2015 International Nuclear Atlantic Conference - INAC 2015
São Paulo, SP, Brazil, October 4-9, 2015
ASSOCIAÇÃO BRASILEIRA DE ENERGIA NUCLEAR - ABEN
ISBN: **978-85-99141-06-9**

EVALUATION OF THE ACTIVITY CONCENTRATION OF ^{226}Ra, ^{228}Ra AND ^{210}Pb IN SEDIMENTS FROM ANTARCTICA IN THE ADMIRALTY BAY REGION

Tamires de A. Mora[1], Joselene de Oliveira[1], Rubens C. L. Figueira[2], Michel M. Mahiques[2] and Silvia H. M. Sousa[2]

[1] Instituto de Pesquisas Energéticas e Nucleares (IPEN/ CNEN –SP)
Av. Professor Lineu Prestes 2242
05508-000 São Paulo, SP
tamires.mora@usp.br
jolivei@ipen.br

[2] Instituto Oceanográfico da Universidade de São Paulo
Cidade Universitária
05508-120 São Paulo, SP.
rfigueira@usp.br
mahiques@usp.br
smsousa@usp.br

ABSTRACT

In this study, we performed the radiochemical characterization of a sedimentary record (1B profile), collected in the vicinity of Admiralty Bay, King George Island in Antarctic Operation XXXI (January/2013). The activities of ^{226}Ra, ^{228}Ra and ^{210}Pb were determined by the counting of gross alpha and gross beta activities in the precipitates of Ba(Ra)SO_4 and $PbCrO_4$. Those measurements were carried out in a low background gas flow proportional detector. The ^{226}Ra and ^{228}Ra were measured after 21 days of the final precipitation. The ^{210}Pb activity was determined after 10 days of the precipitation date by gross beta counting of its ^{210}Bi decay product. The activity concentration of ^{226}Ra ranged from 11±1 (mBq g^{-1}) to 54±3 (mBq g^{-1}), and the ^{228}Ra ranged from 48±5 (mBq g^{-1}) to 155±16 (mBq g^{-1}). The activity concentration of ^{210}Pb varied from 8±1 (mBq g^{-1}) to 458±46 (mBq g^{-1}), while unsupported ^{210}Pb ranged from 6±1 (mBq g^{-1}) to 434±65 (mBq g^{-1}). The ^{210}Pb concentrations in sediments have often been used to dating events like deposition and accumulation in various marine environments. Taking into account the results of ^{210}Pb and ^{226}Ra activities obtained in testimony-1B it was estimated the unsupported ^{210}Pb activity which was applied to the CIC geochronological dating model (Constant Initial Concentration). Based in these data, the sedimentation rate obtained was 0.63±0.02 cm $year^{-1}$.

1. INTRODUCTION

Natural radionuclides have been widely used as tracers in research of oceanic processes and management of the coastal region [1-6]. Their applications as tracers in oceanographic and marine pollution studies have helped scientists to elucidate processes occurring in the water column (particle transport, carbon cycle, biogeochemical cycles, drag processes) or sediments (deposition, accumulation, transport and resuspension). The use of natural radionuclides allows often understand the material transfer and estimate various biogeochemical flows between the various compartments of assessed ecosystem.

Over the past 30 years, human activities have increased in the Antarctic environment. As a result, Admiralty Bay has been considered an Antarctic Specially Managed Area in order to avoid and minimize the cumulative environmental impacts due to activities undertaken by different countries in the region [7]. The understanding of local pollution processes concerning trace metals and isotopes in sediments is still scarce in Admiralty Bay.

In 2009, it was established the project "Identification of the sedimentary record of abrupt changes in the Antarctic climate over the Superior Quaternary: Defrost pulses" - coordinated by the Oceanographic Institute of the University of São Paulo (IO /USP) as a part of the Brazilian Antarctic Program - PROANTAR: Process nº 557044 / 2009-0 [8], sponsored by CNPq, 2009. In this project sampling campaigns were developed in the Austral summer during the Antarctic Operation XXXI (from 6 to 25, January 2013). The overall project aims to understand the sedimentary environment, past tense climate and its dynamics from paleoenvironmental reconstructions by means of geological, geochemical and geophysical analysis, and contribute to the understanding of the influence of marine glacio-ocean environments, resulting in a glacial historical geological record of the South Shetland archipelago - Antarctica. Within the broad study there were tested various chemical tools, including the determination of radioisotopes (^{226}Ra, ^{228}Ra and ^{210}Pb) in sedimentary records of the Antarctic Peninsula, specifically in a sediment profile named 1B, about 248 cm long, collected in the Admiralty Bay. Thus, the radiochemical characterization of the sedimentary record and the application of a model for calculating the sedimentation rate and geochronological age would allow evaluate the influence and its variations in climate events and possible environmental impacts from human activities in this region.

2. NATURAL RADIONUCLIDES IN THE MARINE ENVIRONMENT

The three natural radioactive series of the ^{238}U, ^{235}U and ^{232}Th have nuclides with short, intermediate and long half-lives in which they cease with the stable isotope Pb [9]. The ^{210}Pb is a beta emitter with half-life of 22.3 years, provided to the sea waters by ^{222}Rn alpha decay (3.8 days), daughter product of ^{226}Ra (1,600 years). Its contribution is also derived from the atmosphere, where the ^{222}Rn as a gas have the ease of escape from terrestrial interstices migrating to the upper layers. The ^{222}Rn, being a short half-life radionuclide, decays to ^{210}Pb and via precipitation process ends up depositing in the waters of the oceans. Since ^{210}Pb presents a particulate-reactive behavior, it is dragged into the depths by particulate matter and is deposited in the sediments seabed [10]. The precipitation of ^{210}Pb occurs by means of both wet and dry deposition, and it ends up being removed by particulate matter when in contact with water and is taken to the ocean floor compartment being deposited in the sediment. The ^{210}Pb which comes from the earth layer is known as supported lead and the ^{210}Pb that comes from the atmosphere is known as unsupported lead. In the surface ocean layer we measure the concentration of total ^{210}Pb, which means the total concentration of supported and non-supported ^{210}Pb.

The ^{226} Ra is an alpha emitter produced by the decay of ^{230}Th also present in the sediment. Its release to sea water takes place from marine sediments, being spread throughout the water column [10]. Its regeneration in the sediment is time consuming due to the long half-life of 75,400 years of ^{230}Th [11]. It has been used for a long time for monitoring studies of water masses movements in the ocean [12]. The ^{228}Ra is a beta emitter (5.75 years) produced by the radioactive decay of ^{232}Th, which is widespread in the sediment [10]. The ^{228}Ra has been applied as a tracer to determine horizontal turbulent diffusion coefficients in sea water as well as to track waters that had contact with the mainland [10].

3. MATERIALS AND METHODS

The experimental part was divided in locating the region of interest, collecting the sedimentary profile, pre-treatment of sediment samples, homogenization, leaching, radiochemical separation of natural radionuclides ^{226}Ra, ^{228}Ra, and ^{210}Pb and finally the counting of samples in a low background radiation gas flow proportional detector. This part of the experimental radiochemical procedure was carried out in the Environmental Radiometrics Laboratory (LRA) of IPEN.

3.1. Description of the study area

The Admiralty Bay is located in the King's George Basin and has an approximately area of 138 km², presenting waters with variable depths and maritime communication with the fjord which integrates Admiralty Bay in the Bransfield Strait [13]. This site features three large bays, which are: Martel cove, where is located the Brazilian station Comandante Ferraz (EACF); the Mackelar cove, where is placed the Peruvian station Macchu Picchu; and the Ezcurra cove, where the Polish Henryk Arctowski station is found [14]. The hydrodynamic of this region is determined by the heterogeneity of the seabed along with the circulation in Bransfield Strait [15].

The King George Basin is a deep, small morphotectonic depression in the northeast of the Bransfield Strait. Active submarine and subaerial volcanism, associated with back-arc spread in the basin, have generated numerous islands and seamounts and contributed volcanic ash to the basin floor in the form of gravity-flow and/ or airfall deposits [16]. The contribution of clastic sediments discharged from the adjoining landmasses is relatively insignificant except during periods of episodic meltwater streams.

During the Austral summer, the basin receives large amounts of biogenic materials seasonally produced by high planktonic activity in the surface water, with an average production rate of 1.6 g cm^{-2} d^{-1} [17]. On a regional scale, sea ice generally undergoes seasonal growth and decay, controlling surface water productivity; during the winter (July-October) the Bransfield Strait remains ice-covered with limited productivity, whereas during summer (December-April) the area is completed ice-free, giving rise to increased productivity. These processes are expected to be recorded by the sedimentary structures, textures, mineral composition and chemical properties of the sediments in the basin.

3.2. Sampling of the sediment profile

Samples were collected by the Oceanographic Institute of the University of São Paulo (IO/ USP) during Antarctic Operation XXXI and were transferred to IPEN for radiochemical analysis of ^{226}Ra, ^{228}Ra and ^{210}Pb. The sampling was conducted in Admiralty Bay (Fig. 1), where the 1B profile was obtained using a Piston corer (Fig. 2), with an approximate recovery of 250 cm of sedimentary column. The samples were collected in PVC tubes and conveniently described and stored for achieving the specific laboratory tests.

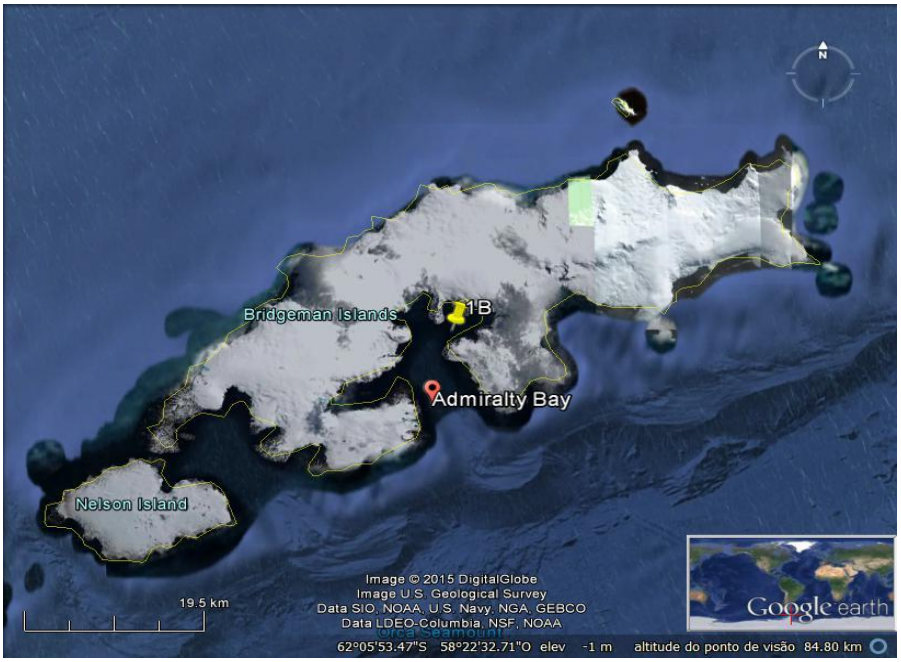

Figure 1: Map of the region studied in Admiralty Bay. The yellow marker indicates geographical location of profile 1B sampling site.

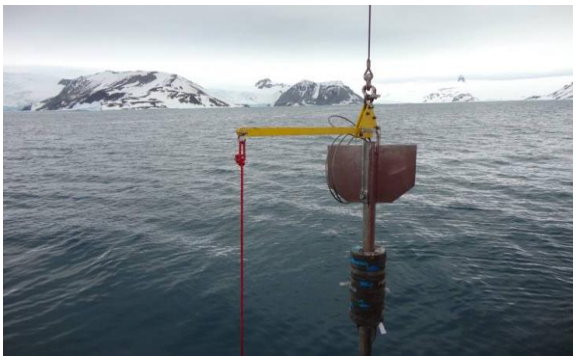

Figure 2: Piston Corer witnessed (Photo courtesy Marine Inorganic Chemistry Laboratory of São Paulo University).

3.3. Sample preparation for analysis

After opening and description of the sedimentary profile in PVC pipes, the removal of samples (aliquots) for testing was taken every 2 cm. These aliquots were stored in labeled polyethylene flasks with its respective information. The samples were frozen and subsequently submitted through the freeze-dried process.

For the radiochemical determination of ^{226}Ra, ^{228}Ra, and ^{210}Pb in the sediment, aliquot samples were obtained by weighting in an analytical balance, with masses ranging from 2.2 to 2.5 g (Fig.3).

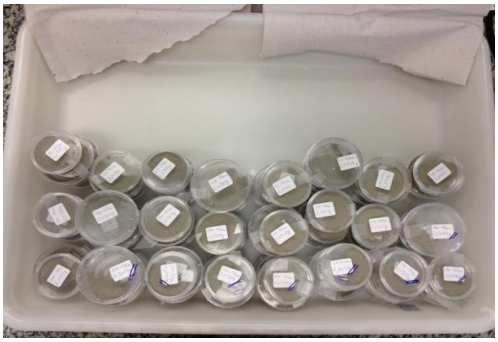

Figure 3: Aliquot samples of sediment profile 1B.

Following the sample homogenization, each sample was passed through an acid leaching method, which provides the total solubilization according to the EPA-3052 protocol [18], suitable for applications that require complete digestion of samples for research purposes. This protocol is applicable to assisted digestion of sediment matrices in a microwave, with acidic digestion pressurized pipe. The acid combination chosen used a ratio 3:1 nitric acid (HNO_3), hydrofluoric acid (HF) and hydrochloric acid (HCl). This is suitable for stabilizing high concentrations of Fe^{3+}, Al^{3+}, Ag^{+1}, Ba^{+2} and Sr^{+2} in solubilized samples. The total sample digestion was required for decomposition of the sedimentary matrix and stabilization of specific elements in high temperature reactions [18], with the help of brand digestor CEM microwave, MARS-6 model. All the acids used were concentrated with high purity to minimize interferences in the samples, and the use of hydrogen peroxide (H_2O_2) (30%) helped in the elimination of organic matter [18].

3.4. Sequential radiochemical separation of ^{226}Ra, ^{228}Ra and ^{210}Pb

Aliquots of 20 ml obtained after the leaching process of the samples were diluted to 1 L with Milli-Q purified water to initiate the radiochemical procedure.

The determination of ^{226}Ra, ^{228}Ra, and ^{210}Pb was done by the counting of gross alpha and gross beta activities in a low background gas flow proportional detector. The methodology is suitable to quantify these radionuclides in a 1 g marine sediments in which they are present mostly in low concentrations or as traces. The chosen technique has as positive the low background radiation and thus a considerably lower limit of detection.

The ^{226}Ra and ^{228}Ra were determined by co-precipitation with barium sulphate at pH 4.5-5.0 in the presence of EDTA, after separation from its daughters by complexation with NTA at pH 12.5-13.0 [19, 20]. The ^{226}Ra was determined by gross alpha counting of the $Ba(Ra)SO_4$ precipitate, after decay of ^{223}Ra and ^{224}Ra, that is, after 21 days. The determination of ^{228}Ra was done by measuring the gross beta activity of its daughter product ^{228}Ac, because it emits beta rays of higher energy (1.2 MeV) in contrast to the lower energy of ^{228}Ra beta particles (40 keV). Both measurements were carried out in a low background gas flow proportional counter Berthold LB-770, at 1650 V.

After its separation by complexation with NTA, ^{210}Pb was co-precipitated with $PbCrO_4$ [19, 20]. The ^{210}Pb was determined through the ^{210}Bi 10 days after the final precipitation, since this time interval is enough for the secular equilibrium to be reached. The gross beta counting

was also performed in a low background gas flow proportional counter Berthold LB-770, at the 1650 V.

The chemical yield was determined gravimetrically by weighing the final mass of $Ba(Ra)SO_4$ and $PbCrO_4$ precipitates, considering the initial additions of 1 mL of Ba^{2+} and Pb^{2+} carrier solutions, respectively. As for the Ra isotopes determination, 100% chemical yield corresponded to a mass of 34.1 mg of $Ba(Ra)SO_4$. For the radiochemical separation of Pb, 100% chemical yield matches a mass of 31.4 mg of $PbCrO_4$.

Typical lower limits of the detection (LLD) for these methods were 2.2 mBq g^{-1} for ^{226}Ra, 3.7 mBq g^{-1} for ^{228}Ra, and 4.9 mBq g^{-1} for ^{210}Pb, at a 95% confidence level. Samples were measured in replicates. The overall uncertainties of the sequential radiochemical determination of ^{226}Ra, ^{228}Ra and ^{210}Pb were below 10%.

The methodology was validated through the participation in proficient test organized by Instituto de Radioproteção e Dosimetria (IRD/CNEN) and Analytical Quality Control Services of The International Atomic Agency (IAEA). The proficient test called "The Interlaboratory Study on Determination Of Ra and U Radionuclides in Water" included a measurement of six water samples (3 natural and 3 synthetic) using the methodology described above indicated the uncertainties were lower than 5%. The final evaluation of our reported results in this intercomparison indicated they were in good agreement with IAEA reference values and were not biased by a systematic error, both for samples with low and high Ra activities. As for ^{210}Pb, the results of proficient test organized by IRD/CNEN also showed a good agreement with reference values. The uncertainties were below 10%.

4. RESULTS AND DISCUSSION

Activity concentrations of ^{226}Ra, ^{228}Ra, $^{210}Pb_{total}$ and $^{210}Pb_{uns}$ (not supported) determined in the sedimentary profile 1B are presented in (Fig.4-7). Activity concentrations of ^{226}Ra varied from 11 ± 1 (mBq g^{-1}) to 54 ± 3 (mBq g^{-1}), while ^{228}Ra activity concentrations ranged from 48 ± 5 (mBq g^{-1}) to 155 ± 16 (mBq g^{-1}). The $^{210}Pb_{total}$ activities ranged from 8 ± 1 (mBq g^{-1}) to 458 ± 46 (mBq g^{-1}), while $^{210}Pb_{uns}$ activities varied from 6 ± 1 (mBq g^{-1}) to 434 ± 65 (mBq g^{-1}).The Fig 4-7 shows the results as concentration activity vertical profiles of ^{226}Ra, ^{228}Ra, and $^{210}Pb_{total}$ $^{210}Pb_{uns}$ vs the depth of the sedimentary column studied. It is evident that the ^{226}Ra and ^{228}Ra distributions are almost uniform throughout the sedimentary column.

Fig. 6 and 7 present the vertical distribution of the activity concentrations of total ^{210}Pb and $^{210}Pb_{uns}$ along the sediment depth. Those figures evidenced a fairly high concentration of both $^{210}Pb_{total}$ and $^{210}Pb_{uns}$ from the top of the sedimentary column to approximately 78 cm deep, where it was observed experimentally that from this point, the ^{210}Pb activities exhibit practically constant trend, thus indicating that the secular equilibrium with its precursor ^{226}Ra was achieved.

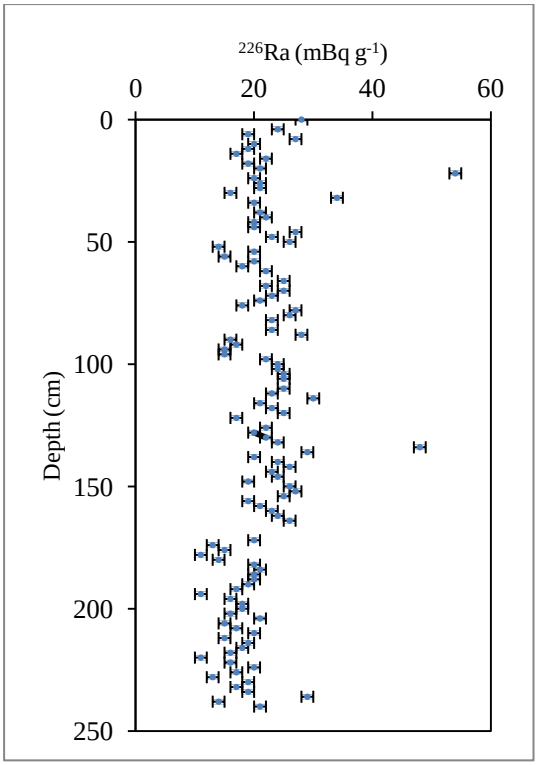

Figure 4: Distribution of activity concentration of ^{226}Ra along the 1B sedimentary column (248 cm).

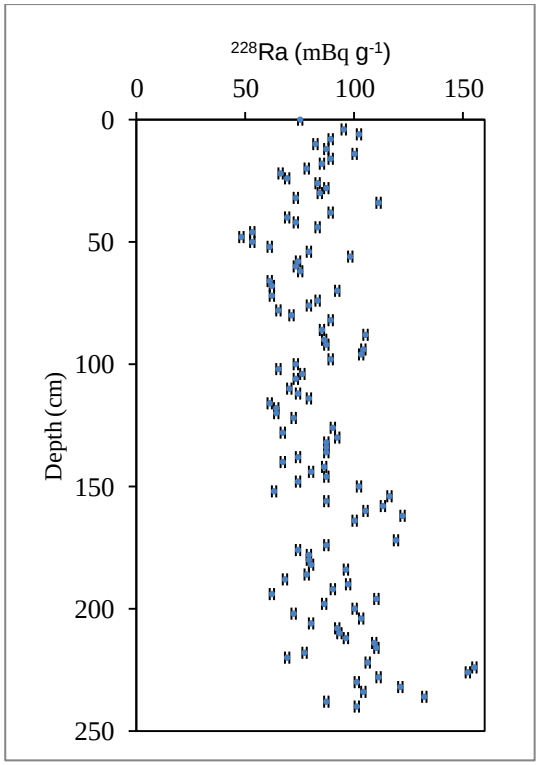

Figure 5: Distribution of activity concentration of ^{228}Ra along the 1B sedimentary column (248 cm).

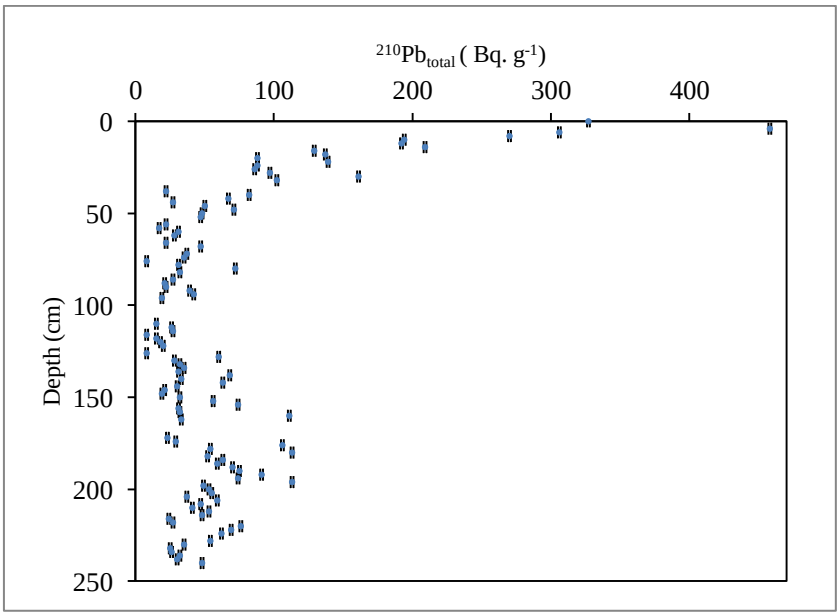

Figure 6: Distribution of activity concentration of $^{210}Pb_{total}$ along the 1B sedimentary column (248 cm).

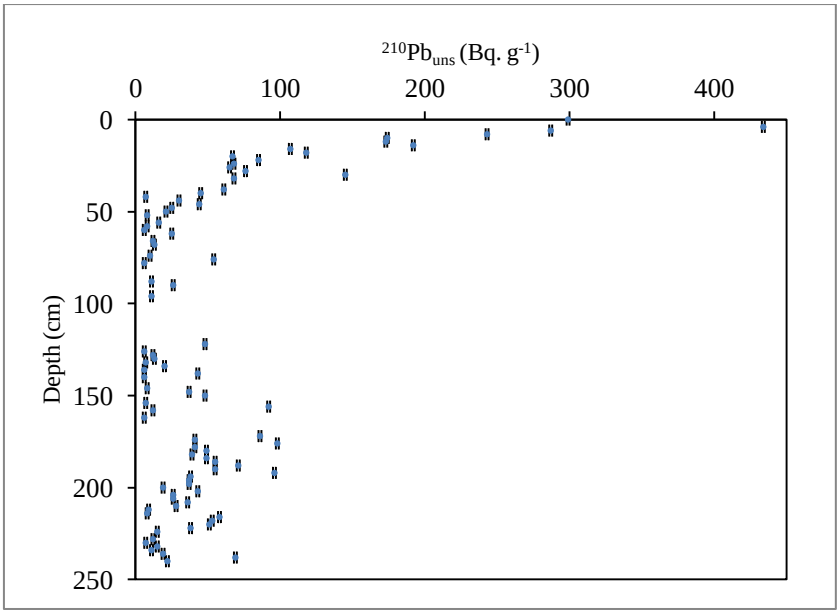

Figure 7: Distribution of activity concentration of $^{210}Pb_{uns}$ along the 1B sedimentary column (248 cm).

Considering the data available, we built the linear regression plot of the ln $^{210}Pb_{uns}$ as a function of the estimated depth of 78 cm, in order to obtain the corresponding angular coefficient and the correlation coefficient. The value of 0.049 was determined for the line slope, while the estimated correlation coefficient was 0.75. There is thus a correlation of 75% with respect to $^{210}Pb_{uns}$ to a depth of 78 cm from the studied sedimentary column (Fig.8).

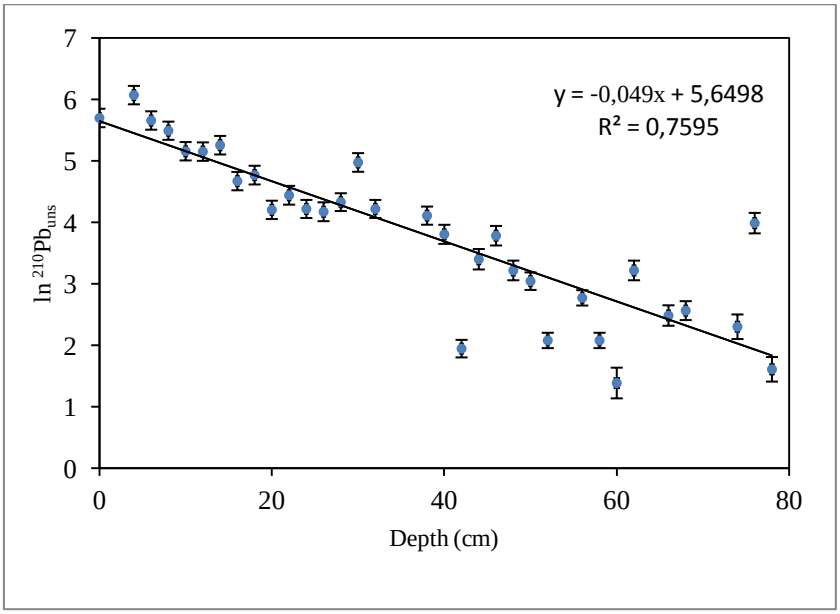

Figure 8: Linear regression plot of ln $^{210}Pb_{uns}$ vs depth (78 cm).

Since activity concentrations of ^{210}Pb in sediments has often been used to date events of deposition and accumulation of sediments in various marine environments, from the results of ^{210}Pb activities obtained in the profile sedimentary 1B and using the dating geochronological model CIC ("Initial Concentration Constant") [21, 22], it was possible to determine the sedimentation rate (cm/ year) by using the following equation:

$$Ts = \frac{\lambda}{b} \quad (1)$$

λ: radioactive decay constant for ^{210}Pb (0.031076 years $^{-1}$)
b: the linear regression coefficient in a $\ln^{210}Pb_{uns}$ cm^{-1} (0.049)

Since CIC model takes into account the constant initial activity concentration of $^{210}Pb_{uns}$ and also a continuous and constant input to the system, the sedimentation rate was of 0.63 ± 0.02 cm / year.

5. CONCLUSIONS

In this study, there was performed the radiochemical characterization of a sedimentary profile (1B), collected in the Admiralty Bay, King George Island, in Antarctica Operation XXXI (January/2013). The radiochemical separation of the of ^{226}Ra, ^{228}Ra, and ^{210}Pb in those sediment samples was done via coprecipitation, in the forms of $Ba(Ra)SO_4$ and $PbCrO_4$, respectively. The activities of ^{226}Ra, ^{228}Ra, and ^{210}Pb were determined by the counting of gross alpha and gross beta measurements in a low background gas flow proportional detector [19, 20].

The activity concentration of ^{226}Ra in the sedimentary profile (1B) ranged from 11 ± 1 (mBq g^{-1}) to 54 ± 3 (mBq g^{-1}), while ^{228}Ra varied from 48 ± 5 (mBq g^{-1}) to 155 ± 16 (mBq g^{-1}). The $^{210}Pb_{total}$ activities ranged from 8 ± 1 (mBq g^{-1}) to 458 ± 46 (mBq g^{-1}), while $^{210}Pb_{uns}$ activities ranged from 6 ± 1 (mBq g^{-1}) to 434 ± 65 (mBq g^{-1}).

The distribution of activity concentrations of ^{226}Ra and ^{228}Ra in 1B vertical profile as a function of depth showed that their distributions are almost uniform throughout the sediment column.

The distribution patterns of $^{210}Pb_{total}$ and $^{210}Pb_{uns}$ in vertical profiles presented increased concentrations on the top of the sedimentary column until to a depth of around 78 cm, where it was observed experimentally that the activity $^{210}Pb_{total}$ and $^{210}Pb_{uns}$ present were practically constant, thus indicating that the secular equilibrium with ^{226}Ra was achieved.

The activity concentration of $^{210}Pb_{uns}$ in sediments was applied to date events of deposition and accumulation of sediments in the Admiralty Bay, according to the dating geochronologic model CIC (English: "Constant Initial Concentration ") [21, 22]. It was estimated a sedimentation rate of 0.63 ± 0.02 cm / year for the sediment profile 1B.

ACKNOWLEDGEMENTS

I thank my supervisor Dr. Joselene de Oliveira of the Institute of Energy and Nuclear Research (IPEN-GMR) for the opportunity to carry out this work, Dr. Rubens C. L. Figueira of Inorganic Chemistry Laboratory (IO/ USP) for having given the testimony sediment, the CNEN/ IPEN the Master's scholarship throughout this graduate period, and my family and friends for their support in achieving this research. This project was funded by CNPq, Brazilian Antarctic Program - PROANTAR: Process nº 557044/ 2009-0.

REFERENCES

1. W.C. Burnett, R. Peterson, W. S. Moore, J. Oliveira. "Radon and radium isotopes as tracers of submarine groundwater discharge – Results from the Ubatuba, Brazil SGD assessment intercomparison". *Estuarine, Coastal and Shelf Science*, **76**, pp. 501-511 (2008).

2. W. S. Moore & Oliveira, J. "Determination of residence time and mixing processes of the Ubatuba, Brazil, inner shelf waters using natural Ra isotopes". *Estuarine, Coastal and Shelf Science*, **76**, pp. 512-521 (2008).

3. J. Oliveira, W. C. Burnett, B. P. Mazzilli, E. S. Braga, L. A. Farias, J. Christoff, V. V. Furtado. "Reconnaissance of submarine groundwater discharge at Ubatuba coast, Brazil, using ^{222}Rn as a natural tracer". *Journal of Environmental Radioactivity*, **69,** pp. 37-52 (2003).

4. J. Oliveira, M. Charette, M. Allen, E. S. Braga, V. V. Furtado. "Coastal water exchange rate studies at the southeastern Brazilian margin using Ra isotopes as tracers". *Radioactivity in the Environment*, **8**, pp. 345-359 (2006).

5. P. P. Povinec, H. Bokuniewicz, W. C. Burnett, J. Cable, M. Charette, J.-F. Comanducci, E. A. Kontar, W. S. Moore, J. A. Oberdorfer, J. Oliveira, R. Peterson, T. Stieglitz, M. Taniguchi. "Isotope tracing of submarine groundwater discharge offshore Ubatuba, Brazil: results of the IAEA-UNESCO SGD project". *Journal of Environmental Radioactivity*, **99**, pp. 1596-1610 (2008).

6. P. P. Povinec, I. Levy-Palomo, J-F. Comanducci, J.F, J. Oliveira, B. Oregioni, A. M. G. Privitera. "Submarine groundwater discharge investigations in Sicilian and Brazilian coastal waters using an underwater gamma-ray spectrometer". *Radioactivity in the Environment*, **8**, pp. 373-381 (2006).

7. I. R. Santos, E. V. Silva-Filho, C. E. G. R. Schaefer, M. R. Albuquerque-Filho, L.S. Campos. "Heavy metal contamination in coastal sediments and soils near the Brazilian Antarctic Station, King George Island". *Marine Pollution Bulletin*, **50**, pp. 185-194 (2005).

8. M. M. Mahique, C. C. Martins, M. C. Bícego, R. C. L. Figueira, I. Wainer, R. C. Montone, S. Taniguchi, R. A. Lourenço, S. H. M. Souza. Identificação do registro sedimentar de mudanças abruptas no clima na Antártica ao longo do Quartenário Superior: Pulsos de Degelo – 2009 – 2012,

http://laqimar.io.usp.br/index.php/pesquisa/projetos/13-projetos-concluidos/24-clima-da-antartica. Instituto Oceanográfico da Universidade de São Paulo. Programa Antártico Brasileiro – PROANTAR, Edital MCT/CNPq – N° 23/2009, processo 557044/2009-0. Operantar XXXI (6-25/01/2013).

9. W. Geibert. "Charts of the ^{238}U, ^{235}U, ^{232}Th and ^{241}Am decay series with principal modes of decay, their intensities and energies". *Radioactivity in the Environment*, **13**, pp. 417-423 (2008).

10. M. M. Rutgers Van Der Loeff & W. Geibert. "U- and Th-Series Nuclides as Tracers of Particle Dynamics, Scavenging and Biogeochemical Cycles in the Oceans". *Radioactivity in the Environment*, **13**, pp. 227-268 (2008).

11. W. S. Moore. "Large groundwater inputs to coastal waters revealed by ^{226}Ra enrichments". *Nature*, **380**, pp. 612-614 (1996).

12. F. F. Koczy. "Natural radium as a tracer in the ocean". United Nations Internat. Conf. Peaceful Uses of Atomic Energy, Geneva, 1958, **18**, pp. 351-357 (1958).

13. R. R. Weber & R. C. Montone. *"Rede 2 - Gerenciamento ambiental na Baía do Almirantado, Ilha Rei George, Antártida"*, Programa Antártico Brasileiro – PROANTAR, MMA/CNPq, Brasil (2006).

14. M. C. Castro, *"Avaliação de Introdução de Esteróis Fecais e Hidrocarbonetos Marcadores Geoquímicos em Sedimentos da Baía do Almirantado, Península Antártica"*, Dissertação de mestrado - IOUSP, São Paulo, Brasil (2001).

15. Z. Szafranski & M. Lipski, "Characteristics of water temperature and salinity at Admiralty Bay (King George Island, South Shetland Islands, Antartic) during the austral summer", *Polish Polar Research*, **3**, pp. 7-24 (1982).

16. J. B. Anderson & B. F. Molnia, "Glacial-marine sedimentation", *American Geophysical Union - Short Course Geology*, **9**, pp. 1-127 (1989).

17. G. Wefer, G.Fischer, D. Füetterer, R. Gersonde R, "Seasonal particle flux in the Bransfield Strait, Antarctica", *Deep Sea Research* **35**, pp. 891-898 (1988).

18. United States Environmental Protection Angency (US. EPA-3052), "Microwave assisted acid digestion of siliceous and organically based matrices", Method 3052, USA, (1996).

19. J. Oliveira, *"Determinação de ^{226}Ra e ^{228}Ra em águas minerais da região de Águas da Prata"*, dissertação de Mestrado, Instituto de Pesquisas Energéticas e Nucleares-Comissão Nacional de Energia Nuclear, São Paulo, Brasil, (1993).

20. S. R. D. Moreira, "Determinação de ^{210}Pb, em águas minerais da cidade de Águas da Prata", dissertação de Mestrado, Instituto de Pesquisas Energéticas e Nucleares-Comissão Nacional de Energia Nuclear, São Paulo, Brasil, (1993).

21. J. A. Robbins, "Geochemical and geophysical applications of radioactive lead", *Bigeochemistry of lead in the environment*, Amsterdam, pp. 285-393 (1978).

22. P. G. Appleby, F. Oldfield, R. Thompson, P. Huttunen, K. Tolonen, "^{210}Pb dating annually laminated lake sediments from Finland", *Nature*, **280**, pp. 53-55 (1979).