranaguá Estuarine Complex.

the bay 58.6 and 23.1 dpm 100 L^{-1}, respectively (Table 1). Total PEC enrichment was then determined by multiplying the enrichment activity by the total volume of the bay 1.4×10^{15} L (Lana et al., 2001), giving values of 6.9 and 2.1 dpm $\times 10^{14}$ for ^{228}Ra and ^{226}Ra, respectively. The fluxes required to support the observed ^{228}Ra and ^{226}Ra enrichment were determined by dividing the total ^{228}Ra and ^{226}Ra enrichment by the PEC water residence time of 84 h (Lana et al., 2001), giving 2.0 and 0.6 $\times 10^{14}$ dpm day^{-1} for ^{228}Ra and ^{226}Ra, respectively. Decay was not a factor because the Bay water residence time is short in comparison to the half-life of ^{228}Ra and ^{226}Ra. The average discharge of all rivers entering the bay is near 200 m^3 s^{-1} (Lana et al., 2001) and the average ^{228}Ra and ^{226}Ra concentrations form the rivers are near 92.6 and 24.3 dpm 100 L^{-1} (Table 1). Therefore, a first order estimate of ^{228}Ra and ^{226}Ra contribution from the rivers is approximately 1.6 and 0.2 dpm $\times 10^{10}$ for ^{228}Ra and ^{226}Ra, respectively. Diffusion from sediments, release from sediment via resuspension, and a tidally driven

Table 1
Radium isotope concentration (dpm 100 L^{-1}) in the surface and bottom waters of the estuary and rivers as well as the groundwater samples in this study. Note that the ^{223}Ra measurements were not taken at the estuary and river bottom.

Station	^{224}Ra	^{223}Ra	^{228}Ra	^{226}Ra	^{224}Ra	^{228}Ra	^{226}Ra
Estuary surface					Estuary bottom		
PA1	0.94	0.04	19.74	0.52	na	na	na
PA2	1.56	0.05	15.18	1.6	34.4	30.4	15
PA3	2.72	0.11	56.22	2.09	33.6	33.4	24.8
PA4	4.82	0.24	45.18	9.78	100.6	63	43
PA5	5.25	0.23	95.64	4.73	56.2	48	31.6
PA6	5.25	0.22	44.22	5.18	112	89.4	49
PA7	6.68	0.27	80.04	6.54	97.8	71.8	45.6
PA8	10.44	0.54	143.52	10.44	61.8	48.3	34
PA9	7.5	0.51	76.68	9.29	12.8	13	15.6
PA10	5.96	0.35	46.2	9.59	13.6	9.6	9.2
PA11	6.11	0.47	98.22	10.86	3.8	9.6	10
PA12	4.99	0.31	61.32	6.53	4	6.4	7.8
LA1	8.27	0.26	94.5	6.63	45	45.4	28
LA2	6.53	0.25	37.26	3.89	62.2	48.2	32.4
LA3	14.75	0.31	80.16	4.49	51	47.4	28.2
LA4	15.82	0.26	122.7	7.14	65.8	49.4	35
LA5	20.15	0.2	68.28	3.75	62.4	48.8	31.8
LA6	23.76	0.52	63.06	9.19	76	46.8	31
LA7	29.98	0.67	91.92	13.26	33.4	31.4	20.6
LA8	22.34	0.37	39.6	9.96	23	22.2	19.2
LA9	12.86	0.3	57.96	9.7	53	42.8	23.8
LA10	29.66	0.72	86.34	12.93	21.4	19.6	17.2
Rivers surface					Rivers surface		
R1	11.03	0.09	74.46	2.04	36.8	37.4	28.4
R2	7.57	0.03	131.76	1.24	81.6	72.8	32.2
R3	54.12	0.11	80.7	5.56	354.8	154	72
R4	81.07	0.22	72.48	4.27	145.2	120.6	71.8
R5*	72.14	0.14	63.42	1.94	96.8	83.2	28.8
R6	74.64	0.1	145.44	4.42	97.6	75.4	39
Groundwater							
PO1 (well)	22.99	0.31	68.1	1.64	na	na	na

Table 2
Apparent water ages in the east-west and north-south axis of the Paranaguá Estuarine Complex (see Fig. 1 for locations).

	$^{224}Ra/^{223}Ra$	$^{223}Ra/^{228}Ra$	
East-west axis			
Sample	Apparent age (days)	Apparent age (days)	Distance (km)
Pa1	2.21	0.00	0.00
Pa2	0.00	14.82	1.41
Pa3	1.81	6.24	7.37
Pa4	3.44	18.79	11.64
Pa5	2.44	7.54	15.85
Pa6	2.09	9.77	20.08
Pa7	1.81	10.24	2.60
Pa8	3.73	6.53	29.17
Pa9	5.87	5.55	32.28
Pa10	4.72	11.74	35.97
Pa11	6.83	9.46	40.49
Pa12	5.16	7.94	47.71
North-south axis			
La3	5.85	1.78	0.00
La5	0.00	6.02	5.70
La6	6.17	5.05	10.34
La7	6.33	6.91	12.41
La8	3.99	11.97	15.58
La9	6.67	14.99	18.36
La10	6.98	5.31	21.75

groundwater are other potential sources of radium that can support the observed radium enrichments in the PEC.

In summary, radium isotope tracers demonstrate high variability in rivers that discharge into the PEC water column and in sediments. The large variations are likely due to the inputs from different sources, such as diffusion process from sediments, groundwater discharge from mangrove sediments and the flow from rivers originating from the Serra do Mar. mountain range. The high radium activities were found in sections with a large influence from turbidity, indicating that the unsupported isotope concentrations in these locations are derived from desorption of suspended particles in the water column. The high Ra isotopes concentrations in the sediments were located in areas with predominantly fine sediments, potentially influencing the diffusion of Ra isotopes from sediments to the water column. Based on the apparent age calculation, the principal rivers originating for the Serra do Mar mountain range took between 6 and 10 days to reach a distance of 48 km at the east west axis, while the rivers along the north south axis took from 7 to 11 days along to reach 22 km.

Acknowledgements

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