Journal of Radioanalytical and Nuclear Chemistry, Articles, Vol. 157, No. 1 (1992) 37–45

MEASUREMENTS OF ^{210}Pb AND ^{210}Po IN JAPANESE HUMAN HAIR

M. YAMAMOTO,* Y. YAMAUCHI,* H. KAWAMURA,**
K. KOMURA,* K. UENO*

**Low Level Radioactivity Laboratory, Kanazawa University, Tatsunokuchi, Ishikawa 923-12 (Japan)*
***Division of Radioecology, National Institute of Radiological Science, 3609 Isozaki, Nakaminato, Ibaraki 311-12 (Japan)*

(Received January 7, 1991)

^{210}Pb and ^{210}Po in human hair have been measured to serve as an aid in order to estimate the dietary intake and body burden of these radionuclides of Japanese. The ^{210}Po concentrations found in 83 hair samples were ranging from 4.0 to 59.3 mBq/g with a mean (median) value of 18.2±12.2 (14.9) mBq/g as compared to the ^{210}Pb concentrations from 0.7 to 6.5 mBq/g with a mean (median) value of 2.3±1.1 (2.0) mBq/g. The ^{210}Po/^{210}Pb activity ratios (mean: 8.7±5.1, median: 7.1) were surprisingly higher compared with the available literature value of about 2. The high concentration of ^{210}Po in human hair of Japanese may be due to the ingestion of animal protein mainly in the form of seafood.

Introduction

^{210}Pb (22.3 y, β^-) and ^{210}Po (138.38 d, α) occur widely in nature as the radioactive family of ^{238}U-series, and especially alpha-ray emitting ^{210}Po makes a substantial contribution to the internal radiation dose to man. The measurement of this pair of radionuclides has, therefore, been made for a number of environmental samples such as air, water, foodstuffs and human tissues. The numerous data obtained[1–5] have provided information such as the level and distribution of these nuclides in the environment, daily dietary intake levels and hazard evaluation to man.

According to the 1977 UNSCEAR Report,[1] Japanese living on seafood may be expected to have a high amount of ^{210}Po intake, particularly in view of the fact that higher ^{210}Po levels have been measured for seafood such as fish, seaweed and shellfish, and that ^{210}Pb intake from foodstuffs in Japanese[6,7] is about 4 times higher than those in Europe and USA.[4] However, no data has been reported for estimating fully the intake and body burden of ^{210}Po and ^{210}Pb in people consuming large amounts of seafood.

Elsevier Sequoia S. A., Lausanne
Akadémiai Kiadó, Budapest

In this context, it is of interest and importance to estimate a reasonable values of intake and body burden of ^{210}Pb and ^{210}Po for Japanese. Prior to the measurements of these nuclides in foodstuffs and human tissues, we first intended to measure ^{210}Pb and ^{210}Po contents in human hair of Japanese, by taking into consideration that human hair could be a good indicator for the estimation of radionuclides in the human body. Various elements contained in human hair have been used as an index of environmental pollution effect of heavy metals to human beings.[8]

This paper describes the measurement of ^{210}Pb and ^{210}Po contents in human hair collected from three areas in Japan, with emphasis on particular efforts with respect to the accuracy and precision of the measurement of ^{210}Pb and ^{210}Po activities.

Materials and methods

Samples

Human hair samples were collected from barbershops in Ishikawa Prefecture facing to the Sea of Japan (June 1988), Shizuoka Prefecture located near central region of Japan (August 1988) and Ibaraki Prefecture facing the Pacific Ocean (January–February 1989). Sampling date, sex, age and smoking habits of the subjects were known for each sample. Unfortunately, for the samples of Ibaraki Prefecture, the weight of each sample was only about 1–2 g. Therefore, each sample from Ibaraki Prefecture was classfied by sex and five years of age interval.

The sample was cut into short length (2–3 mm in length), and then washed by the recommended method of IAEA with acetone and distilled water to remove external contamination. After air-drying, 5 to 10 g of the sample was stored for the analyses of ^{210}Pb and ^{210}Po.

Analytical procedures

An aliquot of hair sample, 3–5 g in weight, was subjected to radiochemical analyses of ^{210}Pb and ^{210}Po, using ^{209}Po as a yield tracer of polonium. The sample was completely decomposed by wet ashing with HNO_3, H_2O_2 and $HClO_4$, and the residue was transformed to chlorides with HCl. The resultant residue was then dissolved in 50–100 ml of 0.5M HCl with warming, and several 10 mg's of ascorbic acid was added to the solution. A polished thin silver disc (about 20 mm in diameter, the backside of which was coated with heat-resistant tape) was put into the solution. Polonium was electrochemically deposited on a silver disc with occasional stirring and heating of solution (70–90 °C) on a hot plate for more than 6 hours.

In order to determine ^{210}Pb, the residual solution after electrochemical deposition of polonium was gently evaporated to dryness with the addition of small amounts of HNO_3 and H_2O_2. The residue was dissolved in 10–20 ml of 10M HCl, and the solution was passed through a Dowex 1-X8 anion exchange resin column (ϕ 8 mm × 70 mm) to remove completely the polonium remaining after its deposition onto the silver disc. The column was thoroughly washed with 80–90 ml of 10M HCl. Pb is not adsorbed on the column under these conditions, while Po is strongly adsorbed. ^{209}Po tracer was spiked to the effluent (about 100 ml) from the column, and the solution was stored for 3–6 months to allow the growth of ^{210}Po from ^{210}Pb. The ^{210}Po grown by the decay of ^{210}Pb was coprecipitated with $Fe(OH)_3$, and deposited onto a silver disc by the above mentioned procedures.

Measurement of ^{210}Pb and ^{210}Po activities

The activity of polonium (^{210}Po and ^{209}Po) deposited on the silver disc was measured by using a Si(Au) surface barrier detector with a 450 or 500 mm^2 active area, coupled with a 1K channel pulse-height analyzer.

The concentrations of ^{210}Pb (A_0^D) and ^{210}Po (A_0^F) at the sampling date (t_0) were calculated using the following equations:

$$A_0^D = f_1 \cdot A_2^F$$

$$A_0^F = f_2 \cdot (A_1^F - f_3 \cdot A_0^D)$$

where A_1^F – activity of ^{210}Po (mBq/g) measured at time t_1,
A_2^F – activity of ingrowth ^{210}Po (mBq/g) measured at time t_2 for the measurement of ^{210}Pb,
f_1 – factor to correct for the growth of ^{210}Po from ^{210}Pb in the period from t_1 to t_2,
f_2 – factor to correct for ^{210}Po decay in the period from t_0 to t_1,
f_3 – factor to correct for growth of ^{210}Po from ^{210}Pb in the period from t_0 to t_1.

The factors f_1 and f_3 were calculated by using Beteman's equation for the decay and growth of three radionuclides, that is, ^{210}Pb, ^{210}Bi and ^{210}Po.

Results and discussion

Accuracy and precision of measurement

For the determination of ^{210}Po, since ^{209}Po was spiked to the sample solution as a yield tracer at the beginning of chemical analysis, there seems to be no problem unless the sample solution is evaporated to dryness at higher temperature during wet ashing. On the other hand, no tracer for determining the chemical yield of ^{210}Pb is spiked to the sample solution because of the lack of a suitable radioactive tracer of this element. Therefore, the accuracy and precision of the present method were certified by analysis of the human hair sample spiked with known amount of ^{210}Pb and non-spiked human hair sample. Since ^{210}Pb (E_γ = 46.5 keV, abundance = 4.05%) can be measured directly by non-destructive gamma-ray spectrometry using Ge-LEPS (low energy photon spectrometer), the value measured directly by Ge-LEPS and the value measured indirectly by alpha-ray spectrometry for the ^{210}Po ingrowth from ^{210}Pb were compared using the ^{210}Pb fraction obtained throughout the analytical procedure.

At first, the analysis was made using about 1 g of human hair spiked with a known amount of ^{210}Pb. Secondly, the determination of ^{210}Pb was performed using a large amount of human hair to make it possible to detect ^{210}Pb activity by Ge-LEPS.

Results of analyses are given in Tables 1 and 2, respectively. As can be seen from the results of ^{210}Pb obtained by Ge-LEPS in Table 1, ^{210}Pb spiked was recovered quantitatively. Furthermore, the ^{210}Pb activity found by alpha-spectrometry of ^{210}Po from ^{210}Pb showed good agreement with spiked ^{210}Pb activity within the uncertainty of one sigma of propagated counting error. In the case of actual samples shown in

Table 1
Comparison of ^{210}Pb measurements by Ge-LEPS
with that by ^{210}Po ingrowth method using hair samples spiked with a known amount of ^{210}Pb

Sample	^{210}Pb content	
	^{210}Pb by Ge-LEPS, dpm	^{210}Po ingrowth, dpm
1 ml of ^{210}Pb standard solution*	65.4±2.6	64.8±1.3
Run-1**	63.0±3.0	66.8±1.6
Run-2**	67.8±3.3	63.3±1.5
Run-3**	64.2±3.6	64.3±1.7

*^{210}Pb standard solution in radioactive equilibrium with ^{210}Po: (^{210}Pb 65.4±2.6 dpm/ml).
**Analysis was performed using about 1g of a hair sample spiked with 1 ml ^{210}Pb standard solution.

Table 2
Comparison of ^{210}Pb measurements by Ge-LEPS with that by the ^{210}Po ingrowth method, using large amounts of non-spiked human hair samples

Sample No.	Weight, g	^{210}Pb content, mBq/g	
		^{210}Pb by Ge-LEPS	^{210}Po ingrowth
H-1	68.1	2.51±0.37	2.37±0.18
H-2	84.2	1.73±0.22	1.85±0.11
H-3	61.9	1.74±0.21	1.70±0.15
H-4	90.6	2.04±0.25	2.07±0.18

Table 3
Replicate analyses of ^{210}Pb and ^{210}Po, using about 2.5 g of a hair sample

Run	Weight, g	Concentration, mBq/g		Activity ratio: ^{210}Po/^{210}Pb
		^{210}Pb	^{210}Po	
1	2.49	0.90±0.07*	6.9±0.5	7.6±0.8
2	2.52	0.78±0.06	6.6±0.4	8.4±0.8
3	2.52	0.96±0.06	6.7±0.4	7.0±0.6
4	2.56	0.89±0.07	7.0±0.5	7.9±0.8
5	2.53	0.81±0.07	6.7±0.4	8.3±0.9
Mean ± S.D.		0.87±0.07	6.8±0.2	7.8±0.6

*1 σ of counting error.

Table 2, the results of two methods agreed quite well within the limit of counting error, although the counting error of Ge-LEPS is a little larger. The replicate analyses of both ^{210}Pb and ^{210}Po applied to the same sample are given in Table 3. As known from these tables, the accuracy and precision of the present method were confirmed to be sufficiently good for the measurement of ^{210}Pb and ^{210}Po in human hair samples.

^{210}Pb and ^{210}Po contents of human hair samples

Prior to the determination of ^{210}Pb and ^{210}Po in hair samples, a preliminary experiment was carried out to evaluate the effect of washing by the IAEA recommended method of treating with acetone and distilled water. The analytical results of ^{210}Pb and ^{210}Po contents measured for both washed and untreated samples are compared in Table 4. Most of the samples showed nearly the same values for both ^{210}Pb and ^{210}Po, indicating that the contribution of surface contamination of hair by airborne dust containing ^{210}Pb and ^{210}Po is quite small or negligible.

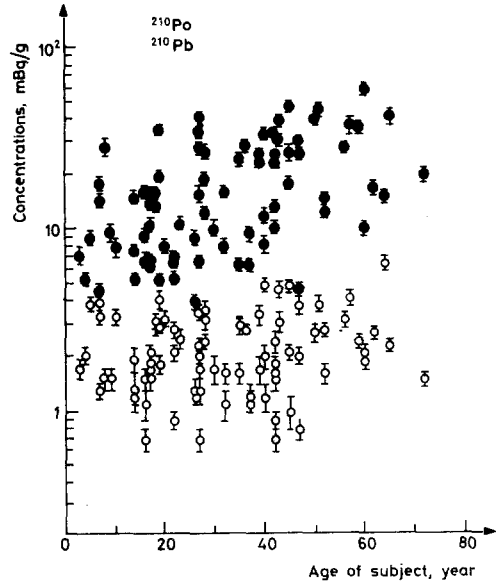

Fig. 1. Concentrations of ^{210}Pb and ^{210}Po in human hair as a function of age of subjects

Table 4
Removal of ^{210}Pb and ^{210}Po by hair-washing with acetone and distilled water (IAEA recommended method)

Sample No.	^{210}Pb concentration, mBq/g		^{210}Po concentration, mBq/g	
	Washed	Unwashed	Washed	Unwashed
1	3.3±0.3	3.4±0.4	7.9±0.8	7.2±0.6
2	1.1±0.2	1.2±0.2	6.8±0.6	6.8±0.5
3	2.9±0.2	2.0±0.2	19.5±1.3	22.1±1.8
4	1.8±0.2	1.7±0.2	5.3±0.4	5.0±0.4
5	2.9±0.2	3.2±0.2	6.4±0.5	7.2±0.6
6	5.0±0.4	5.5±0.3	33.2±2.6	31.5±2.6

The results of ^{210}Pb and ^{210}Po measurement of hair samples in Ishikawa, Shizuoka and Ibaraki Prefectures are plotted as a function of the age of subjects in Fig. 1. The frequency distributions of ^{210}Pb and ^{210}Po contents, and their activity ratios are shown in Fig. 2. As can be seen from Fig. 1, ^{210}Pb and ^{210}Po contents in 83 hair samples varied considerably regardless of age, and no significant relationship between the contents of these nuclides and age was found. Both ^{210}Pb contents (0.7–6.5 mBq/g) and ^{210}Po contents (4.0–59.3 mBq/g) vary ranging almost 10-fold. Any

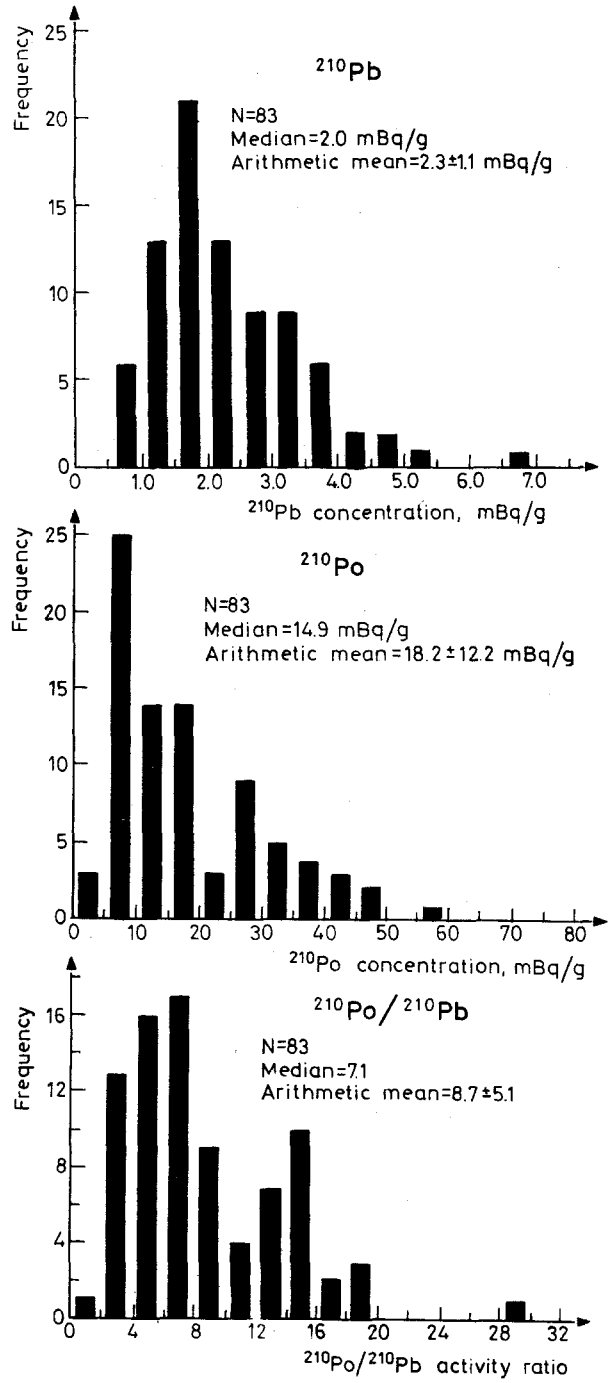

Fig. 2. Frequency distributions of ^{210}Pb and ^{210}Po contents and ^{210}Po/^{210}Pb activity ratios in human hair

difference between males and females, smokers and non-smokers, and among different locations was examined both for ^{210}Pb and ^{210}Po activities, but apparent differences were not found for any cases.

As shown in Fig. 2, the mean and median of ^{210}Pb contents were 2.3±1.1 mBq/g and 2.0 mBq/g, and those of ^{210}Po 18.2±12.2 mBq/g and 14.9 mBq/g, respectively. The ^{210}Po/^{210}Pb activity ratios were found to be in a wide range from 1.9 to as high as 28.3, and the mean and median were 8.7±5.1 and 7.9. According to the review of PARFENOV,[4] the ^{210}Po contents in human hair of the general population in various areas range from 1.4 to 18.5 mBq/g, and the mean ^{210}Po content is approximately 5.6 mBq/g. The mean and median values of ^{210}Po obtained in this study (18.2 and 14.9 mBq/g, respectively) are close to the highest ^{210}Po value reviewed of PARFENOV, but are about 3 times higher than the mean ^{210}Po value (5.6 mBq/g).

As for ^{210}Pb, data are very scarce in the literature. LANDINSKAYA[5] reported the mean ^{210}Pb and ^{210}Po contents, and their activity ratios in hair (n = 33) of the Rostov-to-the-Don inhabitants (USSR): ^{210}Pb, 1.48±0.28 mBq/g, ^{210}Po, 3.31±0.71 mBq/g and ^{210}Po/^{210}Pb ratio, 2.2±0.4. Although direct comparison of our data and LANDINSKAYA's data is difficult mainly because of the differences of eating habits, it is of interest to note that the mean ^{210}Pb value in Japanese hair is nearly the same as that for the Rostov-to-the-Don inhabitants, but the mean ^{210}Po content is about 5 times higher in Japanese hair. ERMOLAEVA-MAKOVSKAYA et al.[9]

Table 5
^{210}Pb and ^{210}Po contents in different parts of hair*

Sample No.	Total weight of sample, g	Concentration, mBq/g		Activity ratio: ^{210}Po/^{210}Pb
		^{210}Pb	^{210}Po	
1	6.74	0.52±0.04	10.5±0.8	20.2±2.2
2	13.64	0.45±0.04	9.4±0.6	20.9±2.2
3	14.70	0.43±0.03	8.3±0.7	19.3±2.2
4	13.64	0.52±0.05	7.8±0.6	15.0±1.8
5	6.51	0.62±0.05	5.6±0.4	9.0±1.0

*Hair of about 20 cm in length was cut into five sections.
Sample No. 1 is close to the root of hair.

reported that the ^{210}Po contents in the hair of Petropavlovsk-Kamchatsky inhabitants (USSR) in whose diet sea fish is an important component amount to as high as 22.2 mBq/g. This ^{210}Po value is close to the mean ^{210}Po value in the Japaneses hair studied at present.

At this stage it seems difficult to know the exact reason why the ^{210}Po contents in the hair of Japanese is higher than in other countries. The fact that Japanese people ingest animal protein mainly through seafood may be pointed out as one of the possible reasons. However, it must be further considered that the distribution of ^{210}Pb and ^{210}Po, especially short-lived ^{210}Po, may not be uniform, because that human hair grows at a rate of 1–2 cm per month.[10] Such an example is shown in Table 5, in which hair of about 20 cm in length was cut into five sections, and each section was measured for ^{210}Pb and ^{210}Po contents. As expected, the ^{210}Po/^{210}Pb activity ratio of the tip of hair is about half of that of the root part, indicating the decay of ^{210}Po.

Now we are measuring ^{210}Pb and ^{210}Po contents in various foodstuffs and human tissues to estimate the dietary intake and body burden of these nuclides for Japanese population.

*

The authors express their heartly thanks to Mr. K. KUROSAWA and Mrs. H. SAWAHATA, Laboratory for Radioecology, Nakaminato, N. I. R. S., and Mr. M. ARAKI, barber in Ishikawa Prefecture, for the collection of human hair samples.

References

1. UNSCEAR Report, Sources and Effects of Ionizing Radiation, United Nation, New York, 1977.
2. R. B. HOLTZMAN, Health Phys., 9 (1963) 385.
3. NCRP Report No. 45, National Background Radiation in the United States, 1975.
4. Y. D. PARFENOV, At. Energy Rev. IAEA, 12 (1974) 75.
5. L. A. LANDINSKAYA, Arch. Environ. Health, 27 (1973) 254.
6. N. TAKADA, N. WATANABE, R. ICHIKAWA, J. Radiat. Res., 9 (1968) 29.
7. K. KAMETANI, H. IKEBUCHI, T. MATSUMURA, H. KAWAKAMI, Radioisotopes, 30 (1981) 681.
8. IAEA Report, Activation Analysis of Hair as an Indicator of Contamination of Man by Environmental Trace Element Pollutants, IAEA/RL/50, Vienna, 1978.
9. ERMOLAEVA-MAKOVSKAYA et al., Radioecological Research Methods (in Russian), Atomizdat, Moscow, 1971, p. 182.
10. Private communication with barber.