

Measurements of Transuranium Nuclides in the Environment at the Institute for Radiation Protection of the Gesellschaft für Strahlen- und Umweltforschung mbH, Munich

G. Rosner, H. Hötzel and R. Winkler

Institut für Strahlenschutz der Gesellschaft für Strahlen- und Umweltforschung mbH, München,
Federal Republic of Germany

Work on environmental transuranium nuclides at the Institute for Radiation Protection of the Gesellschaft für Strahlen- und Umweltforschung mbH, Munich, is briefly described and standard needs are discussed. Fallout plutonium measurements in air dust and precipitation samples started in 1970/1971. The procedure is outlined and results are presented as annual mean and sum values, respectively. Since 1973, transuranium nuclides in primary coolant, stack effluent air and waste-water samples from nuclear power stations are measured. Nuclides detected are $^{239/240}\text{Pu}$, ^{238}Pu and/or ^{241}Am , ^{242}Cm and ^{244}Cm . Examples of alpha particle spectra are given. Needs for standards in environmental transuranium analysis are discussed.

(Transuranium nuclides; environment; fallout; nuclear reactor effluents; primary coolant; coprecipitation; cold ashing; Frisch-grid ionization chamber)

Introduction

Environmental levels of transuranium nuclides are of continuously growing public interest. There are, roughly, two main sources for the entry of transuranium nuclides into the biosphere: (i) from nuclear explosions via the fallout, and (ii) from nuclear installations, as e.g. power reactors, reprocessing plants, via the effluents. Accordingly, the Radiochemical-Analytical group at the Institute for Radiation Protection of the Gesellschaft für Strahlen- und Umweltforschung mbH Munich has started their transuranium measurements with fallout plutonium in 1970, and continued later, in collaboration with the Federal Health Office – Bundesgesundheitsamt – with a study on levels of transuranium nuclides in emissions from nuclear power reactors.

Fallout plutonium

Fallout plutonium has been determined monthly in air and precipitation samples since 1970 and 1971, respectively.

Procedure and results

Aerosol samples from about 40 000 m³ of ground level air are taken continuously every month on micro-sorbane filters and are dry ashed at 450°C. Monthly deposition samples are taken in two collectors of 0.6m² surface each, and are then evaporated and ashed with nitric acid.

The subsequent treatment is identical for both kinds of samples: They are treated with perchloric, hydrofluoric and again perchloric acid, iron is removed by extraction from

hydrochloric acid and polonium is deposited on silver foil. Plutonium is then separated by coprecipitation on iron cupferronate, and finally the plutonium is electrodeposited on stainless steel discs from weakly acid ammonium sulphate solution. Alpha-particle spectrometric measurements are then carried out in a 20-cm-dia. Frisch-grid ionization chamber.

Detection limits of our procedure are 40 fCi/m² month $^{239/240}\text{Pu}$ in deposition and 1 aCi/m³ in air; for ^{238}Pu the values are 70 fCi/m² month in deposition and 2 aCi/m³ in air. The detection limits are based on three times the standard deviation of the background. More details on the entire procedure are given in (Hötzel, 1976a).

An example of the spectra obtained is shown in Fig. 1. The spectrum shows the lines of $^{239/240}\text{Pu}$, ^{238}Pu and tracer ^{236}Pu ; other lines can not be detected. The results of our monthly measurements, summarized as mean annual values, are shown in Table 1.

A comparison of the results on the plutonium content in ground level air at some independent stations may be of some interest, as shown in Table 2. Here, the very few available concentration values from stations of similar geographic latitude (Wehner, 1971; EJNR, 1970; Cambray, 1974; HASL, 1976) are compared by calculating the ratios of activity concentrations, normalized by putting the Neuherberg values = 1. The overall agreement can be considered as good. However, a more rigorous analysis of these data, such as for the results of an intercomparison run, does not seem to us to be appropriate because of the large number of parameters involved in the overall procedure. Such a comparison, although of interest by its own right, can, however, not replace an intercomparison run, which certainly would be of considerable help to many workers in this field.

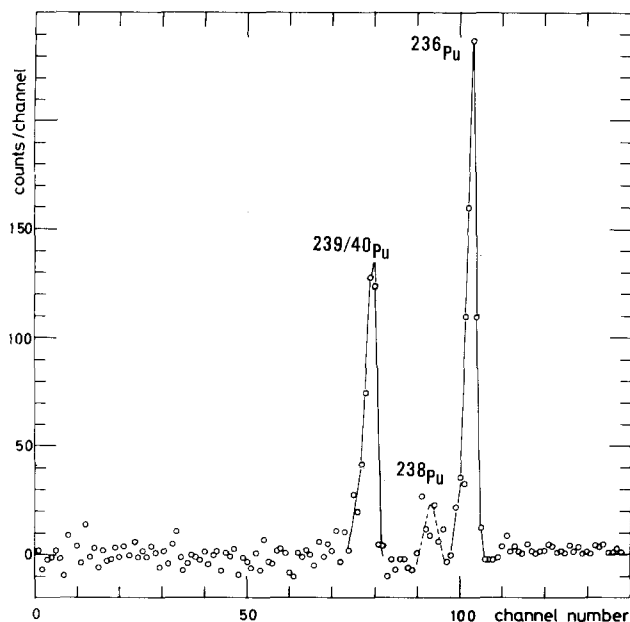


Fig. 1. Alpha-particle spectrum of plutonium isolated from an air dust sample (sampling period: June 1972; counting time 22.2 hours; $^{239/240}\text{Pu}$ content 0.37 pCi).

Table 1. Plutonium 239/240 in ground level air and precipitation at the GSF site, Neuherberg near Munich

Year	Concentration in ground level air aCi/m ³	Concentration in precipitation pCi/l	Deposition to ground pCi/m ² year
1970	50 ⁺	—	—
1971	61	62	40
1972	23	19	13
1973	12	7,5	5,5
1974	32	15	14
1975	24 ⁺	7,1	6,1
Mean	35	22	16

⁺: 7 months only

—: Measurements started in 1971

Table 2. Comparison of Neuherberg Plutonium-239/240 values to those of other authors at sites of similar geographic latitude, normalized to Neuherberg values = 1

Station	AIR				DEPOSITION
	PTB Bunswick	INRC Ispra	AERE Chilton	HASL New York	INRC Ispra
1970 ⁺	0,73	0,98	1,39	1,75	=
1971	1,08	1,09	1,14	1,03	0,41
1972	1,17	0,86	0,70	0,85	0,48
1973	—	—	1,05	1,16	—
1974	—	—	—	1,16	—
1975 ⁺	—	—	—	1,19	—

⁺: 7 months

=: No Neuherberg values available

—: Values from other stations not or not yet available

Standard used

The tracer nuclide used in our plutonium fallout work is ^{236}Pu . We consider it as very well suitable for this purpose, and our sincere thanks are due to Dr. J. Harley for having made it available to us.

Alpha emitters in power reactor effluents

From the radiological point of view, the question of the presence of alpha emitters in airborne and liquid effluents from nuclear power stations is now becoming of more and more interest, while the plutonium fallout values continue to serve as background information. In our institute, since 1973, primary coolant water samples from one, stack aerosol samples from several and effluent water samples from all nuclear power stations in the Federal Republic of Germany have been investigated for alpha emitters.

Procedure and results

While the sampling is done by the reactor operators themselves, samples are prepared by us for counting and are then counted in a 20-cm Frisch-grid ionization chamber. Stack aerosol samples on their asbestos filters are "cold ashed" in an excited oxygen plasma, primary coolant samples are evaporated on stainless steel planchets, and to waste water samples a technique consisting of coprecipitation on iron hydroxide, subsequent filtration and "cold ashing" is applied. Some alpha-particle spectra are shown in Figs. 2, 3 and 4. Figure 2 is an alpha-particle spectrum of a

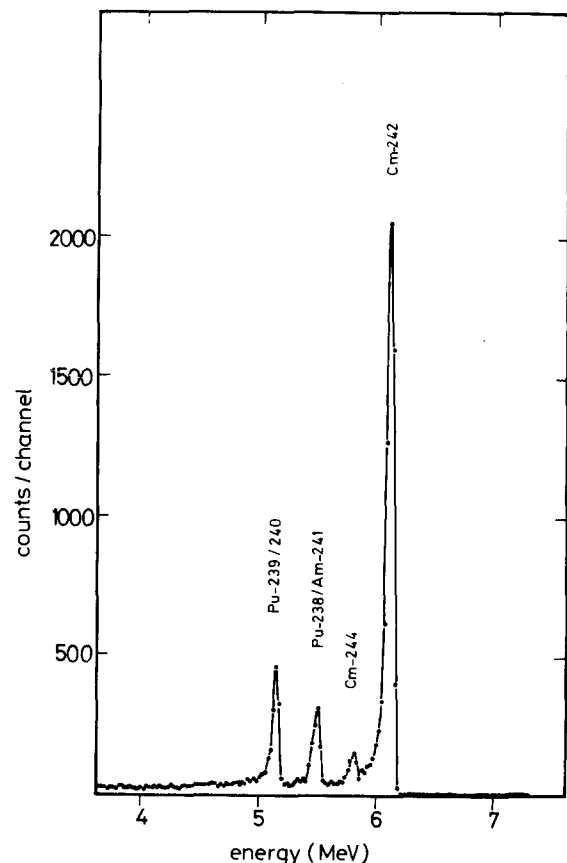


Fig. 2. Alpha-particle spectrum of a 200 ml primary-coolant-water sample evaporated onto a stainless steel planchet. Counting time: 80 000 s in a 20-cm-diameter Frisch-grid ionization chamber.

primary coolant sample, illustrating the occurrence of alpha emitters and taken in order to learn which transuranium nuclides can in practice be expected to occur in reactor emissions; Fig. 3 shows the alpha-particle spectrum from a waste water sample, and in Fig. 4 the alpha-particle spectrum of a stack aerosol sample can be seen. Alpha emitters detected are $^{239/240}\text{Pu}$, ^{238}Pu and/or ^{241}Am , ^{242}Cm and ^{244}Cm . More details are given in (Hötzl, 1976b).

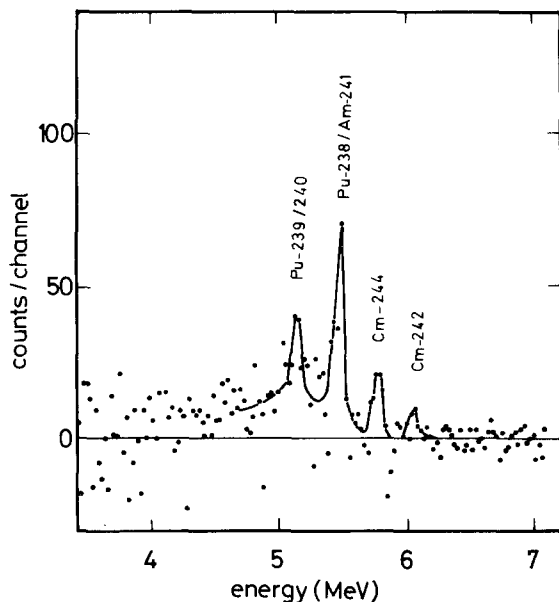


Fig. 3. Spectrum of alpha emitters in a power-reactor waste-water sample. Sample preparation: coprecipitation on iron hydroxide and "cold ashing" in excited oxygen. Counting time: 130 hours in a 20-cm-dia Frisch-grid ionization chamber.

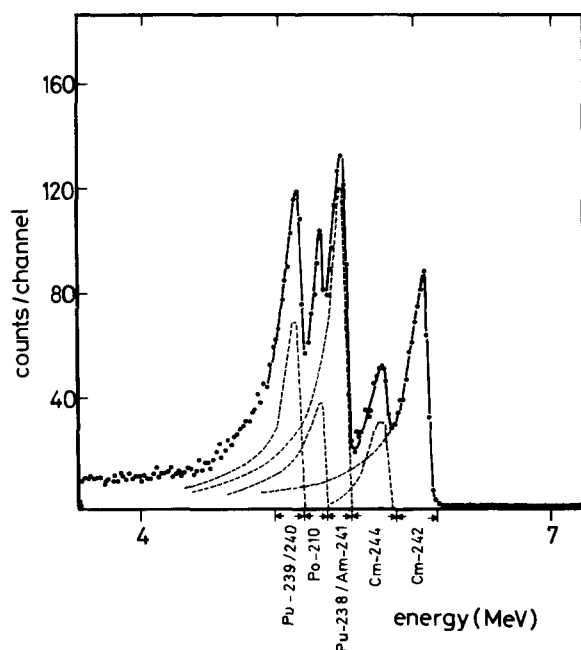


Fig. 4. Alpha-particle spectrum from a stack aerosol sample on asbestos filter. Sample preparation: "cold ashing" in excited oxygen. Counting time: 80 000 s in a 20-cm dia Frisch-grid ionization chamber.

Standards needed for future work

When discussing the needs for standards in an environmental transuranium analysis, one must keep in mind that the term "standard" covers a broad range of meanings. In many instances, preference is given to standards in the natural matrix. For our own work, standards of interest would be air filters and fresh-water samples, the latter comprising also fresh water sediments and biota, as well as some foodstuff samples.

In a somewhat different manner, the term "environmental standard" has been used, e.g. in the National Bureau of Standards catalogue (NBS, 1975–76), for a calibrated solution of a radionuclide of low activity. In our opinion, an important problem, especially for smaller laboratories not associated to large nuclear research centers, is the availability of suitable transuranium nuclides, particularly in that form.

In plutonium analysis, a useful tracer nuclide would be ^{242}Pu . Due to its low alpha energy, ^{242}Pu does not cause interferences in the $^{239/240}\text{Pu}$ and ^{238}Pu energy region, as ^{236}Pu may do especially in the ^{238}Pu region. Although in principle, after sufficient chemical separation, preparation of practically carrier-free sources with negligible tailing corrections always remains possible, there may be practical reasons requiring measurements of samples of finite thickness, e.g. in surveillance. In this case, the use of ^{242}Pu would be greatly advantageous. Moreover, because of its low specific activity ^{242}Pu may serve as a kind of isotopic carrier for the shorter lived $^{239/240}\text{Pu}$ and ^{238}Pu .

We appreciate very much, that in the NBS (1976) catalogue ^{242}Pu is announced as "in preparation". The standardized form we propose would be a solution similar to those already existing as environmental standards for other transuranium nuclides, e.g. NBS ^{239}Pu or LMRI ^{241}Am (LMRI, 1974).

For americium-241 analysis, the necessary ^{243}Am standard is listed already in the NBS (1976) catalogue as environmental standard.

Finally, ^{242}Cm , although less important from the radiological point of view than plutonium or americium, has often the highest activity concentration in power reactor emissions. Often it is accompanied by smaller quantities of ^{244}Cm . For a simultaneous determination of both ^{242}Cm and ^{244}Cm in the same sample, a tracer other than these currently available nuclides would be needed. A suitable tracer for such a simultaneous determination is ^{245}Cm or ^{246}Cm . We do not know how difficult it would be to prepare these nuclides, in practice, as standards, but we do not hesitate to add them to our list of wanted nuclides – although not with the same priority as ^{242}Pu .

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Discussion

Bowen: In considering our needs for tracer nuclides and for standard nuclide samples, it is important to bear in mind the uses that are to be made, in reference to the requirement for radiochemical purity of the materials. A nuclide standard that is to be used for calibration of energy dispersion or of detector efficiency in spectrometry can often be quite impure in respect to other nuclides. Contrariwise, a nuclide that is to be used as a yield monitor, and consequently that will be actually added to samples being analyzed, should be of the highest radiochemical purity.

In the latter case also problems may arise simply because of the non-availability of suitable nuclides that are not also to be sought in analysis. Curium is a good example, offering only two yield monitor possibilities, both of which are environmental contaminants. We have, by serendipity, met this problem by developing a method that

separates curium together with americium, and with identical yields; thus we can use ^{243}Am as yield monitor for both these transuranics.

It is a pleasure to see that the Federal Republic of Germany is so far ahead of us in examining its reactor effluents. We have only just begun, in my laboratory, with support from our Nuclear Regulatory Commission, a study of the transuranic nuclides in reactor effluents, their biological availability and their fate in aquatic environments. *Gans:* The investigations on the isotopic composition of the α -activity in liquid effluent from nuclear power stations in Germany were carried out in 1973–74 on monthly composite samples. The specific nuclide analysis was dropped in 1975 and only gross α -measurements are done since then. The reason is that the total α -activity of each of the nuclear power plants in the Federal Republic of Germany, that is released each year, is less than 1 mCi. This activity is not important in public health respects. Nevertheless we are thinking about taking up specific measurements maybe on quarterly composite samples.

Bowen: Although one can understand the reasons, both financial and practical, for such a reduction in program as Dr. Gans describes, there is another context in which it appears unfortunate. This is the problem posed by anticipated massive increases in the amounts of waste curium radioactivity and by our need to predict the biological and environmental behaviour of this transuranic element. Curium has not been measurable in world-wide fallout; so the only environmental data available as bases for prediction come from a variety of waste disposal operations. Curium is readily measurable in power reactor effluents, and serious thought should, I am convinced, be given to continued study of the transuranics in these situations just to extend our knowledge of the behaviour patterns of this problem element of which we now know so very little.