Proceedings of the Korean Nuclear Society Autumn Meeting Seoul, Korea, October 1995

Inventory of Pu-238 and Pu-239,240 in the Soil of Korea

Myung Ho Lee, Chang Woo Lee, Yong Ho Choi, Sang Bok Kim, Sam Rang Kim

Korea Atomic Energy Research Institute

ABSTRACT

The cumulative deposition of Pu-238 and Pu-239,240 by the end of 1994 in undisturbed for the last 40 years, was determined at 9 sites in Korea. The cumulative deposition of Pu-238 and Pu-239 ranged from 0.76 to 3.77 and from 18.42 to 101.84 Bqm^{-2}, respectively. The mean values of the cumulative deposition of Pu-238 and Pu-239,240 were 2.16 and 54.75 Bqm^{-2}, respectively. These values are close to the value of worldwide fallout. No significant contribution to the cumulative deposition of Pu-238 and Pu-239,240 originating from the Chernobyl accident was found at any site.

INTRODUCTION

About 20 years ago, a global inventory of Pu-238 and Pu-239,240 in soil

from atmospheric tests and of Pu-238 from the burn-up of the SNAP-9A satellite was provided by Hardy [1]. Later the cumulative deposition of Pu-238 and Pu-239,240 and factors influenceing this deposition were studied in more detail in various countries[2-5]. Cumulative deposition in undisturbed areas covered by forest is about 30% higher that in those areas covered by grass, due to the interceptive function of forest canopies[2]. On the other hand no influence of soil faculties studied, such as acidity, pedological, lithological, geomorphological parameters, except for organic matter content, was found on the value of the cumulative deposition[3].

For this study, soil samples from 9 different localities of Korea were collected during 1994, in connection with our inventory programme of Pu-238 and Pu-239,240 in the soil. The main objectives of this paper was both to determine the cumulative deposition of Pu-238 and Pu-239,240 over the area of the country up to 1994 and to confirm the assumption that the main source of contamination of Korea soil was atmospheric nuclear tests (and the SNAP-9A burn-up) with the contribution from the Chernobyl accident and other accidents being very low.

EXPERIMENTAL AND METHODS

Soil sampling and pretreatment

Soil samples were collected from 9 sites in south Korea. All sampling points were selected on a flat area, if possible, in order to exclude precipitation run-off. Soil sampling was carried out in 1994. In each site, 5 to 10 soil samples were taken with a core sampler(4.5 cm in diameter)

within an area of about 50 m × 50 m to a depth of about 20 cm. Several preliminary experiments on forest soils showed that a sampling depth of 20 cm is sufficient to account for more than 95% of deposited activities of Pu and Cs from global fallout. The samples were dried at 110 °C for 48 h after pebbles and fragments of plant root were removed and then sieved through a 1.0 mm screen.

Radiochemical analysis of Pu-238 and Pu-239, 240

Radiochemical analyses of Pu-238 and Pu-239,240 were carried out on aliquots of 100g soil according to the method which had been developed in the Karlsruhe Research Center for emission control and environmental monitoring[6]. After the gamma-spectrometry analysis, the soil samples were calcined at 600 °C for 24 h to eliminate organic matter. After adding ^{242}Pu as a yield tracer, a leaching procedure using HNO_3, HF, and $Al(NO_3)_3$ solution is performed. Plutonium is extracted from this dissolved sample material into trioctylphosphine oxide in cyclohexane. This very effective extraction step separates Pu from most of the matrix elements like Si, Fe, alkali and alkaline earth elements. Back extraction is done using ascorbic acid in HCl-solution. The radiochemical purification of this Pu-fraction is performed in two steps: coprecipitation with LaF_3 and anion exchange. Finally, plutonium is electrodeposited from HCl/oxalate solution and measured by α-spectrometry. The chemical yields attained with this analytical procedure are in the range of 60 to 80%. The detection limit was 0.0043 Bq/kg-dry soil for 86,000 seconds of counting times.

RESULTS AND DISCUSSION

Inventory of Pu-239, 240 and Pu-238

The inventories are shown in Table 1. The inventory of Pu-239, 240 was estimated to be 54.8±32.1 (individual data ranged from 18.4 to 101.8) Bqm^{-2}. The average accumulated deposit of Pu-239, 240 in Japan at 1980 is also estimated to be 55-65 Bqm^{-2} by the analysis of surface soils (about 20cm depth)[7]. The inventory of Pu-239, 240 estimated for Korea soils seems a little lower than that for Japan. The difference of the inventory between korea soils and Japan ones may come from deposition influenced by meteorological condition, topogragphical features of the earth's surface and geochemically driven migration.

Also, the average inventory of Pu-238 was estimated to be 2.16±1.21 (individual data ranged from 0.76 to 3.77) Bqm^{-2}. The weapons Pu-238 contribution was obtained by multiplying the Pu-239, 240 values by 0.024 (the average weapons Pu-238 to Pu-239, 240 ratio found for six soils collected before fallout from the SNAP-9A)[1]. The SNAP-9A Pu-238 is then simply the difference between the total measured Pu-238 and the weapons Pu-238. The SNAP-9A Pu-238 has an entirely different distribution pattern. Most of the SNAP 9A debris has deposited in the Southern Hemisphere where the maximum fallout is 2.5 times that in the Northern Hemisphere. The average inventory was estimated to be 1.31± 0.76 Bqm^{-2} for the weapons Pu-238 and 0.85±0.45 Bqm^{-2} for the SNAP-9A Pu-238, respectively. These values are close to values 1.55 and 0.92 Bqm^{-2}, respectively reported by UNSCEAR(1982) for the north temperate zone(40-30°)[8].

Activity ratio of Pu-238/Pu-239, 240

The mean activity ratio of Pu-238/Pu-239, 240 in soils was calculated to be 0.040, a little higher than the value of 0.037 which is characteristic of fallout from nuclear weapon testings[2]. The small difference in the value of activity ratio can be explained by insufficient purification of the Pu-fraction from other alpha-emitters. Unless plutonium is isolated in a pure form, the similar alpha energy emitter naturally abundant like Th-238 in the soil may be mistaken for Pu-238 and the concentration of Pu-238 is overestimated. But, the mean activity ratio of Pu-238/Pu-239, 240 is not very different from the typical value of worldwide fallout. It means that the contribution of the Chernobyl-derived plutonium to the soil in Korea was negligible relative to their inventories from weapon testings, considering the reporting value of 0.47 of Pu-238/Pu-239, 240 in the Chernobyl fallout over Sweden[9].

REFERENCES

1. E. P. Hardy, P. W. Krey and H. L. Volchok, Nature, 241, 444(1973)

2. K. Bunzl and W. Kracke, J. Environ. Radioactivity, 8, 1 (1988)

3. A. A. Cigma, L. C. Rossi and G. Zurlini, J. Environ. Radioactivity, 5, 71(1987)

4. L. Sha, M. Yamamoto, K. Komura, K. Ueno, J. Radioanal. Nucl. Chem., Letters, 155, 45(1991)

5. J. H. Harley, J. Radiat. Res., 21, 83(1980)

6. H. Schuttelkopf, KfK-Report 3035 (1981)

7. M. Yamamoto, Doctoral thesis, University of Tsukuba , 1985

8. "Ionizing Radiation: Sources and Biological Effects", United Nations Scientific Committee on the Effects of Atomic Radiation, Report to the General Assembly, United Nations, New York, 1982.

9. R. W. Perkins, C. W. Thomas, World Wide Fallout. In Transuranic Elements in the Environment, W. C. Hanson, DOE/TIC-22800, National Technical Information Service, Springfield, 1980.

Table 1. The inventory of Pu-239, 240 and Pu-238 and their activity ratios

	Inventory(Bq/m^2)			Pu-238
Location	Pu-239,240	Pu-238(weapons,SNAP-9A)		Pu-239,240
GONGJU	87.92	3.47	(2.11, 1.36)	0.039
BOEUN	71.12	2.92	(1.71, 1.21)	0.041
EUIWANG	101.84	3.77	(2.40, 1.37)	0.037
TAEJON	21.06	0.84	(0.51, 0.33)	0.040
KANGLUNG	50.52	1.97	(1.21, 0.76)	0.039
JUNGSUN	84.08	3.28	(2.02, 1.26)	0.039
SESAN	18.42	0.76	(0.44, 0.32)	0.041
YEIJU	30.09	1.26	(0.72, 0.54)	0.042
YANGGU	27.71	1.14	(0.67, 0.47)	0.041
Average	54.7±32.1	2.16±1.2	(1.31±0.8, 0.85±0.5)	0.040±0.002