A Study on the Excretion of Pb-210 and Po-210

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(Received December 26, 1981)

Uranium mine worker/Faecal and urinary excretion/Pb-210 and Po-210

The amount of Po-210 excreted in urine and feces was more influenced by Po-210 that was taken with food and drink than taken through inhalation.

The amount of Pb-210 in urine of mining workers among uranium mine workers was higher than that of the non-uranium mine workers. It was thought that this fact was due to the working environment in uranium mine the amount of Pb-210 being a few tens times higher than that in normal environment.

The activity ratios of Po-210 of faecal to urinary excretion are widely distributed, however, the average value of many samples approached to 10.

Urinary excretion of Po-210 was highest after 24 hours of ingestion, but for faecal excretion, it was highest after 3 days.

INTRODUCTION

A great many kinds of the primordial radionuclides and their decay products have existed in the lithosphere, hydrosphere and biosphere, and they have been taken by mankind via inhalation and ingestion.

Among these radionuclides, concentrations of Pb-210 and Po-210 (decay products of U-238) in environmental samples and human organs have been investigated for a long time.\(^1\)-\(^5\)

In many uranium mines of the world, in particular, the measurement of Po-210 in the excreta of workers have been conducted due to estimate the exposure dose of workers. In Japan, this has been done and has been reported, too.\(^6\)-\(^8\)

However, these analysis have been conducted mostly on urine, and very few on faecal excretion.

It is considered that a portion of the materials that intaked into gastrointestinal tract are generally absorbed and the remainder are excreted in feces. The materials that entered into blood are transferred to various parts of the body by blood and a part of them are deposited on the various organs and tissues,
and the remainder are excreted in urine via kidney. Therefore, it is important to know the mode of excretion of these nuclides in urine and feces, on the estimation of the uptake of Pb-210 and Po-210 in the body.

With this purpose, measurements of Pb-210 and Po-210 excreted in urines and feces of uranium mine workers and non-uranium mine workers were conducted.

MATERIALS

All urines and feces excreted during 24 hours were collected from several workers in each workshop (mining, refining, analysis and office) among the uranium mine workers. (Uranium mine: Ningyo Pass Mine, Okayama prefecture.)

24hr-urines were collected from several healthy non-uranium mine workers (from 25 to 45 years old) and 24hr-urines and feces were collected from a healthy non-uranium mine worker (43 years old). The concentrations of Po-210 in these samples were measured, only a part of them were analyzed for Pb-210.

The concentrations of Po-210 in various foodstuffs, Japanese cigarettes and air collected in a Japanese uranium mine have been reported. However, measurement for Pb-210 concentrations in some of these samples was also conducted.

ANALYTICAL METHODS

Weighted sample was carefully wet-ashed in a beaker with concentrated HNO₃ and 30% H₂O₂ completely. Because polonium is volatile, particular attention was paid to avoid evaporation to dryness of the residue in the beaker.

In order to change the residue to chloride form, concentrated HCl was added after wet-ashing and the sample was continuously heated. This step was repeated until nitrous fumes cease to be evolved. When the residue has been changed to chloride form, the solution was neutralized with saturated NaOH solution.

This solution was then diluted to 200 ml with 0.5 M HCl solution. After which, 100 mg of ascorbic acid was added.

This solution was transferred to a plating cell. It was made from a commercial baby nursing bottle with the bottom removed and a silver plate fitted inside the cap which was then firmly screwed onto the bottle.

The plating cell was then placed in a water bath at 90-100°C and was stirred with a glass stirrer for 4 hours. Occasional small additions of distilled water were necessary to replace lost water during evaporation in the solution.

The cell was removed from the water bath and solution was then poured into a bottle. This solution was reserved for Pb-210 determination. The cell was dismantled and its silver plate was rinsed with distilled water. It was then dried at room temperature. After which the silver plate was measured for the alpha activity by an alpha scintillation counter with ZnS detector.

Concentration of Pb-210 in the sample was measured as follows.
The solution from where Po-210 had been plated, after storage for about 6 months was then returned to a plating cell and the volume of the solution made up to 200 ml with 0.5 M HCl and 100 mg of ascorbic acid was added. The Po-210 formed from the decay of Pb-210 was deposited on a silver plate by the procedure earlier mentioned.

The concentration of Pb-210 at the time of sampling can be calculated from the amount present in a given sample at two later times $t_1$ and $t_2$ (where $t_1$ is as close as possible to the time of sampling and $t_2$ is about 6 months after $t_1$) from the following equation:

$$A_D = \frac{A_3^F - A_2^F e^{-\lambda (t_2 - t_1)}}{1 - e^{-\lambda (t_2 - t_1)}}$$

$$A_0^F = \frac{A_1^F - A_D (1 - e^{-\lambda t_1})}{e^{-\lambda t_1}}$$

where: $A_D$ is the Pb-210 activity of the sample; $A_0^F$ is the Po-210 activity at time $t_0$, $A_1^F$ is that measured at time $t_1$; $A_2^F$ is that remaining after removal of the Po-210 at $t_1$, and $A_3^F$ is that measured at time $t_2$; $t_0$ is the time at sampling, $t_1$ the time of the first analysis, and $t_2$ the time of the second analysis; and $\lambda$ is the decay constant of Po-210.

RESULTS AND DISCUSSIONS

Concentrations and the ratio of Po-210 in urines and in feces of uranium mine workers are shown in Table 1.

Though concentrations of Po-210 in excreta are widely distributed, it appears that the faecal excretion of Po-210 of uranium mine workers (32±34 pCi/day) were higher than that of non-uranium mine workers (6.4±6.7 pCi/day).

A non-uranium mine worker, who usually excrete 2-10 pCi/day of Po-210 in his feces, excreted 16-20 pCi/day of Po-210 in his feces when he went to an uranium mine for sampling.

As previously reported, the concentrations of Po-210 in the air of the various working areas of an uranium mine were several or several ten times higher than that of the normal environment. Considering working hours, uranium mine workers take more about one or two pCi of Po-210 than non-uranium mine workers via inhalation, and this dose does not reach the high amount of Po-210 excreted in feces. Therefore, it is suggested that the increase of faecal excretion of Po-210 is due to the foodstuffs.
Table 1. Amounts of Po-210 in urines and feces of uranium mine workers and activity ratios of Po-210 of faecal to urinary excretion.

| Workshop | Po-210 in urine pCi/day | Po-210 in feces pCi/day | Activity ratio Feces/urine |
|----------|-------------------------|-------------------------|---------------------------|
| M-1 mining | 1.7 ± 0.1 | 18 ± 0.4 | 11 ± 0.7 |
| 2      | 4.6 ± 0.1 | 14 ± 0.4 | 3.0 ± 0.1 |
| 3      | 2.4 ± 0.2 | 42 ± 1.3 | 18 ± 1.6 |
| 4      | 0.2 ± 0.1 | 22 ± 0.9 | 110 ± 0.5 |
| 5      | 4.7 ± 0.3 | 26 ± 0.9 | 5.5 ± 0.4 |
| 6      | 2.3 ± 0.2 | 6.2 ± 0.3 | 2.7 ± 0.3 |
| 7      | 2.6 ± 0.2 | 98 ± 1.8 | 38 ± 3.0 |
| 8      | 3.5 ± 0.2 | 66 ± 1.4 | 19 ± 1.2 |
| 9      | 3.0 ± 0.2 | 8.1 ± 0.4 | 2.7 ± 0.2 |
| 10     | 1.2 ± 0.1 | 133 ± 4.0 | 111 ± 0.8 |
| 11     | 5.6 ± 0.3 | 33 ± 1.0 | 5.9 ± 0.4 |
| A-1 analysis | 2.4 ± 0.1 | 20 ± 0.4 | 8.3 ± 0.4 |
| 2      | 0.9 ± 0.1 | 21 ± 0.6 | 23 ± 2.6 |
| 3      | 3.0 ± 0.2 | 5.3 ± 0.3 | 1.8 ± 0.2 |
| O-1 office | 2.7 ± 0.2 | 18 ± 0.4 | 6.7 ± 0.5 |
| 2      | 1.1 ± 0.1 | 7.0 ± 0.4 | 6.4 ± 0.7 |
| 3      | 2.4 ± 0.2 | 29 ± 0.8 | 12 ± 1.1 |
| 4      | 1.6 ± 0.1 | 14 ± 0.7 | 8.8 ± 0.7 |

R.L. Blanchard had reported\(^1\) that a Po-210 dietary intake of 2 pCi/day was probably somewhat high for the normal environment. And ICRP has recommended that the amount of intake of Po-210 per day was 2.7 pCi.\(^10\) As shown in Table 1 and in a previous report\(^9\), Po-210 dietary intake of the Japanese is estimated to be several or several ten pCi per day, because this is due to the abundant consumption of marine products that contain high level of Po-210 in Japan.

It was found that the activity ratio of Po-210 in feces to urine are widely distributed. As shown in Table 1, the ratios of uranium mine workers are ranged from 1.8 to 111 with the average value of 12. The ratios for 5 days running on an individual scattered from 5.2 to 15\(^9\) with the average of these values was 10. Activity ratio of Po-210 of faecal to urinary excretion is recommended to be 9 in the publication No. 10 of ICRP,\(^10\) and the measured values are similar to this value.
As it has been reported, when the sampling was done continuously the amount of Po-210 in excreta and the activity ratio of Po-210 of faecal to urinary excretion on an individual are almost constant. It is considered that this finding shows uniformity of the living environment and habits of an individual.

However, when samples were collected in different periods, the results were scattered, and this may show the effect of variations of the living environment. Consequently, the variation of the results of the concentrations of Po-210 in excreta of uranium mine workers in Table 1 shows the differences of the living environments of workers.

The activity ratios of Po-210 of faecal to urinary excretion are widely distributed but when sampling population is numerous, the average of ratios approached to 10, and it is considered that the value of 9 recommended by ICRP is reasonable.

On the other hand, if the metabolism information is studied on only a few samples, conclusion will be misleading.

Average concentrations of Pb-210 and Po-210 and the activity ratios of Po-210 to Pb-210 in the urines of uranium mine workers and non-uranium mine workers are shown in Table 2.

| Workshop   | Number of sample | Po-210 pCi/1 | Pb-210 pCi/1 | Po-210/Pb-210 |
|------------|------------------|--------------|--------------|---------------|
| Mining     | 10               | 1.8 ± 1.1    | 1.3 ± 0.9    | 1.4 ± 1.2     |
| Refining   | 3                | 1.0 ± 0.7    | 0.6 ± 0.2    | 1.8 ± 1.4     |
| Analysis   | 4                | 0.8 ± 0.4    | 0.4 ± 0.4    | 1.7 ± 1.7     |
| Office     | 3                | 0.7 ± 0.4    | 0.4 ± 0.2    | 1.7 ± 1.2     |

Table 2. Concentrations and the ratios of Po-210 and Pb-210 in urine.
There was no significant difference between the concentrations of Po-210 in the urines of uranium mine workers and that in non-uranium mine workers. However, the concentrations of Pb-210 in the urines of miners and refining workers in the uranium mine were higher than that of non-uranium mine workers. And within the uranium mine itself, those of miners were higher than that of the office workers. It is suggested that these high level of Pb-210 in urines are due to the concentration of Pb-210 in the air of working environment as shown in Table 3.

The activity ratios of Po-210 to Pb-210 in the urines of uranium mine workers are 1.4±1.2, 1.8±1.4, 1.7±1.7 and 1.7±1.2 for mining, refining, analysis and office workers, respectively. And the ratio for non-uranium mine workers is 3.8±3.2. There was no significant difference in these ratios.

It has been reported that in the middle latitudes of the northern hemisphere the average concentrations of Pb-210 and Po-210 in surface air have been estimated to be 14 fCi/m³ and 3.3 fCi/m³, respectively. As shown in Table 3 the concentrations of Pb-210 in the air of an uranium mine in Japan are higher than that of Po-210. In such a manner, concentration of Pb-210 in the air is generally higher than that of Po-210.

According to the publication No. 2 of ICRP the transfer rates of Pb-210 and Po-210 from blood to critical organs are not so different whether by inhalation or ingestion.

However, it is recommended that the transfer rate of Pb-210 from blood to kidney and bone is two or three times faster than that of Po-210 and that from blood to liver is slower than that of Po-210. Inspite of these, the concentrations of Po-210 in urines were higher than that of Pb-210. This may be due to the fact that Japanese consume a large quantity of marine foods that contain high concentration of Po-210, and that Po-210 that decayed from Pb-210 deposited in the body is excreted.

Table 3. Concentrations of Po-210 and Pb-210 in the air of a Japanese uranium mine.

| Sampling places | Po-210:10⁻¹⁵ Ci/m³ | Pb-210:10⁻¹⁵ Ci/m³ |
|-----------------|-------------------|-------------------|
| Office          | 1.6               | 10                |
| Intake airway   | 7.6               | 24                |
| Exhaust airway  | 400               | 370               |
| Closed pit      | 190               | 880               |
| Closed pit      | 2300              | 17000             |
| Drilling        | 110               | 190               |
| Drilling        | 90                | 400               |
Biological half-time of Pb-210 and Po-210 for whole body are 1460 days and 30 days, respectively.\textsuperscript{10} Polonium is also assumed to be retained in the body with a biological half-time of 50 days.\textsuperscript{10} These may also have affected amount of excretion of Pb-210 and Po-210 from the body.

In order to investigate the period of Po-210 from ingestion till excretion, the following test was conducted.

Marine products, especially dried sardine contains high level of Po-210 and the stock of dried sardine also contains a large quantity of Po-210. Then the stock of dried sardine that contained of 130 pCi of Po-210, was ingested by a healthy adult man (43 years old) and his excretion was collected for the measurement of Po-210. The concentrations of Po-210 in urines and feces are shown in Table 4 and Fig. 1.

The stock was ingested at 14:00 on the 5th of June, and the amount of Po-210 in urine that was collected from 8:30 on the 6th of June till 8:30 on the 7th of June, increased to 1.2 pCi.

This value was about 4 times higher than that of normal urine (0.26~0.38 pCi/24 hours).

\textbf{Table 4. Faecal and urinary excretion of Po-210 and these ratio following the intake of the stock of dried sardine.}

| Po-210 in 24hr-urine pCi (sampling term) | Po-210 in feces of a day pCi (sampling date) | Activity ratio feces/urine |
|----------------------------------------|-----------------------------------------|---------------------------|
| 0.3 ± 0.03 (2nd~3rd, June)             | 3.4 ± 0.2 (3rd, June)                   | 11 ± 1.3                  |
| 0.3 ± 0.02 (3rd~4th, June)             | 3.9 ± 0.2 (4th, June)                   | 13 ± 1.1                  |
| 0.4 ± 0.02 (4th~5th, June)             | 3.5 ± 0.2 (5th, June)                   | 8.8 ± 0.7                 |

Intake of the stock of dried sardine : 14:00 5th of June

| Po-210 in 24hr-urine pCi (sampling term) | Po-210 in feces of a day pCi (sampling date) | Activity ratio feces/urine |
|----------------------------------------|-----------------------------------------|---------------------------|
| 0.4 ± 0.03 (5th~6th, June)             | 4.5 ± 0.2 (6th, June)                   | 11 ± 1.0                  |
| 1.2 ± 0.05 (6th~7th, June)             | 3.0 ± 0.1 (7th, June)                   | 2.5 ± 0.1                 |
| 0.3 ± 0.02 (7th~8th, June)             | 12 ± 0.4 (8th, June)                    | 40 ± 3.0                  |
| 0.2 ± 0.02 (8th~9th, June)             | 5.0 ± 0.2 (9th, June)                   | 25 ± 2.7                  |
| 0.2 ± 0.02 (9th~10th, June)            | 3.7 ± 0.2 (10th, June)                  | 19 ± 2.2                  |
The amount of Po-210 in urine that was collected from 8:30 on the 7th of June till 8:30 on the 8th of June, decreased to 0.29 pCi and did not increase henceforth.

On the other hand, the amount of Po-210 in feces after the intake of the stock on the 5th of June did not increase till the 7th of June. The amount of Po-210 in feces on the 8th of June increased to 11.8 pCi, and this was about 3 times higher than that of normal value (average value is 3.6 pCi). The change

![Graph showing the concentrations of Po-210 in urines and feces.](image-url)

**Fig. 1.** The concentrations of Po-210 in urines and feces.
of concentrations of Po-210 in urine with elapsed time after intake of the stock of dried sardine is shown in Fig. 2. It was found that the concentration of Po-210 in urine reached the highest value 24 hours after the intake of the stock.

Since the faecal excretion reached the highest value 3 days after the intake of the stock, these faecal and urinary excretion pattern is very interesting.

ICRP has recommended that the whole body retention of polonium is described by a function of the form\(^{11}\);

\[ R(t) = e^{-0.693t/50} \]

where \( R(t) \) is the whole body retention of polonium \( t \) days later. According to this equation, about 8% of total intake of Po-210 is excreted in 6 days after intake.
In case of this experiment, about 11 pCi (about 8.5% of 130 pCi) of Po-210 was excessively excreted during 5 days after intake of the stock of dried sardine. This excretion rate is similar to the recommended value by ICRP.

As shown in Table 4, the activity ratio of Po-210 of faecal to urinary excretion in this period was from 2.5 to 40 and these values were in the range of the values that was shown in Table 1. The average value of these ratios before intake of the stock was 11 and this value was near to the value of 9 recommended by ICRP. However, after intake of the stock, it increased and reached the highest value of 40. From these results, it was found that the daily diet was very influential to the change of the concentrations of Po-210 in urines and feces.

CONCLUSIONS

The amount of Po-210 excreted in urine and feces was more influenced by Po-210 intake with food and drink than the intake through inhalation.

The amount of Pb-210 in urine of mining workers among uranium mine workers was more than that of non-uranium mine workers.

It is concluded that this fact was due to the few ten times higher amount of Pb-210 in the working environment of an uranium mine than that of a normal environment.

The activity ratios of Po-210 of faecal to urinary excretion are widely distributed, and it was suggested that this distribution was caused by food and drink. However, the average value of many samples approach 10.

There was a case that a part of Po-210 had by ingestion was excreted in urine after 24 hours of ingestion and the remainder was excreted in feces after 3 days. Therefore, when the studies of excretion of Po-210 is conducted, it is necessary to investigate thoroughly the habits on food and drink of an individual.

REFERENCES

1. R.L. Blanchard (1967) Concentrations of $^{210}$Pb and $^{210}$Po in human soft tissues. Health Physics, 13: 625-632.
2. Z. Jaworowski (1969) Radioactive lead in the environment and in the human body. Atomic Energy Review IAEA, 7: 3-45.
3. R.L. Blanchard and J.B. Moore (1971) Body burden, distribution and internal dose of $^{210}$Pb and $^{210}$Po in a uranium miner population. Health Physics, 21: 499-518.
4. Y.D. Parfenov (1974) Polonium-210 in the environment and in the human organism. Atomic Energy Review IAEA, 12: 75-143.
5. UNSCEAR Report (1977) Sources and Effects of Ionizing Radiation. United Nations Scientific Committee on the Effects of Atomic Radiation, (United Nations, New York).
6. T. Inouye, Y. Yamada and S. Akiyama (1964) The level of polonium in urine of Japanese uranium miners. Proceedings of the Symposium on Radiological Health and Safety in Mining and Milling of Nuclear Materials. Vol. 2, IAEA: 425-429.
7. H. Okabayashi, M. Suzuki-Yasumoto, S. Hongo and S. Watanabe (1975) On the evaluation of Po-210 bioassay for uranium mine workers in Japan for the personal exposure index to radon daughters. J. Radiat. Res., 16: 142-151.

8. M. Ohira, S. Yoshioka, K. Masuda and S. Matsumoto (1975) Polonium-210 excreted in urine of the workers in a uranium mine. Jap. J. Hyg., 30: 556-561. (in Japanese)

9. R.B. Holtzman (1963) Measurement of the natural contents of RaD(210Pb) and RaF (210Po) in human bone estimates of whole-body burdens. Health Physics, 9: 385-400.

10. A Report by Committee 2 of ICRP (1980) Limits for intakes of radionuclides by workers. ICRP, Publication 30.

11. A Report by Committee 4 of ICRP (1968) Evaluation of radiation doses to body tissues from internal contamination due to occupational exposure. ICRP, Publication 10.

12. A Report by Committee 2 of ICRP (1960) Permissible dose for internal radiation. Health Physics, 3: 217-220.