\(^{137}\)Cs INVENTORY IN SEDIMENTARY COLUMNS FROM CONTINENTAL SHELF OF SÃO PAULO STATE

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ABSTRACT

\(^{137}\)Cs is an artificial radioactive isotope produced by \(^{235}\)U fission. This radionuclide has a high fission yield and a half-life of 30 years. It has been detected in the environment since 1945 and its principal contamination source has been nuclear tests in the atmosphere. There are other sources of \(^{137}\)Cs contamination in the environment, such as: release from nuclear and reprocessing plants, radioactive dumping and nuclear accidents (Chernobyl, for example). This paper presents an inventory of \(^{137}\)Cs on the Continental Shelf of São Paulo State, a region located between Cabo de Santa Marta Grande (Santa Catarina state) and Cabo Frio (Rio de Janeiro state). In this area, 9 cores were collected by the Instituto Oceanográfico da Universidade de São Paulo (São Paulo University Institute of Oceanography). The cores were sliced at every 2 cm; sub-samples were lyophilized, grinded and stored in plastic containers. \(^{137}\)Cs was determined by 661 keV photopeak using a gamma spectrometry detector (Ge hyperpure). The analysis was performed by efficiency and background in different counting times. \(^{137}\)Cs concentration activities varied from 0.3 to 3.6 Bq kg\(^{-1}\) with a mean value of 1.2±0.6 Bq kg\(^{-1}\). The inventory of \(^{137}\)Cs in this area was 13±7 Bq m\(^{-2}\). Values obtained are in agreement with the Southern Hemisphere, a region contaminated by atmospheric fallout due to past nuclear explosions.

1. INTRODUCTION

Since its rise in the 1940s, nuclear energy has been the cause of debate all over the world because of its ambiguous character. It may be used to produce weapons of mass destruction as well as electric power, a necessity in both developed and developing countries.

The importance of monitoring artificial and natural radionuclides is related to the impact of these elements on the environment. Radioactive pollution, as well as other forms of pollution, is extremely harmful, mainly to the marine environment, as seas and oceans have been the major repositories of different kinds of pollution worldwide. Radioactive isotope research has greatly increased since the beginning of nuclear tests: several radionuclides have been used in environmental studies, especially in marine processes, and have become important tools in oceanography. Among the radionuclides produced artificially and released into the marine environment, \(^{137}\)Cs has a great importance because it has a high fission yield and a half-life of 30 years.
In the marine environment, this radionuclide may be retained by sediments through fixation in suspended matter and sedimentation, direct precipitation of colloidal forms and the direct fixation by adsorption and deposition of organic matter which had previously incorporated radionuclides [1].

The Brazilian marine coast has no significant sources of radioactive pollution, apart from the area of Angra dos Reis, where there are two nuclear power plants. The main source of radioactive contamination in Brazilian marine samples has been atmospheric fallout from past nuclear tests. There are some reports of artificial radionuclides in seawater, fish and marine sediments from the Brazilian coastal environment [1-4]. Thus, the objective of this work was to determine $^{137}$Cs levels in marine sediments from cores collected on the Continental Shelf of São Paulo State. Results from this study will enable the establishment of an inventory of this radionuclide in this important region of the South Atlantic Ocean.

2. SAMPLING AREA

The sampling area (Figure 1) is located in the center of an arc-shaped Brazilian margin known as São Paulo Bight which extends from latitude 23°00'S to 28°30'S which extends from Cabo de Santa Marta (28°30'S – 49°00'W) to Cabo Frio (23°00'S – 42°00'S) [5]. In this area, the shelf shows its maximum width (231 km) and a declivity of 1:1333. The shelf break is located between 160 and 180 meters depth and gives way to a relatively gentle continental slope.

![Figure 1: Sampling area and collecting points on the Brazilian southeastern coast](image)

The geological evolution of São Paulo Bight has resulted from the interaction between the Mesozoic-Cenozoic development of the Brazilian margin and the series of climatic cycles, especially those related to the Quaternary. A succession of drownings and desiccations of the shelf have resulted in the sedimentation of a thick layer of transgressive and regressive
deposits. The present sedimentation is essentially terrigenous, being constituted of quartzose, sands and mud. The inner and middle shelf dynamics are determined by the displacement of three water masses, which show strong seasonal variation [6].

In this area, 9 sedimentary columns (cores) were collected using a box core, and the water depths varied from 90 to 260 meters (Figure 1). Each core was described and sampled continuously at 2 cm intervals. Sub-samples were kept frozen for later freeze-drying. Water content was determined by weight difference before and after freeze-drying. Grain size was determined from the sand and mud found in each sample.

3. MATERIALS AND METHODS

3.1. Equipment
Gamma-ray spectrometers, low background HPGe detector, EG&ORTEC, GEM 50P and GMX 25190P models, with a resolution of 1.9 keV at 1332.40 keV in the photopeak of $^{60}$Co. The spectrometers are linked to an 8K MCA with spectrum stabilizer. Spectra were analyzed with MAESTRO® from EG&ORTEC.

3.2. $^{137}$Cs analysis
The samples, containing from 5 to 40 g of marine sediment, were dried, homogenized and transferred to appropriate plastic containers for gamma counting. $^{137}$Cs was assayed by photopeak of 661.6 keV. The method consisted of the following steps: detector calibration, state detector efficiency, cumulative counting of both background and samples in regular intervals of counting time, photopeak smoothing and linear regression. The methods of analysis and data acquisition are described in Figueira et al. [7] and Saito et al. [8,9].

4 RESULTS AND DISCUSSION

$^{137}$Cs concentration activity varied from 0.1 to 3.9 Bq kg$^{-1}$ in all samples analyzed, and mean value was 1.2±0.6 Bq kg$^{-1}$. Figure 1 shows mean values of $^{137}$Cs concentration activity from each core collected in this area.

These results of $^{137}$Cs concentration activities enabled the calculation of the $^{137}$Cs inventory by Equation 2:

$$I = \sum (A_s \times m_s)$$

where,

$I$ is the $^{137}$Cs inventory (Bq m$^{-2}$);  
$A_s$ is the $^{137}$Cs activity concentration in sample (Bq kg$^{-1}$);  
$m_s$ is the sample mass per area

The $^{137}$Cs inventory in cores from the Continental Shelf of São Paulo State varied from 0.3 to 30.6 Bq m$^{-2}$, with a mean value of 13±7 Bq m$^{-2}$. In addition to this region not having any significant source of radioactive pollution, these values are related to fallout from global nuclear tests in the atmosphere carried out in the past (1950s and 1960s). Figure 2 shows the $^{137}$Cs inventory for each core collected in this region.
Table 1. $^{137}$Cs concentration activity, in Bq kg$^{-1}$, according to depth from cores collected in São Paulo State Continental Shelf.

<table>
<thead>
<tr>
<th>Depth (cm)</th>
<th>6669(1)</th>
<th>6678(1)</th>
<th>6735(1)</th>
<th>6651(2)</th>
<th>6654(2)</th>
<th>6683(2)</th>
<th>6692(2)</th>
<th>6704(2)</th>
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</table>

(1) Standard deviation of 9.0% (GEM 50P) is related to counting efficiency.
(2) Standard deviation of 8.0% (GMX 25910P) is related to counting efficiency.

4.1. Minimum detectable concentration (MDC)

When a sample is measured by Gamma Spectrometry, the term usually associated with detection limits is minimum detectable concentration (MDC), which can be expressed by Equation 1 [10]:

$$\text{MDC} = \frac{4.66 \times s_b}{\varepsilon \times p_\gamma \times W}$$

(1)

where,

- $s_b$ is the estimated standard error of the net count rate;
- $\varepsilon$ is the counting efficiency of the specific energy of the nuclide;
- $p_\gamma$ is the absolute transition probability by gamma decay through the selected energy
- $W$ is the sample mass

In this study, MDC values, calculated for both detectors, GEM 50P and GMX 25910P, were 0.1 and 0.3 Bq.kg$^{-1}$, respectively.
The values obtained in this study are in agreement with Figueira et al.[11]. Levels on the Brazilian southeastern coast are lower than those in other world regions that are subject to the release from nuclear or reprocessing plants or even from Chernobyl’s accident [12,13].

5 CONCLUSION

$^{137}$Cs levels obtained from the southeastern coast were those expected in the Southern Hemisphere as the main source of artificial radioactivity is the atmospheric fallout from past nuclear tests. The possibility of $^{137}$Cs measurement in sedimentary columns has made this radionuclide an important tool to determine sedimentation rates for coastal areas and the continental shelf. This survey enabled the creation of a database for $^{137}$Cs levels on the Brazilian southeastern coast and any alterations in the future will be attributed to a specific cause.

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REFERENCES


