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DETERMINATION OF $^{239+240}Pu$ IN SURFACE AIR IN SEVERAL LOCALITIES IN CZECHOSLOVAKIA IN 1986 IN CONNECTION WITH THE CHERNOBYL RADIATION ACCIDENT

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In connection with the radiation accident at the Chernobyl nuclear power plant, the concentration of $^{239+240}Pu$ was determined in surface air at several localities of Czechoslovakia during the year 1986. $^{239+240}Pu$ was found in the surface air in the period of April 29 - May 5 in amounts ranging from 10 to 140 $\mu Bq.m^{-3}$. In the period of June-December, 1986, the $^{239+240}Pu$ concentration in the surface air was comparable with that before the Chernobyl radiation accident.

INTRODUCTION

In connection with the radiation accident at the Chernobyl nuclear power plant, our laboratory performed, apart from determinations of radionuclides in air using γ-spectrometry, determinations of other radionuclides, e.g. $^{239+240}Pu$, that are difficult to determine by the above method. In the period from April 28 to May 31, 1986,

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$^{239+240}$Pu was determined in surface air of Prague /long. 14°26′, lat. 50°06′/, Roztoky near Prague and Dukovany /long. 16°,10′, lat. 49°11′/. Roztoky is situated approx. 15 km to the north of Prague. In June, $^{239+240}$Pu was determined in the surface air of Prague and Dukovany, in the period July-December 1986 only in the air of Prague. The concentration of $^{239+240}$Pu in the surface air of Dukovany was also determined before the Chernobyl radiation accident.

EXPERIMENTAL

Unless stated otherwise, all reagents used were of A.R. quality. Airborne dust in surface air was collected at 1.5-10 m height above ground level with an air filter /types FPP-15-1.7 or FS-S/. Alpha activity of plutonium isotopes was measured with an α-spectrometer consisting of a silicon surface barrier detector and 800-channel pulse height analyzer.

Following γ-spectrometric measurement, a part of the filter /or parts of more filters/ was transferred into a porcelain crucible. The yield monitor /200-400 mBq of ^{238}Pu/ was added and the sample then ashed at 500-600 °C. Carbon free ash was transferred into a teflon dish using 15M HNO_3 and after evaporation to dryness twice fumed with 5 ml 22M HF and twice with 5 ml 15M HNO_3. Plutonium was then leached from the residue by a mixture of 20 ml of 15M HNO_3 and 1 ml 1M $Al/NO_3/_3$. Plutonium was then separated from inorganic macro-components and other alpha emitters and finally electrodeposited on a stainless steel disc by the method described previously[1].

RESULTS

Table 1 shows the values of $^{239+240}$Pu content in surface air of Dukovany and Prague in the time period from April 28 to May 31, 1986. The presence of $^{239+240}$Pu in the surface air of Dukovany was proved in the period of April 30-May 4. The concentration of $^{239+240}$Pu in surface air was within the range of 49 to 140 $\mu Bq.m^{-3}$. In the surface air of Prague $^{239+240}$Pu was proved from April 29 to May 5. The concentration of $^{239+240}$Pu in this time period ranged from 10 to 28 $\mu Bq.m^{-3}$. In air taken at Dukovany after May 4 and in Prague after May 5, $^{239+240}$Pu was not proved any more.

The concentration of $^{239+240}$Pu in surface air of Roztoky was determined in the period of May 1-27, 1986. The results are given in Table 2.

Figure 1 presents the α-spectrum of $^{239+240}$Pu /and ^{238}Pu added to the sample as yield monitor/ isolated from surface air of Prague in two periods in May 1986. Curve 1 shows α-spectrum of $^{239+240}$Pu isolated from 3504 m^3 of air, taken from May 2-4. Curve 2 shows the α-spectrum of $^{239+240}$Pu separated from 1141 m^3 of air, taken during the period of May 9-10. It is obvious that while the first spectrum shows distinct peak of $^{239+240}$Pu, in the second one this peak is absent.

Table 3 presents the values of $^{239+240}$Pu in the surface air of Dukovany in the periods of January to March, and in June 1986, and those for Prague in the period from June to December. The volume of air for determination of plutonium ranged in individual months from 7860 to 30451 m^3.

TABLE 1

The concentration of $^{239+240}Pu$ in surface air of Dukovany and Prague at the end of April and in May 1986

Dukovany			Prague		
Date of sampling	V, m^3	$^{239+240}Pu$ in air, $\mu Bq.m^{-3}$	Date of sampling	V, m^3	$^{239+240}Pu$ in air, $\mu Bq.m^{-3}$
April 28 /12^{00}/-			April 29 /8^{15}/-		
April 29 /14^{30}/	822	<4	April 30 /8^{15}/	428	15
April 30 /5^{30}/-			May 1 /8^{15}/-		
May 1 /16^{00}/	531	49	May 2 /8^{15}/	845	10
May 2 /23^{15}/-			May 2 /8^{15}/-		
May 3 /5^{00}/	88	140	May 4 /8^{15}/	3504	28
May 3 /5^{00}/-			May 4 /8^{15}/-		
May 3 /11^{15}/	92	<32	May 5 /8^{15}/	831	13
May 4 /11^{00}/-			May 6 /8^{15}/-		
May 4 /17^{00}/	90	132	May 7 /8^{15}/	828	<4
May 5 /5^{00}/-			May 9 /8^{15}/-		
May 5 /22^{00}/	280	<11	May 10 /8^{15}/	1141	<3
May 7 /10^{00}/-			May 12-May 19	5607	<1
May 9 /12^{45}/	681	<4			
May 15 /15^{45}/-					
May 16 /6^{30}/	484	<6			
May 19-May 26	2535	<1			

V - Volume of air used for the determination of $^{239+240}Pu$.

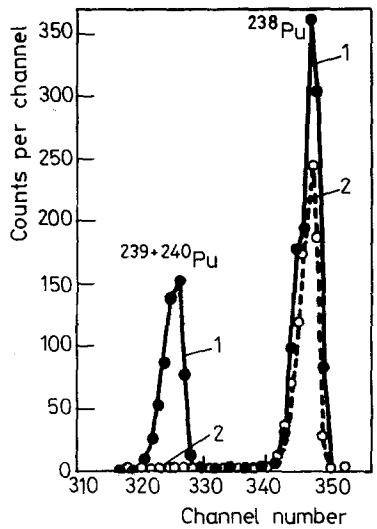

Fig. 1. Alpha spectrum of $^{239+240}$Pu /and ^{238}Pu used as yield monitor/ separated from air of Prague in time period May 2-4 /curve 1/ and May 9-10 /curve 2/, 1986

TABLE 2

Concentration of $^{239+240}$Pu in surface air of Koztoky near Prague in the period of May 1-May 27, 1986

Date of sampling	Volume of air used for $^{239+240}$Pu determination, m^3	$^{239+240}$Pu in air, $\mu Bq.m^{-3}$
May 1-May 6	181	<17
May 7-May 13	410	< 7
May 14-May 27	767	< 4

DISCUSSION

Measurements performed in our laboratory have proved the presence of $^{239+240}$Pu in the surface air of Prague and Dukovany at the end of April and the beginning of

TABLE 3

Concentration of $^{239+240}$Pu in surface air of Prague and Dukovany in 1986

Months	$^{239+240}$Pu in air, μBq.m^{-3}	
	Prague	Dukovany
January		<0.06
February		<0.06
March		<0.12
June	<0.24	<0.20
July	<0.34	
August	<0.16	
September	<0.20	
October	<0.14	
November	<0.08	
December	<0.13	

May 1986. The increased activity of $^{239+240}$Pu /and ^{238}Pu/ has also been proved in some localities of Finland[2] in the period from April 28 to 30 and in West Germany[3] /Munich/ in the period from April 29 to May 2, in both cases in the fall-out. The concentration of $^{239+240}$Pu in surface air of Prague /April 29-May 5/ is comparable with that in Heidelberg[4] /i.e. a locality situated nearest to localities studied by our laboratory/ in the years 1962-1963 when the concentration of these nuclides in the air reached a maximum due to atmospheric nuclear weapons tests. However, in the period of April 30-May 4 in surface air of Dukovany the concentration of $^{239+240}$Pu was definitely higher and at Roztoky in the period of May 1-May 6 the same or lower than that obtained in Prague.

Comparing further values measured in May with those before the Chernobyl radiation accident, we have found that even after May 5 the concentration of $^{239+240}$Pu in surface air could have exceeded the level before this accident. In the following period, i.e. June for Dukovany and June to December 1986 for Prague, the concentrations of $^{239+240}$Pu in surface air were similar to those measured at Dukovany from January to March 1986, i.e. in the period before the Chernobyl radiation accident /Table 3/. These values are comparable with those measured in other laboratories[5-8] in the years 1981-1984. For the period of June-December 1986, no increase in $^{239+240}$Pu concentrations in the surface air due to Chernobyl radiation accident is therefore assumed in localities investigated by our laboratory.

There is a question as to what extent our values of $^{239+240}$Pu concentration in surface air /in the period from April 29 to May 5/ have been influenced by the presence of ^{238}Pu in analyzed air using it simultaneously as a yield monitor. Commonly used yield monitors /^{236}Pu/ have not so far been available in our country. Since the presence of ^{238}Pu in analyzed air was presumed, we have decided /to eliminate its influence on accuracy of $^{239+240}$Pu determination/ to add a greater activity of ^{238}Pu /200-400 mBq/ to the analyzed sample than is usually used in the case of ^{236}Pu /~37 mBq/. Under these conditions, ^{238}Pu activity in the analyzed air formed only a small part of ^{238}Pu added to the sample as a yield monitor. In this way, a minimum error was attained due to the presence of ^{238}Pu in the analyzed air in most of the positive findings given in Table 1. Thus, for instance, supposing ^{238}Pu is present in air in an amount of 35% of $^{239+240}$Pu activity

/as was found in Munich[3]/, $^{239+240}$Pu concentration in surface air of Prague in the period of May 2 to May 4 is then about 17% and that for Dukovany in the period of April 30-May 1 by about 11% higher than the values given in Table 1. For other positive results, this increase is lower than 5%.

REFERENCES

1. Z. Hölgye, Appl. Radiat. Isotopes, 37 /1986/ 1015.

2. Second Interim Report Radiation Situation in Finland from May 5 to 16, 1986, Report STUK-B-Valo 45 /Finnish Centre for Radiation and Nuclear Safety, Helsinki/ 1986.

3. Umweltradioaktivität und Strahlenexposition in Südbayern durch den Tschernobyl-Unfall, Report GSF-Bericht 16/86 /Gesellschaft für Strahlen- und Umweltforschung, München/ 1986.

4. A.T. Jakubick, Migration of Plutonium in Natural Soils, in: Transuranium Nuclides in the Environment, IAEA, Vienna, 1976, p. 47.

5. H. Hötzl, G. Rosner, R. Winkler, Report GSF-Bericht S-956 /Gesellschaft für Strahlen- und Umweltforschung, München/ 1983.

6. K. Hirose, Y. Sigimura, Health Phys., 46 /1984/ 1281.

7. R.S. Cambray, G.N.J. Lewis, K. Playford, J.D. Eakins, Report AERE-R 10859 /Environmental and Medical Sciences Division, AERE Harwell, Oxfordshire/ 1983.

8. R.S. Cambray, K. Playford, G.N.J. Lewis, Report AERE-R 11915 /Environmental and Medical Sciences Division, AERE Harwell, Oxfordshire/ 1985.