Cs-137 in Sand and Seawater Samples from Piraquara Beach, Brazil:

Discharge site of effluents from the Angra dos Reis Nuclear Power Plants

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Abstract

This study presents the Cs-137 radionuclide concentration and activity in sand profile and seawater samples from Piraquara Beach (PB) and from an adjacent small stretch of sand used as a beach. Both sites are located near the discharge point for cooling water and liquid effluents from the Almirante Álvaro Alberto Nuclear Power Station (CNAAA) in Brazil. The chemical composition of the sand samples was determined by granulometric analysis, elemental chemical characterization, and X-ray diffraction (XRD). Mineral sorbents for the Cs-137 radionuclide were found close to the discharge point. The presence of Cs-137 radionuclides in sand and seawater samples was determined by gamma spectrometry analysis, which was also used for seawater samples after absorption by ammonium phosphomolybdate (APM). The Cs-137 radionuclide activity measurements were lower than the minimum detectable activity (MDA): sand < 0.31 Bq·kg^{-1}, direct analysis of seawater < 0.42 Bq·L^{-1}, and seawater with APM < 0.004 Bq·L^{-1}. The aim of this study was to measure the amount of the Cs-137 radionuclide in the deeper layers of PB. A determination of radioactive exposure to individuals was not performed because the result values were lower than the MDA values. Our results indicate that the study site shows no contamination by the Cs-137 radionuclide.

Keywords: beach sand, ^{137}Cs, radioactive pollution, seawater

1. Introduction

1.1 Cs-137 Radionuclide

Radioactive pollution comprises more than 200 active radionuclides, and effluents from nuclear power plants are one source of radionuclides in the seas and oceans (Aarkrog, 2003; International Atomic Energy Agency [IAEA], 1995, 2001, 2004). Other sources of radionuclide contamination in the seas and oceans are fallout (Collins, Jardim, & Collins, 1998; Aarkrog, 2003; IAEA, 1995, 2004), nuclear tests (Aarkrog, 2003; IAEA, 1995), nuclear submarine accidents (Aarkrog, 2003; IAEA, 2004), dumping (IAEA, 1999), effluents from reprocessing plants (Aarkrog, 2003; IAEA, 2001), radiochemical source losses (IAEA, 1988), nuclear accidents (Aarkrog, 2003; IAEA, 1995, 2004), and detonation of nuclear weapons (Aarkrog, 2003; IAEA, 2004).

In considering the environmental impact of radionuclides, the Cs-137 radionuclide is very important, not only because it has a relatively long half-life of 30 years, but also because of its chemical behavior, which causes Cs-137 to be disseminated in its ionic form by fluids in the human body, in a similar manner to that of sodium and potassium ions (Collins et al., 1998; IAEA, 1995; Agency for Toxic Substances and Disease Registry [ATSDR], 2004; Figueira & Cunha, 1998; National Research Council [NRC], 1961). Owing to this feature, the Cs-137 radionuclide is one of the main contributors to radiation from artificial sources in the environment, and is a fundamental indicator of radioactive pollution. Thus, Cs-137 is an important tool for the evaluation of disseminated radiation resulting from nuclear accidents and for monitoring radiation around nuclear power plants (IAEA, 2004, 2011; United Nations Scientific Committee on the Effects of Atomic Radiation [UNSCEAR]; International Union of Radioecology [IUR]). The severity of the effects of radiation exposure to the Cs-137

radionuclide on the human body depends on the absorbed energy per unit of mass and the parts of the body directly affected. Thus, aspects that should always be considered are what the dose absorbed by the body was and which organs received the highest doses (IAEA, 1988; National Nuclear Energy Commission [CNEN], 2005; CNEN; Haber & Rothstein, 1969). Such issues, associated with the "Law of Bergoni é and Tribondeau" (CNEN; Haber & Rothstein, 1969) indicate the danger of exposure to radiation doses for people, especially in infants and children. Radionuclides released into the marine environment may be dispersed, diluted, redistributed, and finally accumulated in specific ecosystem compartments. The processes of dispersion, accumulation, and transport of radionuclides in the marine environment are influenced by physical, chemical, and biological factors (Aarkrog, 2003; IAEA, 2001, 2004).

1.2 Angra dos Reis Nuclear Power Plants

The Almirante Álvaro Alberto Nuclear Power Station (CNAAA) is located on the southeastern coast of Brazil, in the city of Angra dos Reis, 100 km west of the city of Rio de Janeiro, and is the site of two nuclear power plants: Angra I and Angra II (Angra III is under construction), both operated by Eletrobras Termonuclear—ELETRONUCLEAR (ETN). The two plants, Angra I and Angra II, located on Itaorna Beach, began operations in 1985 and 2000, respectively. The water used for cooling in the plants is collected at Itaorna Beach and discharged, along with liquid effluents, through an underground channel in Saco da Piraquara de Fora Bay (SPF) (ETN), where Piraquara Beach (PB) is located (Figure 1).
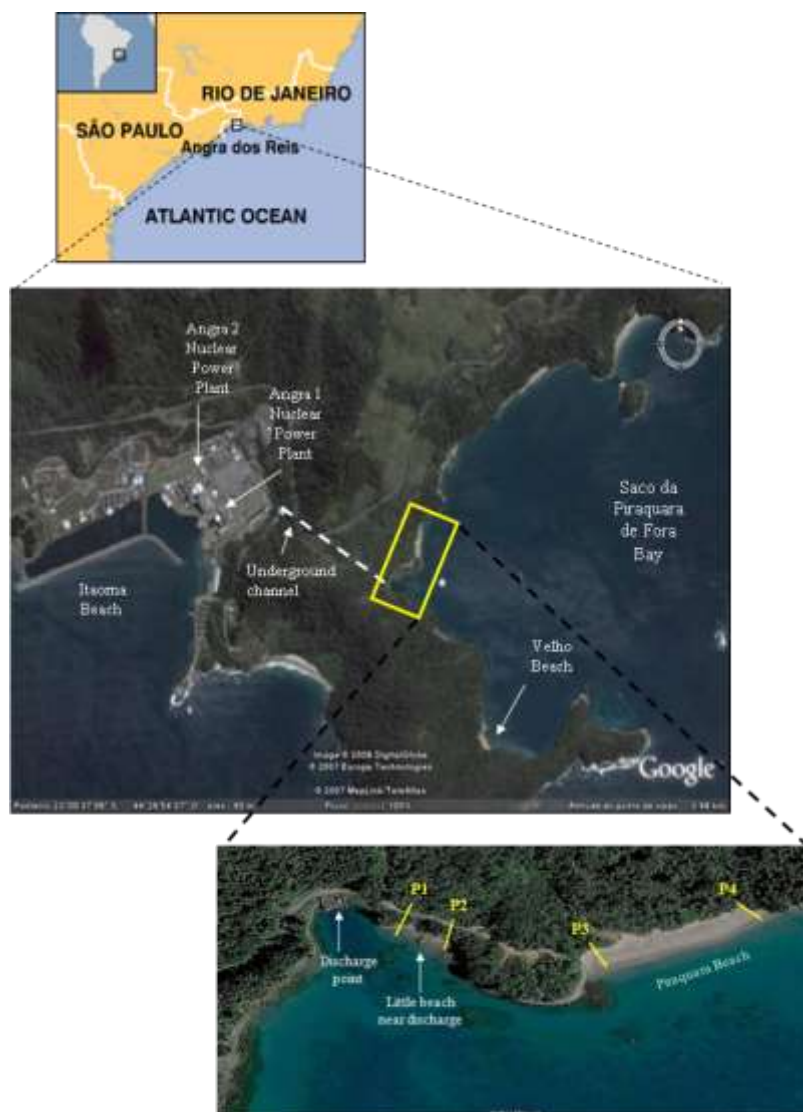

Figure 1. Location of the Angra 1 and 2 nuclear power plants on Itaorna Beach, Angra dos Reis, Brazil. The expanded view shows the underground channel, the discharge point, and Piraquara Beach in Saco da Piraquara de Fora Bay. The main sand sampling points (P1, P2, P3, and P4) are shown in yellow.

In 1985, a landslide, which occurred on PB, modified the composition of the sand at PB, and almost blocked the CNAAA discharge point (Figure 2). After restoration, land advancement separated a small stretch of sand, which is used as a beach and is located beside the CNAAA discharge point, from PB (Figure 1). Today, this small stretch of sand and PB are often used for leisure and recreation.
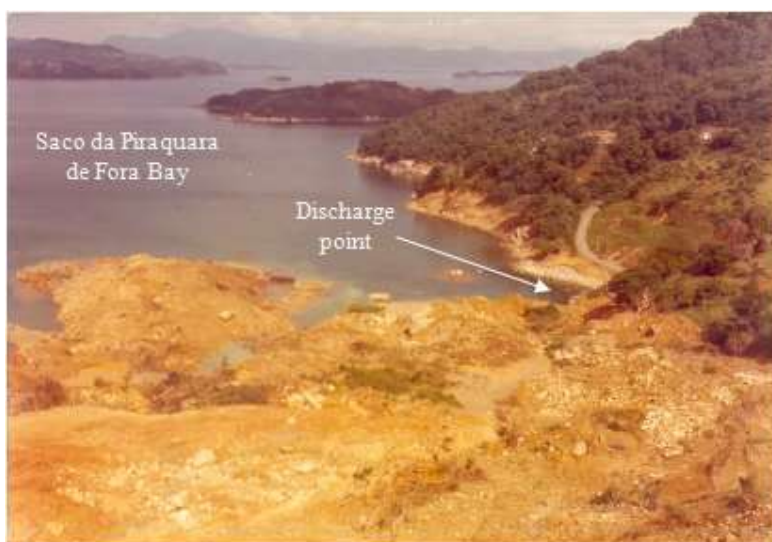

Figure 2. Photo of the 1985 landslide that almost blocked the Almirante Álvaro Alberto Nuclear Power Station (CNAAA) discharge point.

1.3 Cs-137 in Beach Sand

In comparing with the studies of Cs-137 in soils (Dion, Romanek, Hinton, & Bertsch, 2005; Bostick, Vairavamurthy, Karthikeyan, & Chorover, 2002; Flury, Mathison, & Harsh, 2002; Lujanienė, Vilimaitė-Šilobritienė, & Jokšas, 2005; McKinley et al., 2004; Todorović, Milonjić, & Čomor, 1992; Chowdhury, Kamal, Alam, Aftabuddin, & Zafar, 2004; Komarneni, 1985; Kanai et al, 2013; Rajec, Šucha, Eberl, Środoń, & Elsass, 1999; Sawhney, 1970; Environmental Protection Agency [EPA], 1999; Wahlberg & Fishman, 1962), the adsorption of the Cs-137 radionuclide in beach sand has not yet been well explored (Flury, Czigány, Chen, & Harsh, 2004; McKinley et al., 2001; Nyarko et al., 2011). The migration of the Cs-137 radionuclide through soil was observed in the Goiânia accident (IAEA, 1988), and also surrounding the Hanford plant (Flury et al., 2004) where high levels of Cs-137 caused by seepage were detected in sediments and in other studies (McKinley et al., 2001; Bostick et al., 2002; Flury et al., 2002; McKinley et al., 2004; Todorović et al., 1992; Chowdhury et al., 2004). Several studies have assessed the factors that cause adsorption of Cs-137, and the type and location of Cs^+ ion adsorption in the inner layers and the frayed edge sites (FES) of phyllosilicate minerals, such as vermiculite, illite, kaolinite, muscovite, and others (Figueira & Cunha, 1998; Flury et al., 2004; McKinley et al., 2001; Nyarko et al., 2011; Dion et al., 2005; Bostick et al., 2002; Flury et al., 2002; Lujanienė et al., 2005; McKinley et al., 2004; Todorović et al., 1992; Chowdhury et al., 2004; Komarneni, 1985; Kanai et al, 2013; Rajec et al., 1999; Sawhney, 1970; EPA, 1999; Wahlberg & Fishman, 1962).

McKinley et al. (2004) reported that the absorption of Cs^+ ions by phyllosilicate minerals, such as illite and kaolinite, is favorable. The exchange of K^+ ions for Cs^+ ions in the internal structural layers of tetrahedral-octahedral-tetrahedral (T-O-T) type structures results in a strong retention of Cs^+ ions within phyllosilicates. Another interesting study presented by the Environmental Protection Agency of the United States of America (EPA-USA) measured significant partition coefficient values for the Cs^+ ion in minerals composed of phyllosilicates, such as vermiculite and kaolinite, among others (EPA, 1999).

1.4 Study Site in Saco da Piraquara de Fora Bay

In our study area, the sand from the beach at PB was mixed with the soil from the landslide, which altered the mineralogical composition, and could possibly result in an increased adsorption of the Cs-137 radionuclide in the recreation areas of the beaches near the CNAAA liquid effluent discharge point. Liquid effluents from CNAAA are released in batches of cooling water into SPF. The geography of this inlet and the presence of islands at the entrance to the inlet, create a natural barrier against the entry of big currents and waves, and retard the dispersion of radionuclides out of SPF. Thus, the accumulation of the Cs-137 radionuclide in the sand is directly related to its concentration in the seawater. This release of effluents can lead to increased amounts of radionuclides in the

marine water, and may exceed the values recommended by the United States Nuclear Regulatory Commission [US-NRC] and the safe limit of radioactive dose exposure recommended by the National Nuclear Energy Commission (CNEN, 2005), which is the Brazilian regulator of ETN.

The current generated by the discharge of cooling water added to the liquid effluents from CNAAA mixed with the waters of SPF, prevents the formation of waves on PB and, particularly, on the small stretch of sand used as a beach close to discharge point. This lack of waves causes the sand morphology of the beach to not be significantly altered (Pilkey et al., 1993; Suguio, 1992), and this could possibly enhance the adsorption of the Cs-137 radionuclide by the sand because dispersion in SPF is slow and the grains that make up the sand layers are only slightly changed. This situation could lead to a localized concentration of Cs-137 with possible migration occurring mainly after the sand mixed with the soil from the 1985 landslide.

Franklin, Rosman, and Fernandes (2004) modeled the dispersion of Cs-137 and H-3 radionuclides in SPF and reported that CNAAA, according to its environmental licensing, can release up to 8×10^{11} Bq·year^{-1} of the Cs-137 radionuclide. The authors determined that SPF is not affected by wind or by external currents, and that the speed of the cooling water and disposal of effluents from CNAAA is the main current source in SPF.

Carvalho, Ferreira, Azevedo, Martins, and Lauria (2013) showed that the Cs-137 radionuclide was present in sediments that were in the direction of the mixing zone of SPF. The authors highlighted that the start of operations at the Angra 2 plant was responsible for increasing the flow of the discharge stream, which caused clay materials to be dragged from the bottom sediments to the discharge point, and increased the capacity for adsorption of the radionuclides released by CNAAA.

Lucca et al. (2005) studied the effect of the thermal plume from the CNAAA cooling water and discharged effluents in SPF using rhodamine. In their study, the dissemination of rhodamine in SPF mainly occurred along the short stretch used as a beach. This fact led us to study this area.

Therefore, considering the facts presented above, and because of the lack of studies performed on this short stretch, the determination of the concentration of the Cs-137 radionuclide in the SPF seawater, in the short stretch used as a beach, and in the PB leisure area is important because these are the first sites to receive the CNAAA cooling water and liquid effluents.

2. Method

2.1 Materials and Equipments

Materials used are   nitric acid P.A. (Merck), cesium chloride P.A.(Fluka), APM (Aldrich) and 1% nitric acid solution. The equipments used are a Atomic Absorption Spectrometer VARIAN, model Spectr AA558, for chemical analyses; a Bruker-D4 Endeavor XRD for XRD analyses and the gamma-ray spectrometer manufactured by Canberra for the radiometric analysis (for energy efficiency of the detector used standard cocktail made by Eckert & Ziegler Analytics, traceable by the National Institute of Standards and Technology (NIST).

2.2 Sampling

Samples were collected from the recreation zone, where people (children, in particular) have contact with the wet sand on the beach (National Environment Council [CONAMA], 2000), and in the spreading zone, where morphological changes of the sand occur due to wave action (Pilkey et al., 1993; Suguio, 1992). Profile samplings were carried out using collecting tubes, with the goal of identifying the presence of Cs-137 radionuclides in deeper layers. Seawater was sampled from the surface in front of the profile sampling sites at a distance of approximately 5 m from each collection point in the sand. The sampling points were located on PB, which is used as a recreation zone near the CNAAA discharge point, the short stretch used as a beach next to the discharge point, and the spreading area. The main sampling points near the discharge point are shown in Figure 1 and the coordinates of each sampling point are presented in Table 1. Figure 1 shows Velho Beach, the location of point 5, which is also used for recreation. This beach is not highlighted because it is farther away from the CNAAA discharge point; however, Velho Beach is close to the sediment collection points where the presence of Cs-137 radionuclides was detected by Carvalho et al. (2013). The blank samples (sand and seawater) were collected at Mambucaba Beach (point 6), which is located 9 km southeast of CNAAA. The coordinates were determined using a Map 276C Global Positioning System (GPS) instrument manufactured by Garmin.

For sampling of the sand profiles, two iron cylindrical collectors were built, each with a transparent acrylic tube placed in its interior, similar to those used in previous studies (IAEA, 1989, 2003; Boyd, 1995; Boyd & Tucker, 1998). The sampling depth was about 20 cm at points 1 and 2 (next to the discharge point), 40 cm at points 3 and 4, and 60 cm at points 5 and 6.

The sand samples were collected in the recreation area in a location between the high and low tide, but where the water from the waves was always in contact with the sand. A circular sampling design was used, based on one central sample with eight further samples collected at a radius of 30 cm from the central sample, simulating a compass. About 5 kg of sand was collected, drained by gravity for 6 h, dried at 105 °C until a constant weight was reached, and stored in Marinelli beakers. The samples were subsequently sealed, stored under refrigeration, and analyzed by gamma spectrometry (IAEA, 2003; Vianna et al., 1995; Michael, 1998; Cunha & Fabra, 1995; Godoy et al., 2003; Cunha, Munita, Paiva, & Teixeira, 1993).

Table 1. Coordinates of sand and seawater* sampling points

Sampling points	Latitude	Longitude
Point 1	23 °00′42″ S	44 °26′46″ W
	23 °00′42″ S(*)	44 °26′46″ W(*)
Point 2	23 °00′41″ S	44 °26′45″ W
	23 °00′41″ S(*)	44 °26′44″ W(*)
Point 3 - Piraquara Beach	23 °00′38″ S	44 °26′41″ W
	23 °00′38″ S(*)	44 °26′40″ W(*)
Point 4 - Piraquara Beach	23 °00′33″ S	44 °26′40″ W
	23 °00′33″ S(*)	44 °26′39″ W(*)
Point 5 - Velho Beach	23 °01′12″ S	44 °26′26″ W
	23 °01′12″ S(*)	44 °26′25″ W(*)
Point 6 - Mambucaba Beach (blank samples)	23 °01′40″ S	44 °31′56″ W
	23 °01′42″ S(*)	44 °31′56″ W(*)

2.3 Chemical characterization

Chemical analyses were carried out in the laboratories of the Center for Mineral Technology (CETEM). Granulometric curves were constructed for each point according to the classification of the Brazilian Technical Standards Association (ABNT, 1995) and Vargas (1977) for identification of the sand profiles (Figure 3).
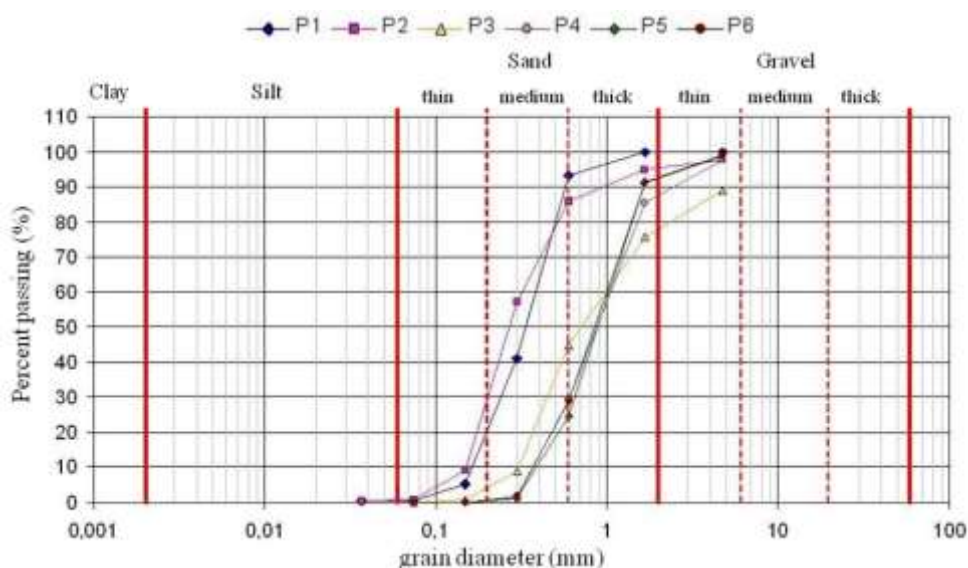

Figure 3. Granulometric curves for sampling points 1–6

The techniques employed for the elemental quantitative chemical analyses were: gravimetry for SiO_2 and ash; flame atomic absorption using acetylene/nitrous oxide for CaO, MgO, TiO_2, and Al_2O_3; flame atomic absorption using acetylene/air for Na_2O, K_2O, MnO, and Fe_2O_3. The measurements were performed in triplicate, and the results are presented in Table 2.

Table 2. Chemical characterization of sand samples from points 1–6

Parameters (%)	Point 1	Point 2	Point 3	Point 4	Point 5	Point 6
SiO_2	73.8	68.0	82.0	86.7	94.0	95.0
CaO	0.34	0.37	1.4	1.3	0.12	0.10
MgO	0.60	0.61	0.38	0.25	0.09	0.09
Na_2O	0.65	0.66	0.61	0.38	0.30	0.31
K_2O	1.6	1.5	1.4	1.3	0.70	0.64
TiO_2	3.9	4.3	0.81	0.35	0.20	0.18
MnO	0.14	0.19	441 mg·kg^{-1}	185 mg·kg^{-1}	66.8 mg·kg^{-1}	40.2 mg·kg^{-1}
Al_2O_3	4.9	4.3	4.2	3.5	2.1	2.0
Fe_2O_3	11.7	18.8	3.9	1.0	0.55	0.15
ash	0.94	0.86	2.0	1.7	0.53	0.33

X-ray diffraction (XRD) analysis of each sample was also performed at the laboratories of the CETEM, to complement the chemical profiling of the sand samples. The diffractograms obtained for points 1–5 are presented in Figures 4–8, respectively. The XRD powder diffractograms were obtained using a Bruker-D4 Endeavor system. The qualitative interpretation of the spectra was performed by a comparison with standards using the Bruker DiffracPlus software. The XRD results are discussed in section 3.2.
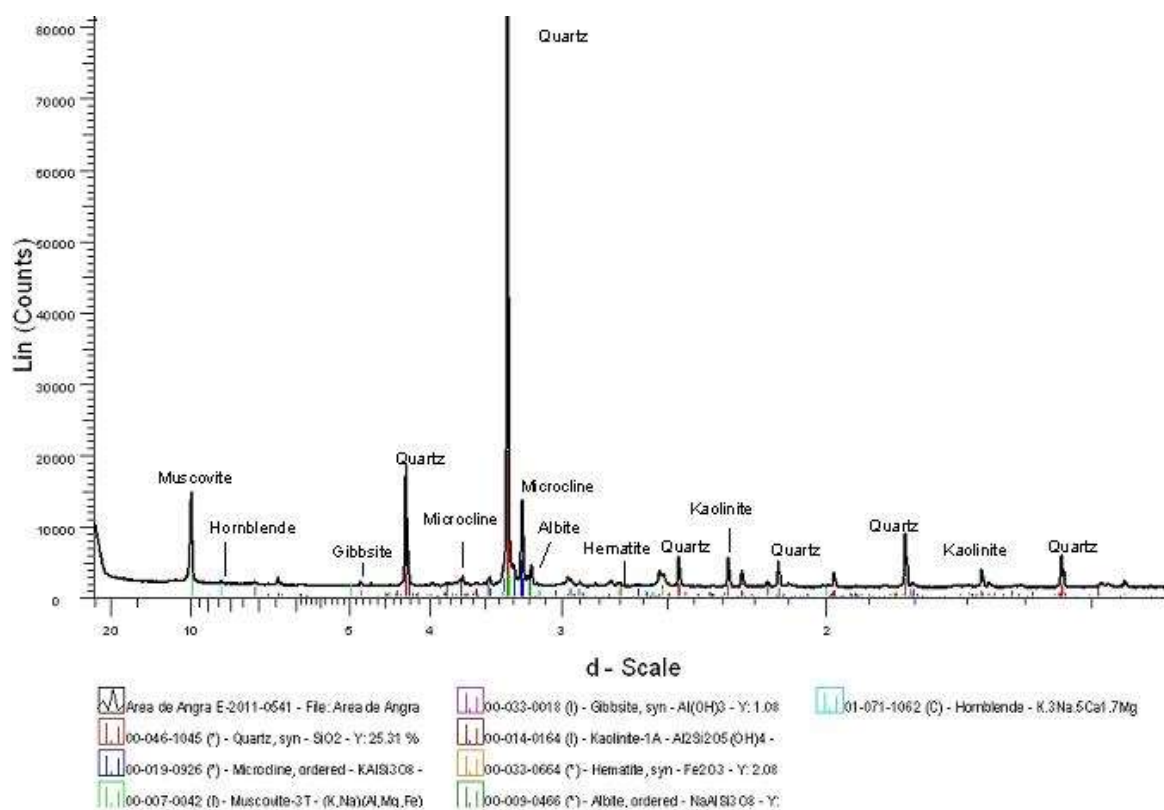

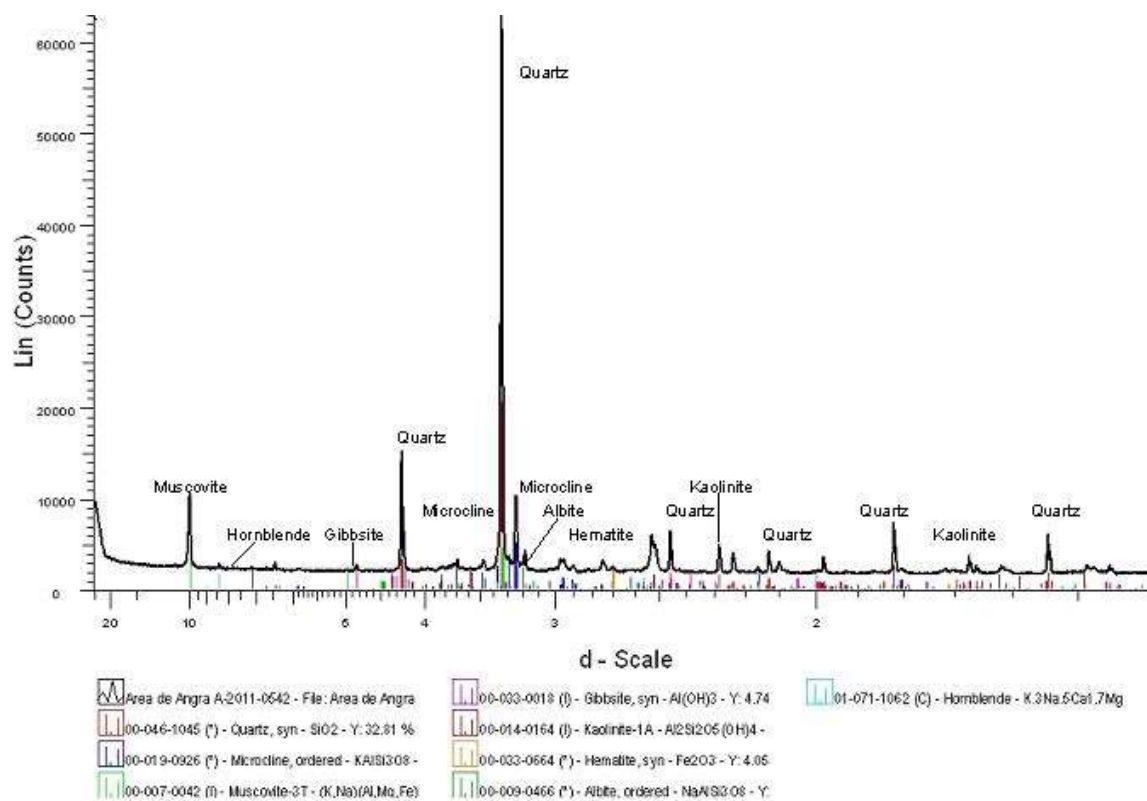Figure 4. X-ray diffractogram of the point 1 sample

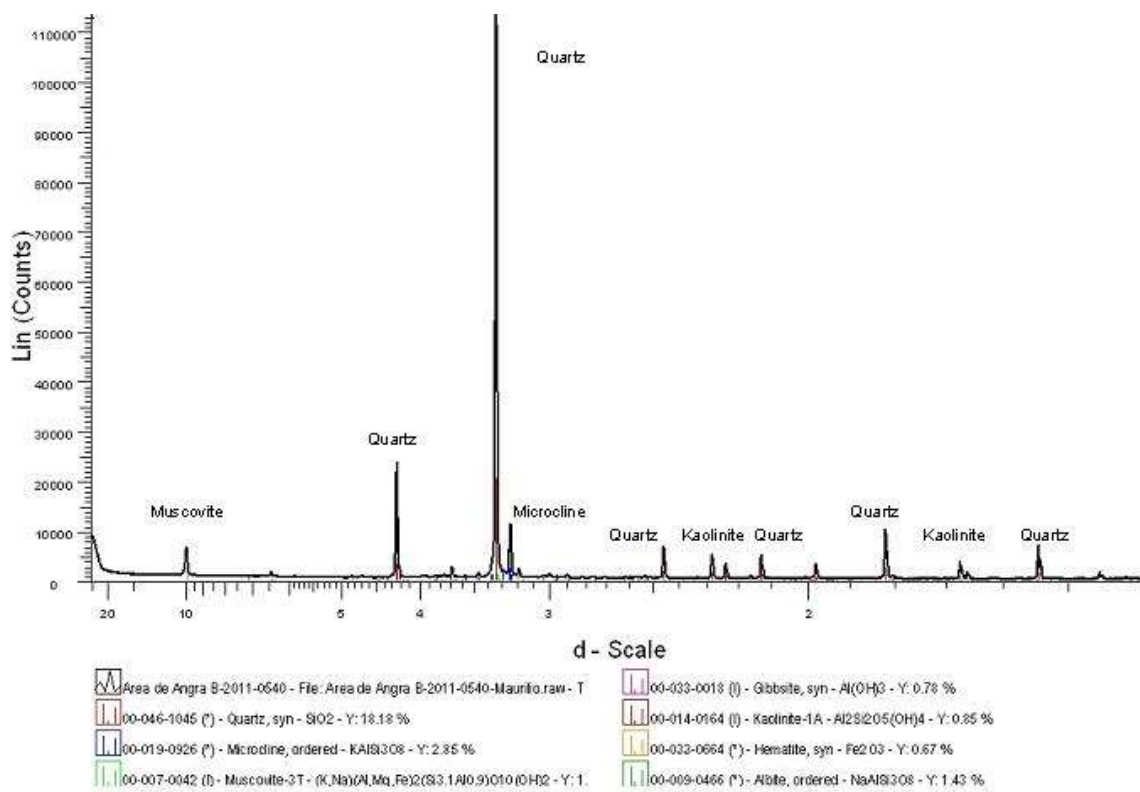Figure 5. X-ray diffractogram of the point 2 sample
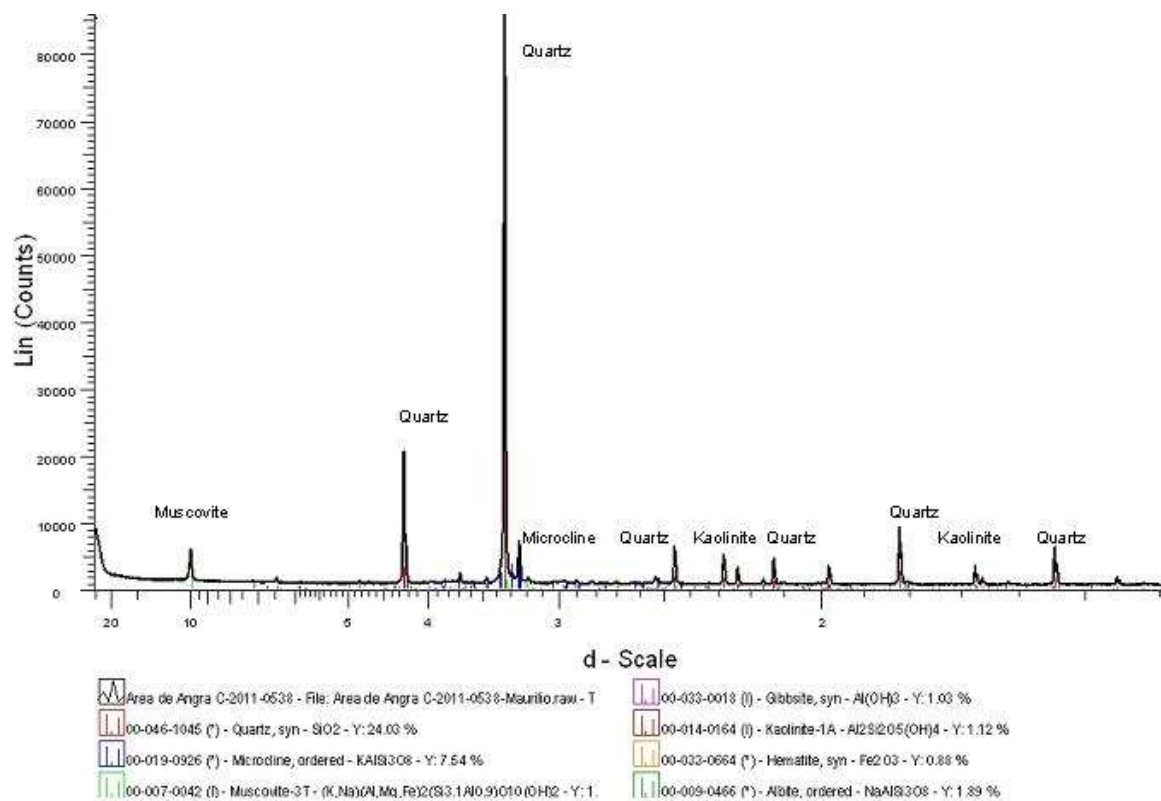

Figure 6. X-ray diffractogram of the point 3 sample

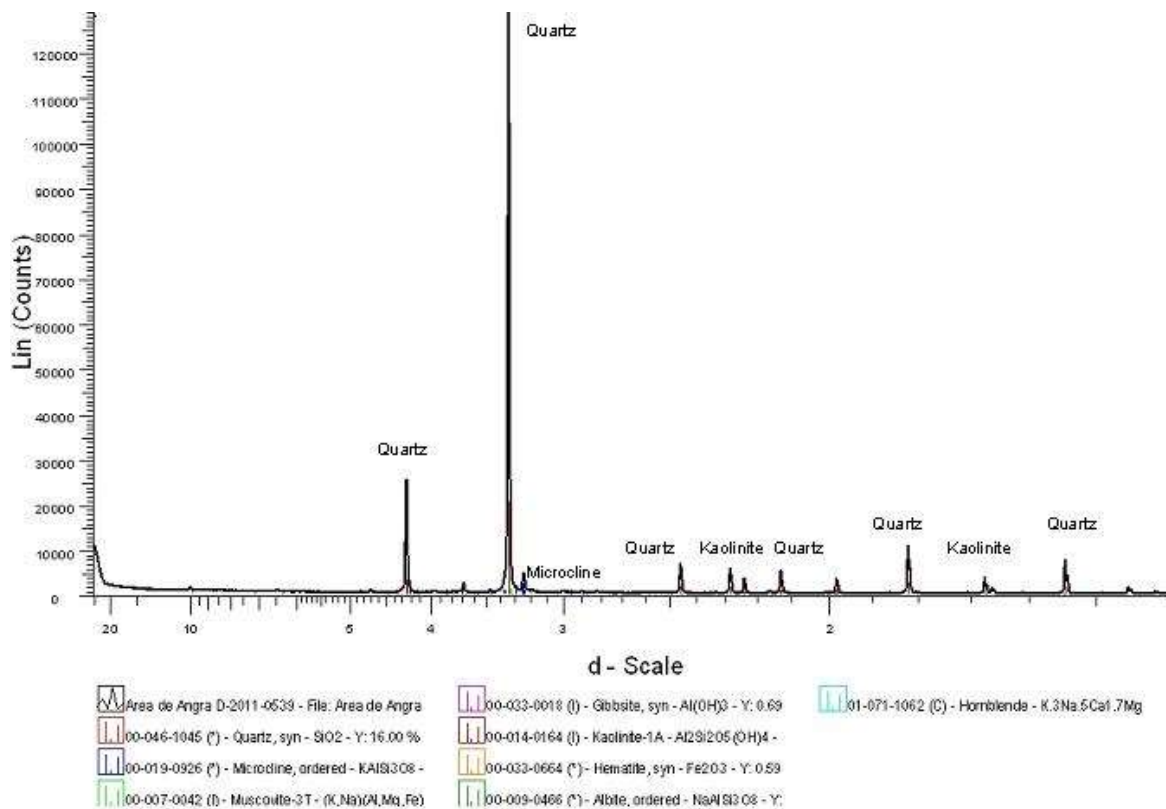Figure 7. X-ray diffractogram of the point 4 sample

Figure 8. X-ray diffractogram of the point 5 sample

2.4 Gamma Spectrometry Analysis

The radiochemical analyses were performed in the ETN laboratory that regularly participates in inter-laboratory exercises promoted by the CNEN. Gamma spectrometry was used for direct analysis of the sand samples. Two methods were used for gamma spectrometry analysis of the seawater samples: direct analysis and analysis of the precipitate resulting from adsorption of the Cs-137 radionuclide by ammonium phosphomolybdate (APM).

Method 1: About 5 L of seawater was collected from the surface, filtered with a 0.45 mm filter, and placed in polyethylene bottles. These samples were transferred to Marinelli beakers, sealed, and stored under refrigeration until the radiochemical analysis was performed (IAEA, 2003; Vianna et al., 1995; Michael, 1998; Cunha & Fabra, 1995; Godoy et al., 2003; Cunha et al., 1993).

Method 2: About 20 L of seawater was collected from the surface, filtered with a 0.45 mm filter, and the pH was adjusted to 1.6 using nitric acid. Cesium chloride P.A. (0.26 g) was added, and the liquid was agitated for 10 min. Subsequently, APM (4.0 g) was added, the liquid was agitated for 1 h, and then left to rest for 24 h. The sample was then filtered with a 0.45 mm filter and washed with a 1% nitric acid solution. After drying at 60–70 ℃ for 48 h, the filters were counted for 60 000 s using the same geometry as the certified standard (Nyarko et al., 2011; Buesseler, Aoyama, & Fukasawa, 2011; Honda et al., 2012; Aoyama, Tsumune, Uematsu, Kondo, & Hamajima, 2012; Kaeriyama et al., 2013; Cunha, Figueira, & Saito, 1999; Aoyama, Hirose, Miyao, & Igarashi, 2000; Figueira, Saraiva, & Cunha, 2001; Inoue et al., 2012). The recovery of added cesium was around 98%.

The gamma-ray spectrometer was equipped with a high purity germanium detector. This system has a 20% relative efficiency and an energy resolution of 1.8 keV for the Co-60 radionuclide at the reference energy of 1.33 MeV. The coaxial germanium detector was mounted vertically and coupled to a digital 3 keV high voltage source. The detector was placed inside a thick lead shielding with an inner layer of cast copper to reduce background radiation. The output signal of the detector was directed to a computer equipped with a Model 9660 multichannel analyzer (Canberra). The Genie-2000 software package from Canberra was used for data acquisition, and in the scanning and nuclide identification modules. The system was calibrated for energy and efficiency in the same geometry as that of the samples. The energy calibration was performed using a certified calibration source with known energies: Co-60, $E\gamma$ = 1332.5 keV and Cs-137, $E\gamma$ = 661.6 keV. The energy efficiency of the detector was determined using a standard cocktail made by Eckert & Ziegler Analytics, traceable by the National Institute of Standards and Technology (NIST) in the same geometry as that of the samples.

The Marinelli beakers containing the samples (sand and seawater) were placed in direct contact with the detector. Each sample was counted for 60 000 s. The decays were corrected to July 2012 for both the sand and seawater samples. Measurements were also carried out with an empty Marinelli beaker, under identical conditions, to determine the background count. The background count was subtracted from each sample measurement to obtain the actual activity of the radionuclides.

To obtain gamma spectrometry measurements for the Cs-137 radionuclide adsorbed by APM, the seawater samples were filtered using a filter of the same dimensions as the standard filter cocktail of radionuclides manufactured by Eckert & Ziegler Analytics. The concentration of the Cs-137 radionuclide was measured directly at 661.6 keV (Vianna et al., 1995; Cunha & Fabra, 1995; Buesseler et al., 2011; Honda et al., 2012; Aoyama et al., 2000, 2012; Kaeriyama et al., 2013; Figueira et al., 2001; Inoue et al., 2012; da Silveira, Schmidt, Campos, de Godoi, & Ikeda, 2000).

3. Results and Discussion

3.1 Sampling

We observed that the mixture of soils from the landslide affected the sampling locations differently, because the points that did not experience effects from the landslide had greater depth values in the sampling. The new composition of the beach sand, especially in points 1 and 2, generated greater resistance to the entrance of the profile sampler at the collection points. The points that were moderately or weakly affected by the landslide, points 3 and 4, had greater depth with the profile sampler compared with points 1 and 2. The results of the elemental analysis (Table 2) are in agreement with this view, as higher concentrations of Fe_2O_3, TiO_2, and Al_2O_3 were observed in points 1 and 2, which were most affected by the landslide (see Section 3.2).

3.2 Chemical Analyses

The granulometric curves (Figure 3) show that there is little difference in the sample composition at points 1 and 2. These locations were mostly composed of particles smaller than 0.04 mm, i.e., silt(0.6 and 0.8%, respectively), whereas points 3–6 were mostly fine granulated sand according to the ABNT 6502/95 standard (ABNT, 1995) and Vargas (1977). Figure 2 shows that the landslide occurred predominantly in the area where points 1 and 2 are

located; this explains the differentiated composition observed (Table 2) and the different grain sizes observed (Figure 3). Using this reasoning, points 3 and 4 were moderately affected by the landslide; compared with points 1 and 2, points 3 and 4 have a higher concentration of SiO_2 and a similar concentration of Al_2O_3, whereas the highest concentration of CaO is observed for points 3 and 4 (Table 2). Points 5 and 6 have similar compositions and are characterized by a reduction in Fe_2O_3 concentration when compared with the other points.

The chemical characterization of the samples (Table 2), supported by the granulometric curves, shows the differences in the chemical compositions of the samples collected at the different points. The XRD analyses of points 1 and 2 (Figures 4 and 5, respectively) complement these experiments, as the presence of phyllosilicate minerals is observed. These minerals, such as microcline, muscovite, gibbsite, kaolinite, hematite, albite, and hornblende, are Cs^+ cation exchangers. The XRD analyses of points 3 and 4 (Figures 6 and 7, respectively) also show the presence of microcline, muscovite, gibbsite, kaolinite, hematite, albite, and hornblende, but with lower intensities. This result confirms that the main area affected by the landslide was where points 1 and 2 are located. The XRD analysis of point 5 (Figure 8) shows the predominant presence of quartz, with an insignificant presence of phyllosilicates, which indicates that the beach sand at this point is less prone to contamination by the Cs-137 radionuclide.

3.3 Cs-137 Gamma Spectrometry Analysis

The measured concentration of the Cs-137 radionuclide in the seawater samples for all sampled points by both methods (<0.42 $Bq \cdot L^{-1}$ for direct analysis by gamma spectrometry and <0.004 $Bq \cdot L^{-1}$ for analysis using pre-concentration with APM) was below the minimum detectable activity (MDA) values for gamma spectrometry analysis. The measured concentration of the Cs-137 radionuclide in the sand for all sampled points was below the MDA value of 0.31 $Bq \cdot kg^{-1}$.

The amount of radionuclides from nuclear tests, nuclear accidents, effluents from nuclear fuel reprocessing plants and nuclear power plants, and nuclear weapons detonations are linked to fallout and ocean currents and have distinctive values around the world (Table 3). A factor that could have contributed to the low concentrations of Cs-137 found in this study is the dilution effect that occurs when creeks, streams, and rivers, which are commonly present in the southern region of the State of Rio de Janeiro, are present next to coastal waters.

Table 3. Cs-137 radionuclide activity in seawater from Piraquara Beach and other regions in Brazil and the world

Region	Reference	Cs-137 ($Bq \cdot m^{-3}$)
Brazilian Coast	Figueira & Cunha, 1998	1.4
Brazilian Southeastern Shelf	Godoy et al., 2003	0.9–4.0
São Paulo Coast (Brazil)	Cunha et al., 1993	1.7–1.9
Whole Brazilian Coast	Cunha et al., 1999	0.8–1.7
Brazilian Southern Coast	Figueira et al., 2001	1.0–4.7
Around the Japanese archipelago	Inoue et al., 2012	0.93–52.53
Mediterranean Sea	IAEA, 1995	5.0–11.8
Northeastern Atlantic	IAEA, 1995	2.8–125
Pacific Ocean	IAEA, 2005	1.4–2.0[1]
South and Southeastern Atlantic	IAEA, 2005	1.4
Piraquara Beach (PB)	This study*	<4.0
Small stretch of beach next to the CNAAA discharge point	This study*	<4.0
Velho Beach	This study*	<4.0

* results from the methodology using ammonium phosphomolybdate (APM)

With respect to the results presented in Table 3, it is important to note that the seawater samples were collected in the open sea, far from the coast, where there is significant mixing with ocean currents along the Brazilian coast. The position of the southern coast of Rio de Janeiro, in relation to marine currents, is somewhat north of the region of convergence of two major ocean currents (the warm Brazil current and the cold Falklands current), and this region is not influenced by the northern hemisphere ocean currents (da Silveira et al., 2000). Lower values are detected for the Cs-137 radionuclide along the coast of Brazil (Table 3) because accidents, nuclear testing, and the discharge of radioactive waste in the oceans are more concentrated in the northern hemisphere, according to reports from the IAEA (IAEA, 1995, 1999, 2001, 2004).

This study comprised sampling of water in bathing beaches and recreational areas. The current created by the discharge of effluents and cooling water in the opposite direction of the waves creates a resistance to the arrival of waves on the sands of PB. This fact retards the dispersion of the Cs-137 radionuclide and promotes its deposition on the beach sands next to the CNAAA discharge point. Thus, it can be assumed that the values

determined at these sampling points were not affected by the influence of the concentration of the Cs-137 radionuclide in the oceanic currents.

The importance of correlating the values of Cs-137 found in the sand with the radiation to which humans will be exposed was highlighted in Section 1.1. However, such a correlation can only be validated by calculating the amount of radiation to which the public is exposed (Alencar & Freitas, 2005; Freitas & Alencar, 2004; UNESCAR, 1988, 2000). The calculation of the radioactive dose exposure to the public at PB is important to verify the proper functioning of the CNAAA liquid effluent discharge. However, this study shows that the Cs-137 radionuclide values found are below the MDA values and are within the limit of release for effluents, which is 37 Bq·L^{-1}, recommended by NRC-USA for the Cs-137 radionuclide (Wahlberg & Fishman, 1962). Therefore, the values obtained in this study are not harmful to the public. This data is important to local beachgoers and to the general population.

4. Conclusion

The measurements of the Cs-137 radionuclide in sand samples from PB, the small stretch of sand used as a beach next to the discharge point, and Mambucaba and Velho Beaches confirm that these locations do not contain detectable radioactive contamination by the Cs-137 radionuclide. Granulometric and chemical composition analyses showed that the landslide at PB introduced minerals composed of phyllosilicates, which are known as Cs^+ cation exchangers, into the sand composition, especially close to the discharge point of effluents from CNAAA (points 1 and 2). However, the amount of phyllosilicates found in the beach sands was not enough to adsorb significant amounts of the Cs-137 radionuclide. The profile samples show that migration of the Cs-137 radionuclide did not occur to the deeper layers that were sampled because all the results were below the MDA values. The values of Cs-137 found in the seawater samples were similar to those found by other researchers who conducted studies in Brazilian waters at points farther away from the coast, where a dilution effect by ocean currents is present.

The values of the Cs-137 radionuclide measured in the samples collected in this study were below the MDA values and did not allow the radiation dose exposure of the public to be calculated. Therefore, we conclude that the beaches studied do not contain detectable amounts of the Cs-137 radionuclide. Thus, the public can enjoy the peace, tranquility, and beauty of these locations without the risk of exposure to the Cs-137 radionuclide. Conversely, we must increase our concern and radiochemical monitoring of oceans to acquire further knowledge about the extent of the effects along the Brazilian coast from the Fukushima radioactive accident. This incident compels us to continue with new radiological monitoring studies because of the large quantity of radionuclides being released into the atmosphere, seas, and oceans.

Acknowledgments

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