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PLUTONIUM ISOTOPES IN THE SURFACE AIR AT VINČA-BELGRADE SITE IN MAY 1986

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Surface air concentrations of plutonium isotopes at the Vinca-Belgrade site for the period May 1-15, 1986, are reported. The increase in $^{238}Pu/^{239,240}Pu$ ratios confirms that the source of plutonium in surface air was the Chernobyl accident.

INTRODUCTION

After the Chernobyl accident numerous measurements of γ- and β-emitters in fallout were performed, but very few data on air concentrations of plutonium nuclides were reported in the literature.

Since 1960 beta and gamma aerosol activity is continually measured in the Belgrade-Vinca Laboratory. The concentrations of plutonium radionuclides in air are especially measured at the time of nuclear weapons tests and local accidental situations. Total β- and γ-spectrometry measurements[1] as well as α-spectrometry measurements were also performed using the aerosol samples collected during the Chernobyl accident. The concetra-

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tions of ^{238}Pu and $^{239,240}Pu$ were determined in surface air at the Vinča site (12 km from Belgrade, Long. 20° 30', Lat. 44° 50') measuring the α-activity of Pu-isotopes in air in the period from May 1 until the end of 1987.

EXPERIMENTAL

Aerosol samples of 630 m^3/24 h of ground level air were taken each day starting from 8 h a.m. on May 1, 1986. The aerosol filter efficiency was about 80%. The analytical procedure applied for the separation of plutonium isotopes from air samples was nearly the same as that described in the previous paper[2]. The procedure includes thermal mineralization of aerosol filters, fusion with ammonium hydrogen sulfate for dissolution of insoluble plutonium oxide[3], plutonium coprecipitation with iron(III) hydroxide and separation by anion exchange resin (Dowex 1-X8, chloride form, 100-200 mesh). The ^{236}Pu tracer was added to the samples after thermal mineralization.

The eluent was finally electroplated on stainless steel discs by a procedure described by Halstadius[4], providing thin sources which made separation of close alpha energies possible. The radioactive sources obtained were measured by a silicon surface barrier detector spectrometer (ORTEC-676A). The counting time was about 5 d.

RESULTS AND DISCUSSION

The α-spectrum of $^{239,240}Pu$, ^{238}Pu and ^{236}Pu separated from the air sample taken on May 5 is given in Fig. 1. In addition to Pu-peaks, a 5.3 MeV α-peak is

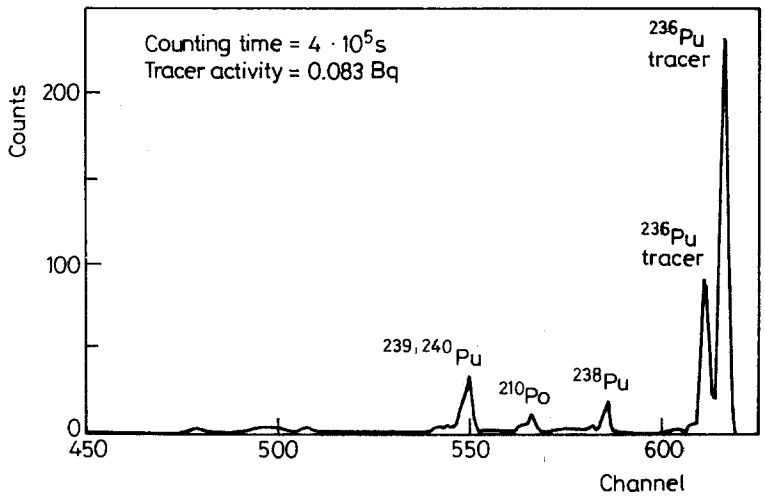

Fig. 1. Alpha spectrum of plutonium isotopes separated from the air sample taken on May 5, 1986

also found in the spectrum from the natural α-emitter ^{210}Po.

The measured concentrations of $^{239,240}Pu$ and ^{238}Pu in daily air samples collected from May 1 to May 15, 1986, and the concentrations of $^{239,240}Pu$ in the composite three-month samples collected in 1987 and 1989 are given here. The aerosol filter efficiency of 80% was included in the determination of absolute activity of Pu-isotopes. Total yield of the method (40-80%), determined for each sample, was also included in the calculations. In normal (nonaccidental) situations the content of Pu-isotopes is below the detection limit in daily samples, so the three-month samples are used. According to the recommended HASL procedures, the detection limit for the counting time of 4×10^5 s was found to be 10 nBq when the confidence level was set at 95%. Maximum counting error near this detection limit is ±10%.

TABLE 1

Daily concentrations of plutonium isotopes in surface air at Belgrade-Vinča site in the first half of May 1986 and $^{238}Pu/^{239,240}Pu$ and $^{239,240}Pu/^{137}Cs$ ratios

Date May 1986	Concentration, μBq m^{-3} $^{239,240}Pu$	^{238}Pu	$^{238}Pu/^{239,240}Pu$	$^{239,240}Pu/^{137}Cs$
01	0.9	0.52	0.578	4.02×10^{-5}
01-02	4.9	3.6	0.735	1.9×10^{-6}
03	10.2	5.4	0.529	3.5×10^{-6}
04	4.3	3.8	0.884	8.1×10^{-6}
05	10.6	5.7	0.538	3.9×10^{-5}
06	6.7	5.5	0.821	3.5×10^{-6}
07	4.7	4.2	0.894	3.1×10^{-6}
08	2.8	2.4	0.857	4.7×10^{-6}
09	3.5	2.8	0.800	8.3×10^{-6}
10	1.8	1.4	0.778	3.673×10^{-4}
11	0.4	0.3	0.868	6.78×10^{-5}
12	2.9	2.8	0.966	1.074×10^{-3}
13	3.6	1.9	0.528	3.00×10^{-3}
14	6.0	3.6	0.600	1.20×10^{-2}
15	5.2	3.9	0.754	6.75×10^{-4}

Standard statistical uncertainty = ±10%,
Air efficiency = 80%,
Total yield = 40-80%.

The $^{239,240}Pu/^{137}Cs$ activity ratios (Table 1) were obtained using ^{137}Cs activity data from a previously published paper[1].

Maximum concentrations of $^{239,240}Pu$ and ^{238}Pu were registered in the samples collected in the period of May 1-7 (Fig. 2). The maximum concentration of ^{137}Cs was observed at this location in the same period. The

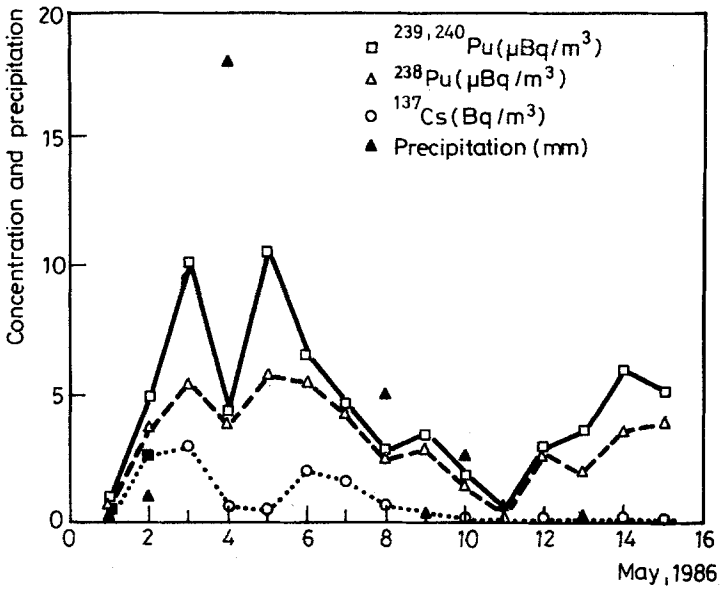

Fig. 2. Concentrations of $^{239,240}Pu$, ^{238}Pu and ^{137}Cs in air at Belgrade-Vinča site and precipitation for the period of May 1-15, 1986

$^{239,240}Pu$ concentrations in the surface air increased to 10.2 μBq m^{-3} and 10.6 μBq m^{-3} on May 1 and May 3, respectively. These concentrations are slightly lower than the concentrations of $^{239,240}Pu$ registered at the same location in the period of intensive nuclear weapons tests, during March-May 1965[2]. The concentrations of $^{239,240}Pu$ in the surface air at the Vinča-Belgrade site for the period of May 1-5, 1986 are comparable with those observed at Prague[6], Minich-Neuhenberg[7], Paris[8], and Risø[9] at the beginning of May 1986. Some regularity between rainfall quantity and plutonium concentration in surface air is observed (see Fig. 2). After a few days with heavy rainfall the concentration of plutonium in air decreased. Plutonium isotopes from the Chernobyl

TABLE 2

The concentrations of plutonium isotopes in composite air samples for 1987 and 1989

Quarter 1987	Concentration, $\mu Bq\ m^{-3}$	Quarter 1989	Concentration, $\mu Bq\ m^{-3}$
I	n.d.*	I	20 ± 14
II	226 ± 52	II	n.d.
III	n.d.	III	13 ± 5
IV	n.d.		

*Not detected.

fallout were hardly detected in the surface air in 1987. $^{239,240}Pu$ was detected, as shown in Table 2, only in the so-called "spring peak" of the fallout. The concentrations of plutonium isotopes in the composite air samples for the first, third and the fourth quarter of 1987 were below detection limits (Table 2).

The observed $^{238}Pu/^{239,240}Pu$ activity ratios range from 0.53 to 0.97 (Table 1), with an average value of 0.74. According to the Soviet report on the Chernobyl accident[10], the $^{238}Pu/^{239,240}Pu$ activity ratios in soil samples taken in the 30 km zone around the damaged reactor ranged from 0.4 to 0.7, while this ratio in the air samples taken at a height of 200 m was estimated to be 0.55. The plutonium isotopic ratio obtained from air samples collected in Minich during the first few days of May was 0.042 ± 0.03[11].

The values of $^{238}Pu/^{239,240}Pu$ activity ratios given in Table 1 are slightly higher than those given elsewhere. The ^{238}Pu α-line is at 5.5 MeV and the only other actinide which could be considered at this energy is ^{241}Am (α-line at 5.48 MeV) the daughter of ^{241}Pu. Since

the sources were measured immediately after electroplating, the 5.5 MeV α-line intensity is assumed to be pure ^{238}Pu activity. On the other hand, the increase of the activity ratios $^{238}Pu/^{239,240}Pu$ is interpreted as the consequence of the α-decay of the ^{242}Cm (T = 162.8 d) during the period of 4.5 y which elapsed between the sampling and the radiochemical separation of plutonium.

The $^{239,240}Pu/^{137}Cs$ activity ratios (Table 1) in the air samples from 1 to 9 May vary between 1.9×10^{-6} and 3.9×10^{-5} and they are consistent with the values obtained in München (2.5×10^{-6})[11], Chilton, U.K. (1.7×10^{-6})[12] and Tsukuba, Japan (1.7×10^{-6})[13], measurements.

The first Chernobyl radioactive plume which contained material released on April 26 in the reactor explosion reached our country on May 1. The second plume containing material released April 29-30 arrived here on May 2, and the third radioactive plume containing material, released mainly on May 5, reached Yugoslavia on May 9[1,14]. According to our measurements, the $^{239,240}Pu/^{137}Cs$ activity ratios observed in the air samples taken from May 9 to 15 are found to be significantly higher than those measured in the period of May 1-9. This result leads to the conclusion that the relative enrichment of plutonium isotopes occured in the additional release of the Chernobyl accident.

Unfortunately, these results were not obtained earlier since our laboratory started development of an α-spectrometry method for measurement of α-radionuclides in environmental air in 1989 and completed it in 1993. The aerosol filters from the Chernobyl period used for γ-spectrometry measurements[1] were saved and exploited for α-measurements.

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