

**APPLICATION OF RADIOCHEMICAL METHODS AND DISPERSION  
MODEL IN THE STUDY OF ENVIRONMENTAL POLLUTION IN BRAZIL**

Ieda I.L.Cunha, Rubens C.L.Figueira and Roberto T.Saito

Instituto de Pesquisas Energéticas e Nucleares - IPEN-CNEN/SP

Radiochemistry Supervision

Caixa Postal 11049 - CEP 05422-970 - Pinheiros - São Paulo - SP - Brazil

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**ABSTRACT**

Methodologies for analysis of anthropogenic and natural radionuclides in marine samples were developed and applied in environmental samples. Results of systematic measurements of these radionuclides have showed that artificial radioactivity levels are in agreement with the values from the regions not affected directly by nuclear accidents or nuclear reprocessing plant discharges and are due to the global fallout. The average concentration of Cs-137 is of  $1.4 \text{ Bq.m}^{-3}$  in seawater, ranges from 13 to  $220 \text{ mBq.kg}^{-1}$  in fish, and from  $0.4$  to  $1.8 \text{ Bq.kg}^{-1}$  for sediments. Sr-90 levels in seawater are of  $1.8 \text{ Bq.m}^{-3}$  and in fish vary from 19 to  $75 \text{ mBq.kg}^{-1}$ . Sediments present concentrations of Sr-90 lower than  $0.8 \text{ Bq.kg}^{-1}$  and for Pu-239+240 of  $0.03$  to  $0.18 \text{ Bq.kg}^{-1}$ . Po-210 levels in fish range from  $0.5$  to  $5.3 \text{ Bq.kg}^{-1}$ . The data generated represent reference values for our country and are used to estimate the intake levels of these radionuclides by consuming of marine products.

Fax number: 55 11 8169188, e-mail: [figueira@net.ipen.br](mailto:figueira@net.ipen.br)

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## **INTRODUCTION**

In the last years there has been an increasing interest in artificial and natural radionuclides present in the environment and their possible effects on human health. The radionuclides can be transported across long distances from their source of emission, removed from the atmosphere, deposited in the biosphere and hydrosphere and enter to the human population by several pathways and one of them is the marine environment.

Considering these problems and the lack of information concerning our country, monitoring programs have been carried out in order to obtain the radionuclide levels in marine samples. Radiochemical methods for precise determinations of Cs-137, Sr-90, Pu-239+240 and Po-210 in seawater, fish and sediment have been already developed. Marine samples are been collected at different locations from the Brazilian coast.

A study of Cs-137 radionuclide dispersion in surface water was also developed. The Ocean Model simulates the surface water contamination caused by routine or accidental releases. For the model simulation, it was applied in the North Sea, based on the published transfer coefficient data. Results obtained show that the model provides a good response to evaluate the radionuclide dispersion in the marine environment[1].

This work is part of a research programme that aims at providing technology for reliable assessment of radionuclide contamination, at ensuring the capability of our laboratory to perform radionuclide analysis present in low concentration in environmental samples and at maintaining a data base of radionuclide levels.

## **EXPERIMENTAL**

### **1. Sampling and sample preparation**

Surface seawater and fish were sampled at fixed stations from the Brazilian coast since Rio Grande do Sul (latitude 32° 11'S) to Para State ( latitude 00° 26'S).

After sampling, seawater samples were acidified to pH 1 by adding conc. hydrochloric acid and then stored in 20 L polyethylene containers. Cesium-137 was preconcentrated from 100 liters of sample using ammonium phosphomolybdate (AMP). The precipitate was decanted overnight. After filtering and drying, the precipitate was transferred to a plastic pot [2].

For fish analysis, the edible part (about 1.5 kg) was weighed, dried at 110° C for one week and later on the temperature was gradually increased up to 450° C, until ashes were obtained. Ashes were weighed in plastic pots (sample height: 3.0cm). The main fish consumed by our population, such as sardine, weak fish mullet, saw fish, lane snapper, atlantic croaker and white grunt were analyzed. Fish samples were purchased from fishermen's cooperative associations, that know the exact points of collecting.

Marine sediments were collected at the water - sediment boundary at several characteristic depths. Samples were dried at 60° C, homogenized, sieved, weighed and transferred to plastic pots.

## **2. Equipment**

Hyperpure Ge detector (ORTEC, GEM 60120P Model), resolution of 1.9 keV at 1332.40 keV photopeak of Co-60. The software of the data acquisition was MAESTRO II. A detector calibration was performed using several gamma rays emitting nuclides, specially Cs-137 (661.6 keV) and Co-60 (1332.4 keV). To determine the detector counting efficiency in the Cs-137 photopeak region, the IAEA-375 reference material was employed, being obtained the value  $1.923 \pm 0.013$  %, by counting the sample in a standard plastic pot with 3.0 cm of height.

Dual alpha spectrometer, surface barrier Si detector (ORTEC 576, 450 RV Model), efficiency 30%.

Low level Geiger-Müller multi counter system (Model RISO GM-25-5), gas flow, efficiency 28% to Y-90.

### 3. Background Radiation

The environmental samples present low activity, so any background radiation causes influence in the detection of the radionuclide. A study of the background radiation variation in the region of Cs-137 photopeak, in function of the time was performed [3].

By MAESTRO II software, it was possible to record the background counting in intervals of 10 000 to 200 000 seconds, in a total time of counting that varied from 70 000 to 250 000 seconds. The counting varied according to Cs-137 level in the sample, in order to improve counting statistics.

The gamma spectrum was smoothed according to a binomial algorithm, which replaces the original data, channel by channel, with the smoothed data as follows:

$$S_i = \sum_{j=1}^5 C_j \cdot O_{i-3+j} \quad (1)$$

where:

$S_i$  are smoothed data in the channel  $i$ ;

$C_j$  is smoothing coefficient;

$O_i$  are original data in channel  $i$ .

It was noticed that the ideal number of channels for the analysis was eleven. The Cs-137 photopeak was fixed to be positioned in the channel, from 2641 to 2651, and any variation in the channel versus energy function could lead to error in the analysis (change in the photopeak of Cs-137). Therefore, the spectrum must be often recalibrated to the channel position previously specified.

## 4. Analysis Methods

### 4.1 Cesium-137 Analysis

For all the matrices studied, cesium-137 was assayed by gamma counting as described below [3].

Standard plastic pots containing the sample were counted in the Ge detector. Accumulative countings were recorded in intervals of 10 000 seconds, being that the total time of counting ranged from 25 000 to 150 000 seconds. The Cs-137 activity was calculated according to the following equation:

$$(A_{Cs} \pm \sigma_{Cs}) = \frac{(C_S \pm \sigma_S) - (C_{BG} \pm \sigma_{BG})}{m_A \cdot t \cdot (\varepsilon \pm \sigma_\varepsilon)} \cdot 1000 \quad (2)$$

where,

$A_{Cs}$  is the Cs-137 activity in the sample ( $Bq \cdot kg^{-1}$ );

$\sigma_{Cs}$  is the standard deviation of Cs-137 activity in the sample;

$C_S$  is the Cs-137 gross counting in the sample;

$\sigma_S$  is the standard deviation of Cs-137 counting in the sample;

$C_{BG}$  is the background counting;

$\sigma_{BG}$  is the standard deviation of background counting;

$m_A$  is the mass of sample (g);

$t$  is the counting time (s);

$\varepsilon$  is the counting efficiency to the Cs-137 photopeak (661.6 keV).

In order to verify if the method here developed is adequate to the Cs-137 analysis, it was applied to reference materials. Table 1 presents the results in the Cs-137 analysis in reference samples. Samples containing Cs-137 level of about  $0.8 Bq \cdot kg^{-1}$  present a relative error of 23%, but decreases to 0.6% as Cs-137 level reaches

50 Bq.kg<sup>-1</sup>. The method showed good precision and accuracy and the methodology is favourable to routine monitoring purposes, specially in our case, as small photopeaks appear in the gamma spectrum.

#### 4.2 Strontium-90 Analysis

To 50 liters of seawater, 6.0 g of strontium carrier (strontium nitrate), strontium-85 tracer, 500 g of ammonium chloride and 875 g of sodium carbonate were added, in order to precipitate, strontium, calcium and only a little amount of magnesium carbonate.

The supernatant was retired by suction and discarded, the strontium carbonate was dissolved with conc.nitric acid. Further, for the elimination of the calcium interference, sulphuric acid ( 10% of solution volume) was added.

The strontium sulphate was filtered and converted to carbonate, by addition of 60g of sodium carbonate. The supernatant was retired by suction and discarded.

The strontium carbonate was dissolved with conc.nitric acid. Iron carrier (10 mg of Fe) was added and the pH was increased up to 7 in order to precipitate ferric hydroxide. The retention of strontium in the hydroxide was lower than 15%.

To the strontium present in the supernatant 60 g of sodium carbonate were added, the strontium carbonate was stored by 14 days in order to attain the radioactive equilibrium. The separation of Sr-90 from Y-90 was carried out in several steps.

Strontium carbonate was dissolved with conc.nitric acid and yttrium carrier (10 mg of Y) was added. The solution was heated by fifteen minutes. The yttrium was precipitated as hydroxide, by addition of conc.ammonium hydroxide (pH 8). After filtering, the precipitate was dissolved with conc.hydrochloric acid, in the presence of 10 mg of strontium as hold back carrier. The yttrium was again precipitated as hydroxide, as here described for three times.

Finally, the yttrium hydroxide was dissolved with conc. hydrochloric acid, and 3 ml of the solution 1 M oxalic acid (pH 1.5) were added. The yttrium oxalate was assayed by beta counting, that was corrected by building up and decay of Y-90, counter background, detector efficiency, strontium and yttrium yields and blank value in order to obtain the Sr-90 activity. Analysis of the blank was performed simultaneously to the seawater analysis [4].

About 20 to 50 g of muscle and bone ashes (1 to 2 kg of fish wet) and 50 g of sediment were taken for the analysis. Then, 20 mg of strontium carrier per gram of sample, strontium-85 tracer, 100 mL of 8M nitric acid and drops of hydrogen peroxide were added. The leaching was carried out for 16 hours [5].

The residue was separated by filtration. The other radiochemical steps of the analysis are the same above described for seawater analysis.

#### **4.3 Po-210 Analysis**

Samples were dissolved by using 8 M nitric acid and hydrogen peroxide by 20 hours, in the presence of 20 mg lead carrier and Po-208 tracer. The nitric acid was removed by evaporating the solution and adding hydrochloric acid. The residue was dissolved in 30 mL of 0.5 M hydrochloric acid, 300 mg of ascorbic acid was added. The solution was transferred into plating cell (disposable plastic tube), which contains a silver disc (20 mm diameter). This cell was placed in water bath at 70-90° C, under mechanical agitation, and the plating time was 6-8 hours. The Po-210 was counted by alpha spectrometry.

The lead and polonium recoveries were calculated by gravimetric analysis and Po-208 tracer, respectively. Analysis of the blank has also been runned periodically [6].

#### 4.4 Plutonium-239+240 Analysis

The method of analysis established in this work consists of lixiviation of sediment sample (8 M nitric acid and hydrogen peroxide in the presence of plutonium tracer by 20 hours), ferric hydroxide coprecipitation, dissolution in 8M nitric acid and addition of sodium nitrite, purification by an anionic exchange column (Bio RAD AG1-X8, 100-200 mesh, 8 M nitric acid medium); electrodeposition in sulphuric medium onto a polished stainless steel disc at pH 2.0, and constant current of 1.8 ampere for 75 minutes, counting by alpha spectrometry.

### RESULTS AND DISCUSSION

The methods developed for Sr-90, Po-210 and Pu-239+240 were applied to the reference materials from the International Atomic Energy Agency (IAEA). Results obtained are presented in the Tables 2, 3 and 4. Results show a good accuracy for the methods here established.

Cs-137 levels in Brazilian coastal seawaters ranged from 0.8 to 1.7 Bq.m<sup>-3</sup> and are presented in Table 5 [2]. Cs-137 has been monitored by the European Community Member States and the Gesamp Report [7] gathered data relative to the level of this radionuclide in seawater and fish over the period from 1980 to 1985. Levels in Pacific Ocean varied to 0.3 to 4.0 Bq.m<sup>-3</sup> (Antartic), in the Indian Ocean is of about 3.0 Bq.m<sup>-3</sup>. The Atlantic Ocean presents values of 2.4 (W Central), 2.9 (N.W) up to 7.6 Bq.m<sup>-3</sup> in the Artic. Areas influenced by input from reprocessing plants and Chernobyl accident contain higher concentrations. Mediterranean (5.4 Bq.m<sup>-3</sup>), Black sea (52 Bq.m<sup>-3</sup>), Irish Sea (55 Bq.m<sup>-3</sup>), Danish Straits (73 Bq.m<sup>-3</sup>) and Baltic Sea (125 Bq.m<sup>-3</sup>) [8].

Cs-137 concentrations in Brazilian fish varied from 0.01 to 0.22 Bq.kg<sup>-1</sup> [9] while the values for European countries ranged from about 0.1 to 13.0 Bq.kg<sup>-1</sup> (wet), the higher levels being found in the Baltic and Irish Sea.

Concentrations of Cs-137 in Brazilian environmental samples are very low and its analysis by an instrumental method, without performing the procedures of detector recalibration, smoothing and accumulative countings, is impracticable or presents errors so high as 200 % . By carrying out such procedures, it was possible to analyse Cs-137 levels as low as  $0.8 \text{ Bq.kg}^{-1}$  with a standard deviation of 14%.

Cs-137 levels obtained for seawater and fish were used to calculate the dose received by the population from consumption of fish. The dose values varied from 0.8 to  $31.7 \text{ nSv.y}^{-1}$ , and were low compared to the values recommended by the ICRP for the limit of annual dose to members of the public ( $1 \text{ mSv.y}^{-1}$ ) [9].

Sr-90 levels in Brazilian Seawater is of  $1.8 \pm 0.3 \text{ Bq.m}^{-3}$ . This concentration is typical value due to fallout deposition and it is lower compared to areas influenced by input from discharges of nuclear plants or Chernobyl accident, such as Cap La Hague [10] ( $29 \text{ Bq.m}^{-3}$ ), Yellow Sea [11] ( $14 \text{ Bq.m}^{-3}$ ) and Baltic Sea [12] ( $17.6 \text{ Bq.m}^{-3}$ ).

Sr-90 levels in Brazilian coastal fish varied from 3 to  $12 \text{ mBq.g}^{-1}$  of Ca (Table 6) for bone and 19 to  $75 \text{ mBq.kg}^{-1}$  for muscle. These concentrations are typical values due to fallout for the southern hemisphere and are lower when compared to others regions of the world such as Sellafield [13], ( $230$  to  $320 \text{ mBq.kg}^{-1}$ ), Baltic Sea [12] ( $20$  to  $410 \text{ mBq.kg}^{-1}$ ), Cap la Hague [10], ( $130 \text{ mBq.kg}^{-1}$ ) and Yellow Sea [11], ( $370 \text{ mBq.kg}^{-1}$ ), for fish muscles, and Baltic Sea [12] ( $8.3$  to  $27 \text{ mBq.g}^{-1}$  of Ca), and Japan Sea [14], ( $2$  to  $100 \text{ mBq.g}^{-1}$  of Ca), for fish bones, being these areas influenced by input from discharges of nuclear reprocessing plants or Chernobyl accident.

In Brazil, available data on Po-210 in fish are very sparse. This radionuclide is concentrated by most marine organisms and in several cases provides the major natural dose for marine organisms. The methods here developed were applied in fish from both Mediterranean (Monaco) and Brazilian coast. Po-210 levels in fish from Monaco varied

from 0.7 to 1.5 Bq.kg<sup>-1</sup> of edible part and in Brazil from 0.5 to 5.3 Bq.kg<sup>-1</sup> (wet) (Table 7).

According to the Mardos Project [8], data available for polonium show that there are no significant differences in concentration from one ocean to another. The uncertainty in Po-210 concentration is high, since the methods are complex and only a few international intercalibration exercises have been performed. A global concentration for this radionuclide is of 2.4 Bq.kg<sup>-1</sup> in fish.

Radionuclides analyses in Brazilian sediments presented levels of 0.4 to 1.8 Bq.kg<sup>-1</sup> for Cs-137, lower than 0.8 Bq.kg<sup>-1</sup> for Sr-90 and about 0.03 to 0.18 Bq.kg<sup>-1</sup> for Pu-239+240. Table 8 presents the levels of Cs-137, Sr-90 and Pu-239+240 in marine sediments in different regions of the world [15-19].

## CONCLUSIONS

In this work, levels of artificial radionuclides in marine samples are in agreement with the values from regions not affected directly by nuclear accidents or nuclear reprocessing plant discharges and can be considered reference levels to Brazil. The anthropogenic radionuclide levels in Brazilian marine samples are due to global fallout, where the radionuclides released from atomic explosions were distributed over the world through atmospheric currents.

Relative to Po-210 analyses, the background of the natural radioactivity in Brazilian marine samples has not been routinely studied. So, the levels here published represent reference values for our country. Assessment of Po-210 concentrations in environmental samples is very important to determine its contribution to the radiation background as well as to estimate the intake levels of this radionuclide by consumption of marine products.

Data of artificial and natural radionuclide levels are used to calculate the doses received by the Brazilian population due to the radiation from marine food chain.

The methods here developed can be applied in another research fields, such as dating of sediments, sediment accumulation rate and atmospheric cycling studies.

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## TABLES

**Table 1 - Analysis of Cs-137 in reference materials, in Bq.kg<sup>-1</sup>.**

Reference Material	Certified Value	Obtained Value	S.D. <sup>(1)</sup> (%)	Error <sup>(2)</sup> (%)
Soil-6	53.65 (51.43-57.91)	54 ± 1	1.8	0.6
IAEA-307	4.9 (4.5 - 5.2)	4.7 ± 0.5	10.6	4.1
IAEA-352	2.7 (2.5- 2.8)	3.3 ± 0.5	15.1	22.2
IAEA/SD-N-2	0.8 (0.5-1.0)	0.61 ± 0.09	14.7	23.8

(1) relative standard deviation

(2) relative error

**Table 2 Sr-90 levels in reference materials.**

Air Filter (IAEA-083) <sup>(a)</sup>	Sr yield (%)	Y yield (%)	Sr-90 Activity
Mean Value and Standard Deviation	94 ± 4	92.8 ± 7	226 ± 18
Certified Value (Reference date:01/01/86)			231 (222-241)
Marine Sediment (IAEA-367) <sup>(b)</sup>			.
Mean Value and standard Deviation	89 ± 5	100	118 ± 25
Certified Value (Reference date:01/01/90)			102 (62-129)
Soil ( Soil-6) <sup>(b)</sup>			
Mean Value and standard Deviation	77 ± 6	99 ± 2	46 ± 6
Certified Value (Reference date: 01/30/83)			30.34 (24.2-31.67)

(a) activity in Bq.filter<sup>-1</sup>

(b) activity in Bq.kg<sup>-1</sup>

**Table 3 - <sup>210</sup>Po levels in reference materials**

Reference Material	Yield (%) (range)	<sup>210</sup> Po (Bq.kg <sup>-1</sup> )*	Certified Value (confidence interval)
IAEA-300	44 - 63	342.9 ± 36.1	360 (339 -395)
IAEA-307	40 - 46	56.1 ± 5.4	58.5 (40 - 91)
IAEA-308	50 - 70	82.0 ± 8.7	73 (66 - 75)

\* Values corrected to the reference date.

Table 4 - Pu-239+240 levels in reference materials

Sample	Mass (g)	Certified Value	Activity (Bq.kg <sup>-1</sup> )
IAEA-300 (marine sediment)	3	3.55 (3.44 - 3.65)	3.6 ± 0.6 (n=6)
IAEA-367 (marine sediment)	1.3	38 (34.4 - 39.8)	40 ± 1 (n=6)
IAEA-368 (marine sediment)	1.8	31 (29 -34)	27 ± 2 (n=2)
IAEA-375 (marine sediment)	10.6	0.299 ( 0.245 - 0.339)	0.34 ± 0.04 (n=1)
Soil-6 (soil)	10.5	1.04 ( 0.962 - 1.110)	1.2 ± 0.3

\* Values corrected to the reference date.

Table 5 - Cesium-137 levels in Seawater ( Bq.m<sup>-3</sup>)

State	Latitude	Longitude	Annual Average 1992 year	Annual Average 1993 year	Annual Average 1994 year
Rio G. Sul	32° 11'S	52° 02'W	1.1 ± 0.1	0.8 ± 0.2	1.0 ± 0.1
Paraná	25° 37'S	48° 16'W	1.2 ± 0.1	1.2 ± 0.1	1.4 ± 0.1
São Paulo	23° 00'S	44,5° W	1.4 ± 0.1	1.2 ± 0.1	1.2 ± 0.1
Rio de Janeiro	22° 57'S	43°55'W	1.0 ± 0.1	0.8 ± 0.2	1.0 ± 0.1
Bahia	12° 57'S	38° 32'W	1.4 ± 0.1	1.7 ± 0.1	1.7 ± 0.1
Pernambuco	08° 02'S	34°51'W	1.4 ± 0.1	1.6 ± 0.1	1.7 ± 0.1
Ceará	03° 42'S	38°29'W	1.6 ± 0.1	1.4 ± 0.1	1.5 ± 0.1
Pará	00° 26'S	47° 49'W	1.0 ± 0.1	1.1 ± 0.1	1.0 ± 0.2

Table 6 - Data obtained in the analyses of Brazilian coastal fish bone samples. (date of sampling: August and September 1991)

Fish (Specie)	Region	Sr yield (%)	Y yield (%)	Sr-90 mBq.g <sup>-1</sup> ash	Activity mBq.g <sup>-1</sup> Ca
<i>Mugil liza</i>	Belém	90.1 ± 0.8	96 ± 6	8 ± 2	3 ± 1
<i>Scomberomorus bras.</i>	Recife	83.1 ± 0.3	86 ± 5	42 ± 9	12 ± 3
<i>Mugil liza</i>	Itacuruçá	86.4 ± 0.8	94 ± 5	18 ± 7	5 ± 2
<i>Cynosciium sp.</i>	Paranaguá	90.1 ± 0.8	100 ± 6	42 ± 11	11 ± 3

**Table 7 -  $^{210}\text{Po}$  levels in fish samples (date of sampling: July 1994)**

<b>Fish</b>	<b>Mass (g)</b>	<b>Yield (%)</b>	<b><math>^{210}\text{Po}</math> (Bqkg<sup>-1</sup>)</b>
<i>Cynoscium sp</i>	23.7	47.7	3.1 ± 0.2
<i>Cynoscium sp</i>	19.5	48.5	1.4 ± 0.1
<i>Bagre sp</i>	19.9	74.5	3.9 ± 0.2
<i>Sardinella braziliensis</i>	23.3	58.7	2.6 ± 0.2
<i>Caranx hippos</i>	12.5	61.3	5.3 ± 0.3
<i>Archosargus rhomboidalis</i>	19.3	63.3	0.51 ± 0.03
<i>Paralanchurus brasiliensis</i>	21.7	46.1	2.3 ± 0.1

**Table 8: Levels of anthropogenic radionuclides in Brazilian sediments samples compared to other regions of the world (Bq.kg<sup>-1</sup>).**

<b>Region</b>	<b>Cs-137</b>	<b>Sr-90</b>	<b>Pu-239+240</b>
Brazil	0.4 - 1.8	< 0.8	0.03 - 0.18
China[10]	14	11	-
United Kingdom [15]	0.2 - 408	-	0.044 - 373
Irish Sea [16]	7.7 - 179	-	0.012 - 1.4
Central Pacific [17]	40	1.5	73
Syria [18]	0.75 - 1.2	-	0.147 - 0.72
Black Sea [19]	124	-	10