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Evaluation of ^{210}Pb levels in the estuarine region of an industrial complex

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ABSTRACT

Currently, the increase in the environmental radioactivity levels was due to anthropic activities, such as the oil industries that produce around 2.5×10^4 to 2.25×10^5 tons of contaminated materials per year. Thus, the study aimed to determine the ^{210}Pb concentrations in the estuarine region of the SUAPE industrial complex in the state of Pernambuco. The ^{210}Pb concentrations were determined in soil, sediment, fish, and leaf samples, using ion exchange and gamma spectrometry methods. ^{210}Pb concentrations in soils, sediments, fish, and mangrove leaves ranged from less than LD (Detection Limit) to $992.35 \text{ Bq.kg}^{-1}$. The results showed that the area related to the preoperational situation found values above those estimated for regions considered natural, presenting anthropogenic interference.

Keywords: Bioaccumulation, trace metal, Brazilian estuary.

Introduction

Studies of environmental radioactivity levels have been of great importance and interest, mainly of natural radionuclides (^{238}U , ^{226}Ra) and their descendants (^{222}Rn , ^{210}Pb) present in the natural environment (Salles et al., 2019). Among the natural resources, the mangrove areas represent a remarkable heritage, since they have considered zones of high biological productivities being responsible for the productive dynamics of the tropical estuaries and adjacent areas (Kaewtubtim et al., 2018).

Industrial processes involving mining and metal extraction, phosphate mining, and oil production all contribute to increasing concentrations of radionuclides or radioactive materials in the estuarine region (Jia & Jia, 2012). Lead contamination in the SUAPE estuarine region can be attributed to the installation of the oil refinery, caused by the storage and refining of

oil. These tailings from extractions and through atmospheric deposition cause severe effects on human health (Santos et al., 2019) and the environment (Amaral et al., 2019).

The radioactive decay of ^{232}Th generates radioactive lead isotopes (^{212}Pb), ^{235}U (^{211}Pb) and ^{238}U (^{210}Pb and ^{214}Pb) (Fernández et al., 2017). The ^{212}Pb (half-lives ($t_{1/2}$) short 10.64 hours), ^{211}Pb (half-lives ($t_{1/2}$) short 36.1 minutes), and ^{214}Pb (half-lives ($t_{1/2}$) short 26, 8 minutes) and therefore they do not migrate to great distances in the environment. On the other hand, ^{210}Pb with a half-life of 22.26 years was found in many environmental samples (Santos Júnior et al., 2018).

^{210}Pb can be found adsorbed to atmospheric particles and can return to the soil by the action of rain, in which they can form compounds and rapidly distributed in the environment (Silva et al., 2018). Contamination of

food is recognized as the most important route; once ingested, its radionuclide present in food is deposited in bones and liver can induce various types of health damage, such as cancer (Silva et al., 2018). In this context, the study aimed to determine the ^{210}Pb concentrations in the estuarine region of the SUAPE industrial complex in the state of Pernambuco.

Material and Methods

The experimental area is located on the southern coast of the state of Pernambuco, between the municipalities of Cabo de Santo Agostinho and Ipojuca in the estuarine region, restricted to the Massangana, Ipojuca and Tatuoca rivers, with the following geographical coordinates 08°20'58.8" S and 34°58'23.4" W. The climate of the area is hot and humid, pseudo-tropical, of the 'As' type, characterized by motion rains almost all year round, with a well-defined and relatively short dry season occurring in the fall (Köppen, 1931). Rainfall ranges from 1,850 to 2,364 mm per year. Average annual temperatures vary between 25°C minimum and 30°C maximum, with an annual average relative humidity greater than 80% (Cordeiro et al., 2019).

Samples of soils, sediments, mangrove leaves (*Avicena schaueriana* - mangrove or seriuba, *Rhizophora mangle* - mangrove or red mangrove, *Laguncularia racemosa* - mangrove or tame) and fish (Mullet - *Mugil platanus*, Catfish - *Catfish marinus* Ariidal, Carapitinga - *Lutjanus jocu*, and Salema - *Anisotremus virginicus*) were collected in the estuarine region, where there is the influence of the installation of an oil refinery through the stratified random sampling method.

Soil samples were collected at eight surface points, and sediments were collected at seven points at a depth of 2 cm surface sediment, with three repetitions below the water depth, using an Eckman steel sampler. The samples were dried in a forced-circulation oven at 60°C on average for about one week until a constant mass was obtained. After this stage, the samples were sieved through a 300 µm mesh to discard roots, pebbles, gravel, and pebbles (Rodriguez et al., 2017). After sieving, the resulting mass of 200 g with a particle size of fewer than 300 µm was placed in a polyethylene container and labeled.

Mangrove leaf samples that were healthy and mature were collected according to the method established by (Markert et al., 1996), that is, twenty leaves of each branch in five trees, with a distance between them of 50 m. Then the samples were ground and placed to dry in a forced-circulation oven at 60° C until constant mass. The fish samples were washed to remove

solid residues and stored in cooled polyethylene containers, then placed in a freezer at -3°C. The samples were dried on a hot plate at 70°C without treatment, that is, with the scales and viscera, to remove all excess moisture. The dry matter was obtained using an oven, where the temperature was gradually increased over 48 hours until reaching 450°C to avoid rapid combustion, then left for 48 hours at this temperature, until the ashes were obtained, and 20g mass was used for analysis.

The determination of ^{210}Pb concentrations in soils and sediments was performed using the ion exchange method (Godoy et al., 2011). The ^{210}Pb concentration (in Bq.kg⁻¹) was determined using Equation 1 (Jia & Jia, 2012).

$$C_{Pb}^{210} = \frac{C_{Am} - C_{Bg}}{Rq \cdot m \cdot \varepsilon (1 - e^{-\lambda_{Bi} t})} \quad \text{Eq.(1)}$$

where C_{Am} = sample count rate (cpm); C_{Bg} = background radiation counting rate (cpm); Rq = chemical yield; m = mass of sample analyzed (kg); ε = counting system efficiency; t = time elapsed between precipitation of PbCrO_4 and - date of counting (days); λ_{Bi} = ^{210}Bi disintegration constant.

The chemical yield was calculated by the ratio between the experimental average concentration and the theoretical concentration multiplied by 100. The determination of ^{210}Pb concentrations in mangrove and fish leaf samples was firstly used the ion exchange method, but due to calcium interference, it was not possible to precipitate ^{210}Pb . Subsequently, the gamma spectrometry method was used, as (Długosz & Bem, 2013) showed that this method could be applied not only to soil and sediment matrices but also to plant samples. However, due to the low power of ^{210}Pb for such determination, some parameters were changed.

The masses of each sample were standardized to 20 g, packed in acrylic containers, with a volumetric capacity of 54 cm³, with a counting time of 200,000 s, where they were hermetically sealed and stored for a minimum of 30 days, necessary for secular radioactive balance occurs between ^{226}Ra and its children, ^{214}Pb and ^{214}Bi , that is, between short-lived radionuclides of interest for the analysis. After this period, the samples were analyzed in a previously calibrated Canberra hyperpure germanium detector.

The ^{210}Pb was determined directly through its 46.5 keV line. The self-absorption calculation was performed as a function of the attenuation of low energy gamma radiation by the sample itself. In this analysis, a fish and a mangrove leaf pattern

were prepared using 20 g of each sample by doping it with 1 mL IRD-calibrated liquid lead carrier expressed in Bq.mL^{-1} with activity 1.265871 Bq, where high surface tension and low initial absorption could be observed. Then the samples were dried in a forced circulation oven at 60°C for 12h to obtain constant mass, and after 35 days, the determination of counting efficiency and specific activity of ^{210}Pb was performed.

The counting efficiency was determined through a net standard of ^{152}Eu with $46,729 \text{ Bq.mL}^{-1}$ was used, whose physical half-life is approximately 12.7 years, obtained from the Institute of Radioprotection and Dosimetry - IRD / CNEN. The activity of the reference standard was calculated by Equation 2, taking into account the activity adjustment for the reference date.

$$A = A_0 * e^{-\lambda t} \quad \text{Eq.(2)}$$

where A = activity to be calculated; A_0 = initial activity; λ = radioactive decay constant, t = elapsed time.

The specific activities of ^{226}Ra radionuclides were calculated by applying Equation 3 (Santos Júnior et al., 2018):

$$A = \frac{C - C_B}{\varepsilon * t * I_{\gamma} * m} \quad \text{Eq.(3)}$$

where A = specific activity determined for the radionuclide under consideration (Bq.kg^{-1}); C = sample count; C_B = background count; ε = counting efficiency for specific energy (cps / dps); t = count time (s); I_{γ} = percentage gamma abundance of radionuclide in the energy under consideration; m = sample mass (kg).

The detector efficiency was determined by Equation 4 using the integrated counts.

$$\varepsilon = \frac{C}{A * t * I_{\gamma} * m} \quad \text{Eq.(4)}$$

where ε = counting efficiency for the specific energy (cps / dps); C = sample count; A = specific activity determined for the radionuclide under consideration (Bq.kg^{-1}); t = count time (s); I_{γ} = percentage gamma abundance of radionuclide in the energy under consideration; m = sample mass (kg).

Data of ^{210}Pb concentrations in soil, sediment, leaves, and fish of the study were subjected to Shapiro and Wilk normality tests and submitted to analysis of variance to evaluate the difference between the concentrations at the collection points. The Tukey test performed the comparison of means at 5% significance, and the statistical software used was SISVAR (Ferreira, 2019).

Results and Discussion

The samples showed a significant difference, and the sample P6 showed the highest concentrations when compared to the other collection points (Figure 1). The concentrations obtained for ^{210}Pb in soil samples ranged from 40.75 ± 12.67 to $622.89 \pm 35.49 \text{ Bq.kg}^{-1}$, with an average of $256.89 \text{ Bq.kg}^{-1}$. Among the results found, Porto et al. (2014) point out that the average concentration of ^{210}Pb in soils worldwide is 104 Bq.kg^{-1} . In the study of Ramona et al. (2011), the concentrations presented values below the detection limit and the highest average concentration around 260.6 Bq.kg^{-1} .

These high concentrations may have been influenced by siltation, erosion, and organic matter (Fungaro et al., 2019). The progressive introduction of industrial contaminants, associated with the natural sedimentation of the river-marine environment, may also have contributed to the increase of these concentrations. Preston et al. (2016) reported that the more intensified the anthropic activity in the region, the higher the metal concentrations, such as the ^{210}Pb , thus justifying the values found in the referred study.

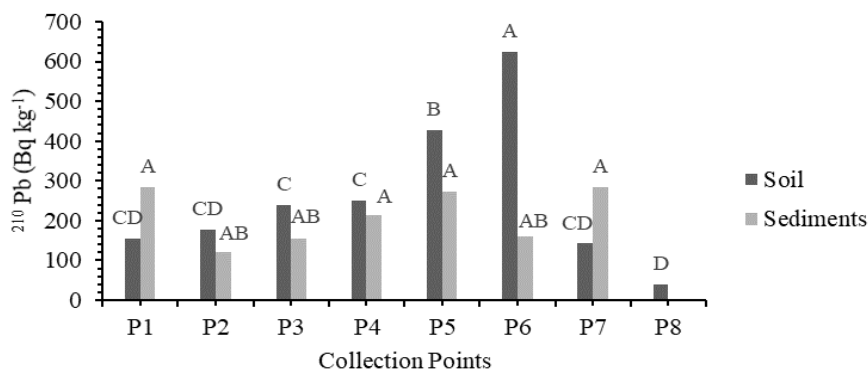


Figure 1. ^{210}Pb de concentrations in soils and sediments in the estuarine region of Cabo de Santo Agostinho and Ipojuca municipalities, Pernambuco State. Font: Cunha et al. (2019).

In estuaries, sediment deposition depends on the type of mixing process. This study refers to a partially mixed estuary in which the sediments are circulating and are deposited in the same location (Ribeiro et al., 2016). Similar quotes from the same phenomenon, in the field of studies, are endorsed according to the values obtained. The ^{210}Pb concentrations in sediment samples ranged from 121.42 ± 24.65 to 284.87 ± 75.54 , with an average of $207.80 \text{ Bq.kg}^{-1}$, P1 presented a higher mean value than the others, as can be observed in Figure 1. According to Silva (2006), the Mogi and Cubatão rivers estuary in the state of São Paulo presented concentrations in the range 26 to 538 Bq.kg^{-1} , showing to be higher than the levels considered normal for the region, this due to the influence of inputs derived from the fertilizer industry found in the region. Machado et al. (2008) presented concentrations in the range of 13.5 to 158 Bq.kg^{-1} on the southeast

coast of Brazil. According to Machado et al. (2008), high average concentrations were found in the Morrão estuary in the state of São Paulo when compared to this study, they are not natural, and the high levels of these radionuclides are due to tailings from the fertilizer industries launched in the estuary.

Fluvial and marine sediments are also environments highly subject to metal contamination (Helal et al., 2016), including natural radionuclides.

^{210}Pb concentrations were observed in samples of mangrove leaves, where F1 presented higher values about the others presenting significant differences (Figure 2). The concentration ranges found for ^{210}Pb were high, ranging from 202.29 ± 9.06 to $1043.33 \pm 25.78 \text{ Bq.kg}^{-1}$. The concentrations found for the evaluated leaf samples presented levels above the maximum limits recommended by Uncear (2017).

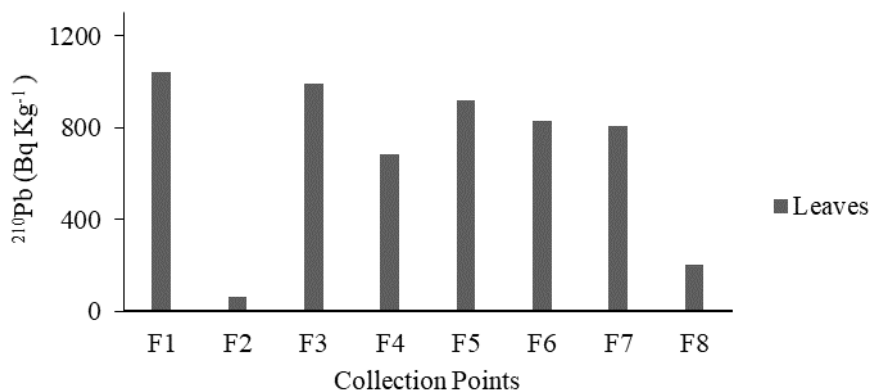


Figure 2. ^{210}Pb concentrations in leaves in the estuarine region of Cabo de Santo Agostinho and Ipojuca municipalities, Pernambuco State. Font: Cunha et al. (2019).

Mangrove leaves accumulate high amounts of radionuclides, and it means a possibility to be used as a bioindicator of contamination in estuarine regions (Garcia et al., 2019). Environmental risks are related to the high degree of mobility of these radionuclides, causing greater mobility in the soil by water and saline solutions, making them available to plants (Mierzwa et al., 2018). Mobility depends on soil pH, the hydrology of each terrain, the presence of clay minerals, and the characteristics of the vegetation and organisms present in the environment (Zangrande et al., 2018), where the vegetation retains long-term radionuclides, making them available for migration into the environment.

It is explained due to the mobility of this radionuclide in the soil, letting it available to plants, causing bioaccumulation (Santos et al., 2017). Silveira et al. (2013) found, in the Tatuoca estuary in SUAPE in the Pernambuco State, values ranging from 214.39 ± 4.82 to $237.44 \pm 0.54 \text{ Bq.kg}^{-1}$; it is observed that these concentrations increased significantly over the years as a result of landfills, dredging, dams and out-of-state materials for the installation of the oil refinery, occurring until the partial disappearance of the Tatuoca River.

Figure 3 shows the values of ^{210}Pb concentrations in different fish species, and it was found that higher values were presented for Mullet with a significant difference in the other species.

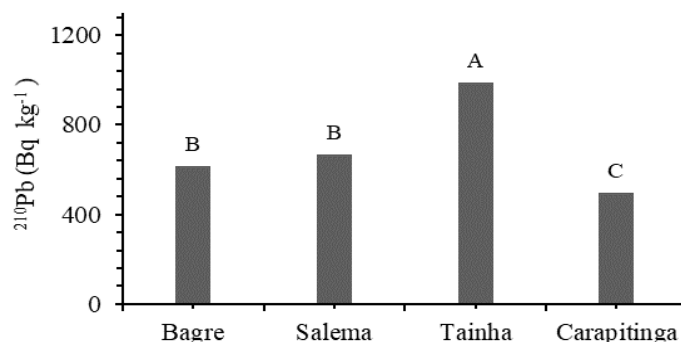


Figura 3. ^{210}Pb concentrations in fish species (bagre, salema, tainha, and carapitinga) in the estuarine region of Cabo de Santo Agostinho and Ipojuca municipalities, Pernambuco State. Font: Cunha et al. (2019).

Average concentrations of ^{210}Pb in fish samples ranged from 495.38 ± 20.65 to 989.17 ± 23.09 Bq.kg⁻¹. According to Pennington (2006), the limit concentrations for fishery products for ^{210}Pb radionuclides is 0.2 Bq.kg⁻¹ ash. In most results found in Brazilian studies and worldwide, the values were lower than those found in this study. Literature states that lower values were observed in fish samples from the Cananéia-Iguape system. The ^{210}Pb concentrations ranged from 0.17 to 15.1 Bq.kg⁻¹. According to Yamamoto et al. (1994), analyzing fish collected in the Sea of Japan, concentrations ranged from 0.04 to 0.54 Bq.kg⁻¹. According to Másico et al. (2014), ^{210}Pb concentrations in different fish species off the coast of Niterói ranged from 1.68 ± 1.23 to 190 ± 31.54 Bq.kg⁻¹.

Radionuclide bioaccumulation processes by different fish species can be explained due to factors of a physical, chemical, biological, ecological nature, eating habits, species metabolism, fish size, and place of collection Pouil et al. (2018). Levels of ^{210}Pb in mullet samples were possibly increased due to its feeding habits, this species seeks its food source in the mangroves, removing a thin layer of microorganisms, mainly algae, and debris as a result of decomposition of foliage (Garcia, 2016).

All interventions in the environment generate impacts, positive or negative, causing qualitative changes in the floristic and fauna components, in the geomorphological, sedimentological, and hydrological characteristics (Aquino, 2012). In the case of Suape-PE, with the construction of jetties for mooring ships in the Port Complex, communication between the Ipojuca and Merepe rivers with the sea was blocked, causing flooding in the areas used by the sugarcane agribusiness (Neumann et al., 1998).

A minimizing of this impact through opening access to the reef, close the mouth of the Ipojuca River, to allow the sea to flows in, not only conditioned changes in the tidal cycle but

also maintained high levels of salinity and caused considerable sedimentation in the area, with consequences in the decrease of water transparency (Koenig et al., 2002).

According to Devassy & Goes, (1988), any disturbance in the environment produces changes and many growth variables for an organism that often leads the community to become reorganized. Particular morphological and physiological adaptations allow a species to flourish. The abrupt changes lead to the replacement of a group of organisms by others or a decrease in its reproductive rate. For this reason, it can be said that the constant impacts in the area led to changes in the ecosystem and affected the dynamics of the phytoplankton community structure (Mazzilli et al., 2019).

Conclusion

The ^{210}Pb concentrations in the soil, sediment, and mangrove leaf samples indicate that it is essential to investigate the source of this contamination. Fish sampling needs new definitions of critical concentration limits to be released for human consumption. The concentrations found in the estuarine region of the SUAPE Industrial Pole indicate that the preoperational situation was altered due to substantial anthropogenic interference.

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