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Application of radiochemical methods and dispersion model in the study of environmental pollution in Brazil

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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 ^{137}Cs is of 1.4 Bq·m^{-3} in seawater, ranges from 13 to 220 mBq·kg^{-1} in fish, and from 0.4 to 1.8 Bq·kg^{-1} for sediments. ^{90}Sr levels in seawater are of 1.8 Bq·m^{-3} and in fish vary from 19 to 75 mBq·kg^{-1}. Sediments present concentrations of ^{90}Sr lower than 0.8 Bq·kg^{-1} and for $^{239+240}$Pu of 0.03 to 0.18 Bq·kg^{-1}. ^{210}Po levels in fish range from 0.5 to 5.3 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.

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 ^{137}Cs, ^{90}Sr, $^{239+240}$Pu and ^{210}Po in seawater, fish and sediment have been already developed. Marine samples are been collected at different locations from the Brazilian coast.

A study of ^{137}Cs 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

Sampling and sample preparation

Surface seawater and fish were sampled at fixed stations from the Brazilian coast from 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: 3.0 cm).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.

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Equipment

Hyperpure Ge detector (ORTEC, GEM 60120P Model), resolution of 1.9 keV at 1332.40 keV photopeak of ^{60}Co. The software of the data acquisition was MAESTRO II. A detector calibration was performed using several gamma-rays emitting nuclides, specially ^{137}Cs (661.6 keV) and ^{60}Co (1332.4 keV). To determine the detector counting efficiency in the ^{137}Cs photopeak region, the IAEA-375 reference material was employed, being obtained the value 1.923±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 ^{90}Y.

Background radiation

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

It was possible by MAESTRO II software 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 ^{137}Cs 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:

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

where S_i are smoothed data in the channel i, C_j is the smoothing coefficient, and O_i are original data in channel i.

It was noticed that the ideal number of channels for the analysis was eleven. The ^{137}Cs 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 ^{137}Cs). Therefore, the spectrum must be often recalibrated to the channel position previously specified.

Analysis methods

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, the total time of counting ranged from 25 000 to 150 000 seconds. The ^{137}Cs activity was calculated according to the following equation:

$$(A_{\mathrm{Cs}}\pm\sigma_{\mathrm{Cs}})=\frac{(C_s\pm\sigma_s)-(C_{\mathrm{BG}}\pm\sigma_{\mathrm{BG}})}{m_A\cdot t\cdot(\varepsilon\pm\sigma_\varepsilon)}\cdot 1000 \qquad (2)$$

where A_{Cs} – ^{137}Cs activity in the sample ($\mathrm{Bq\cdot kg^{-1}}$), σ_{Cs} – standard deviation of ^{137}Cs activity in the sample, C_S – ^{137}Cs gross counting in the sample, σ_s – standard deviation of ^{137}Cs counting in the sample, C_{BG} – background counting, σ_{BG} – standard deviation of background counting, m_A – mass of sample (g), t – counting time (s), ε – counting efficiency to the ^{137}Cs photopeak (661.6 keV).

In order to verify if the method here developed is adequat for the ^{137}Cs analysis, it was applied to reference materials. Table 1 presents the results in the ^{137}Cs analysis in reference samples. Samples containing ^{137}Cs level of about 0.8 $\mathrm{Bq\cdot kg^{-1}}$ present a relative error of 23%, but decreases to 0.6% as ^{137}Cs level reaches 50 $\mathrm{Bq\cdot kg^{-1}}$. 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.

Strontium-90 analysis

To 50 l 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 removed by suction and discarded, the strontium carbonate was dissolved in conc. nitric acid. Further, for the elimination of the calcium interference, sulphuric acid (10% of solution volume) was added.

Table 1. Analysis of ^{137}Cs in reference materials (in $\mathrm{Bq\cdot kg^{-1}}$)

Reference material	Certified value	Obtained value	S.D.,[1] %	Error,[2] %
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.

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

The strontium carbonate was dissolved in 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 for 14 days in order to attain the radioactive equilibrium. The separation of ^{90}Sr from ^{90}Y was carried out in several steps.

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

Finally, the yttrium hydroxide was dissolved in conc. hydrochloric acid, and to 3 ml of the solution 1M oxalic acid (pH 1.5) was added. The yttrium oxalate was assayed by beta counting, that was corrected by building up and decay of ^{90}Y, counter background, detector efficiency, strontium ad yttrium yields and blank value in order to obtain the ^{90}Sr 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 as above described for seawater analysis.

Polonium-210 analysis

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

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

Plutonium-239+240 analysis

The method of analysis established in this work consists of lixiviation of sediment sample (8M 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, 8M 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 ^{90}Sr, ^{210}Po and $^{239+240}$Pu 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.

^{137}Cs levels in Brazilian coastal seawaters ranged from 0.8 to 1.7 Bq·m^{-3} and are presented in Table 5.[2] ^{137}Cs 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 from 0.3 to 4.0 Bq·m^{-3} (Antarctic), in the Indian Ocean it is of about 3.0 Bq·m^{-3}. The Atlantic Ocean presents values of 2.4 (W Central), 2.9 (N.W) up to 7.6 Bq·m^{-3} in the Arctic. Areas influenced by input from reprocessing plants and Chernobyl accident contain higher concentrations. Mediterranean (5.4 Bq·m^{-3}), Black Sea (52 Bq·m^{-3}), Irish Sea (55 Bq·m^{-3}), Danish Straits (73 Bq·m^{-3}) and Baltic Sea (125 Bq·m^{-3}).[8]

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

Concentrations of ^{137}Cs 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 ^{137}Cs levels as low as 0.8 Bq·kg^{-1} with a standard deviation of 14%.

Table 2. ^{90}Sr levels in reference materials

Sample	Sr yield, %	Y yield, %	^{90}Sr activity
Air filter (IAEA-083)[a]			
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)[b]			
Mean value and standard deviation	89 ± 5	100	118 ± 25
Certified value (reference date: 01/01/90)			102 (62–129)
Soil (Soil-6)[b]			
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^{-1}.
[b] Activity in Bq·kg^{-1}.

Table 3. ^{210}Po levels in reference materials

Reference material	Yield, % (range)	^{210}Po,* Bq·kg^{-1}	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. $^{239+240}$Pu levels in reference materials

Sample	Mass, g	Certified value	Activity,* Bq·kg^{-1}
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.

^{137}Cs 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 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 mSv·y^{-1}).[9]

^{90}Sr levels in Brazilian seawater is of 1.8± 0.3 Bq·m^{-3}. This concentration is a 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 Bq·m^{-3}), Yellow Sea[11] (14 Bq·m^{-3}) and Baltic Sea[12] (17.6 Bq·m^{-3}).

^{90}Sr levels in Brazilian coastal fish varied from 3 to 12 mBq·g^{-1} of Ca (Table 6) for bone and 19 to 75 mBq·kg^{-1} for muscle. These concentrations are typical values due to fallout for the southern hemisphere and are lower when compared to other regions of the world such as Sellafield,[13] (230 to 320 mBq·kg^{-1}), Baltic Sea[12] (20 to 410 mBq·kg^{-1}), Cap la Hague[10] (130 mBq·kg^{-1}) and Yellow Sea[11] (370 mBq·kg^{-1}), for fish muscles, and Baltic Sea[12] (8.3 to 27 mBq·g^{-1} of Ca), and Japan Sea[14] (2 to 100 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 ^{210}Po in fish are very scarce. 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. ^{210}Po levels in fish from Monaco varied from 0.7 to 1.5 Bq·kg^{-1} of edible part and in Brazil from 0.5 to 5.3 Bq·kg^{-1} (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 ^{210}Po 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^{-1} in fish.

Radionuclide analyses in Brazilian sediments presented levels of 0.4 to 1.8 Bq·kg^{-1} for ^{137}Cs, lower than 0.8 Bq·kg^{-1} for ^{90}Sr and about 0.03 to 0.18 Bq·kg^{-1} for $^{239+240}$Pu. Table 8 presents the levels of ^{137}Cs, ^{90}Sr and $^{239+240}$Pu in marine sediments in different regions of the world.[15–19]

Table 5. ^{137}Cs levels in seawater (in Bq·m^{-3})

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 by the analyses of Brazilian coastal fish bone samples. (Date of sampling: August and September 1991)

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

Table 7. ^{210}Po levels in fish samples. (Date of sampling: July 1994)

Fish	Mass, g	Yield, %	^{210}Po, Bq·kg^{-1}
Cynoscium sp.	23.7	47.7	3.1 ± 0.2
Cynoscium sp.	19.5	48.5	1.4 ± 0.1
Bagre sp.	19.9	74.5	3.9 ± 0.2
Sardinella braziliensis	23.3	58.7	2.6 ± 0.2
Caranx hippos	12.5	61.3	5.3 ± 0.3
Archosargus rhomboidalis	19.3	63.3	0.51 ± 0.03
Paralonchurus brasiliensis	21.7	46.1	2.3 ± 0.1

Table 8. Levels of anthropogenic radionuclides in Brazilian sediment samples compared to other regions of the world (in Bq·kg^{-1})

Region	^{137}Cs	^{90}Sr	$^{239+240}$Pu
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

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 ^{210}Po 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 ^{210}Po 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 field, such as dating of sediments, sediment accumulation rate and atmospheric cycling studies.

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