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Analysis of ^{210}Po in marine samples

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A method for determining ^{210}Po in marine samples by wet dissolution, deposition onto silver disc and counting by α-spectrometry is described. Recovery of polonium was obtained with ^{208}Po tracer. ^{210}Po levels in fish from the Brazilian coast and sediments from Antarctic region were determined. Levels in fish ranged from 0.5 to 5.3 Bq · kg^{-1} wet and for sediments, mean values of 10 mBq · kg^{-1} were obtained.

Introduction

^{210}Po is the decay product of ^{210}Pb ($T_{1/2}$ = 22.3 y) and it is an α-emitter with 138 days half-life. These radionuclides continuously enter the marine environment as daugther products of ^{222}Rn, which is released from the earth's crust to the atmosphere. ^{210}Po can be also introduced by radioactive decay of ^{226}Ra and ^{222}Rn that diffuse from the sediment. Part of it is scavenged from the water by particles and removed to the sediment.

^{210}Po is included in the group of most highly toxic radioisotopes and provides the major natural radiation dose for many marine organisms. Most of the ^{210}Po analysis methods are based on deposition onto a silver disc.[1–4]

In this work, a radiochemical procedure for ^{210}Po analysis in fish and sediments was developed. The accuracy of the method has been verified by the analysis of reference material from the IAEA.

Experimental

Reference material

Standard samples (IAEA-300 marine sediment, IAEA-307 sea plant, IAEA-308 seaweed) having know ^{210}Pb and ^{210}Po concentrations were obtained from the International Atomic Energy Agency (IAEA). The samples were dried at 90 °C by 12 hours before analysis.

Environmental samples

Fish samples of species most consumed by the population were purchased from markets in Sao Sebastiao (Northern Coast of Sao Paulo State, Brazil). Edible parts were used for the measurements.

Sediment samples from the Antarctic were collected during the period from December 1992 to January 1993, at different points of Admiralty Bay, as Thomas Pta., Ullman Pta. and Plaza Pta. Sediments were collected at the water – sediment boundary at several characteristic depths. Sampling was performed by the Oceanographic Institute of the Sao Paulo University. The samples were dried at 60 °C. After homogenisation, sieving and weighing, the samples were submitted to the radiochemical procedure for ^{210}Po analysis.

Radiochemical procedure

The samples were dissolved by using 8M nitric acid and hydrogen peroxide for 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, 300 mg of ascorbic acid was added. The solution was transferred into a plating cell (disposable plastic tube), which contains a silver disc (20 mm diameter). This cell was placed in a water bath at 70–90 °C, under mechanical agitation, and the plating time was of 6–8 hours. ^{210}Po was counted by alpha spectrometry.

The reminiscent solution of the plating was evaporated near dryness. Then, it was diluted with 8M nitric acid to a final volume of 20 ml. This solution was passed through an ion exchange column (Dowex Ag 1-X 8, 100–200 mesh, height 6 cm, diameter 1 cm, preconditioned with 50 ml of 8M nitric acid) at a flow rate of 0.03 ml · s^{-1}. The column was washed with 80 ml of 8M hydrochloric acid, and this solution was used for subsequent ^{210}Pb determination (time of separation is zero). The lead fraction was evaporated near dryness, dissolved in 25 ml of 0.5N hydrochloric acid and stored for 3 to 6 months, to obtain radioactive equilibrium. By carrying out another ^{210}Po deposition, it was possible to determine the activity of ^{210}Pb.

After plating, the lead solution was heated (90 °C) and concentrated sulphuric acid was added (10% v/v). Lead and polonium recoveries were calculated by gravimetric analysis and ^{208}Po tracer, respectively.

Analysis of the blank has also been runned periodically.

Results and discussion

The problems of this analysis method are related to the determination of radionuclides present in low con-

Table 1. ^{210}Po levels in reference material

Reference material	Yield, %	^{210}Po, Bq · kg^{-1}*	Certified value (confidence interval)
IAEA-300	44–63	342.9 ± 36.1	360 (339–395) Reference date: 01-01-93
IAEA-307	40–46	56.1 ± 5.4	58.5 (40–91) Reference date: 01-01-88
IAEA-308	50–70	82.0 ± 8.7	73 (66–75) Reference date: 01-01-86

*Values correct to the reference date.

Table 2. ^{210}Po levels in fish samples (sampling date: July/1994)

Fish	Mass, g	Yield, %	^{210}Po, Bq · kg^{-1}*
Cynoscion sp	23.7	47.7	3.1 ± 0.2
Cynoscion sp	19.5	48.5	1.4 ± 0.1
Bagre sp	19.9	74.5	3.9 ± 0.2
Sardinella brasiliensis	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

*Number of determination = 3.

Table 3. ^{210}Po levels in marine sediments (sampling date: January/1993)

Local	Yield, %	^{210}Po, Bq · kg^{-1}
Thomas Pta	70.9 ± 2.5	8.2 ± 0.5
Ullman Pta	67.5 ± 2.5	9.8 ± 1.9
Plaza Pta	74.4 ± 2.5	8.0 ± 0.1

*Number of determination = 3.

Table 4. Detection limits (LID) for the different matrices

Sample	LID	
IAEA-300	1.6 ± 0.1	Bq · kg^{-1}
IAEA-307	0.39 ± 0.01	Bq · kg^{-1}
IAEA-308	0.90 ± 0.03	Bq · kg^{-1}
Fish	22.7 ± 0.7	mBq · kg^{-1}
Marine sediment	87.9 ± 2.8	mBq · kg^{-1}

centration. Loss of polonium may occur by adsorption on glassware and by volatilisation (during the process of deposition, or taking the sample near dryness). It was verified that an efficient deposition depends on elevated temperature, low volume of solution and silver disc rotation during deposition.

^{210}Po concentrations were determined in three reference materials from the IAEA. Mean values obtained for this radionuclide in the analysed samples are given in Table 1. Results obtained are in good agreement with the certified values. The precision was about 10%.

Table 2 presents the ^{210}Po levels in fish samples. The values ranged from 0.5 to 5.3 Bq · kg^{-1} and are reference values for the Brazilian samples. The chemical yield was between 46 to 75%. The error in the analysis was about 6%.

According to an Agency Report,[5] data available for polonium show that there are no significant differences in its 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.

The concentration of ^{210}Po in marine sediments from Antarctic region are reported in Table 3. The values varied from 8 to 10 Bq · kg^{-1}. In these sediments (more than 3 years old) ^{210}Po is in secular equilibrium with ^{210}Pb, and they present equal activities.

CARVALHO[6] measured ^{210}Pb–^{210}Po in bottom sediments in the Tagus Estuary, Portugal. Analysis of sediments from outside zone of phosphate waste releases showed that these radionuclide were in radioactive equilibrium. The ^{210}Po concentration in < 63 μm size fraction of sediments was of 68 ± 19 Bq · kg^{-1}. Sediments from Syria[7] present about 20 Bq · kg^{-1} of ^{210}Po and marine sediments from United Kingdom[8] are 1 to 57 Bq · kg^{-1}. The great variation of the ^{210}Po concentration in sediments is related to the level of organic material, sediment grain size, releases of wastes by the phosphate fertiliser industry, coprecipitation of lead with iron–manganese hydroxides in the fresh water-salt water mixing zone and efficiency of scavenging of soluble naturally occurring ^{210}Pb onto suspended matter.

Table 4 presents the detection limits (LID) (95% confidence level) for the studied matrices. LID

determination was based on a USNRC Report.[9] Data show that the radiochemical method here developed can be applied for low level ^{210}Po analysis.

Conclusions

The background of the natural radioactivity in Brazilian marine samples has not been routinely studied. Available data of ^{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 these organisms. The levels published here represent reference values for our country. The assessment of ^{210}Po concentrations in environmental samples is very important for determining its contribution to the radiation background as well as for estimating the intake levels of this radionuclide by consuming of marine products. Data of ^{210}Po are used to calculate the radiation doses received by the Brazilian population due to the radiation from marine food chain.

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