BOREAL ENVIRONMENT RESEARCH 7: 99–104
Helsinki 19 June 2002
ISSN 1239-6095

Transuranic elements in fishes compared to ^{137}Cs in certain lakes in Finland

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Ikäheimoinen, T. K. & Saxén, R. 2002. Transuranic elements in fishes compared to ^{137}Cs in certain lakes in Finland. *Boreal Env. Res.* 7: 99–104. ISSN 1239-6095

Transuranic elements from Chernobyl fallout were detected in freshwater fish sampled in Finland after the accident. Maximum concentrations of ^{239,240}Pu, ^{241}Pu, ^{241}Am and ^{242}Cm were 3.3, 110, 2.6 and 340 mBq kg^{-1}, respectively. In certain samples, ^{238}Pu/^{239,240}Pu ratios were between 1.0 and 7.2 while the expected value was 0.5. This could indicate differences in environmental mobilization between ^{238}Pu and ^{239,240}Pu. The concentration factor (CF) for Am from water to fish flesh, 230, was more than 2 times higher than the figure of 84 for Pu. The CF for Cm, 5100, was considerably higher than that of Am and does not confirm the presence of similar biological behaviour for Am and Cm. The average transfer factor (TF) from deposition to fish was highest for ^{137}Cs, 44×10^{-3} m^2 kg^{-1}. This was somewhat higher than the TF for Am but more than 10 times higher than that for Pu. Transuranic depositions that could cause the European Council regulation limit in freshwater fish to be exceeded are, Pu and Am respectively, 3000 and 1500 times higher than the highest fallout of Pu and Am recorded in Finland following the Chernobyl accident.

Introduction

^{137}Cs was the most important nuclide for radiation doses during the long period of radioactive fallout originating from the Chernobyl accident. In Finland the fallout also consisted of transuranic elements, albeit, in much smaller amounts than ^{137}Cs. Deposition of these nuclides was very unevenly distributed (Arvela *et al*. 1992, Paatero 2000). Rain showers washed most radiocaesium into soil and watersheds in central Finland, while deposition in eastern and northern Finland was low. Most transuranic elements were deposited with associated particles. Dry deposition also occurred. The deposition of transuranics was correlated with that for other nonvolatile nuclides, e.g. ^{95}Zr, but not with ^{137}Cs. The highest ^{239,240}Pu deposition found in northeastern Finland was about 8 Bq m^{-2} (Paatero 2000). The ratios of transuranics released from Chernobyl were about 0.55 for ^{238}Pu/^{239,240}Pu, 82 for ^{241}Pu/^{239,240}Pu, 12 for ^{242}Cm/^{239,240}Pu and 0.4 for ^{241}Am/^{239,240}Pu (USSR 1986). However, in deposition samples and other samples representing the deposition these ratios varied widely in Finland (Saxén *et al*. 1987, Reponen *et al*. 1993, Paatero 2000).

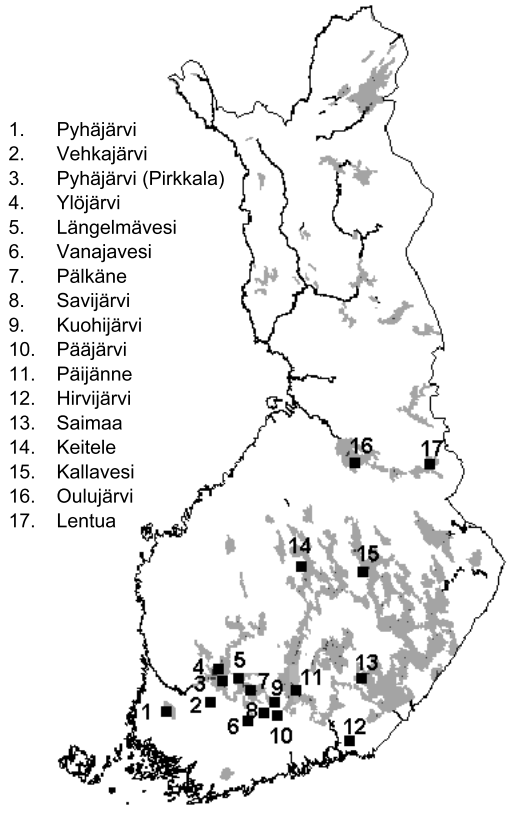

Fig. 1. Sampling sites of fish. Samples were taken in 1986–1988.

Uneven deposition and environmental factors cause large variation in ^{137}Cs contents in fishes from various lakes. Therefore, freshwater fishes were analysed after the Chernobyl accident with a rather extended sampling programme for ^{137}Cs (Saxén *et al*. 2000). Some of the programme samples were also taken for analyses of transuranic radionuclides (Fig. 1). The aim of the present study was to determine if any transuranic elements originating from the Chernobyl fallout could be seen in freshwater fishes from Finland within a period of 2.5 y after deposition. Little information is available on behaviour of transuranic elements in freshwater fishes. The study was focused on lake Päijänne where lake water samples were also analysed. The transfer factors (TFs) for transuranic elements from the lake water and deposition into the fish were calculated and compared with the TFs for ^{137}Cs in the southern lake Päijänne area.

Materials and methods

Transuranic elements and ^{137}Cs were analysed in 19 fish flesh, 9 liver and 5 spawn samples. The species studied were perch (*Perca fluviatilis*), pike (*Esox lucius*), whitefish (*Coregonus lavaretus*), vendace (*Coregonus sp*.) and burbot (*Lota lota*). Water samples from lake Päijänne were also analysed. Samples were taken between August 1986 and November 1988.

Fish were cleaned and gutted. Large fish were filleted and their heads, viscera, scales or skin, fins, backbone and other large bones were removed. Only the heads and viscera were removed from perch, vendace, roach and other small fish, thus only edible parts were taken for analysis. Fish liver and spawn samples were pooled from several individuals in each sample. The cleaned flesh samples were cut into small pieces for ^{137}Cs determination and packed into 560-ml Marinelli beakers. If only a small amount of tissue was available, a smaller 30-ml container was used. Some liver and spawn samples were partly analysed while fresh and some after drying and homogenization. Most tissue samples, however, were analysed in the smaller containers after ashing of the dried samples at 450 °C. Water samples (20 l) were concentrated by evaporation to about 500 ml and analysed for ^{137}Cs in Marinelli beakers.

Gammaspectrometric analysis was performed using either lithium-drifted or high-purity detectors with relative efficiencies ranging between 15% and 39%. The activity concentrations of the samples were calculated using the computer program GAMMA-83 (Sinkko 1981, Sinkko and Aaltonen 1985). The measurement time was usually 2–3 h.

For transuranic analysis the cleaned fish flesh, liver and spawn samples were dried overnight at 105 °C. Homogenized dried samples were wet-ashed with strong nitric acid and a mixture of nitric and hydrochloric acids. Tracers of ^{242}Pu and ^{243}Am were added before wet ashing. The fresh weights of the flesh samples were about 250 g, while those of liver and spawn samples were between 20 and 60 g. Pu separation from samples was based on anion exchange and changes in the oxidation states of Pu. After separation pure Pu was electrodeposited onto a stain-

less-steel disc and measured with alphaspectrometry by either silicon surface barrier semiconductor detectors or no-background PIPS detectors. (Taipale and Tuomainen 1985). Beta-emitting ^{241}Pu was measured with the ingrowth of its daughter nuclide ^{241}Am in the same disc after 12–13 y (Ikäheimonen 2000).

Am and Cm were purified together from a separated fraction coming from Pu analysis. After coprecipitation with oxalate, extraction with HDEHP was performed. The sample solutions were then passed through several ion-exchange columns. After electrodeposition, Am and Cm were measured with alphaspectrometry. Due to their chemical similarity, ^{243}Am could be used to determine the yield of both elements, this was verified experimentally. The lake water samples (100–200 l) were analysed using a similar procedure, except that preconcentration with ferric hydroxide was used instead of wet ashing. The detailed method has been published separately (Taipale and Tuomainen 1985).

Results

Variations in the concentrations of ^{239,240}Pu, ^{241}Pu, ^{241}Am, ^{242}Cm and ^{137}Cs in fish flesh, liver and spawn samples are presented in Table 1. The detection limit for ^{239,240}Pu, ^{241}Am and ^{242}Cm was 0.6 mBq kg^{-1} for fish flesh and 3 mBq kg^{-1} for liver and spawn samples. For ^{241}Pu the limits were 20 and 200 mBq kg^{-1}, respectively. The highest concentrations of transuranic elements in fish flesh were found in lakes Päijänne and Oulujärvi, while liver concentrations were highest in Pääjärvi and Pyhäjärvi and those of spawn in lakes Lentua and Keitele (Fig. 1). In lake Saimaa, all concentrations were below the detection limits. No clear differences were found between fish species.

^{238}Pu could also be detected in 4 flesh and 3 liver samples; ^{238}Pu/^{239,240}Pu was between 1.0 and 7.2 in these samples, being highest in the liver. Uncertainties for these ratios varied 10%–25%.

The average ^{241}Am/^{239,240}Pu ratios in fish flesh and liver were 1.6 and 12, respectively. In 5 of the samples only ^{241}Am could be detected. ^{242}Cm was found in fish flesh in 1986 and in early 1987. The liver and spawn samples were analysed later, when ^{242}Cm was no longer detectable. ^{137}Cs in fish flesh from lake Päijänne varied widely. ^{137}Cs in the spawn from lake Päijänne varied from 230 to 1720 Bq kg^{-1}, showing an average of 790 Bq kg^{-1} in vendace and 850 Bq kg^{-1} in whitefish.

Concentrations of ^{239,240}Pu, ^{241}Pu and ^{241}Am in water of lake Päijänne were 13, 800 and 8 mBq m^{-3} during the summers of 1986 and 1987, respectively. The concentration of ^{242}Cm was measured once in late summer 1986, and when corrected for decay to May 1986 it was 18 mBq m^{-3}. In addition, variation in ^{137}Cs concentrations between Finnish lakes has been reported in detail elsewhere (Saxén and Rantavaara 1987, Saxén and Rantavaara 1990, Saxén *et al*. 2000).

Variation in and average values for the concentration factor (CF) from lake water to fish flesh, liver and spawn in lake Päijänne are presented in Table 2. Lake water concentrations were corrected to the sampling dates of fish before calculations. The calculated values for ^{239,240}Pu and ^{241}Pu were in good agreement with each other and are presented together, representing a CF value for Pu. The mean CF value for Pu was 84. The CF for Am was more than 2

Table 1. Variations in the activity concentrations of ^{239,240}Pu, ^{241}Pu, ^{241}Am, ^{242}Cm and ^{137}Cs in fish flesh, liver and spawn samples in Finnish lakes (m Bq kg^{-1} f.w., for lakes *see* Fig. 1). Concentrations were corrected to the sampling dates (28 August 1986–1 November 1988).

Nuclide	Fish flesh	Fish liver	Fish spawn
Pu-239,240	< 0.6–3.3	< 3–6.9	< 3
Pu-241	< 20–110	< 200–780	< 200–400
Am-241	< 0.6–2.6	8.7–140	5.4–29
Cm-242	< 0.6–340	–	–
Cs-137	(190–3300) × 10^3	–	(230–1720) × 10^3

times higher than that for Pu, when the mean was 230. A surprisingly high value of CF was calculated for ^{242}Cm, showing an average of 5100. The concentration of ^{242}Cm in lake water was first corrected to the fish sampling dates. The CFs for ^{137}Cs varied and were dependent on the fish species. The highest value was found in pike.

The TFs from deposition to fish flesh were calculated for Pu, Am and ^{137}Cs (Table 3). The deposition values used for calculations were for ^{137}Cs from Arvela *et al.* (1992) and for Pu and Am from Paatero (2000). The highest TF was for ^{137}Cs, showing an average of 44×10^{-3} m^2 kg^{-1}. This was somewhat higher than the mean TF for Am, but more than 10 times higher than that for Pu.

Discussion

No reference values exist for Pu concentrations in Finnish freshwater fishes before the Chernobyl accident, but a value of 0.6 mBq kg^{-1} was thought to indicate the presence of Chernobyl fallout. This estimation is based on concentrations found in Baltic Sea fishes from earlier studies (Kanisch *et al.* 1995, Rissanen and Ikäheimonen 2000). The presence of the fresh fallout nuclide ^{242}Cm and the high ratio of ^{241}Pu/^{239,240}Pu were clear signs of transuranics originating from Chernobyl. These signs of Chernobyl fallout were detected in 11 fish flesh, 6 liver and 1 spawn samples.

The expected value for ^{238}Pu/^{239,240}Pu in fish was approximately 0.5, while it varied 1.0–7.2 when detected. One reason could be the considerable variation in ratio of the nuclides in fresh fallout although the mean was about 0.5 (Saxén *et al.* 1987, Paatero 2000). A small part of the ^{238}Pu could be ingrown from short-lived ^{242}Cm, but this alone cannot explain the high ratios. The results could indicate differing environmental mobilization for ^{238}Pu compared with ^{239,240}Pu. Some other authors have also recognized greater biological uptake of ^{238}Pu over ^{239,240}Pu (Fresquez *et al.* 1999. Hokanson and Johnson 1973, Suchanek *et al.* 1996, Booher *et al.* 1998, NCRP 2001).

The ratio of ^{241}Am/^{239,240}Pu in fish flesh and liver (1.6 and 12, respectively) was considerably higher than that in lake water, 0.6. This indicates a higher accumulation of Am from water to fish. The higher bioavailability of Am over Pu in the environment has been detected in several studies (Hetherington *et al.* 1976, Wahlgren *et al.* 1976, Raabe 1980, Suchanek *et al.* 1996, Paatero 2000).

The large variation in ^{137}Cs contents in fish is

Table 2. Variations and averages of concentration factors (CF*) from lake water (lake Päijänne) to fish flesh, liver and spawn calculated to the sampling dates of fishes.

Nuclide	CF for fish flesh	(aver.)	CF for fish liver	CF for fish spawn	(aver.)
Pu-239,240 and Pu-241	< 45–250	(84)	< 200	< 200	(–)
Am- 241	< 85–370	(230)	–	1400	(–)
Cm-242	2100–9800	(5100)	–	–	(–)
Cs-137	250–5800	(2600)		110–820	(390)

$$^{*}\,CF = \frac{\text{Activity concentration of a radionuclide in fresh fish (Bq kg}^{-1})}{\text{Activity concentration in lake water (Bq kg}^{-1})}$$

Table 3. Variations and averages of transfer factors (TF*) from deposition to fish flesh (lake Päijänne).

Nuclide	TF m^2 kg^{-1}	Average
Pu-239,240 and Pu-241	(< 2.4–13) $\times 10^{-3}$	3.0×10^{-3}
Am-241	(< 6–26) $\times 10^{-3}$	18×10^{-3}
Cs-137	(6.9–160) $\times 10^{-3}$	44×10^{-3}

$$^{*}\,TF = \frac{\text{Activity concentration of a radionuclide in fresh fish (Bq kg}^{-1})}{\text{Average deposition (Bq m}^{-2})}$$

due to the uneven deposition of ^{137}Cs in the lake area. After deposition, contamination of fishes with different feeding habits occurs at different times. Nonpredatory fishes are contaminated most rapidly after deposition, while ^{137}Cs in predatory fishes reaches the highest levels 1–2 y after deposition in various lakes (Saxén *et al.*1997).

In the southern part of lake Päijänne, which was one of the lakes most contaminated by Chernobyl deposition, the concentration of ^{137}Cs in the water had declined to 300 Bq m^{-3} in June 1988 compared with a value in August 1986 of 1700 Bq m^{-3} (Saxén and Aaltonen 1987). ^{137}Cs levels in water of this lake decreased after the Chernobyl accident in 1986 with a half-life of 50 days. For about the next 2 y ^{137}Cs decreased with a half-life of one year thereafter the decrease was still slower (Saxén *et al*. 1997).

The ^{242}Cm concentration in the lake water of Päijänne was surprisingly low compared with the deposited amounts. One reason may be differences in the behaviour of Pu and Cm in the freshwater system during the short period following deposition.

The CF value for Pu, 84, was in good agreement with earlier studies. Eyman and Trabalka (1980) estimated values of between 5 and 250 for both marine and freshwater fish, with highest values for bottom feeders. The NCRP (2001) reports values of 5–300 for freshwater fishes.

The greater CF for Am over Pu is due to the greater bioavailability of Am compared with that for Pu. The chemical and biological behaviour of Am and Cm has generally been assumed to be similar. However, in the present study the CFs for Am and Cm differed greatly from each other. The correction for ^{242}Cm water concentration applied to the sampling dates of fish samples could have introduced some error for the calculated values if the ^{242}Cm concentration in lake water was not constant. However, there is still a marked difference between the calculated CFs for Am and Cm. A lack of previous environmental data is apparent for calculation of CFs for transplutonium elements, especially for Cm of freshwater fish. Eyman and Trabalka (1980) reported estimated CF values for Am, Cm and Np of between 50 and 2500, i.e. 10 times higher than that for Pu. In the present study the data were very limited and additional studies are needed to verify the possible differences in biological behaviour of Am and Cm indicated by these results.

Transfer of deposited ^{137}Cs to fishes in lakes is affected by many factors and varies with time after deposition. One of the most important factors is nutrient level of lake water, transfer being inversely proportional to the nutrient level of water (Kolehmainen *et al*. 1966).

In the event of future accidents, the limit proposed by the EU for Pu and transplutonium elements in foods other than milk or liquid foods is 80 Bq kg^{-1} (EURATOM 1989). Pu and Am depositions that could cause the limit to be exceeded in fish were estimated to be 27 000 Bq m^{-2} and 4500 Bq m^{-2}, respectively, the estimation is based on the TFs for Pu and Am. These values for Pu and Am are, respectively, 3000 times and 1500 times higher than the highest fallout of Pu and Am recorded in Finland following the Chernobyl accident (Paatero 2000).

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Received 30 August 2001, accepted 10 March 2002