SHORT COMMUNICATION

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Analysis of ^{210}Pb and ^{210}Po in Brazilian foods and diets

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Radiochemical procedures for the analysis of ^{210}Pb and ^{210}Po in foods and diets are presented. Because of the low beta energy of ^{210}Pb, its analysis was based on a separation of the daughter radionuclide ^{210}Bi by precipitation of lead sulphate, ^{210}Bi ingrowing and beta counting of this nuclide. ^{210}Po analysis was based on wet dissolution of the sample, deposition onto silver disc and counting by alpha-spectrometry. Levels of these radionuclides in individual items and diets of selected university students were determined in order to evaluate the intakes of ^{210}Pb and ^{210}Po as well as the dose due to ingestion of foods and diets in São Paulo city.

Introduction

^{210}Pb and ^{210}Po radionuclides are included in the group of most highly toxic radioisotopes and provide the major natural radiation for the man. Consumption of food is usually the most important route by which natural radionuclides can enter the human organisms. Assessment of natural radionuclide levels in different foods and diets is important to estimate the intake of these radionuclides by man.[1–2] From the public health point of view, it is important to ensure the population that the contaminant levels on a diet do not exceed the permissible limits. So the aim of this paper was to establish radiochemical methods for ^{210}Pb and ^{210}Po analysis, to determine the levels of these radonuclides in the most common individual foods present in diets of São Paulo city as well as in diets of selected university students. Based on the results, the daily dietary intakes of these radionclides as well as the effective dose resulting from the consumption of diets were evaluated. Results were compared to other published data in different countries.

Experimental

Food samples

Food samples of most consumed by the population were purchased from markets in São Paulo city. The samples were washed with water, dried at 105 °C and weighed. The fruits and legumes were peeled before washing and drying. The radionuclides were analysed in ten categories of foods, consisting of marine organisms (fish and seafoods) tea, cereals, vegetables, macaroni, meat, leguminose, fruits, milk and root vegetables. The number of samples analysed for each variety of food was at least 3. About from 10 to 30 g of food were taken for analysis.

Diets

The diet samples were collected from selected university students, aged 20 to 23 years. The duplicate portion method was employed to study the radionuclide intake by the students.

The diets consisted of all the components of foods, including beverages (exception of drinking water), that were consumed during 24 hours. Samples were homogenized in a mixer, freeze-dried and powdered. About 10 g of the diet were taken up for the analysis. The sampling method was applied in diets collected in two days of the week. The samples were collected in September, l997.

Sample dissolution

Samples were dissolved by using a microwave digestor (open system, Star Cem Coorporation) using 8M nitric acid solution and hydrogen peroxide. The quantities of reagents, temperature, ramp time, hold time and the number of steps necessary to the total dissolution were established for every food and diet analysed.

Radiochemical procedures

^{210}Pb analysis: After food or diet dissolution, 10 mg of lead, bismuth, barium and strontium carriers were added. Lead was precipitated with concentrated sulphuric acid (10% vol) and heated. After decantation, filtration, the precipitate was converted to carbonate by addition of 1 g sodium carbonate at of 90 °C. The precipitate was dissolved in 8M nitric acid solution and lead precipitated as hydroxide, dissolved in 8M nitric acid solution and again precipitated as sulphate.

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After decantation and filtration, the precipitate was dried (infra red lamp) and weighed. After waiting of time to reach radioactive equilibrium, ^{210}Bi beta-counting was carried out by a low background Geiger-Müller detector (Risoe, Denmark, GM-25-5 Model). Lead recovery was calculated by gravimetric analysis. Blank analysis was also done for every kind of sample.

^{210}Po analysis: After sample dissolution, in the presence of 20 mg polonium carrier and ^{208}Po tracer, the nitric acid was removed by evaporating the solution and adding concentrated hydrochloric acid. This process was repeated several times. The residue was dissolved by 10M hydrochloric acid and diluted to 40 ml, final pH of 1.5–2.0, in the presence of 300 mg of ascorbic acid. The solution was transferred into a plasting cell (disposable plastic tube), which contains a silver disc (20 mm diameter). This cell was placed in a water bath at 80–90 °C, under mechanical agitation, and the plating time was of 6 hours. ^{210}Po was counted by alpha-spectrometry (Si detector, Ortec, 576 A Model). Polonium recovery was calculated by ^{208}Po tracer. Analysis of the blank has also been runned periodically.

Results and discussion

The analysis method described in this paper presented typical problems of the radionuclide determination in low concentration. Loss of radionuclides may occur by adsorption on glassware and by volatilization. But, this was overcomed by using the radionuclide tracer.

The main problems occurred in the steps of the sample dissolution and of the change of the acid medium (nitric acid to hydrocloric acid) due to the carbonization of any matrices such as rice, macaroni and bean. This was avoided by using the microwave digestor, that destroys the organic matter.

Radionuclides concentrations were determined in reference materials from the IAEA. Results obtained were in good agreement with the certified values. The precision and accuracy were about 10%.

^{210}Po analysis: The highest concentrations of ^{210}Po were obtained for marine organisms. The levels in muscles of fish varied from 0.17 to 6.7 Bq/kg and in seafood from 2.17 to 5.5 Bq/kg. The other groups of foods presented the following levels: tea (0.23–1.62 Bq/kg), cereals (0.3 Bq/kg), vegetables (0.07–0.31 Bq/kg), rice and macaroni (about 0.27 Bq/kg), leguminose (0.04–0.17 Bq/kg), meat (0.09–0.025 Bq/kg), fruits (0.05–0.07 Bq/kg), root vegetables (0.025–0.05 Bq/kg) and finally milk (0.02–0.07 Bq/kg). The error in the analysis was of about 10%.

The ^{210}Po levels found in Brazilian individual foods are in good agreement with the values published in the literature. According to PARFENOV,[3] the values are: tea (1.7 Bq/kg), vegetables (0.018–3.73 Bq/kg), meat (0.44 Bq/kg), cereals (0.04–0.4 Bq/kg) and seafoods (2 Bq/kg). Date of the MARDOS Project[4] shows a global medium concentration of ^{210}Po in fish muscle of 2.4 Bq/kg. CUNHA et al.[5] measured this radionuclide in various species of Brazilian fish (such as sardinella braziliensis, caranx hippos, cyanoscium sp, bagre sp, archosagus rhomboidalis) and the values ranged from 0.5 to 3.9 Bq/kg.

Intake and dose resulting from the ^{210}Po ingestion

Based on the official food consumption statistics,[6] the annual intake per capita was estimated by multiplying the ^{210}Po level present in the individual food by the amount of food consumed by year. Data were used to calculate the annual effective dose received by the population from food consumption. Doses were evaluated considering the dose conversion factor for ingestion exposure of ^{210}Po (Sv/Bq) and the f_1 value (fraction), recommended by the ICRP.[7] Data are presented in Table 1.

The ^{210}Po intake varied from 14.6 to 37.6 Bq/year (from 0.040 to 0.103 Bq/d), that is in good agreement with values obtained in other countries, such as: Argentine (0.05 Bq/d), USA (0.07 Bq/d) and Europe (0.04 to 0.4 Bq/d) determined by PARFENOV.[3]

In this paper, the annual effective dose varied from 8.7 to 22.6 μSv. The foods responsible for the highest doses were fish, cereals, milk and fruits. In spite that milk presents a low level of ^{210}Po, its contribution to the dose is high, compared to other foods, due to its high consumption. Fish also contribute significantly to the dose, its consumption in Brazil is low, but present high concentration of ^{210}Po.

^{210}Po analysis in diets

Table 2 summarizes the results on ^{210}Po levels in diets. The ^{210}Po concentration in Brazilian diets varied from 25 to 85 mBq/d while the annual effective dose ranged from 5.5 to 18.7 μSv. These values present good agreement with the obtained data for the individual foods, showed in Table 1.

KRANDEKAR[8] has estimated the ^{210}Po intake in diets consumed by the population, of Bombay India, as 56 mBq/d, and the author concluded this value is low compared to the values obtained in USSR and Germany (152 and 170 mBq/d, respectively), but comparable to Argentine (48 mBq/d).

Table 1. Intake and effective dose by the consumption of ^{210}Po

Groups of foods	^{210}Po level (range), Bq/kg	Annual food consumption per capita, kg/year	Intake, Bq/year	Dose range, µSv/year
Fish and seafood	0.17–6.73	2.446	0.42–16.46	0.25–9.88
Tea	0.23–1.62	0.078	0.02–0.13	0.01–0.08
Cereal	0.3	25.845	7.75	4.65
Vegetable	0.1–0.3	3.213	0.32–0.96	0.19–0.58
Macaroni	0.15–0.25	4.006	0.60–1.00	0.36–0.60
Meat	0.025	12.734	0.32	0.19
Pork meat	0.085	1.302	0.11	0.07
Leguminose	0.04–0.17	7.762	0.31–1.32	0.19–0.79
Fruits	0.05–0.07	44.581	2.23–3.12	1.33–1.87
Milk	0.02–0.07	72.782	1.46–5.09	0.87–3.06
Root vegetables	0.025–0.05	14.207	0.36–0.71	0.22–0.43
Chicken meat	0.04	16.597	0.66	0.40
			Total=from (14.6±1.5) to (37.6±3.6)	Total=from (8.7±0.9) to (22.6±2.3)

Table 2. Intake and effective dose due to the consumption of ^{210}Po

Student	Diet	Diet dry weight, g	^{210}Po intake, mBq/diet or mBq/day	^{210}Po effective dose, µSv/year
1	1	126.0	48.7 ± 4.7	10.7 ± 1.0
	2	322.9	52.1 ± 5.8	11.4 ± 1.3
2	1	320.9	73.7 ± 7.1	16.2 ± 1.6
	2	248.4	85.2 ± 10.6	18.7 ± 2.3
3	1	275.9	66.6 ± 9.0	14.6 ± 2.0
	2	204.9	32.6 ± 3.3	7.1 ± 0.7
4*	1	441.9	52.0 ± 4.9	11.4 ± 1.1
5	1	359.9	53.6 ± 4.7	11.7 ± 1.0
	2	317.3	40.0 ± 3.8	8.8 ± 0.8
6	1	215.7	25.3 ± 2.2	5.5 ± 0.5
	2	308.1	31.8 ± 2.8	7.0 ± 0.6

* Diets of two days were mixed.

Table 3. Intake and effective dose resulting from the ingestion of ^{210}Pb

Food	^{210}Pb level range, Bq/kg	Annual consumption, kg	Intake range, Bq/year	Effective dose range, µSv/year
Fish and seafood	1.3–1.8	2.446	3.2–4.4	0.4–0.6
Root vegetables	0.15–1.8	14.207	2.1–25.6	0.3–3.5
Tea	0.5–0.6	0.078	0.04–0.05	0.005–0.007
Vegetables	0.15–0.20	3.213	0.48–0.64	0.07–0.08
Leguminose	0.11	7.762	0.85	0.1
Fruits	0.10	44.581	4.5	0.6
		Total	From 11.2±3.5 to 35.9±8.5	From 1.5±0.5 to 4.9±1.2

^{210}Pb analysis

The highest ^{210}Pb levels were observed in fish and seafood (1.3 to 1.8 Bq/kg) followed by root vegetables (0.15 to 1.8 Bq/kg), vegetables (0.15 to 0.2 Bq/kg) and tea (0.5 to 0.6 Bq/kg), leguminose (0.11 Bq/kg) and fruits (0.10 Bq/kg). Meat, lentil, apple, macaroni, milk, rice, tomato and potato present ^{210}Pb levels lower than the detection limit. Literature values are: fish (0.03 to 0.54 Bq/kg),[2] vegetables (1.4 to 2.4 Bq/kg), and root vegetables (0.4 to 1.8 Bq/kg).[9]

Table 3 shows the ^{210}Pb intake as well as the effective dose resulting from the food ingestion. The ^{210}Pb intake varied from 11.2±3.5 to 35.9±8.5 Bq/year. The effective dose for food ingestion ranged from 1.5± 0.5 to 4.9±1.2 μSv/year. The highest doses were observed in marine organisms, root vegetables and fruits.

^{210}Pb levels in diets

The diets were collected in 1997, kept in plastic pots and frozen. They were analysed in 1999, so ^{210}Pb and ^{210}Po reached the radioactive equilibrium. The activities and intake obtained for ^{210}Po (25 to 85 mBq/d) are equal to ^{210}Pb. Comparing the ^{210}Pb intake values due to the consumption of marine organisms (from 0.48 to 0.69 Bq/day)[2] by the Japanese population that consumes great quantities of these products with the values here obtained (about 0.01 Bq/day), it can be concluded that the Brazilian population is submitted to a lower dose than the Japanese population.This can be also verified with the worldwide measurements of ^{210}Pb dietary intake summarized by HOLTZMAN,[9] that ranged from 52 mBq/d to 1.48 Bq/d. The effective dose for ^{210}Pb resulting from diet intake varied from 1.3 to 4.3 μSv/year, corresponding to 25% of the ^{210}Po dose.

Conclusions

The methods developed in this paper for ^{210}Po and ^{210}Pb determination allowed the analysis of these radionuclides in low concentration and it was possible to evaluate that the levels obtained were lower than the established values by the UNSCEAR.[10]

The highest concentrations of ^{210}Po were observed in marine organisms, tea, cereal and vegetables. ^{210}Po intake due to the food consumption (from 40 to 103 mBq/day) is in agreement with the values obtained for diet (from 25 to 85 mBq/day). This was also noticed for the effective dose that ranged from 8.7 to 22.6 (μSv/year) for foods and from 5.5 to 18.7 (μSv/year) for the diet. For ^{210}Pb levels in Brazilian foods, the highest concentrations were found in fish, seafoods, legumes, tea and vegetables.

Considering both radionuclides, ^{210}Po and ^{210}Pb, the effective dose ranged from 10.2 to 27.5 μSv/year for foods and from 6.8 to 23.0 μSv/year for diets. These values are lower than the annual medium value estimated by the UNSCEAR which is 60 μSv.[10]

Values here obtained of ^{210}Pb and ^{210}Po dietary intakes for university students of São Paulo city are similar to the worldwide measurements published in the literature.

This paper presents a great contribution to the public healt of the area. The assessment of natural radionuclides in foods and diets allowed to evaluate the items that present the highest risks to the population and if the natural radionuclide intakes are lower than the limits established by the UNSCEAR. In Brazil, these data are very scarse.

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References

1. M. B. COOPER, P. C. CLARK, W. ROBERTSON, I. R. MCPHARLIN, R. C. JEFFREY, J. Radioanal. Nucl. Chem., 194 (1995) 379.
2. M. YAMAMOTO, T. ABE, J. KUWABARA, K. KOMURA, K. UENO, Y. TAKIZAWA, J. Radioanal. Nucl. Chem., 178 (1994) 81.
3. Y. D. PARFENOV, At. Energy Rev., 12 (1974) 75.
4. International Atomic Energy Agency (IAEA) Sources of Radioactivity in Marine Environment and their Relative Contributions to Overall Dose Assessment from Marine Radioactivity (Mardos Project), (IAEA-MEL-R2/94) Vienna, 1994.
5. I. I. L. CUNHA, A. I. SANTOS, R. T. SAITO, Annals of III ENAN Joint Nuclear Conferences, Brazil, 1995.
6. IBGE, Brazilian Institute of Geography and Statistics, Research of Familiar Budget, 1996.
7. International Commission on Radiological Protection (ICRP), Publication No. 72, 1996.
8. R. N. KRANDEKAR, Health Phys., 33 (1977) 149.
9. R. B. HOLTZMAN, J. Radioanal. Nucl. Chem., 115 (1987) 628.
10. United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). Sources and Effects of Ionizing Radiation, United Nations Report, New York, USA, 1993.