

# Americium in the Finnish environment

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This paper reviews studies on environmental americium,  $^{241}\text{Am}$ , in Finland. There are two sources of americium in the Finnish environment: fallouts from nuclear weapons tests in the 1950s and 1960s and from the Chernobyl accident in 1986, the former constituting around 98% of the total environmental load. The weapons test fallout was distributed more or less uniformly over Finland, while the Chernobyl fallout was deposited on a sector from southwestern coast to northeast. The total deposition of  $^{241}\text{Am}$  in Finland is approximately  $20 \text{ Bq m}^{-2}$  and the amount is still somewhat increasing due to decay of  $^{241}\text{Pu}$ . In this paper, the distribution and migration of americium in forest and aquatic environments is described. Americium concentrations in natural waters, sediments, soils, vegetation and fishes are given. In addition, the behaviour of americium in the food chain from lichen via reindeer into man is discussed. Radiation doses to humans due to the environmental americium in Finland are of no practical importance.

## Introduction

Americium is an artificial actinide element formed in nuclear reactors and in nuclear explosions. The most important radionuclide of americium,  $^{241}\text{Am}$ , is an alpha emitter with a half-life of 433 years. It is formed by beta decay of  $^{241}\text{Pu}$ , which is the most prevailing plutonium nuclide in fresh fallout from nuclear explosions and accidents. Americium is a highly radiotoxic element that together with plutonium forms most of the alpha activity in fallout situations.

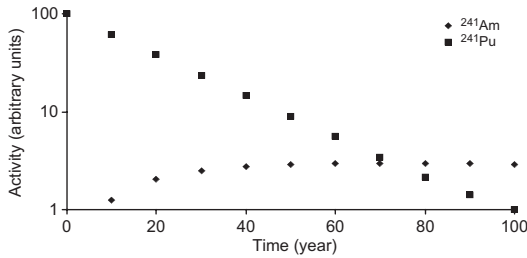
This paper reviews the studies on americium in the environment and in the food chains in Finland. These studies started in the early 1970s at the University of Helsinki. Two other important Finnish institutes in the research of environmental transuranium elements have been the Finnish Authority for Radiation and Nuclear Safety (STUK) and the Finnish Meteorological Institute. The sources of americium in the Finnish

environment have been fallouts from the nuclear weapons tests and the Chernobyl accident, the former constituting around 98% of the total environmental americium.

In this paper, I (1) describe sources and distribution of americium in Finland, (2) explain behaviour and distribution of americium in aquatic (sea and lake waters, sediment profiles as well as in fishes) and terrestrial (forest soil horizons, forest plants as well as in the food chain from lichen via reindeer into man) environments. For comparison, corresponding data on plutonium in most cases and on cesium in some cases will be given as well.

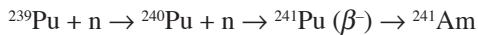
## Nuclear and chemical properties of americium

Americium is a transuranium element formed in nuclear fuel and in nuclear explosions. Ameri-



**Fig. 1.** In-growth of  $^{241}\text{Am}$  from  $^{241}\text{Pu}$  as a function of time.

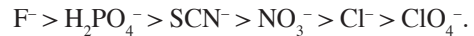
cium has altogether thirteen isotopes, of which  $^{241}\text{Am}$  is the most important. The lighter americium isotopes are short-lived while the heavier ones, some of which having rather long half-lives, are not formed to such a high extent as  $^{241}\text{Am}$ . The half-life of  $^{241}\text{Am}$  is 433 years. It is formed by neutron absorption and beta decay from  $^{239}\text{Pu}$  in the following way:



The half-life of  $^{241}\text{Pu}$  is rather short, 14.4 years, and thus the in-growth of  $^{241}\text{Am}$  is accordingly rather rapid (Fig. 1). At the year 73 the in-grown activity of  $^{241}\text{Am}$  equals that of  $^{241}\text{Pu}$ , after which the activity of  $^{241}\text{Am}$  starts to decrease gradually.  $^{241}\text{Pu}$  is the most abundant plutonium nuclide in spent nuclear fuel and in the fallout of nuclear explosions. In spent fuel, the abundance of  $^{241}\text{Pu}$ , and thus also the activity of  $^{241}\text{Am}$ , is higher because there is more time for the successive neutron captures than in explosions.  $^{241}\text{Am}$  decays by alpha mode to  $^{237}\text{Np}$ . There are altogether five alpha emissions, of which the most intensive have energies of 5.486 MeV (86%) and 5.442 MeV (13%).  $^{241}\text{Am}$  also emits 59.5 keV gamma radiation with 36% intensity.  $^{241}\text{Am}$  emits also conversion electrons and X-rays.

Americium is the seventh member of the actinide series for which filling of the 5f orbital is chemically characteristic. The preceding actinides (Th, Pa, U, Np and Pu) behave in a transition metal manner, having several oxidation states and forming strong complexes. From americium onwards the actinides behave lanthanide-like: their prevailing oxidation state is +3 and their compounds are ionic in nature. Also americium has several oxidation states, but in environmental conditions +3 is the only one

present. In natural waters at neutral and slightly basic pH ranges americium forms complexes with carbonate and hydroxide:  $\text{Am}(\text{CO}_3)_x^{3-2x}$  and  $\text{Am}(\text{OH})_x^{3-x}$ .  $\text{Am}^{3+}$  is also readily adsorbed by mineral and humics surfaces in waters, soils and sediments. In acidic media, where carbonate and hydroxide are absent, americium forms complexes with other inorganic ligands and the strength of these complexes decreases in the order



In the lanthanide series, the closest homolog is europium, which can be used as an analog in americium studies. Using europium as an analog is necessary when macro amounts are needed, for example, in spectroscopic speciation studies. The amount of americium in environmental samples is so low that no direct analysis of americium species is available.

## Sources and distribution of americium in Finland

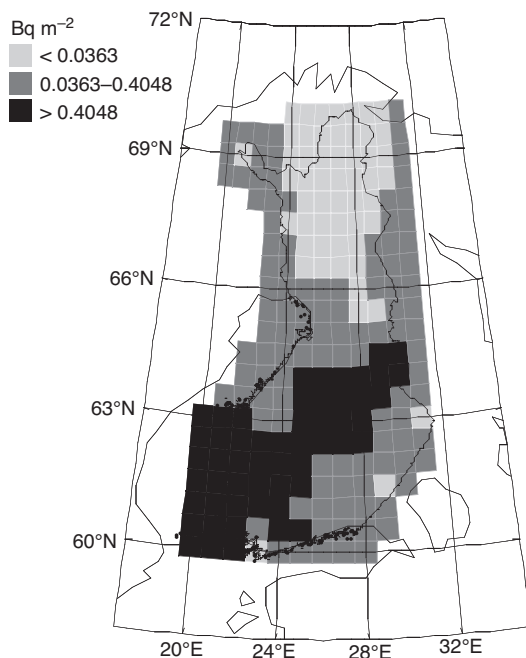
There are two sources of environmental americium in Finland: atmospheric nuclear weapons tests and Chernobyl accident. As will be described below, the former is far more important than the latter. The share of releases from Finnish nuclear power plants (NPP) to  $^{241}\text{Am}$  contamination in the environment is negligible. For example, in the years 1995–1996 the Olkiluoto NPP on the west coast of Finland released annually only 6100 Bq of  $^{241}\text{Am}$  into the air and 27 000 Bq into the sea (Ilus *et al.* 2003). In order to get a rough estimate of the total release, we assume that both nuclear power plants release this much every year. By multiplying  $2 \times (6100 + 27\,000)$  Bq by the average years of operation of these plants (28), we get the total release of  $2 \times 10^6$  Bq. This is only one millionth of the amount of americium present in the environment from the nuclear weapons test fallout.

## Fallout from nuclear weapons tests

About 500 atmospheric nuclear tests were car-

ried out from the 1940s to 1970s, the most active years being 1958 and 1962. These tests created global radioactive fallout that polluted Finland in the same manner as other countries. Salminen *et al.* (2005) calculated the total  $^{241}\text{Am}$  deposited in Finland from the nuclear weapons tests to be  $2.1 \times 10^{12}$  Bq. This calculation was based on the observed  $^{241}\text{Am}/^{239,240}\text{Pu}$  ratio of 0.107 in lichen samples from the years 1963–1965 (Jaakkola *et al.* 1981). It is, however, somewhat questionable whether all  $^{241}\text{Am}$  from those years was from direct deposition. It is probable that a fraction of  $^{241}\text{Am}$  in the lichen samples originated from the in-growth from  $^{241}\text{Pu}$  deposited from earlier tests during the 1950s, since about 60% of the atmospheric tests had been done already in 1958 and before. Due to the lack of data on airborne and rainwater concentrations of  $^{241}\text{Am}$  from those years, no definite conclusion on the direct deposition from the nuclear weapons tests can be drawn, but we can assume that the value of  $2.1 \times 10^{12}$  Bq might be a slight overestimation.

Calculated from the total  $^{241}\text{Am}$  deposition value given above ( $2.1 \times 10^{12}$  Bq) the deposition of  $^{241}\text{Am}$  in the mid-1960s was on average  $6.2 \text{ Bq m}^{-2}$ . Today, i.e. in the first decade of the 21st century, the deposition of  $^{241}\text{Am}$  in soils and sediments is about  $22 \text{ Bq m}^{-2}$ , which means an increase of about four-fold in about forty years. Outola (2003) gives an average value of  $19 \text{ Bq m}^{-2}$  in Harjavalta soils, Ikäheimonen *et al.* (2006) provide a value of  $22 \text{ Bq m}^{-2}$  in Mänttä soil, I (with collaborators) found a value of  $25 \text{ Bq m}^{-2}$  in Inari soil, and M. Lusa (unpubl. data) gives a value of  $20 \text{ Bq m}^{-2}$  in Päijänne sediment. Inari is situated in the northernmost part of Finland, while the other three sites are in southern Finland. As can be seen from these results,  $^{241}\text{Am}$  deposition from the nuclear weapons tests was rather evenly distributed over Finland. In general this has already been known for a long time and experimentally verified in the case of  $^{137}\text{Cs}$  deposition. In all cases, only a minor fraction of  $^{241}\text{Am}$  is from the Chernobyl accident, which will be discussed in the next section. The  $^{241}\text{Am}/^{239,240}\text{Pu}$  ratio has increased from the value of 0.11 in the mid-1960s to the value of about 0.44 today: 0.43 in Harjavalta, 0.43 in Mänttä, 0.44 in Inari and 0.44 in Päijänne.



**Fig. 2.** Concentration of  $^{241}\text{Am}$  ( $\text{Bq m}^{-2}$ ) from the Chernobyl accident in Finland (source: Salminen *et al.* 2005).

### Fallout from the Chernobyl accident

Salminen *et al.* (2005) determined the  $^{241}\text{Am}$  distribution in Finland from 62 peat and 9 lichen samples taken from various locations in Finland immediately after the accident in 1986. They found depositions ranging between  $0.00115$  and  $9.32 \text{ Bq m}^{-2}$  (Fig. 2). Unlike in nuclear weapons tests fallout, the distribution was very uneven. The highest deposition took place in the sector from the southwestern coast to northeast, which correlates with an air mass trajectory associated with the initial release of radionuclides from Chernobyl (Pöllänen *et al.* 1997). The deposition pattern of  $^{241}\text{Am}$  resembles those of other refractory nuclides, such as plutonium isotopes  $^{95}\text{Zr}$  and  $^{144}\text{Ce}$  (Arvela *et al.* 1990, Reponen *et al.* 1993, Paatero *et al.* 2002).

Salminen *et al.* (2005) also calculated that the total deposition of  $^{241}\text{Am}$  in Finland from the Chernobyl accident was  $3.7 \times 10^{10}$  Bq, which is only 1.7% of the total americium deposited in Finland, the rest originating from the nuclear weapons tests. This is an average value and in the most Chernobyl-contaminated areas the Cherno-

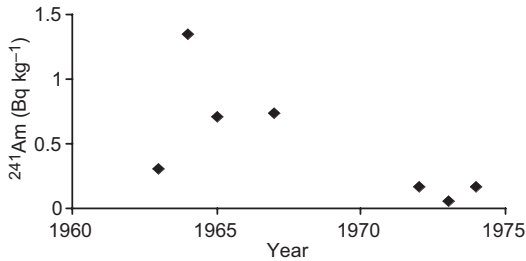


Fig. 3. Concentration of <sup>241</sup>Am in lichen (*Cladonia alpestris*) in Lapland in 1964–1975 (source: Harva 1977).

byl fraction was higher but still below 10%. From the total <sup>241</sup>Am released from the Chernobyl reactor 0.8% was deposited in Finland.

In the Chernobyl fallout, the fraction of <sup>241</sup>Pu in the total plutonium deposited was considerably higher than in the weapons test fallout. The <sup>241</sup>Pu/<sup>239,240</sup>Pu ratio was 95 in the former and 15 in the latter (Jaakkola *et al.* 1981, Paatero *et al.* 1994). This also means that the Chernobyl-derived fraction of <sup>241</sup>Am in the total transuranium pollution will be higher than in nuclear weapons test fallout. Due to ongoing decay of <sup>241</sup>Pu the amount of <sup>241</sup>Am in the environment is still increasing. In 1986 the <sup>241</sup>Am/<sup>239,240</sup>Pu ratio in the Chernobyl deposition was 0.37 (Salminen *et al.* 2005). Now, in 2007, it is 2.3 and in 2059 it will have a maximum value of 2.8. Thus, in the Chernobyl-derived transuranium contamination, americium is the main component. In the overall contamination, plutonium still forms the major fraction, since the Chernobyl contamination was so low compared with that of the weapons tests. The overall <sup>241</sup>Am/<sup>239,240</sup>Pu ratio is currently 0.44, and by taking into account the fraction of Chernobyl-derived americium and present <sup>241</sup>Am/<sup>239,240</sup>Pu ratio in this fraction, we can calculate that in the weapons test fraction the <sup>241</sup>Am/<sup>239,240</sup>Pu ratio is 0.41, only slightly lower than the overall ratio. This ratio will still increase to a maximum value of 0.43 by the mid-2030s.

<sup>241</sup>Am was detected in the air over Finland during two weeks following the Chernobyl accident (Sinkko *et al.* 1987). On 28 April 1986 the ground-level air in Nurmijärvi in southern Finland contained 10–19  $\mu\text{Bq m}^{-3}$  of americium but the concentration decreased very rapidly already the next day. Still in the first half of May americium could be measured in concentrations

ranging from 0.09 to 0.22  $\mu\text{Bq m}^{-3}$ , but at the end of May it was already undetectable. In a later measurement in 1988, americium in the air was undetectable but in deposition samples it could be determined with the monthly-average amount of 0.4 mBq m<sup>-2</sup> (Saxen *et al.* 1994).

## Americium in the food chain lichen–reindeer–man

In the 1960s, a special food chain from lichen via reindeer into reindeer-herding Sami people was intensively studied at the Department of Radiochemistry, University of Helsinki. It was found to heavily enrich radioactive cesium. In the 1970s we started the study of transuranium elements plutonium and americium in this food chain.

### Lichen

The highest concentration of <sup>241</sup>Am in lichen (*Cladonia alpestris*) in Lapland being 1.5 Bq kg<sup>-1</sup> was measured in 1964, which was followed by a more or less systematic decrease (Fig. 3). The calculation of the effective half-life of americium in lichen from this curve is not possible. We can, however, conclude that this half-life is considerably shorter than that of plutonium (two to three years *see* Jaakkola *et al.* 1981), since the <sup>241</sup>Am/<sup>239,240</sup>Pu ratio decreased systematically from 0.37 in 1965 to 0.19 in 1974 (Harva 1977), even though the in-growth from <sup>241</sup>Pu generated increasing amounts of americium in lichen. Compared with concentrations of other radionuclides, the <sup>241</sup>Am concentration was fairly low. In 1964 the <sup>239,240</sup>Pu concentration was 5.5 times higher, <sup>241</sup>Pu concentration 80 times higher and <sup>137</sup>Cs concentration about 1500 times higher than that of <sup>241</sup>Am (Jaakkola *et al.* 1981).

After the Chernobyl accident in 1986–1987, <sup>241</sup>Am concentrations were determined from lichen samples taken from thirteen locations in southern, middle and northern Finland (Paatero *et al.* 1998). <sup>241</sup>Am concentrations varied widely. The highest concentration was found in the southern Finland in Uusikaupunki (1.8 Bq kg<sup>-1</sup>) and in the middle Finland in Sotkamo (1.6 Bq kg<sup>-1</sup>) in 1986. These sites are located in

the high deposition sector (Fig. 3). The lowest values were found in Inari in Lapland:  $0.04 \text{ Bq kg}^{-1}$  and  $0.12 \text{ Bq kg}^{-1}$ . It is rather surprising that highest values of  $^{241}\text{Am}$  in lichen after the Chernobyl accident were at the same level as in the peak years 1963–1964 of the weapons test fallout period, although the Chernobyl fallout was considerably lower.

Paatero *et al.* (1998) calculated the ecological half-life of  $^{241}\text{Am}$  in lichen in 1986–1988, i.e. after the Chernobyl fallout. They obtained the value of 0.88 years, which is a rather low value compared with those for other radionuclides ( $^{137}\text{Cs}$ ,  $^{134}\text{Cs}$ ,  $^{106}\text{Ru}$ ,  $^{110\text{m}}\text{Ag}$ ,  $^{125}\text{Sb}$  and  $^{144}\text{Ce}$ ) (Lehto *et al.* 2008). Since the physical half-life is so long, this value represents also the effective half-life.

In the Chernobyl fallout, the variability of activity results was high even in samples collected close to each other, which complicates the evaluation of the results. This is because large radioactive particles with higher activity concentrations, the so-called hot particles including fuel fragments, were more abundant in the Chernobyl fallout than in the weapons test fallout.

## Reindeer

In the 1970s, a systematic study on plutonium in various reindeer organs and tissues was accomplished, but the data on  $^{241}\text{Am}$  are much more limited.  $^{241}\text{Am}$  was determined from reindeer liver, bones and lungs (Jaakkola *et al.* 1981). In 1976, the  $^{241}\text{Am}/^{239,240}\text{Pu}$  ratio in liver was 0.18, while in lichen the corresponding ratio was higher (0.25). This indicates that americium is not accumulated in liver to such a high extent as plutonium is. In bone, however, the ratio was 1.0, which indicates the americium buildup in bones. The same ratio in lungs was 0.51, which was expected to indicate a higher residence time of americium as compared with that of plutonium in the lungs of reindeer (Tähtinen *et al.* 1977).

After the Chernobyl accident, a more thorough study on americium absorption and distribution in reindeer was carried out (Paatero *et al.* 1998). Reindeer samples were taken in the next year following the accident from Kuhmo, which is the southernmost reindeer-herding district in

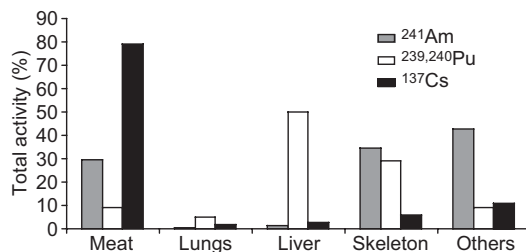


Fig. 4. Distribution of  $^{241}\text{Am}$ ,  $^{239,240}\text{Pu}$  and  $^{137}\text{Cs}$  in reindeer tissues and organs (sources: Rahola *et al.* 1975, Jaakkola and Paatero 1991, Paatero *et al.* 1998).

Finland. Kuhmo is located at the eastern border of middle Finland and belongs to the heavy transuranium fallout sector (Fig. 2).  $^{241}\text{Am}$  deposition in this area from the Chernobyl accident was  $0.6 \text{ Bq m}^{-2}$  and that of  $^{239,240}\text{Pu}$  was  $1.5 \text{ Bq m}^{-2}$ . Lichen concentrations were  $1.04 \text{ Bq kg}^{-1}$  for  $^{241}\text{Am}$  and  $2.09 \text{ Bq kg}^{-1}$  for  $^{239,240}\text{Pu}$ . Figure 4 shows the distribution of americium in various tissues and organs of reindeer. For comparison, the same data for plutonium and cesium are given as well (Rahola *et al.* 1975, Jaakkola and Paatero 1991).

Americium is accumulated mainly in bones and in meat, while cesium is accumulated more or less entirely in meat and plutonium in liver and bones (Fig. 4). Paatero *et al.* (1998) also calculated the transfer factors and absorption coefficients for americium. The transfer factor from ground contamination into reindeer meat was found to be  $7 \times 10^{-3} \text{ m}^{-2} \text{ kg}$ , while the corresponding value for plutonium was more than 100 times lower. The transfer factor for  $^{137}\text{Cs}$  in turn was 100 times higher than that of americium. The absorption coefficient from the gastro-intestinal tract into reindeer meat was calculated to be  $7.5 \times 10^{-4}$  for americium and  $3.0 \times 10^{-5}$  for plutonium. Thus, we can conclude that americium is much more efficiently absorbed into reindeer meat than plutonium, but compared with cesium its absorption and accumulation in reindeer meat is rather low.

## Man

No studies have been done on americium absorption and distribution in humans in Finland. In

order to get a rough picture on the dose from the ingestion of americium-bearing reindeer meat, I performed the following calculation. In the Kuhmo region right after the Chernobyl accident the  $^{241}\text{Am}$  concentration in the reindeer meat is  $0.0035 \text{ Bq kg}^{-1}$  (Paatero *et al.* 1998). The human absorption coefficient for americium is 0.0005, so from each kilogram of ingested reindeer meat  $1.75 \mu\text{Bq}$  of  $^{241}\text{Am}$  was absorbed. The dose conversion factor for americium is  $2 \times 10^{-7} \text{ Sv Bq}^{-1}$  and thus from each kilogram of ingested reindeer meat we get a dose of  $3 \times 10^{-13} \text{ Sv}$ . We assume that the  $^{137}\text{Cs}$  concentration in the reindeer meat was at the same time  $1000 \text{ Bq kg}^{-1}$ . The human absorption coefficient for cesium is 1.0 and the dose conversion factor is  $1.3 \times 10^{-8} \text{ Sv Bq}^{-1}$ . Thus, from each kilogram of ingested reindeer meat we get the dose of  $1.3 \times 10^{-5} \text{ Sv}$ , which is  $4 \times 10^7$  times higher than the dose from americium. In conclusion, we can say that the dose from ingestion of  $^{241}\text{Am}$  is negligible.

## Americium in the forest environment

Before the Chernobyl accident no studies on americium and plutonium in forest soil and their transfer from soil into plants were done. These studies started only recently in the late 1990s. Outola (2003) studied plutonium and americium in pine forest soil in Harjavalta in the southwestern Finland. The objective of the study was to find out the effect of air emissions from a copper smelter on the behaviour of radionuclides in soil and on their transfer from soil into natural plants. In that area the amount of  $^{241}\text{Am}$  was  $19 \text{ Bq m}^{-2}$  and the  $^{241}\text{Am}/^{239,240}\text{Pu}$  ratio 0.43. Only 3% of plutonium originated from the Chernobyl accident and the same is probably true for americium as well. Soil and vegetation samples were collected from increasing distances (0.5, 2, 4 and 8 km) from the smelter. The 22-cm-deep soil cores were divided into four fractions: litter, organic layer and two mineral layers (elluvial and illuvial horizons). At the reference site (8 km away) assumed not to have been affected by the smelter, most of the americium activity (about 75%) was in the organic layer, 5% in the litter and 10% in both mineral layers. When going

closer to the smelter, the fraction in the litter increased to 65% at the distance of 0.5 km, while the fractions in the other horizons decreased accordingly. This was explained by the inhibition of bacterial activity by the airborne pollution from the smelter, which in turn decreased the decomposition of litter. At distances between two and eight kilometres, the concentrations of  $^{241}\text{Am}$  in plants did not vary much. They were, on average,  $1.0 \text{ mBq kg}^{-1}$  in lingonberry twigs,  $2.1 \text{ mBq kg}^{-1}$  in crowberry twigs,  $7.5 \text{ mBq kg}^{-1}$  in mushrooms,  $10.1 \text{ mBq kg}^{-1}$  in reindeer lichen and  $5.4 \text{ mBq kg}^{-1}$  in iceland lichen. The aggregated transfer factors did not depend in a clear way on the distance from the smelter. The transfer factors of americium were, however, clearly higher than those of plutonium, indicating a better bioavailability of americium.

Ikäheimonen *et al.* (2006) determined  $^{241}\text{Am}$  in soil in Mänttä in southern Finland. They found  $22 \text{ Bq m}^{-2}$  of americium in a 15-cm-deep soil column and the  $^{241}\text{Am}/^{239,240}\text{Pu}$  ratio was 0.43, indicating that the main source was nuclear weapons test fallout. Both americium concentration and the  $^{241}\text{Am}/^{239,240}\text{Pu}$  activity ratio were very close to those obtained by Outola (2003) in Harjavalta. The peak concentration of americium was at the depth of 3 cm, which probably was the organic layer. They also studied the concentrations of americium in several plant species. In birch leaves and wild berries, including fruit, twigs and leaves, americium was undetectable. They could, however, find americium in ferns and *Cantharellus* mushrooms (concentrations were  $0.11 \text{ Bq kg}^{-1}$  and  $0.03\text{--}0.10 \text{ Bq kg}^{-1}$ , respectively). In ferns the average activity concentration of  $^{239,240}\text{Pu}$  was three times lower than that of americium. In another study by Ikäheimonen *et al.* (2004) they found  $^{241}\text{Am}$  in *Parmelia physodes*, an epiphytic lichen growing on tree stems, in concentrations varying between  $0.03$  and  $0.08 \text{ Bq kg}^{-1}$ . The study was carried out in 2001 close to the Loviisa nuclear power plant on the south coast of Finland and its objective was to find radionuclide-enriching bio-indicators in which the contamination could be found and measured.

In 2001, I (with collaborators) collected soil and plant samples from Inari in Lapland and measured their americium, plutonium and

cesium activities (*see* Fig. 5 for the fractions of the radionuclides found in five soil horizons). Soil samples were taken from pine forest where the soil is of stratified podzolic nature, the same type as in the study by Outola (2003).

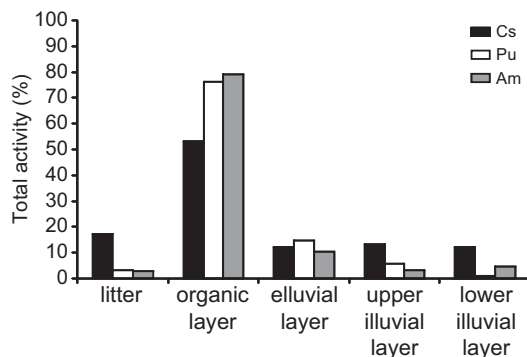
Most of the  $^{241}\text{Am}$  activity (76%) could be found in the organic layer. The value is more or less identical with that obtained by Outola (2003) in Harjavalta. Americium behaves in a very similar manner as plutonium in soil. Only about 3% of these two elements were in the litter and their concentrations in mineral layers decrease when going deeper into the soil.  $^{137}\text{Cs}$ , however, behaves quite differently. A large fraction (17%) was found in the litter, which probably indicates recycling of cesium into surface via root uptake by the plants. In addition, larger fractions of cesium have been migrating into mineral layers due to the higher solubility of cesium as compared with those of americium and plutonium.

The americium concentrations in plants in Inari were very low: in blueberry and lingonberry fruit they were on average  $2.1 \text{ mBq kg}^{-1}$ , in their twigs they were  $5.2 \text{ mBq kg}^{-1}$  and the aggregated transfer factors were 0.0001 and  $0.0003 \text{ m}^2 \text{ kg}^{-1}$ , respectively. These values are somewhat higher than those obtained by Outola (2003). In *Russula* mushrooms the americium activity was below the detection limit.

## Americium in the aquatic environment

### Americium in the Baltic Sea

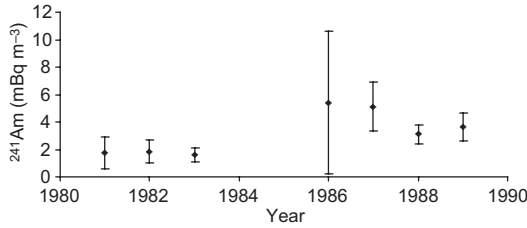
In 1980–1981, S. Leskinen (unpubl. data) collected several seawater samples from three sites in the Baltic Sea: the Gulf of Finland between Helsinki and Tallinn, the Gulf of Bothnia north of Åland and Baltic Proper between Gotland and Latvia. Water activities of  $^{241}\text{Am}$  and  $^{239,240}\text{Pu}$  varied in a rather narrow range, being on average  $1.36$  and  $4.48 \text{ mBq m}^{-3}$ , respectively. The  $^{241}\text{Am}/^{239,240}\text{Pu}$  ratio was thus 0.30, which is a typical value in fallout at that time. The particle fraction ( $>0.45 \mu\text{m}$ ) contained an average of 16% americium and 11% plutonium. The  $^{241}\text{Am}/^{239,240}\text{Pu}$  ratio in the particle fraction was higher than in water phase: 0.50.



**Fig. 5.**  $^{137}\text{Cs}$ ,  $^{239,240}\text{Pu}$  and  $^{241}\text{Am}$  in soils horizons in 20-cm-thick soil profiles from Inari collected in 2001 (source: Lehto *et al.* 2008).

A few years earlier somewhat different data on americium and plutonium concentrations in seawater from four locations in the Gulf of Finland were obtained by M. Lax (unpubl. data). The average concentration of  $^{239,240}\text{Pu}$  in filtered ( $0.45 \mu\text{m}$ ) water was approximately the same as those detected by S. Leskinen (unpubl. data), being  $5.2 \text{ mBq m}^{-3}$ , but the  $^{241}\text{Am}$  concentration of  $4.1 \pm 1.5 \text{ mBq m}^{-3}$  was considerably higher than that measured by S. Leskinen (unpubl. data). Thus the  $^{241}\text{Am}/^{239,240}\text{Pu}$  ratio in water was 0.79, which was clearly higher than the 1980 fallout ratio that can be estimated to be then about 0.3. The particulate fraction ( $>0.45 \mu\text{m}$ ) contained 5% of the total americium and 11% of the total plutonium, and the  $^{241}\text{Am}/^{239,240}\text{Pu}$  ratio in the particulate fraction was 0.35. So, the  $^{241}\text{Am}/^{239,240}\text{Pu}$  ratio in the water and particulate phases had an opposite order in these two studies.

Säteilyturvallisuuslaitos 1982, 1983, 1984, Ikäheimonen *et al.* 1988, Saxen *et al.* 1989, Ilus *et al.* 1993 provide data on the concentrations of  $^{241}\text{Am}$  in surface waters of the Baltic Sea in the 1980s (Fig. 6). The average concentration in the years 1981–1983 was  $1.7 \text{ mBq m}^{-3}$  (range  $0.7$ – $3.1 \text{ mBq m}^{-3}$ ), which increased to about  $5 \text{ mBq m}^{-3}$  (range  $2$ – $14 \text{ mBq m}^{-3}$ ) in 1986. In 1987–1989, the concentrations were between  $3$  and  $5 \text{ mBq m}^{-3}$ . Although the number of data points was limited (24 in total) and their variations were considerable, especially in 1986, it looks reasonable to assume that the Chernobyl accident increased the americium concentration in the Baltic Sea by a factor of two to three.



**Fig. 6.** Annual concentrations of  $^{241}\text{Am}$  in the Baltic Sea. The values are averages of two to six determinations from surface water samples taken from various parts of the Baltic Sea (Baltic Proper, Gulf of Finland and the Gulf of Bothnia) (sources: Säteilyturvallisuuslaitos 1982, 1983, 1984, Ikäheimonen *et al.* 1988, Saxen *et al.* 1989, llus *et al.* 1993).

In 1981–1983, water samples were taken also from the vicinities of the nuclear power plants in Loviisa and Olkiluoto. Compared with the water concentrations in the other parts of the Baltic Sea (average  $1.8 \pm 0.7$  mBq m<sup>-3</sup>), the americium concentrations ( $2.3 \pm 1.4$  mBq m<sup>-3</sup>) in the vicinities of the nuclear power plants were not elevated. When comparing the values given by Säteilyturvallisuuslaitos (1982, 1983, 1984), Ikäheimonen *et al.* 1988, S. Leskinen (unpubl. data) and M. Lax (unpubl. data), those of the first five studies (i.e. Säteilyturvallisuuslaitos 1982, 1983) are very close to each other regarding the americium and plutonium concentrations but the data of M. Lax (unpubl. data) differ from these considerably.

The distribution of americium in sediment profiles from two sites in the Baltic Sea in 1982 are given by Säteilyturvallisuuslaitos (1983). One of these sites was Teili in the northern part of Baltic Proper and the other one was in the Gulf of Bothnia north of Åland. At the former site, the total americium activity in the sediment profile was 20 Bq m<sup>-2</sup> and at the latter one it was 76 Bq m<sup>-2</sup>. At both sites, the peak concentration was found in the 5–10-cm layer but only at the latter site americium could be found deeper in the sediment, even below 20 cm.

### Americium in lakes

Only a few data exist on americium in lakes in Finland before the Chernobyl accident. Hakanen *et al.* (1979) reported that the top 10 cm of a

sediment core taken from Pääjärvi in southern Finland contained 10.4 Bq m<sup>-2</sup> of  $^{241}\text{Am}$  and 52 Bq m<sup>-2</sup> of  $^{239,240}\text{Pu}$ . These values are in good agreement with the amounts obtained in later studies when taking into account the increase of  $^{241}\text{Am}$  due to decay of  $^{241}\text{Pu}$ .

After the Chernobyl accident, Suutarinen *et al.* (1989, 1993) studied the behaviour of plutonium and americium in Lake Päijänne. This lake is located in southern Finland and the study was important because Päijänne is the water source for the Helsinki metropolitan area. One year after the accident, the  $^{241}\text{Am}$  concentrations in water varied in the range from 3.7 to 22 mBq m<sup>-3</sup>. No time trend can be drawn from the given americium data, but the plutonium concentrations showed a very low, if any, decrease in the water phase during the two years time after the accident. In the particulate phase ( $> 0.45$  μm) the activity was 1.6 mBq m<sup>-3</sup> in 1986 and decreased to half of that (0.8 mBq m<sup>-3</sup>) in the following year. In August 1986, the maximum activity in the particulate phase was seen at the depth of 30–40 metres in a 50-m-deep basin. Also, in August 1986 the 1-cm top layer of sediment samples collected from depths varying between 19 and 51 metres contained on average  $1.0 \pm 0.4$  Bq m<sup>-2</sup> of  $^{241}\text{Am}$ . No data on americium distribution between water, particles and sediment were given, but in the case of plutonium the fractions in August 1986 were 24%, 3% and 73%, respectively. We can thus conclude that most of the plutonium and americium activities were deposited into the bottom sediment rather rapidly, but that the soluble fraction had a rather long residence time in the water phase.

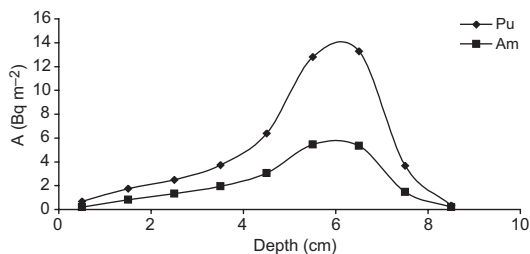
In 2007, M. Lusa (unpubl. data) collected water samples and sediment cores from the same 50-m-deep basin in Päijänne, as in the studies by Suutarinen *et al.* (1989, 1993), and determined their plutonium and americium concentrations (see Fig. 7. for the distribution of  $^{239,240}\text{Pu}$  and  $^{241}\text{Am}$  in the sediment cores). In the unfiltered water phase the concentrations were 4.9 mBq m<sup>-3</sup> ( $^{239,240}\text{Pu}$ ) and 4.1 mBq m<sup>-3</sup> ( $^{241}\text{Am}$ ). It is quite surprising that the decrease in the activities in twenty years has been rather small. In 1986 the average total concentration of  $^{239,240}\text{Pu}$  in the water phase was 14.9 mBq m<sup>-3</sup> and in 1987 it was 10.6 mBq m<sup>-3</sup> (Suutarinen *et al.* 1989). Ikäheimonen



*et al.* (2002) reported the  $^{241}\text{Am}$  concentration of  $8\text{ mBq m}^{-3}$  in Päijänne water 1986–1988.

The peak activities of both  $^{239,240}\text{Pu}$  and  $^{241}\text{Am}$  in sediment could be found at the depth of about six centimetres, which represent weapons test fallout. The total amounts of plutonium and americium in the cores were  $45$  and  $20\text{ Bq m}^{-2}$  (M. Lusa unpubl. data), respectively, which are typical values for weapons test fallout, as is also the obtained  $^{241}\text{Am}/^{239,240}\text{Pu}$  ratio of  $0.44$ . The fraction of  $^{239,240}\text{Pu}$  from the Chernobyl fallout was calculated from the  $^{238}\text{Pu}/^{239,240}\text{Pu}$  ratios in various layers and was found to be  $1.0\text{ Bq m}^{-2}$  ( $2.2\%$ ), which is a typical value for this area (Paatero *et al.* 2002). From the  $^{241}\text{Am}/^{239,240}\text{Pu}$  ratio we can assume that this is approximately the case for americium as well. The fractions of plutonium and americium in the water from their total inventories in Päijänne were  $0.2\%$  and  $0.3\%$ , respectively. In the water phase the  $^{241}\text{Am}/^{239,240}\text{Pu}$  ratio ( $0.84$ ) was twice as high as in the sediment phase ( $0.44$ ), which probably reflects the higher solubility of americium compared with that of plutonium. Another point indicating higher solubility of americium was that the  $^{241}\text{Am}/^{239,240}\text{Pu}$  ratio in the top 1-cm sediment layer was  $0.26$ , while deeper in the sediment (2–10 cm) it was clearly higher at  $0.47$  (M. Lusa unpubl. data).

Ikäheimonen *et al.* (2002) carried out an extensive study on plutonium, americium, curium and cesium concentrations in freshwater fishes from 17 lakes in southern and middle Finland. The fish samples were taken in 1986–1988. The concentrations were determined in flesh, liver and spawn. In flesh, the concentrations of  $^{239,240}\text{Pu}$  and  $^{241}\text{Am}$  were approximately the same:  $< 0.6\text{--}3.3$  and  $< 0.6\text{--}2.6\text{ mBq kg}^{-1}$ , respectively. The concentration factor, i.e. the ratio of concentration in flesh to that in water, was on average  $2.7$  times higher for americium as compared with that for plutonium. Furthermore, much higher americium than plutonium concentrations could be found in liver and spawn. In liver, the  $^{241}\text{Am}$  concentrations were  $8.7\text{--}140\text{ mBq kg}^{-1}$  while in the case of  $^{239,240}\text{Pu}$  they were  $< 3\text{--}6.9\text{ mBq kg}^{-1}$ . In spawn, the corresponding values were  $5.4\text{--}29\text{ mBq kg}^{-1}$  and  $< 3\text{ mBq kg}^{-1}$ . All this indicates that the uptake of americium by fish is considerably higher than the uptake of plutonium. The highest concentrations of plutonium and



**Fig. 7.** Distribution of  $^{239,240}\text{Pu}$  and  $^{241}\text{Am}$  in sediment cores from Päijänne in 2007. The curve represents average values from four cores from the depths of 30 m and 50 m (source: M. Lusa unpubl. data).

americium were detected in lakes in the high fallout sector from the southwest coast to northeast, while in Lake Saimaa in southeastern Finland the concentrations were below the detection limits. Ikäheimonen (2003) estimated that the consumption of freshwater fish in Finland in the peak activity year 1987 created an annual dose of  $0.05\text{ }\mu\text{Sv}$  due to all transuranium elements, including americium, and  $60\text{ }\mu\text{Sv}$  due to  $^{137}\text{Cs}$ . Even the latter one is rather small but the former is more or less negligible.

## Summary and conclusions

There are two sources of environmental americium in Finland: fallouts from atmospheric nuclear weapons tests and from the Chernobyl accident. The former constitutes 98% of the environmental contamination and the latter the rest. Releases from the Finnish nuclear power plants are negligible compared with these two sources. The total amount of americium in Finland is  $2.1 \times 10^{12}\text{ Bq}$  and it is still somewhat increasing for a few decades due to decay of  $^{241}\text{Pu}$ . The  $^{241}\text{Am}/^{239,240}\text{Pu}$  ratio is currently  $0.44$  and it will increase to  $0.46$  in a few decades. Deposition of americium is about  $20\text{ Bq m}^{-2}$ . Nuclear weapons test fallout was distributed over Finland in an even manner, whereas Chernobyl fallout was distributed into a sector from the southeast coast to northeast. Since the Chernobyl fallout of americium was low, it has not considerably affected the total deposition, not even on the most Chernobyl-affected area. In water systems most of the americium is now in sediments at the peak depth depending on the local sedimentation

rate. Water concentrations are very low, only a few millibecquerels per cubic metre, but the decrease rate is very low. In forest soils, most of americium is now in the organic layer where it is available for the root uptake by plants. The transfer from soil to plants is, however, very low and the concentrations of americium in forest plants are minimal. Radiation doses to humans due to ingestion of americium-containing foodstuffs are negligible as compared with natural radionuclides, such as uranium, radium, polonium and lead, and even as compared with the most important pollution nuclide  $^{137}\text{Cs}$ . So, there are no health effects on humans due to environmental americium.

Although in Finland during the last 35 years quite a good picture on environmental americium has been obtained, new information is needed to understand its behaviour from a scientific point of view. First of all, long-term data are needed to understand the development of americium profiles in soils and sediments. Leaching by water and the effect of humics and low molecular weight organic acids from the organic layer into mineral layers are important factors in understanding transfer mechanisms in soils. Another important problem in soils to be studied is the possible uptake of americium by plant roots. In sediments the interesting factors are the diffusion of americium in the sediment phase and the possibility of remobilisation of americium from sediment back to water.

All these problems are very difficult to study because the concentrations of americium are so low and the processes are complex. Also, the low concentrations of americium in the environment do not enable direct speciation of its chemical forms. However, not even indirect methods, such as sequential leaching techniques, have so far been applied for americium speciation in Finnish environmental samples. Studies on the behaviour of the closest chemical analog europium could shed more light on the behaviour of americium. Naturally-occurring europium in real systems or model systems with added europium could be studied. Using macro amounts of such an analog would also enable the use of modern analytical techniques for direct speciation. Deeper insight into the chemical forms of americium and transfer mechanisms cannot be attained with classical

radiochemical and radioanalytical methods, so new methods and collaboration with experts from other disciplines are needed. In addition, modelling of americium transfer processes in the environment would be needed to obtain an adequate general view on the behaviour of americium.

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