

## PLUTONIUM AND AMERICIUM IN THE FOODCHAIN LICHEN-REINDEER-MAN

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The atmospheric nuclear tests have produced a worldwide fallout of trans-uranium elements. In addition to plutonium measurable concentrations of americium are to be found in terrestrial and aquatic environments. Results of the investigations on transfer of plutonium along the foodchain lichen-reindeer-man have been published earlier (1,2).

In the present study the metabolism of plutonium in reindeer was investigated by analyzing plutonium in liver, bone and lung collected during 1963-1976. To determine the distribution of plutonium in reindeer all tissues of four animals of different ages were analyzed. To estimate the uptake of plutonium from the gastrointestinal tract in reindeer, the tissue samples of elk were also analyzed. Elk which is of the same genus as reindeer does not feed on lichen but mainly on deciduous plants, buds, young twigs and leaves of trees and bushes. The composition of its feed corresponds fairly well to that of reindeer during the summer time.

Studies on behaviour of americium along the foodchain lichen-reindeer-man were started by determining the Am-241 concentrations in lichen and reindeer liver. The Am-241 results were compared with those of Pu-239,240.

The plutonium contents of the southern Finns whose diet does not contain reindeer tissues were determined by analyzing autopsy tissue samples (liver, lung and bone). The southern Finns form a control group to the Lapps consuming plenty of reindeer tissues. Plutonium analyses of the placenta, blood and tooth samples of the Lapps are performed presently.

For separation of plutonium and americium the samples were dried and wet-ashed by  $\text{HNO}_3\text{-HCl}$ . Pu-242 and Am-243 were used as tracers for yield determination. Plutonium was isolated by anion-exchange and electrodeposited on a platinum disc from a dilute  $\text{HNO}_3$ -solution. For separation of americium the anion- and cation-exchange techniques combined with HDEHP extraction or  $\text{BiPO}_4$ -coprecipitation were used (3). Americium was electrodeposited from a  $\text{NH}_4\text{Cl-oxalic acid}$  solution. Alphaspectra of Pu and Am were determined using a surface barrier semiconductor-detector and a 256-channel pulse-height analyzer.

Maximal concentrations of Pu-239,240 and Am-241 in lichen (sp. Cladonia) 220-240 and 40 pCi/kg dry wt. resp., were found in 1964 (Fig. 1). Comparison to Pu-239,240 concentrations in surface air, determined in the United Kingdom by Cambray et al. (4), indicates a short residence time for these radio-nuclides in lichen. The maximal concentration of plutonium in air as well as in grass and birch leaves (100 pCi/kg dry wt.) occurred in 1963 (Fig. 1).

Am-241 is a daughter nuclide of Pu-241 which decays with a half-life of 14.9 a to Am-241. In fresh fallout the Pu-241/Pu-239,240 activity ratio was about 15 in 1963 (5). According to this study this ratio in lichen was 12.0-2.3 in 1963, 10.9-1.8 in 1965 and 7.7-3.7 in 1974. In 1963 the Pu-241 concentration in lichen was 2400 pCi/kg dry wt. The activity ratio Am-241/Pu-239,240 in lichen increases by time. It was about 0.15 in 1965 and 0.25 in 1973. This increase is probably mainly due to the Am-241 produced by decay of Pu-241 in the lichen and does not indicate a higher retention of americium by lichen compared with that of plutonium. It has been estimated that the activity ratio Am-241/Pu-239,240 of integrated fallout on the earth's surface will be at its maximum, 0.42, about the year 2040 (6).

The highest Pu-239,240 concentration measured in reindeer was 56 pCi/kg fresh wt. It was found in the liver of a 12-15 year old reindeer slaughtered in 1973 (Table 1). In general, the plutonium content of liver is higher in old animals which have lived during the period of high plutonium content in lichen. In some cases, however, the high concentrations are likely due to exceptionally high contribution of lichen to the diet of reindeer (Table 1). The major part of plutonium in reindeer occurs in liver. Skeleton contains usually 20-30% of the body burden. Only 1-10% occurs in lung (Fig. 2).

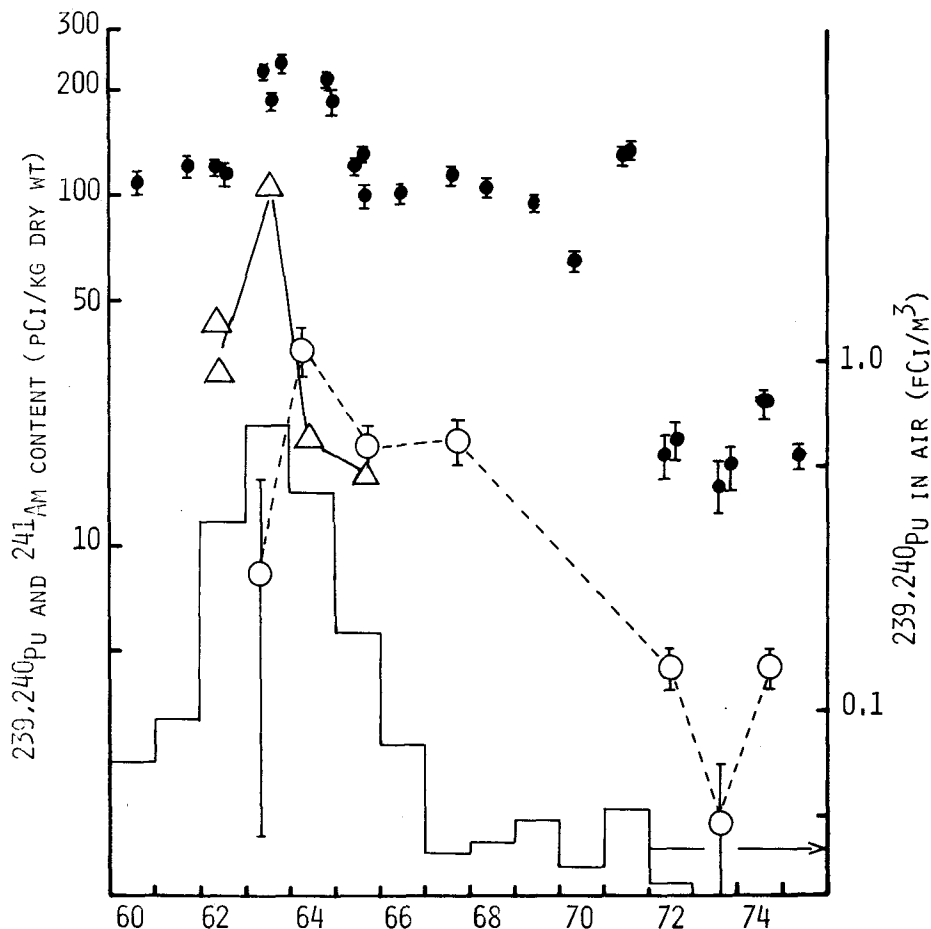


FIGURE 1  $^{239,240}\text{Pu}$  (●) and  $^{241}\text{Am}$  (○) in lichen and  $^{239,240}\text{Pu}$  in birch leaves and grass (△) in Finnish Lapland during 1960-1975. Standard deviation of radioassay (one  $\sigma$ ) is indicated. In addition the  $^{239,240}\text{Pu}$  concentrations in air measured at Chilton, United Kingdom are indicated (Cambray et al. Ref. 4)

Sampling year	No of animals	Age years	pCi/kg fresh wt	
			mean	(min-max)
1963-64	5	3.5- 5	11.4	( 3.5-31.3)
1965-66	5	1.5- 4	7.7	( 1.6-18.6)
1967-68	33	2 - 8	14.6	
1971-72	6	2 - 4	3.9	( 2.4- 5.5)
1973	3	3 - 6	2.1	( 1.6- 2.4)
1973	1	12 -15	55.6	
1974	6	3 -13	3.1	( 0.6- 7.1)
1975	10	3 - 8	2.8	( 0.4- 8.2)
1976	6	1.5- 3	1.2	( 0.7- 2.4)
1976	2	4 - 6	29.9	(28.3-31.4)

Table 1. Pu-239,240 in reindeer liver in Finnish Lapland during 1963-1976. Standard deviation of the radioassay (one  $\sigma$ ) is indicated.

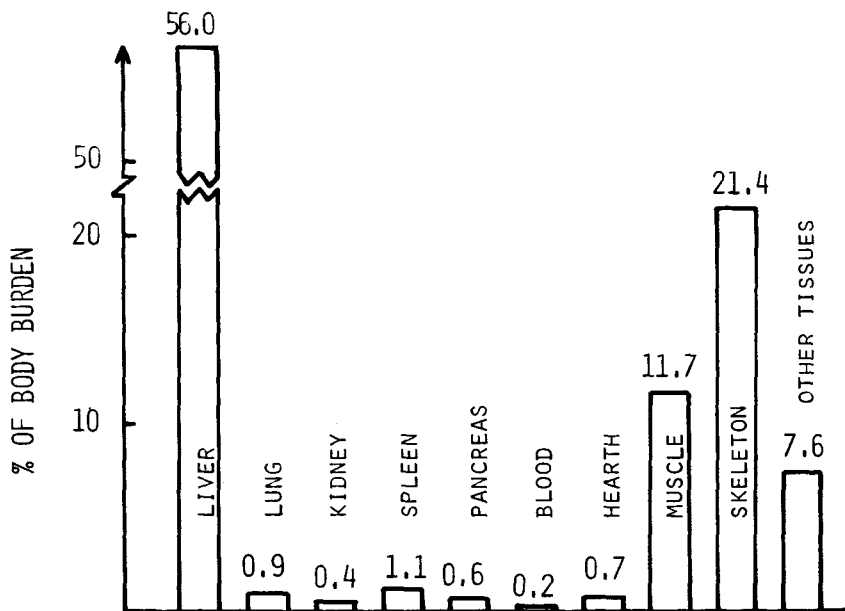


FIGURE 2 DISTRIBUTION OF  $^{239,240}\text{Pu}$  IN REINDEER; AGE 13.5 YEARS, SLAUGHTERED IN NOVEMBER 1974

In 1964-1966, the plutonium concentration in the lung of reindeer was only slightly higher than in that of elk. Instead, the plutonium concentration in the liver of reindeer was 10-50 times, and in bone 5-20 times higher than in those of elk apparently due to the greater dietary uptake of plutonium by reindeer. Assuming that reindeer feeds about 600 kg of lichen (dry wt.) during eight winter months the total dietary intake in its life-time was estimated. The ratio of the sum of plutonium in reindeer liver and bone of reindeer to the total dietary intake was  $5 \times 10^{-7}$ . This gives a rough estimate

Year	Animal age sex years	pCi/kg fresh wt		$^{241}\text{Am}/^{239,240}\text{Pu}$
		$^{241}\text{Am}$	$^{239,240}\text{Pu}$	
1974	13.5 ♂	$1.2^{+0.1}$	$7.1^{+0.2}$	$0.16^{+0.02}$
1975	8 ♀	$1.3^{+0.2}$	$8.2^{+0.6}$	$0.17^{+0.02}$
1976	2 ♂	$0.5^{+0.1}$	$2.4^{+0.1}$	$0.20^{+0.04}$
1976	3 ♀	$0.3^{+0.1}$	$1.3^{+0.1}$	$0.22^{+0.09}$
1976	4 ♀	$4.3^{+0.1}$	$28.3^{+1.2}$	$0.15^{+0.01}$
1976	6 ♀	$5.4^{+0.2}$	$31.4^{+1.3}$	$0.17^{+0.01}$

Table 2. Am-241 and Pu-239,240 in reindeer liver in Finnish Lapland during 1974-1976. Standard deviation of the radioassay (one  $\sigma$ ) is indicated.

for the absorption of plutonium from the diet. The value for the uptake of plutonium from the gastrointestinal tract in man, given by the ICRP is  $3 \times 10^{-5}$  to  $10^{-6}$  (7).

The concentrations of Am-241 in reindeer liver are given in Table 2. The ratio Am-241/Pu-239,240 was quite constant, 0.18-0.03 (mean value of 6 animals). This is slightly lower than the values 0.22-0.25 reported for integrated fallout in soil in 1973 (6).

In 1975 the dietary Pu-239,240 intake of the Lapps was 10 pCi for men and 16 pCi for women. Based on the Am-241 concentrations in reindeer it was estimated that the corresponding values for Am-241 intake were 2 pCi/year and 1 pCi/year for men and women, resp.

The mean values of Pu-239,240 content in liver, bone and lung of southern Finns (13 subjects) were 0.52, 0.20 and 0.03, resp. Assuming that all plutonium is in these tissues, the body burden of Pu-239,240 varied from 0.44 to 1.02 pCi being 0.75 on the average. The radiation dose due to the  $\alpha$ -radiation of Pu is for the southern Finns 0.3, 0.2 and 0.02 mrem/year for liver, bone and lung, resp.

#### ACKNOWLEDGEMENT

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#### REFERENCES

- (1) JAAKKOLA, T., MIETTINEN, J.K., TULIKOURA, J. and MUSSALO, HELENA, Paper presented at the Third European Congress of the International Radiation Protection Association, Amsterdam, The Netherlands, 13-16 May, 1975
- (2) HOLM, E. and PERSSON, R.B.R., (Proc. Symp. San Francisco, 1975) IAEA, Vienna (1976) 435
- (3) NAITO, K., report UCRL-8748, Chemistry-General, TID-4500 (1959)
- (4) CAMBRAY, F.S., EAKINS, J.D., FISHER, E.M.R. and PEIRSON, D.H., A.E.R.E.-R 7832 (1974)
- (5) Global Atmospheric Pu-239 and Pu Isotopic Ratios for 1959-1970, US AEC Report HASL-273 (1973) III
- (6) KREY, T.W., HARDY, E.P., PACHUCKI, C., ROURKEY, F., COLUZZA, J., and BENSON, W.K., (Proc. Symp. San Francisco, 1975), IAEA, Vienna (1976) 671
- (7) The Metabolism of Compounds of Plutonium and other Actinides, ICRP Publication 19 (1972)