Pu-241 AND Am-241 IN THE ENVIRONMENT

E. Holm* and R.B.R. Persson**
*International Laboratory of Marine Radioactivity, IAEA,
Musée Océanographique, Monaco
**Radiation Physics Department, Lasarettet, 221 85 Lund, Sweden

I. INTRODUCTION

The global inventory of plutonium isotopes indicates that the highest activity is represented by Pu-241 of which about 6000 kCi have been released by the atmospheric nuclear weapons tests as world-wide fallout (1,2). Another 2000 kCi of Pu-241 might be locally deposited near the test sites (3). Pu-241 is a rather short-lived beta emitting radionuclide which decays with a half-life of about 13 years to Am-241 which is long-lived (half-life 458 years) and is an alpha-emitter. Thus, due to the decay of fallout Pu-241 the activity of Am-241 in the environment will increase with time and reach the same activity-level as Pu-239 (2000 kCi) in about 70 years. A small amount of Am-241 is, however, present in the fabricated weapons but in the fallout it is mainly derived from the decay of Pu-241. In the effluent from nuclear reprocessing plants the Pu-241/Pu-239+240 activity ratio as well as the Am-241/Pu-239+240 activity ratio can be much higher than that in fall-out from nuclear explosions. Some observations also indicate that americium from reprocessing plants might be more easily available to biota than that originating from nuclear weapons tests (4). In the thermonuclear test explosion 'Ivy Mike' November 1952, the Pu-241/Pu-239+240 activity ratio was reported to be as high as 28 (5).

2. SAMPLING ANALYSIS AND MEASUREMENTS

Samples of lichen (Cladonia alpestris) have been collected since 1961 from the Lake Rogen district in central Sweden (62.3 N, 12.4 E). A standardized technique using a frame over an area of 0.25 m^2 was employed. The samples had previously been analyzed for Pu-238 and Pu-239+240 (6). We have also analyzed one sediment sample collected in July 1975 from the Irish Sea, and one sediment sample collected during 1970 from the Bikini Atoll (Bravo Crater). These sediment samples had been analyzed previously for Pu-239+240 and Am-241 (4) and now about one year later also have been analyzed for Pu-241. The analysis of Pu-241 is performed by separating the Am-241 which is derived from the previously electroplated Pu-241. The Am-241 has been analyzed in the samples by ion-exchange separation in a nitric-acid/methanol medium (7). All samples were electroplated and counted with surface barrier detectors connected to a multi-channel analyzer and the activities of Pu-241 and Am-241 were corrected to the date of collection. The radiochemical yield of the radiochemical separations was monitored by addition of Pu-236 for Pu-isotopes and Am-243 or Cm-244 for Am-isotopes.

3. RESULTS AND DISCUSSION

The activities of Pu-241 and Am-241 lichen corrected to the date of collection during 1961-1975 are given in Fig. 1 together with the Pu-239+240 values determined previously (6). The activity concentrations of all plutonium isotopes in lichen show maxima for the period 1963-1966 of

about 4000 pCi per kg dry weight for Pu-241 and 300 pCi per kg dry weight for Pu-239+240. These maxima are caused by the larger nuclear weapons tests during 1961-1962.

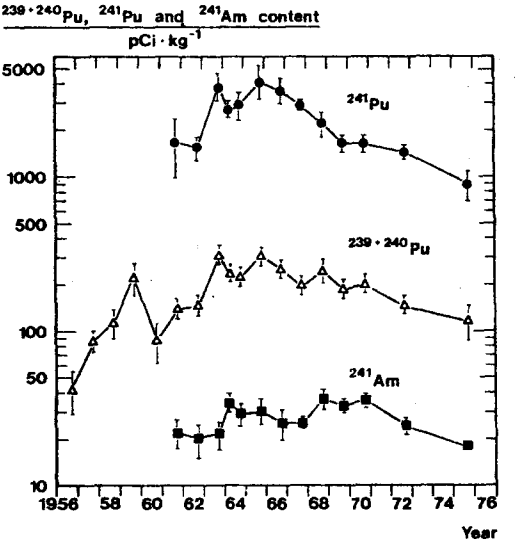

Fig. 1 The temporal variation of the activity-concentrations of Pu-241, Pu-239+240 and Am-241 in lichens collected in central Sweden.

Because most Am-241 is mainly formed in situ by the decay of Pu-241 it does not show a corresponding maximum. The maximum activity concentration of Am-241 appears much later (1968-1970) and is then about 35 pCi/kg dry weight. The activity ratios Pu-241/pu-239+240, Am-241/Pu-239+240 and Am-241/Pu-241 in lichen are presented in Fig. 2. The Pu-241/Pu-239+240 activity-ratio decreases from about 14 in 1964 to about 7 in 1975 which is mainly due to the physical decay of Pu-241. The Am-241/Pu-239+240 activity ratio shows a quite irregular pattern during the period 1961-1963 which is probably due to the presence of Am-241 in the fallout during this period. After 1967 this ratio increased to a more stable value of about 0.17 in the lichen carpet. The Am-241/Pu-241 activity ratio in lichen rises smoothly to a value of about 0.023 in 1970 but then decreased slightly. This might be an indication that Am-241 is more easily accessible in the lichen plant than Pu-241.

The biological mean residence time for plutonium in the lichen carpet was estimated previously as 6 $\pm$ 0.5 years by using a simple compartment model for the area contained in the lichen carpet (1). The deposition-rate of Am-241 was estimated by using the Am-241/pu-241 activity-ratio of surface air and the Pu-241 deposition curve derived previously (1,5). For Am-241, however, one also has to take the in situ build up into consideration and the biological mean residence time for americium in the lichen was estimated as 3 $\pm$ 1 years.

The activity levels and activity-ratios for sediments, lichen and global fallout are summarized in Table 1.

Environmental sample	Pu-239+240	$\frac{\text{Pu-241}}{\text{Pu-239+240}}$	$\frac{\text{Am-241}}{\text{Pu-239+240}}$	$\frac{\text{Am-241}}{\text{Pu-241}}$
Global fallout	1.0±0.1*	7.0±1.0	0.24±0.02	0.034
Reindeer lichen	0.12±0.03**	8.0±1.0	0.16±0.02	0.02
Bikini sediment	83 ± 3**	8.2±0.5	0.58±0.02	0.077
Windscale sediment	110 ± 4**	22±2	2.0±0.1	0.09

(*) nCi/m^2 (**) nCi per kg dry weight

Table 1. The activity content and various activity ratios of Pu-239+240, Pu-241 and Am-241 in various types of environmental samples. (Activity corrected to Oct. 1975).

REFERENCES

(1) HOLM, E., PERSSON, R.B.R. Radiochemical studies of Pu-241 in reindeer lichens. Health Phys. (submitted Aug. 1976).

(2) KREY, P.W., et al. "Mass isotopic composition of global fallout plutonium in soil", p. 671, Transuranium Nuclides in the Environment, IAEA, Vienna (1976).

(3) WRENN, McD. E. "Environmental levels of plutonium and the transplutonium elements", p. 89, EPA Plutonium Standard Hearings Washington D.C., WASH-1359 (1974).

(4) BEASLEY, T.M., FOWLER, S.W., Plutonium and Americium: Uptake from contaminated sediments by the Polychaete Neris diversicolor. Marine Biol. 38 (1976) 95.

(5) THOMAS, C.W., PERKINS, R.W. "Transuranium elements in the atmosphere". Rpt. BNWL-1881, UC-48, Battelle Pacific NW Lab., Richland, Wash., USA (1974).

(6) HOLM, E. PERSSON, R.B.R. Fallout plutonium in Swedish reindeer lichens. Health Phys. 29 (1975) 43.

(7) HOLM, E., FUKAI, R. Determination of americium and curium by using ion-exchange in nitric-acid/methanol medium for environmental analysis. Talanta 23 (1976) 853.

(8) HETHERINGTON, J.A., et al. "Environmental and public health consequences of the controlled disposal of transuranic elements to the marine environment", p. 139, Transuranium Nuclides in the Environment, IAEA, Vienna (1976).

(9) LIVINGSTON, H.D. et al. Pu-241 in the marine environment by a radiochemical procedure, Earth and Plan. Sci. Lett. 25 (1975) 361.

(10) NOSHKIN, V.E., et al. "Transuranics and other radionuclides in Bikini Lagoon: Concentration data retrieved from aged coral sections". UCRL-75785, Lawrence Livermore Lab., Calif. USA (1975).

By analyzing profiles of lichen carpet and soil layers the accumulated area content of Am-241 was determined to be 210 ± 20 pCi/m^2 in 1972; the corresponding value for Pu-241 was 7800 ± 900 pCi/m^2.

The results of present investigations indicate that Am-241 was present in the environment from nuclear weapons tests prior to 1960, though the U.S. and U.S.S.R. tests series in 1961-1962 comprise the major input. In 1975 about 15-20% of the Am-241 in the lichen carpet originated from direct fallout and 80-85% are derived by in situ build up from Pu-241.

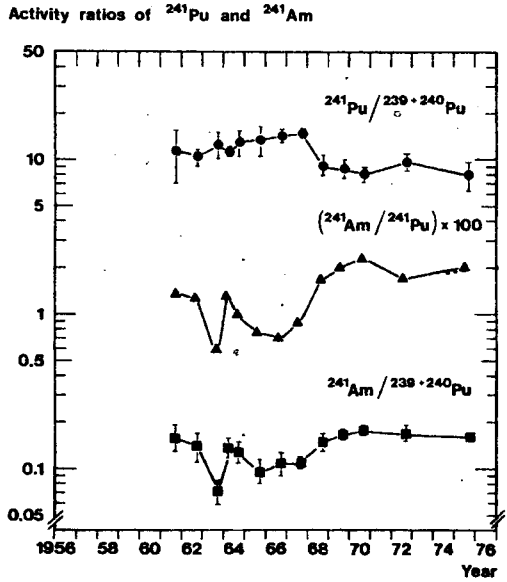

Fig. 2 The temporal variation of the activity-ratios Pu-241/Pu-239+240, Am-241/Pu-241 and Am-241/Pu-239+240 in lichen carpets.

Results of the analysis of the Windscale sediments give an activity ratio for Pu-241/Pu-239+240 of 22 ± 2, in good agreement with the activity ratio in the effluent which is reported to be about 10-100 (8). The corresponding ratio for the IAEA intercalibration samples AG-I-1 (seaweed) and SW-I-2 (seawater) which are collected in the same area as the sediment sample is about 30 (9). For the Bikini sediment samples a lower value of the Pu-241/Pu-239+240 activity ratio was found. In 1976 this was 8.2 ± 0.5, and, corrected for physical decay to 1954 it became 25. This is slightly less than the value 3 which has been observed in coral sections from the same place (10).