

Transfer of plutonium, americium and curium from fallout into reindeer after the Chernobyl accident

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Several reindeer tissue samples were analysed for transuranium elements following the Chernobyl accident in 1986. Pu, Am and Cm were separated using coprecipitation and anion exchange steps, and measured with alpha spectrometry. The activity concentrations of $^{239,240}\text{Pu}$ in reindeer liver found in this study (0.0094–0.062 Bq kg⁻¹ d.w. [= dry weight]) are low compared to the concentrations found in the 1960s and 1970s, even though most of the samples are from the areas with the heaviest Chernobyl-derived fallout of plutonium. On the other hand, the ^{241}Pu concentrations in reindeer liver found in this study (0.7–3.9 Bq kg⁻¹ d.w.) are comparable to earlier results. The calculated transfer factors, $(0.31\text{--}1.61) \times 10^{-5} \text{ m}^2 \text{ kg}^{-1}$ for $^{239,240}\text{Pu}$ and $7 \times 10^{-3} \text{ m}^2 \text{ kg}^{-1}$ for ^{241}Am , show that ^{137}Cs migrates from fallout to reindeer meat 100 times more efficiently than ^{241}Am and 10 000 times more efficiently than $^{239,240}\text{Pu}$. The transfer factor of $2 \times 10^{-3} \text{ m}^2 \text{ kg}^{-1}$ obtained for ^{242}Cm does not take into account the physical decay of ^{242}Cm . Several earlier results concerning plutonium and americium in reindeer could be verified in this study using the fallout from the Chernobyl accident. Americium and curium are accumulated more efficiently than plutonium in bone and lungs. The main pathway of Pu, Am and Cm into reindeer is the gastro-intestinal (GI) tract and not inhalation. The absorption coefficient of $^{239,240}\text{Pu}$ obtained for the GI-tract, 3.0×10^{-5} , is in agreement with earlier results. A GI-tract absorption coefficient of 7.5×10^{-4} for ^{241}Am was obtained in this study.

Introduction

Following the atmospheric nuclear tests in the 1950s and early 1960s several radioecological re-

search projects, focused on the (sub)arctic food chain lichen–reindeer/caribou–man, were initiated in Scandinavia and North America. Lichen collects deposited radionuclides efficiently and is the main

fodder for reindeer during the winter season. The enrichment of radionuclides in this food-chain can lead to exceptionally high body burdens among the indigenous Sami and Inuit (Eskimo) populations consuming large quantities of the meat and edible organs of reindeer and caribou. Initially the research was aimed mainly at ^{137}Cs (Miettinen *et al.* 1963, Svensson and Lidén 1965, Hanson 1967, Rahola *et al.* 1988, Rissanen *et al.* 1990) but later natural radionuclides (Kauranen and Miettinen 1969, Persson 1970) and transuranium elements were also studied (Tulikoura *et al.* 1974, Holm and Persson 1975, Jaakkola *et al.* 1978, Hanson 1980, Mussalo-Rauhamaa 1981, Hakanen *et al.* 1984).

Compared to the global fallout of the atmospheric nuclear tests, the fallout from the Chernobyl accident in April 1986 was very unevenly distributed in Finland (Arvela *et al.* 1990, Reponen *et al.* 1993). In the Chernobyl-derived fallout, volatile elements, including ^{137}Cs as the dominating radionuclide, were differently distributed compared to Pu, Am, Cm and other refractory elements. Even in the most contaminated areas the fallout of $^{239,240}\text{Pu}$ was only some 10 per cent of the nuclear tests' fallout. Among the reindeer-herding districts only Halla, the southernmost reindeer-herding region, received high amounts of deposited radionuclides from the accident. Another difference was the isotopic composition of the transuranium elements. Due to the relatively high burn-up of the nuclear fuel in the Chernobyl reactor, the activity ratio of curium isotopes and ^{241}Pu to $^{239,240}\text{Pu}$ was higher than in the case of global fallout. Only minute amounts of curium isotopes were produced in the test explosions (Holm and Persson 1978). Plutonium-241 only emits low-energy beta particles ($E_{\text{max}} = 21 \text{ keV}$) but it makes the highest contribution to total plutonium activity (Paatero *et al.* 1994). It is also the precursor of ^{241}Am , a nuclide which is highly radiotoxic due to its alpha emission and long physical and biological half-lives in animals and man.

Owing to its unique nuclide composition, the fallout from the Chernobyl accident offered an exceptional opportunity to investigate the behaviour of ^{242}Cm and other transuranium nuclides in reindeer, a large terrestrial herbivorous mammal, in its natural habitat. Also, the source term of the Chernobyl accident was well quantified and its duration limited, which facilitates the interpreta-

tion of the results. The purpose of this study was to investigate the transfer of transuranium elements from the Chernobyl accident along the lichen-reindeer-man food-chain towards man by analysing the Pu, Am and Cm contents of a number of reindeer tissue samples. Most of these samples were from the Halla reindeer-herding district with a relatively high fallout from the Chernobyl plume. For comparison, three elk tissue samples were also analysed. Elk is rather similar to reindeer, except that it does not use lichen as a fodder during the winter season. Two of the elk samples were from Häme, Western Finland, with a plutonium fallout comparable to that of the Halla region. One elk sample was from the coast of Southern Finland with a Pu fallout level lower than in the Halla region.

Materials and methods

The sampling areas for the reindeer (*Rangifer tarandus*) and elk (*Alces alces*) tissue samples are presented in Fig. 1. Both tissue samples from individual animals and pooled samples were analysed. The radiochemical separations were accomplished in 1987–1989. For the soft tissue samples a dry matter content of 30% was assumed if the precise value was unknown.

The method used for the determination of $^{239,240}\text{Pu}$, ^{238}Pu , ^{241}Am and ^{242}Cm was slightly modified from that reported by Yamato (1982). After the addition of ^{242}Pu , ^{243}Am and ^{244}Cm tracers, the dried samples were leached with concentrated nitric and hydrochloric acids. Transuranium elements were coprecipitated with calcium oxalate and $\text{Fe}(\text{OH})_3$. Next, Pu was separated from Am and Cm and purified using anion exchange. The counting preparate of Pu was made with NdF_3 coprecipitation using the method of Hindman (1983). The fraction containing Am and Cm was purified with three subsequent anion exchange steps: the first to remove Fe, the second to remove lanthanides, and the last to remove traces of Th. Finally Am and Cm were electroplated onto platinum or stainless steel discs according to Hallstadius (1984).

The only modification to the separation scheme was made with bone samples. After the leaching, a Fe holdback carrier was added to the sample, which

was then treated with fuming nitric acid to precipitate the bulk of the Ca. The remaining part of the Ca was then used for the oxalate precipitation.

The alpha spectra of the samples were measured in vacuum with semiconductor detectors soon after the radiochemical separation. In the spring of 1996, several years after the separation of Pu, some of the Pu-containing preparates were again assayed alpha-spectrometrically. The ^{241}Pu contents of the samples were assessed by the alpha emissions of the in-grown daughter nuclide ^{241}Am (Holm and Persson, 1977). First the total alpha activity in the energy region of 5.3–5.6 MeV (= sum of ^{238}Pu , 5.449 and 5.457 MeV, and ^{241}Am , 5.486 and 5.443 MeV) was calculated. The ^{241}Am activity was determined by subtracting the decay-corrected ^{238}Pu activity from the total alpha activity. Finally, the ^{241}Pu activity was computed using the formula:

$$A_{\text{Pu}}^0 = \frac{A_{\text{Am}}(\lambda_{\text{Am}} - \lambda_{\text{Pu}})}{\lambda_{\text{Am}}(e^{-\lambda_{\text{Pu}}t} - e^{-\lambda_{\text{Am}}t})} \quad (1)$$

where A_{Pu}^0 is the activity of ^{241}Pu at the time of the radiochemical separation, A_{Am} the in-grown activity of ^{241}Am during the second alpha measurement, λ_{Pu} and λ_{Am} the respective decay constants, and t the time between the radiochemical separation of plutonium and the second alpha measurement. The statistical uncertainties in the ^{241}Pu determinations were high, 17%–100%, due to the error propagation of two measurements (counting errors of 5.3–5.6 MeV total alpha activity and ^{238}Pu).

Results and discussion

The activity concentrations of the transuranium elements in the different organs of reindeer are presented in Table 1. From the high ^{242}Cm : $^{239,240}\text{Pu}$ and ^{241}Am : $^{239,240}\text{Pu}$ activity ratios (Table 2) one can deduce that americium and curium are accumulated more efficiently in bone and lung than plutonium. In kidneys there are no signs of different accumulation behaviour between Pu and Am.

The Pu, Am, and Cm activity contents of a series of reindeer liver samples from the Halla district are reported in Table 3. These reindeer were slaughtered in December 1986. The activity concentrations of $^{239,240}\text{Pu}$ are low compared to



- 1 = Inari
- 2 = Halla reindeer herding district
- 3 = Northern Häme game management district
- 4 = Southern Häme game management district
- 5 = Wessö

Fig. 1. Sampling sites of reindeer and elk tissue samples: 1 = Inari, 2 = Halla reindeer herding district, 3 = Northern Häme game management district, 4 = Southern Häme game management district, 5 = Wessö.

those found in the 1960s and 1970s (Fig. 2) which is in accordance with the smaller amount of deposited plutonium (global fallout: ca. 70 Bq m⁻², Hardy *et al.* 1973; Chernobyl fallout: 1.5 Bq m⁻², Paatero *et al.* 1998). On the other hand, the ^{241}Pu concentrations in reindeer liver found in this study (0.2–1.2 Bq kg⁻¹ f.w. [= fresh weight]) are comparable to those found in 1976–1977, 0.74–8.73 Bq kg⁻¹ (Hakanen *et al.* 1984). A high correlation exists between the concentrations of the transuranium nuclides (Table 4). However, the ^{137}Cs concentrations of the same animals (Rissanen *et al.* 1987) do not show any correlation with the Pu, Am, and Cm concentrations.

If the measured plutonium in the sample originates from two sources with a different isotopic composition (in this case global and Chernobyl-derived fallout), the two portions can be calcu-

Table 1. Activity concentrations of transuranium elements (Bq kg⁻¹ dry weight decay-corrected to 1 May 1986 ± 1 σ propagated counting error) in reindeer tissue samples collected in 1987.

Sample	^{239,240} Pu	²³⁸ Pu	²⁴¹ Am	²⁴² Cm
Liver 04187 (Inari 17 Mar 87)	0.0094 ± 0.0014	< 0.0013	< 0.0005	< 0.006
Liver AB1828/87 (Halla 2 Mar 87)	0.025 ± 0.004	0.008 ± 0.002	0.013 ± 0.003	0.06 ± 0.02
Livers, calves (Halla 15 Feb 87)	0.049 ± 0.007	0.011 ± 0.003	0.023 ± 0.004	0.25 ± 0.04
Livers, adults (Halla 15 Feb 87)	0.052 ± 0.005	0.012 ± 0.002	0.013 ± 0.004	0.13 ± 0.03
Ribs, calves (Halla 15 Feb 87)	0.0012 ± 0.0004	< 0.0010	< 0.003	0.15 ± 0.03
Ribs, adults (Halla 15 Feb 87)	0.0015 ± 0.0004	< 0.0003	0.009 ± 0.002	0.10 ± 0.02
Meat (Halla 15 Feb 87)	< 0.0011	< 0.0011	0.014 ± 0.003	< 0.05
Lungs (Halla 15 Feb 87)	0.0024 ± 0.0008	< 0.002	0.0049 ± 0.0014	0.036 ± 0.012
Kidneys (Halla 15 Feb 87)	0.010 ± 0.002	0.0052 ± 0.0016	0.0043 ± 0.0010	< 0.03
Spines (Halla 15 Feb 87)	< 0.007	< 0.002	0.018 ± 0.004	< 0.02
Horns (Halla 15 Feb 87)	< 0.015	< 0.011	0.016 ± 0.003	< 0.04

Table 2. Activity ratios of transuranium elements (1 May 1986 ± 1 σ propagated counting error) in reindeer tissue samples collected in 1987. The values for the Chernobyl fallout are from the study of Paatero *et al.* (1998).

Sample	²³⁸ Pu: ^{239,240} Pu	²⁴² Cm: ^{239,240} Pu	²⁴¹ Am: ^{239,240} Pu	²⁴² Cm: ²⁴¹ Am
Liver AB1828/87 (Halla 2 Mar 87)	0.34 ± 0.10	2.2 ± 0.8	0.51 ± 0.13	4 ± 2
Livers, calves (Halla 15 Feb 87)	0.24 ± 0.07	5.0 ± 1.1	0.47 ± 0.11	11 ± 3
Livers, adults (Halla 15 Feb 87)	0.22 ± 0.05	2.5 ± 0.7	0.25 ± 0.07	10 ± 4
Ribs, calves (Halla 15 Feb 87)		130 ± 50		
Ribs, adults (Halla 15 Feb 87)		67 ± 22	6 ± 2	11 ± 4
Lungs (Halla 15 Feb 87)		15 ± 7	2.0 ± 0.9	7 ± 3
Kidneys (Halla 15 Feb 87)	0.5 ± 0.2		0.43 ± 0.14	
Chernobyl fallout	0.55	15	0.34	39

Table 3. Activity concentrations of transuranium elements (Bq kg⁻¹ dry weight decay-corrected to 1 May 1986 ± 1 σ propagated counting error) in reindeer liver samples collected from the Halla reindeer herding district in December 1986.

Sample	^{239,240} Pu	²³⁸ Pu	²⁴¹ Am	²⁴² Cm	²⁴¹ Pu
36/705	0.033 ± 0.004	0.007 ± 0.002	< 0.009	0.51 ± 0.05	0.7 ± 0.3
36/734	0.048 ± 0.004	0.014 ± 0.002	0.021 ± 0.004	0.31 ± 0.04	2.3 ± 0.5
36/736	0.043 ± 0.006	0.017 ± 0.004	0.013 ± 0.003	0.30 ± 0.05	1.9 ± 0.6
36/742	0.020 ± 0.003	0.006 ± 0.002	0.012 ± 0.004	0.25 ± 0.07	1.0 ± 0.5
36/748	0.062 ± 0.014	0.034 ± 0.011	0.026 ± 0.005	0.57 ± 0.10	1.3 ± 1.3
36/707	0.052 ± 0.010	< 0.014	0.011 ± 0.003	0.23 ± 0.05	
36/733	0.045 ± 0.008	0.023 ± 0.006	< 0.017	0.62 ± 0.14	3.4 ± 0.8
36/716	0.055 ± 0.009	0.025 ± 0.006	< 0.016	< 0.3	1.8 ± 0.9
36/752	0.036 ± 0.006	0.011 ± 0.003	0.011 ± 0.003	0.22 ± 0.04	0.9 ± 0.5
36/735	0.038 ± 0.005	0.014 ± 0.003	0.008 ± 0.002	0.33 ± 0.05	2.7 ± 0.5
36/740	0.022 ± 0.005	< 0.007	< 0.008	0.24 ± 0.06	
36/738	0.042 ± 0.005	0.015 ± 0.003	0.015 ± 0.003	0.26 ± 0.04	3.5 ± 0.6
30/896	0.060 ± 0.007	0.019 ± 0.005	0.028 ± 0.005	0.59 ± 0.08	3.9 ± 0.9
30/898	0.054 ± 0.005	0.015 ± 0.003	0.020 ± 0.003	0.40 ± 0.03	3.0 ± 0.6

lated from the ^{238}Pu : $^{239,240}\text{Pu}$ or ^{241}Pu : $^{239,240}\text{Pu}$ activity ratios using the formula (Hardy 1972):

$$R = \frac{^{239,240}\text{Pu activity [Chernobyl fallout]}}{^{239,240}\text{Pu activity [global fallout]}} = \frac{C_s - C_f}{C_c - C_s} \quad (2)$$

where C_s , C_f , and C_c are the ^{238}Pu : $^{239,240}\text{Pu}$ or ^{241}Pu : $^{239,240}\text{Pu}$ activity ratios in the sample, in the global fallout and in the Chernobyl fallout, respectively. The values used for global fallout (C_f) and Chernobyl fallout (C_c) were 0.03 and 0.55, respectively, in the case of ^{238}Pu : $^{239,240}\text{Pu}$, and 4 and 95 in the case of the ^{241}Pu : $^{239,240}\text{Pu}$ activity ratio (Paatero *et al.* 1998, Paatero *et al.* 1994). The contribution of Chernobyl-derived $^{239,240}\text{Pu}$ to total measured $^{239,240}\text{Pu}$ (S) is therefore:

$$S = \frac{A(^{239,240}\text{Pu})_{\text{Chernobyl}}}{A(^{239,240}\text{Pu})_{\text{total}}} = \frac{R}{R+1} \quad (3)$$

where $A(^{239,240}\text{Pu})_{\text{Chernobyl}}$ is the Chernobyl-derived and $A(^{239,240}\text{Pu})_{\text{total}}$ the total $^{239,240}\text{Pu}$ activity concentration. The correlation between the calculated Chernobyl-derived $^{239,240}\text{Pu}$ concentrations based on the ^{238}Pu : $^{239,240}\text{Pu}$ and ^{241}Pu : $^{239,240}\text{Pu}$ activity ratios is depicted in Fig. 3. The contribution of Chernobyl-derived $^{239,240}\text{Pu}$ to the total $^{239,240}\text{Pu}$ varied between 35 and 100 per cent based on ^{238}Pu : $^{239,240}\text{Pu}$ activity ratios and between 18 and 88 per cent based on ^{241}Pu : $^{239,240}\text{Pu}$ activity ratios in the liver samples from the Halla region. The use of ^{241}Pu : $^{239,240}\text{Pu}$ activity ratios is complicated by the large statistical uncertainty of the ^{241}Pu activity concentrations.

The efficiency of the transfer of radionuclides from fallout to reindeer meat can be described with transfer factors (TF):

$$\text{TF} = \frac{\text{reindeer meat activity content (Bq kg}^{-1} \text{ f.w.)}}{\text{ground deposition (Bq m}^{-2})} \quad (4)$$

These transfer factors vary depending on the absorption characteristics and physical and biological half-lives of a particular nuclide and on the exposure time. In this study the amount of Chernobyl-derived $^{239,240}\text{Pu}$ in reindeer meat was estimated from the activity contents of liver samples from the Halla district and the ratio (= 0.0015) of $^{239,240}\text{Pu}$ concentrations in meat and liver in 1964–1976 (Jaakkola *et al.* 1981). The value of 1.5 Bq m^{-2} was used for the $^{239,240}\text{Pu}$ ground depo-

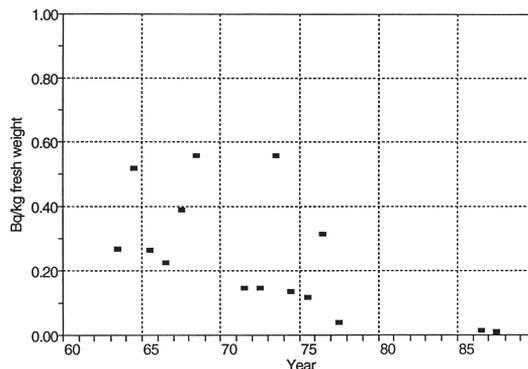


Fig. 2. Activity concentration of $^{239,240}\text{Pu}$ in reindeer liver (Bq kg^{-1} fresh weight) in the period 1962–1987. The data from the 1960s and 1970s are from the study of Jaakkola *et al.* (1981).

sition from the Chernobyl accident in the Halla region, based on analysis of lichen samples (Paatero *et al.* 1998). The transfer factors obtained for $^{239,240}\text{Pu}$ about 7 months after the accident varied between 0.31×10^{-5} and $1.61 \times 10^{-5} \text{ m}^2 \text{ kg}^{-1}$. A corresponding value for ^{241}Am was based on a deposition value of 0.6 Bq m^{-2} , leading to a transfer factor of $7 \times 10^{-3} \text{ m}^2 \text{ kg}^{-1}$ in February 1987. This value, however, does not take into account the pre-Chernobyl ^{241}Am in lichen and reindeer meat. The ^{242}Cm content of reindeer meat in December 1986 was estimated based on the liver samples from the Halla district (average: $0.0994 \text{ Bq kg}^{-1}$ f.w.; Table 3) and the ratio of ^{241}Am concentrations (Bq kg^{-1} f.w.) in meat and liver in February 1987 (0.694; Table 1). A value of 40 Bq m^{-2} was used for ^{242}Cm ground deposition from the Chernobyl accident in the Halla region. These Figures give a transfer factor of $2 \times 10^{-3} \text{ m}^2 \text{ kg}^{-1}$ for ^{242}Cm . This estimate, however, does not take

Table 4. Pearson correlation coefficients between the activity concentrations of transuranium elements and ^{137}Cs (Rissanen *et al.* 1987) in reindeer liver samples collected from the Halla reindeer herding district in December 1986.

	^{137}Cs	$^{239,240}\text{Pu}$	^{238}Pu	^{241}Am	^{242}Cm
^{137}Cs	1				
$^{239,240}\text{Pu}$	-0.1225	1			
^{238}Pu	-0.2931	0.8112	1		
^{241}Am	-0.0089	0.7408	0.6554	1	
^{242}Cm	0.1084	0.5276	0.6004	0.8636	1

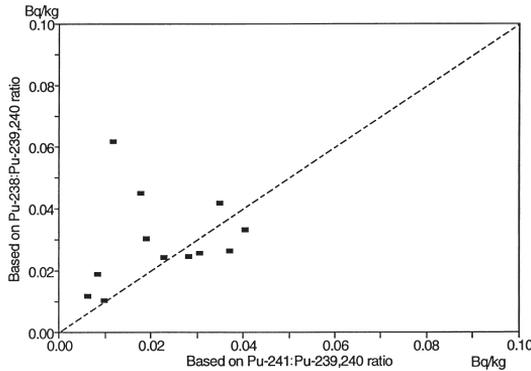


Fig. 3. Correlation between the calculated Chernobyl-derived $^{239,240}\text{Pu}$ activity concentration in reindeer liver based on the $^{238}\text{Pu}:^{239,240}\text{Pu}$ and $^{241}\text{Pu}:^{239,240}\text{Pu}$ activity ratios.

into account the short half-life (163 days) of ^{242}Cm . In their literature review Gaare and Staalnd (1994) give corresponding values for ^{137}Cs ranging from 0.12 to 0.76 $\text{m}^2 \text{kg}^{-1}$. Thus the transfer of deposited ^{137}Cs to reindeer meat is 4–5 orders of magnitude more efficient than the transfer of $^{239,240}\text{Pu}$ and 2 orders of magnitude more efficient than the transfer of ^{241}Am .

One of the factors affecting the radiotoxicity

Table 5. Reindeers' daily intake of lichen and other plants (Gaare and Staalnd 1994), and monthly intake of $^{239,240}\text{Pu}$ and ^{241}Am activity based on the estimated $^{239,240}\text{Pu}$ and ^{241}Am activity concentrations in lichen, grass and birch leaves in the Halla reindeer herding district.

Month	Daily consumption (kg day^{-1})		Monthly intake (Bq month^{-1})	
	Lichen	Other plants	$^{239,240}\text{Pu}$	^{241}Am
1	1.3	0.2	87.59	44.24
2	1.3	0.2	79.12	39.96
3	1.2	0.3	82.80	42.18
4	1.1	0.4	75.49	38.83
5	1.5	1.0	114.02	60.02
6	0.3	2.3	56.28	35.30
7	0.3	3.0	69.94	44.64
8	0.5	2.8	79.53	48.76
9	0.8	1.8	79.48	45.26
10	1.5	1.0	114.02	60.02
11 (1st half)	1.8	0.8	62.95	32.59
11 (2nd half)	1.1	0.5	38.56	19.98
12	1.2	0.3	82.80	42.18

of a nuclide is the absorption coefficient i.e. the fraction of activity in the food which is transferred across the GI tract in the animal (reindeer in this case) or man. Assuming that all the activity absorbed from food into tissues and bones also persists in the body during the exposure time, the absorption coefficient can be calculated from the equation:

$$f_i = \frac{A_{i,\text{tot}}}{mC_i} \quad (5)$$

where f_i is the absorption coefficient of nuclide i , $A_{i,\text{tot}}$ the nuclide's whole-body activity in the reindeer, and m the mass and C_i the activity content of the consumed fodder. According to Holm and Persson (1976), the biological half-life of plutonium in reindeer is 5–11 years which is sufficiently long compared to the exposure time in this study (7.5–9.5 months).

To assess the absorption coefficients for plutonium and americium, the amount of lichen and other plants consumed by reindeer as well as the weights of the different organs of a 70 kg reindeer were estimated using the data of Gaare & Staalnd (1994) (Tables 5 and 6). The values used for the $^{239,240}\text{Pu}$ and ^{241}Am activity concentrations in lichen in the Halla reindeer herding district (2.09 and 1.04 Bq kg^{-1} d.w., respectively) were based on earlier results (Paatero *et al.* 1998). The activity concentrations in other plants were evaluated using the average concentration ratio from four grass or birch leaf and lichen samples collected from same areas in Finland. For $^{239,240}\text{Pu}$ the average ratio was 0.26 and the activity concentration estimate for fodder other than lichen was thus $0.26 \times 2.09 \text{ Bq kg}^{-1} = 0.543 \text{ Bq kg}^{-1}$. Similarly the ^{241}Am activity concentration ratio was found to be 0.36, giving a value of 0.376 Bq kg^{-1} for the activity concentration of fodder other than lichen.

Based on estimates described above, the total $^{239,240}\text{Pu}$ activity ingested by a reindeer between the beginning of May and the middle of December was 656 Bq. According to the study by Jaakkola *et al.* (1981), 60 per cent of a reindeer's whole-body content of plutonium is in its liver. The average $^{239,240}\text{Pu}$ activity content of liver was 0.0126 Bq kg^{-1} f.w. among the reindeers of the Halla district. The liver comprises 1.35 per cent of the reindeer's total mass, i.e. 945 g in the case

of a 70 kg animal. The absorption coefficient for plutonium (f_{Pu}) is thus

$$f_{Pu} = \frac{0.0126 \text{ Bq kg}^{-1} \times 0.945 \text{ kg}}{0.6 \times 656 \text{ Bq}} = 3.0 \times 10^{-5} \quad (6)$$

The absorption coefficient for americium was calculated on the basis of the analyses of tissue samples from reindeer slaughtered on 15 February 1987. If the tissues of adults and calves had been analysed separately, the results concerning adults were used. The total ingested ^{241}Am activity was estimated to be 453 Bq. The analysed tissue samples represented 53.2 per cent of the total mass of a reindeer. The average ^{241}Am activity concentration of all the tissue samples excluding the horns was used to represent the remaining 46.8 per cent of the reindeer's total mass. Based on these assumptions, the whole-body ^{241}Am content was estimated to be 0.338 Bq. These estimates give a value of 7.5×10^{-4} for the absorption coefficient of ^{241}Am in reindeer.

The Pu, Am and Cm concentrations in elk liver samples (Table 7) are low compared to those in reindeer liver samples, despite the comparable amounts of deposition in Häme and Halla. This confirms earlier results (Jaakkola *et al.* 1981) that

the main pathway for the transuranium elements is the gastro-intestinal tract and not inhalation.

Summary and conclusions

The activity concentrations of $^{239,240}\text{Pu}$ in reindeer found in this study are low compared to those found in the 1960s and 1970s, even though most of the samples are from areas with the heaviest Chernobyl-derived fallout of plutonium. This is in accordance with the $^{239,240}\text{Pu}$ deposition values of the two sources, global weapons test fallout and the Chernobyl accident. On the other hand, the ^{241}Pu concentrations in reindeer liver found in this study are comparable to those found in the 1970s. The calculated transfer factors show that ^{137}Cs migrates from fallout to reindeer meat 100 times more efficiently than ^{241}Am and 10 000 times more efficiently than $^{239,240}\text{Pu}$ within the time span of this study, 7.5–9.5 months. The transfer factor of ^{242}Cm is of the same order of magnitude as that of ^{241}Am owing to the chemical resemblance of americium and curium.

Several earlier results concerning plutonium and americium in reindeer could be verified in this study using the fallout from the Chernobyl

Table 6. The relative weights of the different organs of a reindeer with a total mass of 70 kg (Gaare and Staaland 1994), estimated dry matter contents of different tissue types and ^{241}Am activity concentrations (Bq kg^{-1} fresh weight), Halla reindeer herding district 15 February 1987.

Organ	Fraction of total mass (%)	Dry matter content (%)	^{241}Am act. conc. (Bq kg^{-1} f.w.)	^{241}Am activity (Bq)
Meat	36.4	25	0.0035	0.0892
Lungs	1.16	22	0.00107	0.000875
Kidney	0.26	20	0.00086	0.000157
Liver	1.35	28	0.00364	0.00344
Bones	10.6	75	0.0105	0.0779
Horns	3.43	100	0.016	0.03842
Other	46.8		0.00392	0.1283

Table 7. Activity concentrations of transuranium elements (Bq kg^{-1} dry weight decay-corrected to 1 May 1986 $\pm 1 \sigma$ propagated counting error) in elk liver samples collected in 1986.

Sample	$^{239,240}\text{Pu}$	^{238}Pu	^{241}Am	^{242}Cm
Liver T5821 (Southern Häme 15 Nov 86)	< 0.001	< 0.001	< 0.003	0.032 ± 0.007
Liver T5765 (Northern Häme 15 Nov 86)	< 0.001	< 0.001	< 0.0004	< 0.003
Liver N:o 2 (Wessö 18–19 Oct 86)	< 0.002	< 0.001	< 0.006	< 0.06

accident. Americium and curium are accumulated more efficiently in bone and lungs than plutonium. Based on comparisons with elk from Southern Finland, it was confirmed that the main pathway for the transuranium elements is the gastro-intestinal tract and not inhalation. The evaluation of the absorption coefficients of $^{239,240}\text{Pu}$ and ^{241}Am included several error sources: the precise amount and nature of reindeer fodder, the $^{239,240}\text{Pu}$ and ^{241}Am activity contents of the fodder, the fraction of pre-Chernobyl activity, and the amount of ^{241}Am formed *in situ* from ^{241}Pu in reindeer. Even so, the absorption coefficient of $^{239,240}\text{Pu}$ obtained, 3.0×10^{-5} , was in good agreement with the values published by Jaakkola *et al.* (1981), $(4.0 \pm 0.4) \times 10^{-5}$, and Holm and Persson (1976), 0.46×10^{-5} for cortical bone, 0.27×10^{-5} for trabecular bone, 1.4×10^{-5} , for liver, and 0.25×10^{-5} for flesh.

References

- Arvela H., Markkanen M. & Lemmelä H. 1990. Environmental gamma radiation and fall-out levels in Finland after the Chernobyl accident. *Radiat. Prot. Dosim.* 32: 177–184.
- Gaare E. & Staaland H. 1994. Pathways of fallout radiocaesium via reindeer to man. In: Dahlgard H. (ed.), *Nordic Radioecology. The transfer of radionuclides through Nordic ecosystems to man*. Elsevier, Amsterdam, pp. 303–334.
- Hakanen M., Jaakkola T. & Korpela H. 1984. Simultaneous determination of ^{241}Pu , ^{238}Pu and $^{239,240}\text{Pu}$ in low activity environmental samples. *Nucl. Instr. Meth.* 223: 382–385.
- Hallstadius L. 1984. A Method for the electrodeposition of actinides. *Nucl. Instr. Meth.* 223: 266–267.
- Hanson W.C. 1967. Cesium-137 in Alaskan lichens, caribou and Eskimos. *Health Phys.* 13: 383–389.
- Hanson W.C. 1980. Transuranic elements in Arctic tundra ecosystems. In: Hanson W.C. (ed.), *Transuranic elements in the environment*. Technical Information Center/U.S. Department of Energy, Springfield, USA, pp. 441–458.
- Hardy E.P. (ed.) 1972. *Fallout Program Quarterly Summary Report (December 1, 1971 through March 1, 1972)*. HASL-249. U.S. Atomic Energy Commission.
- Hardy E.P., Krey P.W. & Volchok H.L. 1973. Global inventory and distribution of fallout plutonium. *Nature* 241: 444–445.
- Hindman F.D. 1983. Neodymium fluoride mounting for α spectrometric determination of uranium, plutonium, and americium. *Anal. Chem.* 55: 2460–2461.
- Holm E. & Persson B.R.R. 1975. Fall-out plutonium in Swedish Reindeer Lichen. *Health Phys.* 29: 43–51.
- Holm E. & Persson B.R.R. 1976. Transfer of fall-out plutonium in the food-chain lichen-reindeer-man. In: *Transuranium nuclides in the environment*. SM-199. International Atomic Energy Agency, Vienna, pp. 435–446.
- Holm E. & Persson B.R.R. 1977. Radiochemical studies of ^{241}Pu in Swedish reindeer lichens. *Health Phys.* 33: 471–473.
- Holm E. & Persson B.R.R. 1978. Global fallout of curium. *Nature* 273: 289–290.
- Jaakkola T., Harva K., Keinonen M. & Hakanen M. 1978. Studies on the behaviour of transuranic elements in plants. *Radioactive Foodchains in the Subarctic Environment*, Contract EY-76-C-02-3011.A002 of the US DOE, Report C-02-3011.
- Jaakkola T., Keinonen M., Hakanen M. & Miettinen J.K. 1981. Investigations of the transfer of plutonium and americium from plants to reindeer and man in Finnish Lapland. In: Wrenn M.E. (ed.), *Actinides in man and animals. Proceedings of the Snowbird Actinide Workshop, 15–17 October 1979*. RD Press, Salt Lake City, USA, pp. 509–524.
- Kauranen P. & Miettinen J.K. 1969. ^{210}Po and ^{210}Pb in the arctic food chain and the natural radiation exposure of Lapps. *Health Phys.* 16: 287–295.
- Miettinen J.K., Jokelainen A., Roine P., Lidén K. & Naversten Y. 1963. ^{137}Cs and potassium in people and diet — A study of Finnish Lapps. *Ann. Acad. Sci. Fenn., Series A, II Chemica* 120, Helsinki.
- Mussalo-Rauhamaa H. 1981. *Accumulation of plutonium from fallout in Southern Finns and Lapps*. Dissertation, University of Helsinki, Department of Radiochemistry.
- Paatero J., Jaakkola T. & Kulmala S. 1998. Lichen (*Cladonia*) as deposition indicator for transuranium Elements investigated with the Chernobyl fallout. *J. Environ. Radioact.* 38: 223–247.
- Paatero J., Jaakkola T. & Reponen A. 1994. Determination of the ^{241}Pu deposition in Finland after the Chernobyl accident. *Radiochim. Acta* 64: 139–144.
- Persson B.R. 1970. ^{210}Pb -Atmospheric deposition in lichen-carpets in northern Sweden during 1961–1969. *Tellus* 22: 564–571.
- Rahola T., Jaakkola T., Miettinen J.K., Tillander M. & Suomela M. 1988. Radiation dose to Finnish Lapps — Comparison of effects of fallout from atmospheric nuclear weapons tests and from the Chernobyl accident. *Arctic Medical Res.* 47, Suppl. 1: 186–190.
- Reponen A., Jantunen M., Paatero J. & Jaakkola T. 1993. Plutonium fallout in southern Finland after the Chernobyl accident. *J. Environ. Radioact.* 21: 119–130.
- Rissanen K., Rahola T. & Aro P. 1990. Distribution of cesium-137 in reindeer. *Rangifer* 10: 57–66.
- Rissanen K., Rahola T., Illukka E. & Alfthan A. 1987. *Radioactivity of reindeer, game, and fish in Finnish Lapland after the Chernobyl accident*. STUK-A63. Finnish Centre for Radiation and Nuclear Safety, Helsinki, 33 pp.
- Svensson G.K. & Lidén K. 1965. The transport of ^{137}Cs from

- lichen to animal and man. *Health Phys.* 11: 1393–1400.
- Tulikoura J., Jaakkola T. & Miettinen J.K. 1974. Plutonium in lichen in Finland during 1960–1973. In: *Radioactive Foodchains in the Subarctic Environment. Technical Progress Report for the Period Aug. 15, 1973–Aug. 14, 1974*. U.S. Atomic Energy Commission contract CH AT (11-1)-3011.
- Yamato A. 1982. An anion exchange method for the determination of ²⁴¹Am and plutonium in environmental and biological samples. *J. Radioanal. Chem.* 75: 265–273.

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