

ACTINIDE NUCLIDES IN ENVIRONMENTAL AIR AND PRECIPITATION SAMPLES AFTER THE CHERNOBYL ACCIDENT

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The present paper describes the analysis of isotopes of uranium, neptunium, plutonium, americium and curium, in air and deposition samples taken at our laboratory site 10 km north of Munich, subsequent to the Chernobyl accident. Uranium-234, ^{237}U , ^{238}U , ^{239}Np , ^{238}Pu , $^{239} + ^{240}\text{Pu}$ and ^{242}Cm have been identified and upper limits of detection have been established for ^{241}Am and ^{244}Cm . Deposition and air concentration values are discussed.

Introduction

Following the Chernobyl accident, many measurements were made of gamma and beta emitters in fallout, where the emphasis has been on the assessment of population radiation exposure (Devell *et al.*, 1986; Fry *et al.*, 1986; SSK, 1987). However, comparatively few data exist on ground deposition and air concentrations of actinide nuclides.

In the region of southern Bavaria, including Munich, the deposition of radioactivity from Chernobyl was one of the highest in Western Europe. This fact, together with the immediate availability of proven sampling and measurement techniques in the Institute for many years (Hötzl *et al.*, 1983; Hötzl *et al.*, 1976), enabled actinide nuclides to be studied in addition to the strontium isotopes and about 20 gamma emitters (Hötzl *et al.*, 1987).

Methods

Most of the experimental methods have been described in detail elsewhere, (Hötzl *et al.*, 1983) and will only be referred to briefly here. The deposition samples, including dry fallout, were collected in tubs of 0.6 m² area whose bottoms were covered with water during dry periods. Samples were analysed radiochemically for the alpha emitting actinide nuclides. Plutonium-242 and ^{243}Am were used as tracers in the separation procedure which involved coprecipitation with iron hydroxide, fil-

tering, wet ashing the filter, the precipitate, and all insoluble matter with nitric acid under pressure, fuming with hydrofluoric and again with nitric acid, followed by anion exchange separation of uranium and plutonium on BioRad AG 1 $\times$ 4, 100 to 200 mesh, in hydrochloric and nitric acid media. The cation exchange separation of americium and curium in the effluent from the anion column was based on a procedure reported by Hiatt and Hahn (1980). The actinides were electro-deposited onto stainless steel discs and determined by alpha spectrometry.

Aerosol samples were collected on 20 cm diameter metal plates by electrostatic precipitation and were analysed directly by alpha spectrometry in large area Frisch grid ionization chambers (Hötzl and Winkler, 1978).

Neptunium-239 was determined by gamma spectrometry in deposition and air samples collected for several hours. Continuous study of these samples showed that fallout from Chernobyl commenced during the night, April 29th and 30th and reached a maximum on April 30th.

Results

The actinide nuclide results are shown in Tables 1 and 2 and include ^{137}Cs for comparison. With the exception of ^{234}U and ^{238}U , the actinide nuclides deposited exhibit a pattern of variation with time similar to ^{137}Cs and the other nuclides indicating their common link with the Chernobyl accident.

Table 1. Ground deposition (Bq/m^2)* of actinide nuclides at Munich-Neuherberg, (GSF), 29 April to 10 May, 1986.

	29 Apr/9:30 to 30 Apr/9:30	29 Apr/9:30 to 2 May/9:30	2 May/9:30 to 10 May/8:30	Sum
Rainfall (mm)	21.2	27.3	5.4	32.7
U-234	n.d.**	20E-3	17E-3	37E-3
U-237	n.d.	25	1.9	27
U-238	n.d.	22E-3	16E-3	38E-3
Np-239	≤ 170	1000-1200	≤ 100	1000-1300
Pu-238	$\leq 4\text{E}-3$	14E-3	7.4E-3	21E-3
Pu-239,240	$\leq 3\text{E}-3$	40E-3	11E-3	51E-3
Am-241	$\leq 17\text{E}-3$	$\leq 19\text{E}-3$	$\leq 9\text{E}-3$	$\leq 28\text{E}-3$
Cm-242	$\leq 20\text{E}-3$	0.44	0.16	0.60
Cs-137	590	17.5E + 3	1.7E + 3	19.2E + 3

*Corrected for radioactive decay to the middle of each sampling period.

**n.d.: not determined.

Plutonium is the element of potentially greatest public interest. The high ^{238}Pu : $^{239+240}\text{Pu}$ ratio 0.42 ± 0.03 of the plutonium isolated in the Munich laboratory indicated that it was of reactor origin. In weapons fallout this ratio is about 0.03–0.05. The $^{239+240}\text{Pu}$ deposited in the fallout from Chernobyl (Fig. 1) is comparable to the monthly plutonium deposition values observed in Munich during the 1970's (Hötzl *et al.*, 1983). The total soil inventory of $^{239+240}\text{Pu}$ which in Munich (Hötzl *et al.*, 1983; Hardy *et al.*, 1973) amounted to about 110 Bq m^{-2} has increased by about 0.05%. This increase is low compared to the increase by a factor of about 5 in the ^{137}Cs content of total soil inventory in Munich.

The presence of Neptunium-239 has been reported by several authors in Scandinavia (Devell *et al.*, 1986; SSI, 1986). The 103.7 and 106.1 keV gamma lines were detected in the early more active air and deposition samples collected in Munich on April 30th and during the first few days in May. Interference from ^{132}Te precluded the evaluation of the main ^{239}Np 228 keV gamma line. The ratio of ^{239}Np to ^{137}Cs observed in Munich was approximately one order of magnitude smaller than in Scandinavia.

Curium isotopes build up in power reactors and only

small quantities are released in plant effluent (Rosner *et al.*, 1978). However, measurements of curium in the environment are rare and usually restricted to the vicinity of weapons testing sites and nuclear fuel reprocessing plants. As a consequence of the Chernobyl accident, curium has been released into the general environment. However, in our measurements ^{244}Cm and ^{241}Am were both below the limit of detection.

The ^{238}U : ^{234}U ratio of the uranium isolated from the fallout indicated that it was of natural composition. Also, the much smaller variation with time exhibited by the uranium compared to the other radionuclides suggested that the main source was the comparatively constant natural deposition. The cumulative uranium deposition was consistent with the $8\text{--}70 \text{ mBq m}^{-2}$ monthly deposition range observed in Munich during a three year period. However, the short lived ^{237}U ($T_{1/2} = 6.75$ days) observed in the uranium samples by its 59.5 keV gamma line and by its decay, indicated that a part at least of the uranium isolated originated from Chernobyl.

The ratios of the alpha emitters observed in Munich agree with ratios calculated from the information given in the Soviet Union's report to the International Atomic Energy Agency (USSR, 1986). In table 4.14

Table 2. Activity concentrations (Bq/m^3)* of actinide nuclides in ground level air at Munich-Neuherberg (GSF), 28 April to 12 May, 1986.

	28 April to 1 May	1 May to 5 May	5 May to 12 May	Time integral ($\text{Bq} \cdot \text{h}/\text{m}^3$)
Np-239	0.09	≤ 0.06	—	4–7
Pu-239/Pu-240	$\leq 12\text{E}-6$	$\leq 8\text{E}-6$	$\leq 4\text{E}-6$	$\leq 2\text{E}-3$
Pu-238/Am-241	$\leq 6\text{E}-6$	$\leq 7\text{E}-6$	$\leq 2\text{E}-6$	$\leq 1.5\text{E}-3$
Cm-242	10E-6	25E-6	4E-6	3.8E-3
Cs-137	1.3	2.0	0.10	300

*Radioactive decay corrected to the middle of each sampling period.

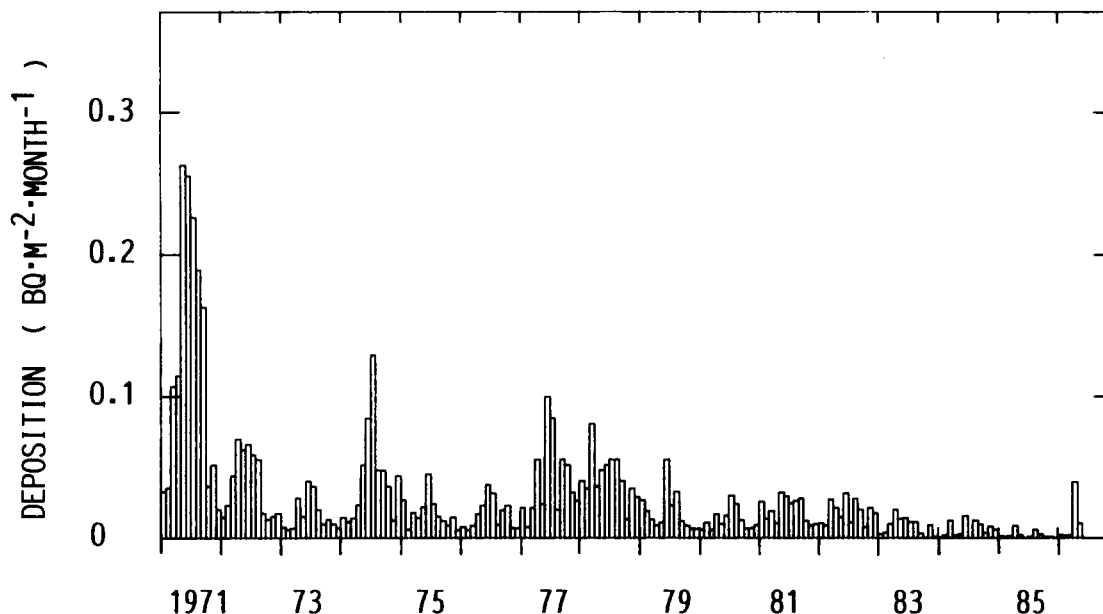


Fig. 1. Monthly ground deposition of Plutonium-239, 240 at Munich from 1971 to May 1986.

of this report the quantities of ^{238}Pu , $^{239+240}\text{Pu}$, and ^{242}Cm released by May 6, 1986, are given as $0.8 \cdot 10^{-3}$, $1.8 \cdot 10^{-3}$ and $2.1 \cdot 10^{-2}$ MCi, giving release ratios $^{238}\text{Pu}:^{239+240}\text{Pu}:^{242}\text{Cm}$ of 0.44:1:12, while our cumulative deposition values by May 10, 1986 at Munich, (Table 2) give 0.42:1:12. This indicates negligible fractionation among these nuclides during their passage to Western Europe.

On the other hand, it appears that the actinide nuclides were depleted strongly with respect to other radionuclides during their passage from Chernobyl. The Soviet Union's report (USSR, 1986) gives the quantity of ^{137}Cs released by May 6, 1986, as 1 MCi. The calculated release ratio $^{239+240}\text{Pu}:^{137}\text{Cs}$ was $1.8 \cdot 10^{-3}$ by May 6, 1986 compared to the deposition ratio $2.5 \cdot 10^{-6}$ measured in Munich by May 10, 1986. The inference is that at locations closer to Chernobyl higher $^{239+240}\text{Pu}:^{137}\text{Cs}$ ratios are likely to be encountered.

Of the actinides reported here, the ratio least depleted appeared to be $^{239}\text{Np}:^{137}\text{Cs}$. The value calculated of the release ratio for $^{239}\text{Np}:^{137}\text{Cs}$ was 7 for April 30th compared to the ratios in deposition and air of 0.06 and 0.07, respectively, measured in Munich on the same date.

Finally, the radiation exposure of the population due to actinides from Chernobyl, is negligibly small compared to doses from the more abundant radionuclides such as ^{137}Cs and ^{131}I . For Southern Bavaria, we estimate that the plutonium isotopes contribute only 0.1% of the total effective dose equivalent from Chernobyl fallout. This dose has been calculated to be 0.5 to 1.1. mSv for a person living in Southern Bavaria during the first year after the Chernobyl accident (SSK, 1987).

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