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Biophysical Aspects of Am-241 and Pu-241 in the Environment

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Summary. Most of plutonium released by nuclear explosions is Pu-241 which decays to Am-241. We have studied the deposition of Pu-241 and Am-241 in lichens collected since 1958 in the central part of Sweden (62.3° N, 12.4° E). Comparative studies with Pu-isotopes, Pu-239 + 240 and Pu-238 were also performed. In 1972 the total accumulated deposition of Pu-241 was 8 mCi/km^2 of Pu-239 + 240 1 mCi/km^2 and of Am-241 0.2 mCi/km^2. About 80% of the Am-241 activity has been formed in situ from decay of Pu-241. The biological mean-residence time for all Pu-isotopes were about 6 years and for Am-241 4 years. The spatial distribution of Am-241 in the lichen carpet is quite different from that of Pu-241.

The activity concentrations of Am-241 and Pu-241 have been studied in reindeer liver and bone. The average concentrations found were in liver 0.6 and 40 pCi/kg, in bone 0.2 and 6 pCi per kg for Am-241 and Pu-241 respectively. The activity content of Am-241 and Pu-241 in the Lapps due to their reindeer diet was estimated to be in liver 1.0 E-4 and 1.0 E-2 pCi/kg, in bone (3–9) E-5 and 1.0 E-2 pCi/kg for Am-241 and Pu-241 respectively. The estimated values for the fractions of ingested activity retained were in liver 7 E-6 and 14 E-6, in bone 20 E-6 for Am-241 and Pu-241 respectively.

The fraction of ingested activity of Pu retained in reindeer liver is about 2–3 times higher than that of Am.

1. Introduction

Most of the transuranium elements now present in the environment has been released from atmospheric nuclear explosions. In Table 1 is given the activity and amount of the transuranium nuclides now present in the environment from nuclear explosions.

There are at present increasing releases of transuranium nuclides into the sea from nuclear fuel reprocessing plants. Great concerns must be taken about the environmental health impact of the transuranium elements because they are mostly

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Table 1. The activity and amount of transuranium nuclides now present in the environment from nuclear explosions (Holm, 1977; Holm and Persson, 1978)

Nuclide	Mode of decay	Half-life	Activity (kCi)
Np-237	α	2.14×10^6 a	0.1
Np-239	β^-	2.35 d	—
Pu-238	α	86.4 a	8.5
Pu-239	α	24 400 a	210
Pu-240	α	6 580 a	140
Pu-241	β^-	14.2 a	2 450
Pu-242	α	3.79×10^5 a	0.6
Am-241	α	458 a	95
Am-242	β^-, EC	16 h	
Am^m-242	IT	152 a	0.02
Cm-242	α	163 d	0.02
Cm-243	α	32 a	
Cm-244	α	17.6 a	0.02

alpha emitters of high potential biological hazard. These releases are therefore often limited by the amount of alpha activity. The release of alpha emitting radionuclides are Am-241 and Pu-239 + 240 and minor amounts of curium isotopes. There is, however, also a significant release of the beta emitter Pu-241 which decays in situ to the alpha emitter Am-241. The Am-241 thus formed might be in form of "hot atoms" of different bioavailability than the Am-241 released directly. The bioavailability of environmental americium is still not very well known and contradictory results appear in the literature. Some investigators found higher uptake of americium than plutonium while others found the contrary (Pentreath and Lovett, 1976; Beasley and Fowler, 1976). Most of such investigations have however been laboratory experiments or not taking in vivo build up of Am-241 from Pu-241 into account.

The investigation of the food chain lichen-reindeer offers an unique opportunity to study the uptake of transuranium elements from fallout under "natural" conditions. We have previously studied the uptake and transfer of various isotopes of fall out plutonium in this food-chain (Holm and Persson, 1975, 1976, 1977a, b). In our investigations of Pu-241 and Am-241 we have found indications of a quite different distribution in the lichen carpet. This might be due to different incorporation rates to biological matter for plutonium and Am-241 formed in situ. These aspects will be described in more detail in the present investigation. By studying both americium and plutonium in lichen, which is the main food for reindeer during winter, and in different tissues of reindeer we are able to compare the uptake and transfer of these transuranic elements in the food-chain lichen-reindeer. We are also able to draw some conclusions concerning the uptake and retention of plutonium and americium in man.

2. Sample Collection, Analysis and Measurements

2.1 Collection of Samples

Samples of lichen (*Cl. alpestris*) have been collected annually since 1961 from the Lake Rogen district in central Sweden (62.3° N, 12.4° E). A standardized sampling technique using a frame with an area of 0.25 m^2 was employed (Svensson and Lidén, 1965). The lichen carpet was fractionated into different layers as shown in Figure 2. The fractionation was made directly upon sample collection, however, prior to 1966 it had been carried out a few weeks after collection. During the storage of the samples some Pu-241 decays to Am-241. Therefore the Pu-241 activity concentration must be known for old samples to calculate the Am-241 activity at the date of collection. The samples have previously been analyzed for Pu-238 and Pu-239 + 240, and the results for the period 1945–1972 have been published elsewhere (Holm and Persson, 1975, 1976, 1977a, b).

2.2 Analysis of Am-241

The samples were dried at 105 °C for 24 h. A suitable quantity (5–10 g) was ashed after 1 pCi of Cm-244 or Am-243 had been added as yield-determinant. The ash was leached with aqua regia and hydrogen peroxide. The ashed samples were dissolved in 9 M HCl and were then first passed through an anion exchanger (Dowex 1X8, 100–200 mesh) which adsorbs U, Fe, Po, and Pu and secondly through an cation exchanger (Dowex 50X8, 200–400 mesh) which adsorb Th. Americium, curium and lanthanides will pass through the system. After evaporation of the americium and curium fraction the trivalent actinides were sorbed on an anionic resin (Dowex 1X8, 100–200 mesh) from a solution of 1 M HNO_3 in 93% methanol. Lanthanides were eluted with 0.5 M ammonium thiocyanate in 80% methanol 0.1 M HCl. Americium and curium were eluted with 1.5 M HCl 86% Methanol. This leaves any traces of Pb-210 and Po-210 on the column. This procedure is a modification of an americium separation procedure previously described by Holm and Fukai (1976).

After evaporation, Am-241 was electroplated on stainless steel discs from 1.5 M ammonium sulfate at pH 2.5 using disposable electrodeposition cells (Talvitie, 1972). The alpha spectra of the samples were recorded with 300 mm^2 surface barrier detectors, connected to a multichannel analyzer. The width of the 5.49 MeV alpha peak at half maximum was 40–60 keV. Since the average energies of the alpha particles from Am-241 and Am-243 are 5.49 and 5.28 keV respectively these peaks are well separated. The average radiochemical yield of Am-241, determined with Am-243 as yield determinant, was found to be 70 ± 20%. The samples were measured during 1000–6000 min in order to obtain a coefficient of variation of about 10% in the counting statistics of Am-241.

At extremely high Po-210 or Pb-210 content, Cm-244, which has the same chemical yield as americium, was used as yield determinant because the 5.3 MeV alpha particles from Po-210 could interfere with the 5.28 MeV alpha particles from

the Am-243 which is used in other cases. The Am-241 activity concentration, CAm, at the collection date can be calculated according to the expression:

$$CAm = A2 - CPu^*[\exp(-\ln 2^*t/T2) - \exp(-\ln 2^*t/T1)]^* \frac{T1}{T2 - T1}.$$

Thus

$$CAm = A2 - \frac{CPu^*(1 - kt)}{31.3}.$$

CAm = Am-241 concentration at collection date (pCi/kg);
A2 = measured Am-241 concentration (pCi/kg);
CPu = Pu-241 activity concentration at date of collection (pCi/kg);
T1, T2 = physical half lives of Pu-241 and Am-241 respectively;
1 − kt = fraction of Pu-241 decayed during storage of the collected sample.

2.3 Analysis of Pu-241

The analytical procedure previously used for Pu-239 + 240 and Pu-238 resulted in complete separation of americium from plutonium. The stainless steel discs, on which all plutonium isotopes (including Pu-241) had been electroplated, were kept for 2–3 years.

Plutonium and Am-241 (built up on the disc during that period) were leached with 8 M HNO_3 and conc. HCl. About 0.2 pCi of Am-243 (half-life 7580a) was added as radiochemical yield determinant of Am-241 and the cleaned discs were checked for the absence of alpha activity. The leaching solution thus obtained was evaporated to dryness and the residue was dissolved in 1 M HNO_3 93% methanol. This solution was then passed through columns filled with Dowex 1X8 resins. Nickel, iron and other impurities from the disc passes through the columns but plutonium and americium were adsorbed on the Dowex 1X8 resin. The americium was first leached with 9 M HCl and then the plutonium with 1.2 M HCl. The americium fraction was further purified from Th-228 and U-232 (decay products from Pu-236 used as yield determinant for Pu) by passing the 9 M HCl solution through Dowex 50X8 to absorb Th and Dowex 1X8 to absorb U. The electrodeposition and counting of the samples were performed as described above (2.2).

The Pu-241 activity concentration, CO_1, at the sampling time given in pCi per kg dry weight can be calculated according to:

$$CO_1 = \frac{A2}{m^*Y} * \frac{T2 - T1}{T1} * \frac{\exp(\ln 2^*t1/T1)}{\exp(-\ln 2^*t2/T2) - \exp(-\ln 2^*t2/T1)}.$$

Thus

$$CO_1 = \frac{A2}{m^*Y} * 31.3 * \frac{Kt1}{1 - Kt2}.$$

A2 = activity of Am-241 on the disc from build up (pCi);
T1, T2 = physical half lives of Pu-241 and Am-241 respectively;
1 − Kt2 = fraction of Pu-241 decayed on disc during t2 (a);
Kt1 = correction for Pu-241 decay from sampling time to time of first plating t1 (a);
Y = the radiochemical yield in the first plutonium separation;
m = weight of sample (kg).

3. Results and Discussions

3.1 Deposition of Pu-241 and Am-241 in Lichen

The deposition per unit area of Pu-241 in the lichen carpet and the curve for the total accumulated Pu-241 deposition are given in Figure 1. In 1972 the total accumulated depositions of Pu-241 and Pu-239 + 240 at the sampling site (62.3° N, 12.4° E) was 7.8 ± 1.0 mCi/km^2 and 1.0 ± 0.1 mCi/km^2 respectively. The deposition of Am-241 from fallout is relatively low and most Am-241 present in the lichen has originated in

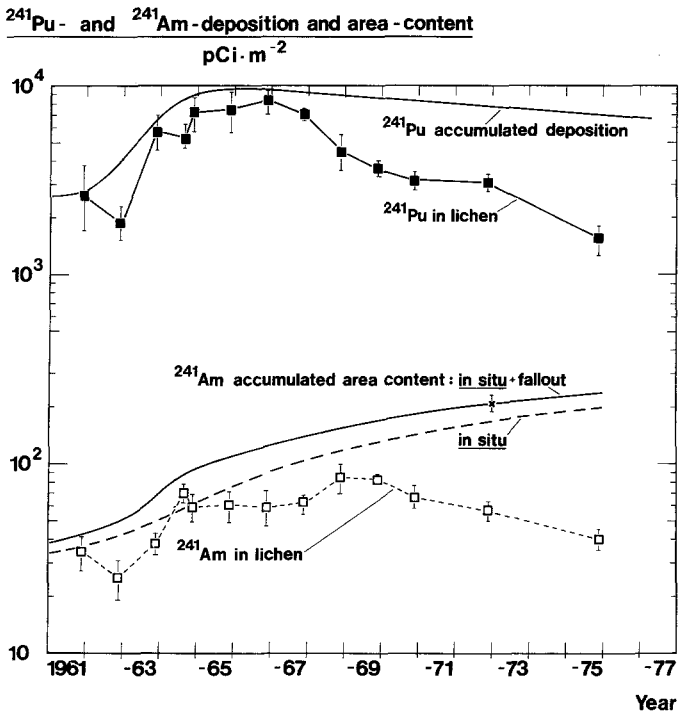

Fig. 1. Experimental results of area content of Pu-241 and Am-241 in the lichen carpet. The upper curves for Pu-241 indicates the calculated total accumulated Pu-241 deposition (pCi m^{-2}) for the sampling place. The dotted curve for Am-241 indicate the calculated in situ ingrowth of Am-241 due to Pu-241 decay and the solid curve also includes the direct fallout of Am-241

situ from Pu-241. However, Pu-241 and thus also Am-241 is present in nuclear weapons (Livingston et al., 1975). The mean residence time for Pu-241 in the stratosphere is about 11 months and additional Am-241 will be formed during this time due to the decay of Pu-241 to Am-241. The area content of Am-241 is also given in Figure 1 together with the accumulated are content of Am-241, from in situ formation and from fallout on the other hand. The total area content of Am-241 at the sampling site was 0.21 ± 0.02 mCi/km^2 in 1972, and of this activity about 80% has been formed in situ.

The Pu-241 deposition curve was estimated from the Pu-239 + 240 values previously estimated and from a Pu-241/Pu-239 + 240 activity ratio in fresh fallout during 1962–1964 of 16 ± 2 (Holm and Persson, 1976; Krey et al., 1976; Livingston et al., 1975). The effective mean residence-time of Pu-241 in the lichen carpet was calculated to 3.9 ± 0.7 years by using the Pu-241 deposition values in Figure 1 in a simple compartment model (Persson, 1972). This corresponds to a biological mean residence time of 5.5 ± 1.0 years, which is not significantly different from the value of 6.1 ± 0.5 years previously found for Pu-239 + 240 (Holm and Persson, 1975).

The deposition rate of Am-241 was estimated on the basis of the Am-241/Pu-241 activity ratio of surface air reported by Thomas and Perkins (1974) and the deposition curve for Pu-241 according to Holm (1977).

3.2 Biological Mean residence Times for Am-241

In estimating the biological mean residence time for americium one must also consider the in situ build up of Am-241 from Pu-241. Thus the annual change of the Am-241 area content can be described by the equation.

$$\mathrm{AAm}(j+1) - \mathrm{AAm}(j) = \mathrm{IN}(j+1) + \mathrm{GAM}^{*}\mathrm{Pu}(j) - 1/\mathrm{TAm}^{*}\mathrm{Am}(j+1)$$

where

AAm(j) = the area content of Am-241 at year j (pCi per m^2);
In(j + 1) = the annual input from fallout at year j + 1 (pCi per m^2);
Pu(j) = the area content of Pu-241 at year j (pCi per m^2);
GAM = the annual ingrowth of Am-241 from Pu-241 decay;
TAm = the mean residence time for Am.

By fitting this equation to the experimental values a mean residence time for americium in the lichen was estimated to be 4 ± 1 year.

3.3 Vertical Distribution

The temporal variation of the spatial distribution of Pu-241 and Am-241 in the lichen carpet are displayed in Figures 2 and 3. The concentrations are expressed as nCi per kg dry weight of the layer of the lichen, and normalized to layer B. The various layers are as follow layer A: 0–3 cm; B: 3–6 cm, C: the rest (about 6 cm

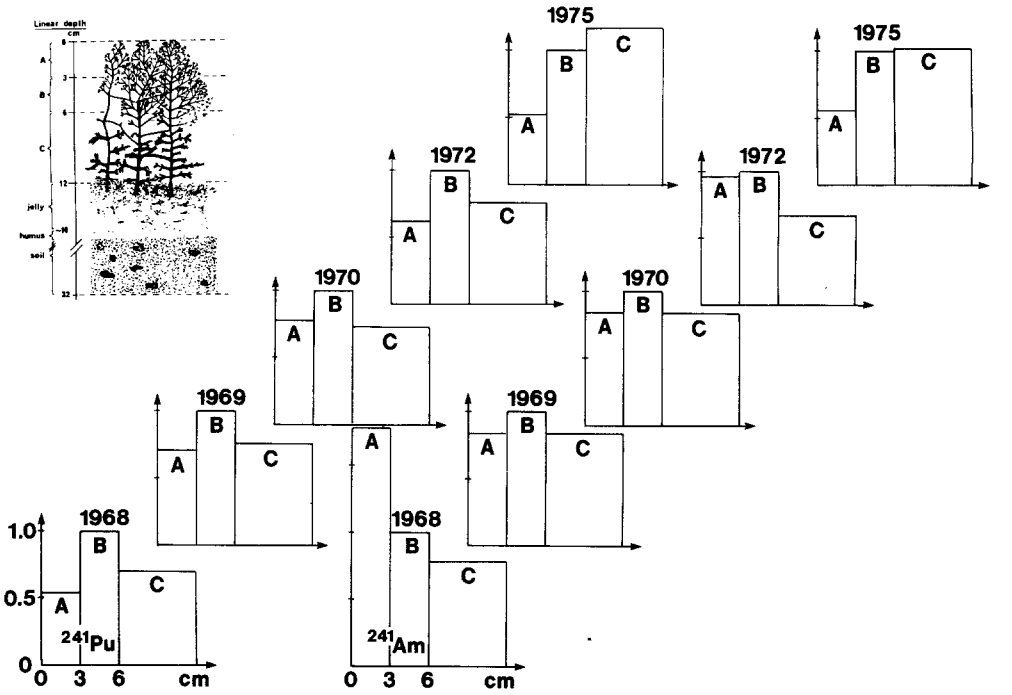

Fig. 2. The vertical distribution of Pu-241 and Am-241 in carpets of *Cl. alpestris* normalized to layer B. One profile is given for each year 1968, 1969, 1970, 1972, and 1975

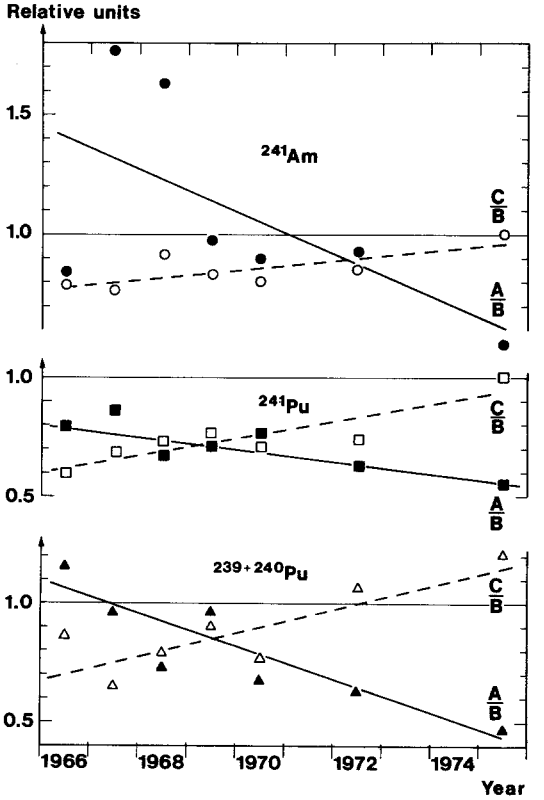

Fig. 3. The ratio of the activity concentration of Am-241, Pu-241, and Pu-239 in lichen layers "A" to "B" and "C" to "B", respectively, at different years

Table 2. Activity concentration (nCi per kg dry weight) of Pu-241 in different layers of lichen carpet and soil (± S.E.)

Part of the depth profile	(nCi per kg dry weight)						
	1966	1967	1968	1969	1970	1972	1975
Lichen layer							
A 0–3 cm	2.3 ± 0.7	3.2 ± 0.3	1.7 ± 0.3	1.5 ± 0.3	1.6 ± 0.3	1.2 ± 0.1	0.6 ± 0.1
B 3–6 cm	2.9 ± 1.0	3.7 ± 0.2	3.0 ± 0.8	2.1 ± 0.2	2.1 ± 0.3	1.9 ± 0.1	1.1 ± 0.3
C > 6 cm	1.7 ± 0.2	2.5 ± 0.7	2.2 ± 0.3	1.6 ± 0.3	1.5 ± 0.2	1.4 ± 0.4	1.1 ± 0.3
Litter	5.3 ± 1.0	5.3 ± 1.0	–	–	1.1 ± 0.2	1.6 ± 0.5	–
Jelly	–	–	1.3 ± 0.2	0.7 ± 0.2	0.7 ± 0.2	1.0 ± 0.2	–
Soil layer							
0–3 cm	–	–	0.4 ± 0.1	–	–	0.3 ± 0.1	–
2–3 cm	–	–	–	0.6 ± 0.1	–	–	–

Table 3. Area content (nCi · m^{-2} eq. mCi · km^{-2}) of Pu-241 in different layers of lichen carpet and soil (± S.E.)

Part of the depth profile	(nCi per m^{-2} or mCi per km^{-2})						
	1966	1967	1968	1969	1970	1972	1975
Lichen layer							
A 0–3 cm	1.4 ± 0.5	2.3 ± 0.2	1.2 ± 0.5	1.1 ± 0.2	1.0 ± 0.3	0.8 ± 0.1	0.3 ± 0.1
B 3–6 cm	1.8 ± 0.6	2.3 ± 0.3	1.6 ± 0.5	1.2 ± 0.1	1.1 ± 0.2	1.0 ± 0.1	0.5 ± 0.1
C > 6 cm	1.5 ± 0.2	2.0 ± 0.4	1.6 ± 0.3	1.1 ± 0.1	1.1 ± 0.3	1.0 ± 0.3	0.6 ± 0.1
Litter	0.6 ± 0.1	0.5 ± 0.1	–	–	0.07 ± 0.02	0.3 ± 0.1	–
Jelly	–	–	1.4 ± 0.2	1.3 ± 0.3	0.9 ± 0.3	1.2 ± 0.1	–
Soil layer							
0–3 cm	–	–	0.6 ± 0.1	–	–	1.6 ± 0.4	–
2–3 cm	–	–	–	1.9 ± 0.4	–	–	–

Table 4. Activity concentration (pCi/kg dry weight) of Am-241 in different layers of lichen carpet and soil (± S.E.)

Part of the depth profile	(nCi per kg dry weight)						
	1966	1967	1968	1969	1970	1972	1975
Lichen layer							
A 0–3 cm	32 ± 5	39 ± 3	52 ± 10	34 ± 3	36 ± 5	25 ± 4	13 ± 3
B 3–6 cm	38 ± 8	22 ± 2	32 ± 5	35 ± 3	40 ± 4	27 ± 3	24 ± 5
C > 6 cm	30 ± 3	17 ± 2	29 ± 3	29 ± 2	32 ± 2	23 ± 2	24 ± 5
Litter	–	–	–	124 ± 20	–	–	–
Jelly	–	–	25 ± 4	26 ± 3	27 ± 3	26 ± 4	23 ± 5
Soil layer							
0– 3 cm	–	–	–	6 ± 1	–	6.2 ± 1.5	3.0 ± 1.0
2– 3 cm	–	–	10 ± 2	–	–	–	–
3– 7 cm	–	–	–	–	–	1.2 ± 0.3	2.0 ± 0.5
7–15 cm	–	–	–	–	–	0.3 ± 0.1	0.4 ± 0.2
> 15 cm	–	–	–	–	–	< 0.1	–

Table 5. Area content pCi · m^{-2} of Am-241 in different layers of lichen carpet and soil (± S.E.)

Part of the depth profile	(pCi per m^{-2})						
	1966	1967	1968	1969	1970	1972	1975
Lichen layer							
A 0–3 cm	20 ± 2	27 ± 2	36 ± 9	24 ± 2	21 ± 3	17 ± 3	8 ± 2
B 3–6 cm	22 ± 4	14 ± 3	19 ± 3	19 ± 2	20 ± 3	15 ± 2	13 ± 3
C > 6 cm	26 ± 3	15 ± 2	25 ± 5	18 ± 2	22 ± 3	18 ± 2	17 ± 4
Litter	–	–	–	26 ± 4	–	–	–
Jelly	–	–	43 ± 4	28 ± 5	39 ± 7	21 ± 6	28 ± 5
Soil layer							
0– 3 cm	–	–	–	–	–	45 ± 6	30 ± 5
2– 3 cm	–	–	18	–	–	–	–
3– 7 cm	–	–	–	–	–	37 ± 7	34 ± 5
7–15 cm	–	–	–	–	–	17 ± 5	26 ± 5
> 15 cm	–	–	–	–	–	< 4	–

Table 6. Am-241/Pu-239 + 240 activity ratio in different layers of lichen carpet and soil (± S.E.)

Part of the depth profile	1966	1967	1968	1969	1970	1972	1975
Lichen layer							
A 0–3 cm	0.14 ± 0.03	0.17 ± 0.02	0.23 ± 0.05	0.18 ± 0.03	0.21 ± 0.04	0.25 ± 0.06	0.17 ± 0.04
B 3–6 cm	0.11 ± 0.05	0.10 ± 0.02	0.12 ± 0.02	0.18 ± 0.03	0.16 ± 0.02	0.15 ± 0.03	0.16 ± 0.04
C > 6 cm	0.11 ± 0.04	0.11 ± 0.02	0.14 ± 0.03	0.15 ± 0.03	0.17 ± 0.03	0.12 ± 0.02	0.14 ± 0.03
Litter	–	–	–	0.33 ± 0.07	–	–	–
Jelly	–	–	0.27 ± 0.08	0.22 ± 0.03	0.21 ± 0.02	0.10 ± 0.03	–
Soil layer							
0– 3 cm	–	–	–	0.19 ± 0.02	–	–	–
2– 3 cm	–	–	0.19 ± 0.06	–	–	–	–
3– 7 cm	–	–	–	–	–	0.47 ± 0.18	0.24 ± 0.07
7–15 cm	–	–	–	–	–	0.39 ± 0.18	0.36 ± 0.10

Table 7. The activity ratio Pu-241/Pu-239 + 240 in different layers of lichen carpet and soil (± S.E.)

Part of the depth profile	1966	1967	1968	1969	1970	1972	1975
Lichen layer							
A 0–3 cm	10 ± 4	14 ± 2	7 ± 1	8 ± 2	9 ± 1	11 ± 1	7 ± 1
B 3–6 cm	15 ± 5	16 ± 1	11 ± 3	11 ± 1	8 ± 1	11 ± 1	6 ± 1
C > 6 cm	10 ± 1	15 ± 3	11 ± 1	8 ± 1	8 ± 1	8 ± 1	5 ± 1
Litter	14 ± 3	18 ± 4	–	–	8 ± 2	9 ± 2	–
Jelly	–	–	16 ± 5	7 ± 1	5 ± 1	6 ± 1	–
Soil layer							
0–3 cm	–	–	7 ± 1	–	–	7 ± 3	–
2–3 cm	–	–	–	10 ± 2	–	–	–

thick) as indicated in Figure 2. In Tables 2–5 the Pu-241 and Am-241 activity concentrations and area contents of jelly (decomposed plant material) and soil are also given.

The distribution of Am-241 in the lichen carpet is quite different from that of Pu-241. No pronounced maximum at any year is observed and a homogeneous distribution is recorded. The activity ratio of Am-241 to Pu-239 + 240 increases with depth in soil (Table 6). This might be an effect of faster downward migration of Am-241 than Pu-239 + 240. Deeper plutonium is also more aged than the Am-241 formed from decay of Pu-241. Table 7 shows the Pu-241/Pu-239 + 240-activity-ratio in different layers of lichen, jelly and soil. This activity ratio decreases with increasing depth mainly due to the decay of Pu-241.

3.4 Transfer of Pu-241 and Am-241 from Lichen to Reindeer

The transfer of Pu-239 to reindeers has previously been investigated by us (Holm and Persson, 1976). We have now also analyzed the Pu-241 levels in some tissues of reindeer slaughtered on the same occasion in 1970, 1971, 1972–1974. The activity ratios of Pu-241 to Pu-239 + 240 are slightly higher in reindeer liver than expected. But no difference in the metabolism of various plutonium isotopes can be concluded.

We have previously used a simple compartment model to derive the most important factors regulating the transfer of plutonium from lichen to reindeer (Holm and Persson, 1976). This model has now been modified to apply also to the Am-241 data. As can be seen from Figure 1, the annual variation of Am-241 content in the lichen decreases quite slowly over the years. Therefore we consider a constant intake by food each year. It is assumed that the uptake of americium and plutonium from food is more significant than the uptake by respiration during 1970–1976.

The change of the average activity concentration of Am-241 in various tissues is given by the following equation:

$$\begin{aligned} AAm(j+1) = {} & AAm(j) + INAm(j+1)*fAm: \\ & M + GAM*APu(j) - kAm*AAm(j+1) \end{aligned}$$

where

AAm(j) = the activity concentration of Am-241 at year j;
INAm(j) = the intake of Am-241 by food at year j;
INAm(j + 1) = RAm*CAm (pCi/a);
RAm = the rate of mass transfer (kg/a);
CAm = the activity concentration of ingested material pCi/kg;
fa = the fraction of ingested activity which is retained in the step in question;
M = the total mass of the tissue in question;
kAm = 1/TAm the elimination rate constant for Am-241;
TAm = the mean residencetime for Am-241;
GAm = the ingrowth of Am-241 from Pu-241 decay.

Table 8. The fraction of ingested activity of Pu-239 and Am-241 retained "fa", and the fractional residence time in liver, bone and flesh

Organ or tissue	Fraction of ingested activity retained "fa"		Fractional residence time τ^a year	
	Pu-239	Am-241	Pu-239	Am-241
Liver	$(14 \pm 5) \cdot 10^{-6}$	$(7.5 \pm 3.0) \cdot 10^{-6}$	$(4.7 \pm 1.1) \cdot 10^{-5}$	$(3.0 \pm 0.5) \cdot 10^{-5}$
Bone: compact	$(4.6 \pm 0.8) \cdot 10^{-6}$	$(21 \pm 10) \cdot 10^{-6}$	$(2.5 \pm 0.8) \cdot 10^{-5}$	$(4.3 \pm 1.0) \cdot 10^{-5}$
trabecular	$(2.7 \pm 0.7) \cdot 10^{-6}$	$(10 \pm 4) \cdot 10^{-6}$	$(1.3 \pm 0.2) \cdot 10^{-5}$	$(2.1 \pm 1.0) \cdot 10^{-5}$
Flesh	$(2.5 \pm 0.5) \cdot 10^{-6}$	–	$(0.9 \pm 0.1) \cdot 10^{-5}$	–

[a] τ = real fractional residence time. Corrected for in situ Am-241 from Pu-241

By applying this equation to our results the fraction of ingested activity which is retained in reindeer liver and bone is presented in Table 8, together with corresponding results for plutonium. We find that less americium than plutonium is taken up in the liver. But for bone we find an enhanced uptake of Am-241 compared to plutonium. In Figure 4 the results for Am-241 and Pu-241 in the top layer of lichen (which is grazed by the reindeer) is given together with the results for Am-241 in reindeer liver and bone. The results for reindeer liver and bone reported by Tähtinen et al.,

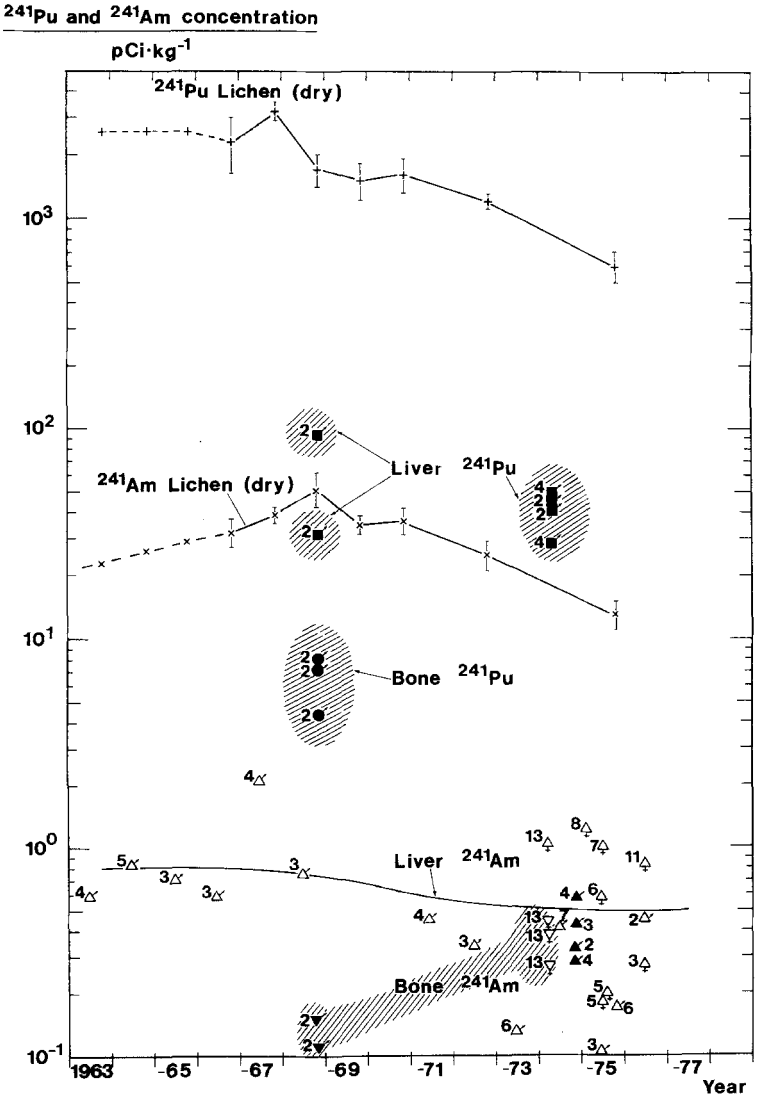

Fig. 4. The activity concentrations of Pu-241 and Am-241 in the top layer of lichen consumed by reindeer and in reindeer liver and bone during the period 1972–1975. The open dots represent values reported by Tähtinen et al. (1976) with correction for physical decay to the date of collection. The figure given at each dot indicates the age of the animal

1977 have been corrected for physical decay to the date of sample collection and are also presented in Figure 4. The fractional residence time (τ'') fpr Am-241 in reindeer liver and bone have been calculated on the basis to the following expression (Persson, 1972):

$$\frac{\text{AAm(j)*M}}{\text{INAm(j)}} = \tau'' = f_a{}^*\tau \,.$$

The values thus obtained are given in Table 8. By correcting for the intake of Pu-241 and in situ ingrowth of Am-241 a fractional residence time was obtained which could be used for the estimation of the Am-concentration in man. By assuming a total annual intake of Am-241 by the Lapps of about 5 pCi the Am-241 concentration in Lapps was estimated to be about 0.1 fCi/kg (f = femto i.e. 10^{-15}) in both liver and bone due to consumption of reindeer.

3.5 Radiation Dosimetry

The absorbed dose to soft tissue with a Pu-239 + 240-activity concentration of 1.0 pCi per kg wet weight is almost 0.1 mrad per year. The reindeer liver which contained the highest concentration of plutonium received a dose-rate due to Pu-239 + 240 which was about 0.5 mrad per year during 1965–1974. Plutonium-241, which is a beta emitter, contributes with only about 1.2% of the absorbed dose from Pu-239 + 240. This applies if the activity ratio of Pu-241 to Pu-239 + 240 is about 10.

With the assumption that plutonium is localized to a thin layer on endosteal surfaces and is not homogenously distributed within the bone the dose rate from Pu-239 + 240 of an activity concentration of 1 pCi per kg in trabecular endosteum and cortical endosteum is 1.4 mrad per year and 0.5 mrad per year respectively.

Americium-241 is introduced into the body partly by ingestion and partly as in situ build up from Pu-241 deposited in the tissues. The absorbed dose-rate from Am-241 will thus also depend on the time that has passed after intake of Pu-241. Because the energy and the fluence rate of the alpha particles from Am-241 and Pu-239 is almost the same the absorbed dose-rate per pCi per kg wt weight is also about 0.1 mrad per year for Am-241. Reindeer liver will thus have received a dose-rate of about 0.07 mrad per year in 1974. The distribution of Am-241 in relation to bone is not known but can be assumed to be about the same as for Pu-241.

The world-wide distribution of fall-out from nuclear explosions in the atmosphere has resulted in an average concentration of Pu-239 + 240 in human liver, bone and lung of about 0.2–1.0 pCi per kg wet weight according to Mussalo et al. (1976). This is mainly due to inhalation of air-born fall-out (Bennet, 1972).

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