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Delayed inputs of hot ^{137}Cs and ^{241}Am particles from Chernobyl to sediments from three Finnish lakes: implications for sediment dating

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Abstract Anomalous peaks in the ^{137}Cs and ^{241}Am records from three lake sediment cores from southern Finland appear to be due to the presence of micron-size hot radioactive particles, almost certainly originating in fallout from the 1986 Chernobyl accident. Since the imbedding sediments all post-date 1986 by several decades, it appears that they were initially deposited on the catchments of these lakes and transported to the lakes some years later. The activities of the particles were determined using a sequential splitting process. Two of the particles were found to contain ^{137}Cs with activities of 64 ± 4 mBq and 266 ± 15 mBq respectively. The third contained ^{241}Am with an activity of 17 ± 2 mBq, but no evidence of significant amounts of ^{137}Cs . The delayed input of such particles into the sedimentary records highlights the need for care in using ^{137}Cs or ^{241}Am

as chronostratigraphic markers in areas subject to significant levels of contamination from Chernobyl fallout.

Keywords Lake sediments · ^{137}Cs dating · Chernobyl fallout · Finland · Hot particles · Post-depositional transport

Introduction

Artificial radionuclides such as ^{137}Cs and ^{241}Am play an important role in dating lake sediments, and the wide range of environmental records they contain. Matches between well dated features in the ^{137}Cs (or ^{241}Am) fallout record and clearly identifiable features in the activity versus depth record are used to date specific sediment layers (Foucher et al. 2021). Although of limited value on their own, these chronostratigraphic dates play an important role in validating the more detailed chronologies obtained from ^{210}Pb records, or correcting ^{210}Pb dates in the event of any discrepancies (Appleby 2001). Since the early 1970s, concentration peaks identified as recording the period of maximum fallout from the atmospheric testing of thermonuclear weapons have been used to identify the 1963 sediment depth (Pennington 1981; Appleby et al. 1991). In many parts of north-west Europe, including Finland (Arvela et al. 1990), high levels of fallout from the Chernobyl reactor fire have resulted in the presence of a second peak that can be

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used to identify the 1986 depth (Erlinger et al. 2008). Fallout from this source took place during a relatively short period of time, from 26th April 1986 through to the middle of May (Persson et al. 1987). Radioactive debris deposited directly onto the surface of a lake will in most cases have a relatively short residence time in the water column. Although some will exit the lake via its outflow, a significant fraction will be delivered to the bed of the lake and incorporated into the sediment record for that year. The record in subsequent years will contain further inputs from fallout deposited on the catchment and delivered to the lake by various catchment/lake transport processes (Appleby et al. 2019). Inputs via this indirect pathway will generally be much lower than direct inputs at the time of the accident, and sediments with highest concentrations of Chernobyl radionuclides can for the most part confidently be dated to 1986. Although concentrations in post-1986 sediments frequently vary in response to factors such as different levels of catchment runoff, or post-depositional migration coupled with preferential absorption onto fine-grained clays, small features in this part of the record are for the most part easily distinguished from the 1986 peak itself. In a recent exception to this, the ^{137}Cs record in a core from a Norwegian fjord was found to contain a very distinct peak in a sediment sample deposited several decades after 1986 (Appleby et al. 2022). Detailed analyses showed that this feature was due to the presence of a single micron-size hot ^{137}Cs particle. Although its activity was just 15 mBq, the ^{137}Cs concentration in this near surface sample (15.6 Bq kg^{-1} , 61% of which was due to the particle) was twice as high as in the slice containing the concentration peak attributed to direct fallout from the 1986 event (7.4 Bq kg^{-1}). The most likely origin of the particle was fallout onto the landscape during the passage of the Chernobyl cloud and transport to the fjord some decades later, though in this case a second potential source was discharge into the marine environment from nuclear installations such as those at Sellafield or Dounreay in the UK.

The study by Arvela et al. (1990) showed that although there were significant amounts of Chernobyl fallout in many parts of Finland, highest levels were mainly recorded in the south of the country (Fig. 1). In a recent palaeolimnological study of lakes in southern Finland, three sites were found to have anomalous features in their Chernobyl fallout records. At two

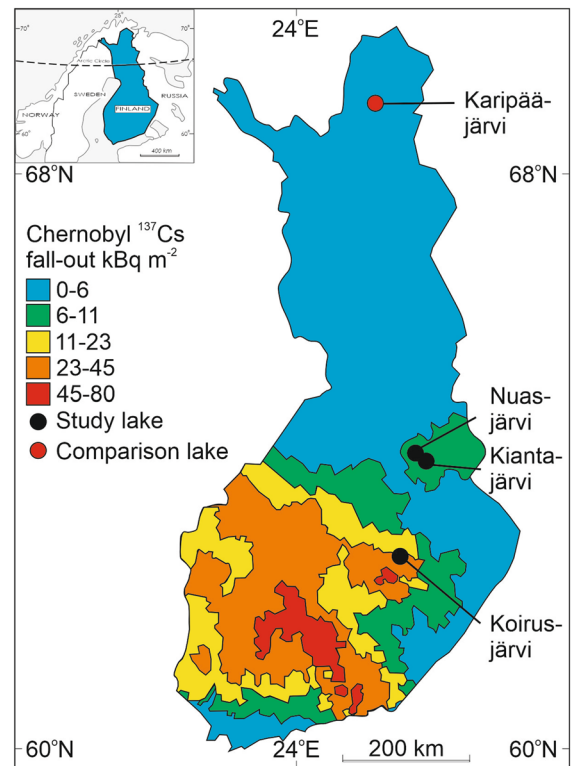


Fig. 1 Distribution of Chernobyl fallout in Finland, modified after Arvela et al. (1990). Also shown are the locations of the study sites, and a comparison lake, Karipääjärvi, from a region with a low level of Chernobyl fallout. Results from Karipääjärvi have been reported earlier in Ruppel et al. (2013, 2015)

sites, Koirusjärvi and Nuasjärvi, the ^{137}Cs record had two distinct peaks. At a third site, Kiantajärvi, a single sample had an unusually high ^{241}Am activity.

Much of the Chernobyl fallout over Finland was carried on particles in the sub-micron range (Kauppinen et al. 1986), though there were reports that it did include “hot particles” of up to $10 \mu\text{m}$ in size (Anttila et al. 1987; Raunemaa et al. 1987; Saari et al. 1989). The term “hot particle” is not well defined (Sandalls et al. 1993), but in this context is normally used to describe highly radioactive particles up to around $10 \mu\text{m}$ in diameter released into the environment from a nuclear accident or device. Although activity levels in particles released by the Chernobyl accident were initially dominated by short-lived radionuclides such as ^{141}Ce , ^{144}Ce , ^{95}Zr , ^{95}Nb , ^{103}Ru and ^{106}Ru , in the longer term they are mainly due to ^{137}Cs , and to a lesser extent, ^{241}Am . The objectives of

the present paper were to investigate the causes of the observed anomalies in the sediment records, the possibility that they might be due to delayed inputs of hot particles from the catchment, and any possible implications for the use of ^{137}Cs and ^{241}Am as chronological markers in dating lake sediments.

Site description

The study lakes (Nuasjärvi, Kiantjärvi and Koirusjärvi) are all located in the southern boreal to middle boreal vegetation zones in central Finland (Fig. 1). Mean annual precipitation (1991–2020) for the area ranges from 585 to 678 mm. Average temperatures (1991–2020) lie between -8.2 and -9.5 °C during January, and 16.2 – 17.1 °C during July (Jokinen et al. 2021). The lakes have ice cover from November to May. All have relatively large catchment areas, though the exceptionally large value for Koirusjärvi reflects the fact that it is hydrologically connected to a much larger lake, Kallavesi. For more detailed physiogeographical and limnological data, see Table 1.

Methods

Sample collection and preparation

Sediment cores were retrieved from the sedimentation basins of the lakes in October 2020 using a HTH-kayak gravity corer (Renberg and Hansson 2008). The cores were subsampled on site at 0.5-cm intervals between 0 and 5 cm depth, and at 1-cm intervals between 5 and 20 cm depth. All samples were stored

Table 1 Physiogeographical and limnological data of the study lakes

	Nuasjärvi	Kiantjärvi	Koirusjärvi
Latitude (N)	64° 9' 30.9"	64° 4' 53.5"	62° 34' 27.0"
Longitude (E)	28° 9' 11.5"	28° 26' 7.7"	27° 42' 53.5"
Mean precipitation (mm)	671	642	624
Water depth (m)	46	29	23
Average water depth (m)	8.5	5.9	9.7
Lake area (km ²)	96	23	6.1
Catchment area (km ²)	7475	3428	16,270

in plastic ziplock bags in a dark cold room at $+4$ °C within eight hours of retrieval. The samples were freeze-dried prior to radiometric analysis. Dry bulk densities were determined from the water content data.

Radiometric analysis

Subsamples from each core were analysed by direct gamma assay using Ortec HPGe GWL series well-type coaxial low background intrinsic germanium detectors (Appleby et al. 1986) in the Liverpool University Environmental Radioactivity Laboratory, preparatory to dating by ^{210}Pb and ^{137}Cs . ^{210}Pb was determined via its gamma emissions at 46.5 keV, and ^{226}Ra by the 295 keV and 352 keV γ -rays emitted by its daughter radionuclide ^{214}Pb following 25 days storage in sealed containers to allow radioactive equilibrium. ^{137}Cs and ^{241}Am were measured by their emissions at 662 keV and 59.5 keV respectively. Detection limits were ~ 8 mBq for ^{210}Pb , ~ 5 mBq for ^{226}Ra and ~ 2 mBq for ^{137}Cs and ^{241}Am . The absolute efficiencies of the detectors were determined using calibrated sources and sediment samples of known activity. Corrections were made for the effect of self-absorption of low energy γ -rays within the sample (Appleby et al. 1992). Supported ^{210}Pb in each sample was assumed equal to the ^{226}Ra activity, and unsupported (fallout) ^{210}Pb calculated by subtracting this from the total ^{210}Pb activity.

Results

Results of the radiometric analyses carried out on each core are shown in Figs. 2, 3, and 4. Table 2 lists some of the basic radiometric parameters for each site, including the maximum unsupported ^{210}Pb activity, unsupported ^{210}Pb inventory, mean ^{210}Pb supply rate, and ^{137}Cs inventory. The mean annual atmospheric ^{210}Pb flux is estimated to be significantly less than $100 \text{ Bq m}^{-2} \text{ y}^{-1}$ (Paatero et al. 2015). Significantly higher ^{210}Pb supply rates at all three core sites can be attributed to factors such as sediment focusing, and allochthonous inputs of fallout deposited on the catchment. All three sites have relatively large catchment areas compared to the size of the lake.

The unsupported ^{210}Pb activity versus depth records all deviated significantly from a simple

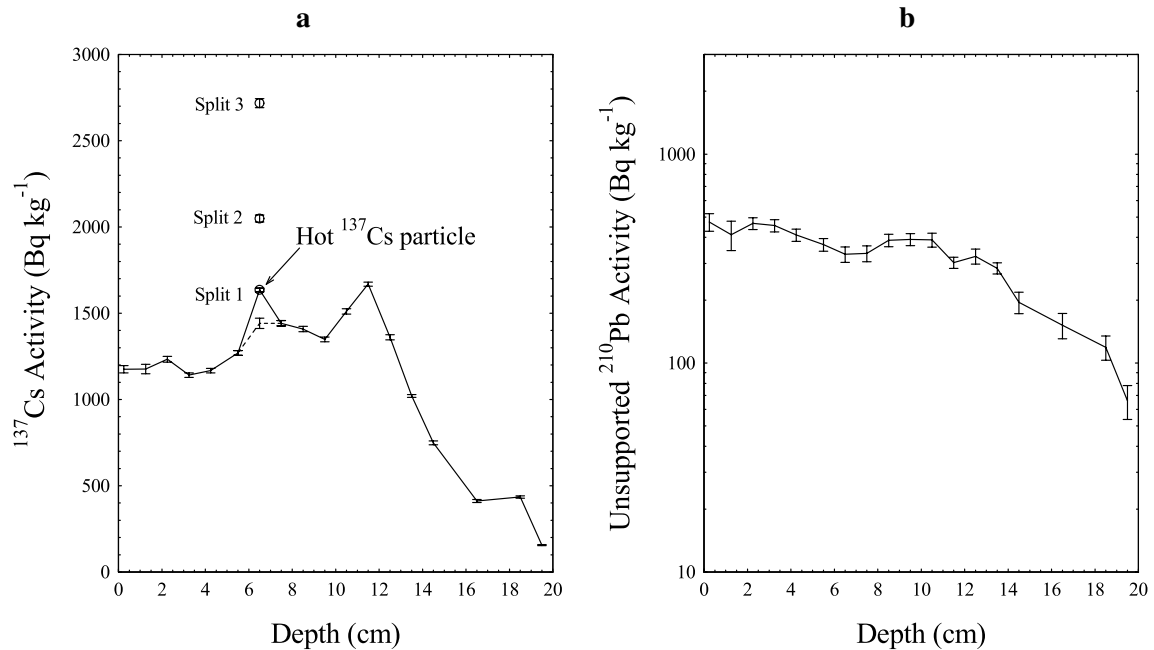


Fig. 2 Fallout radionuclides in the Koirusjärvi sediment core showing **a** ^{137}Cs and **b** unsupported ^{210}Pb concentrations versus depth in the core. ^{241}Am concentrations were below detection limits in this core. The ^{137}Cs record (**a**) has two distinct peaks, in the 6–7 cm and 11–12 cm sections. High concentrations show that both features almost certainly originate in fall-

out from the 1986 Chernobyl accident. The dashed line shows the estimated ^{137}Cs concentration in the anomalous sample attributable to the sediment matrix. The markers show the ^{137}Cs concentrations in the first three splits containing the hot particle

exponential decline, indicating systematic changes in the sedimentation rates. ^{210}Pb dates for Koirusjärvi and Nuasjärvi were calculated using the constant rate supply (CRS) model (Appleby and Oldfield 1978). This has generally proved to be the most reliable means for dating non-exponential records. Because of an apparent hiatus in the Kiantajärvi record, for this core the model was applied in a piecewise way using the methods outlined in Appleby (2001). A table of dates for each site, including their uncertainties, is given in Viksted et al. (2022, p. 68).

The ^{137}Cs inventories have been corrected for decay since 1986, the year of fallout from the Chernobyl nuclear accident. The high ^{137}Cs inventories, most notably at Koirusjärvi, confirm that all three sites were significantly impacted by fallout from the 1986 Chernobyl accident. This is further supported by the fact that the values of the inventories are consistent with the fallout data shown in Fig. 1. Data from Finnish sites less affected by Chernobyl fallout suggest that contributions from nuclear weapons test fallout are unlikely to be more than around 1000 Bq m^{-2} .

The ^{137}Cs record in the Koirusjärvi core (Fig. 2a), the most heavily impacted site, has two well-defined peaks, in the 6–7 cm and 11–12 cm sections. Although these features would normally be assumed to record two distinct events, fallout from the 1986 Chernobyl accident and the 1963 fallout maximum from the atmospheric testing of nuclear weapons, the high concentrations ($> 1500 \text{ Bq kg}^{-1}$) suggest that both peaks contain ^{137}Cs originating in fallout from the Chernobyl accident, with the deeper peak in all probability dating from that time. This is confirmed by the ^{210}Pb dates, which place 1986 within the 11–12 cm section. Sediments in the 6–7 cm section containing the more recent ^{137}Cs peak were dated to 2009 ± 2 years, more than two decades after the Chernobyl fallout event. The year of maximum fallout from the atmospheric testing of nuclear weapons, 1963, is placed within the 14–15 cm section. The absence of a ^{137}Cs peak at this depth can be attributed to downwards migration of the very high levels of Chernobyl ^{137}Cs (Klaminder et al. 2012).

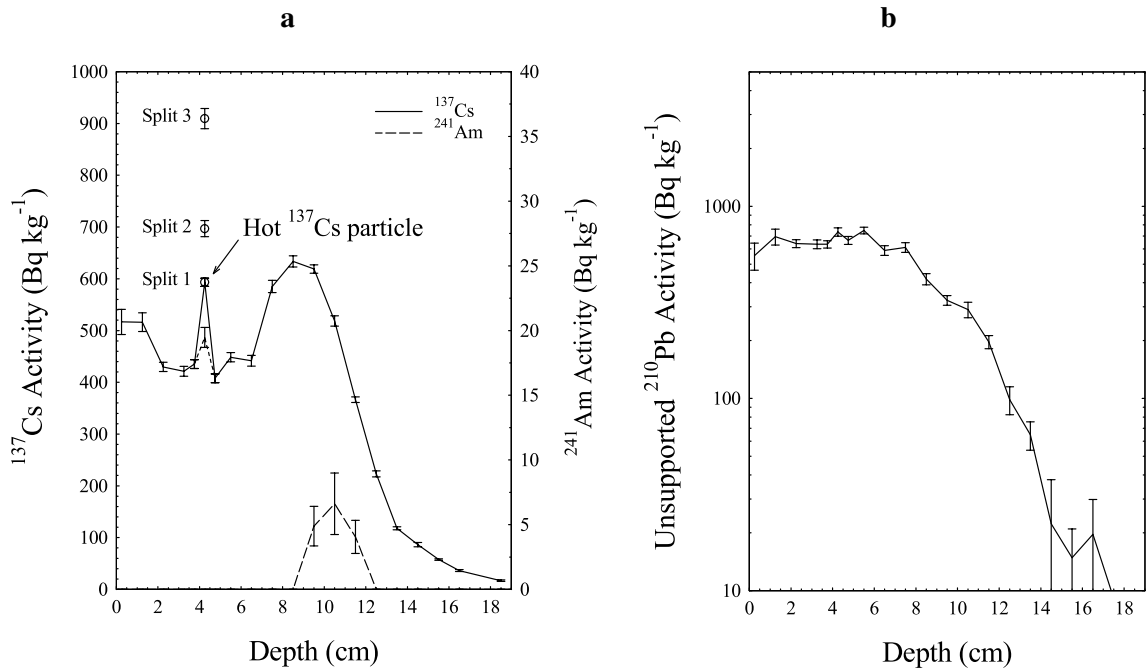


Fig. 3 Fallout radionuclides in the Nuasjärvi sediment core showing **a** ^{137}Cs and ^{241}Am and **b** unsupported ^{210}Pb concentrations versus depth in the core. The ^{137}Cs record (**a**) has two distinct peaks, in the 4–4.5 cm section, and more broadly between 7 and 10 cm. High concentrations again show that

both features almost certainly originate in fallout from the 1986 Chernobyl accident. The dashed line shows the estimated ^{137}Cs concentration in the anomalous sample attributable to the sediment matrix. The markers show the ^{137}Cs concentrations in the first three splits containing the hot particle

To determine whether the original 6–7 cm subsample was representative of the slice as a whole, a second subsample from the same slice was analysed and found to have a similar ^{210}Pb activity but an 11% lower ^{137}Cs activity. The possibility that the discrepancy was due to the presence of a hot particle was examined using a sequential splitting method (e.g. Falk et al. 1988; Pöllänen et al. 1999). The original higher activity subsample was divided into two equal parts that were then reanalysed. They again had similar ^{210}Pb activities, but this time there was a 40% discrepancy in the ^{137}Cs concentrations, $2049 \pm 20 \text{ Bq kg}^{-1}$, compared to $1477 \pm 20 \text{ Bq kg}^{-1}$. Repeated splitting of the high activity subsamples followed a similar pattern. By the fifth split the ^{137}Cs concentration in the high activity half had increased to $5994 \pm 109 \text{ Bq kg}^{-1}$, compared to $1334 \pm 64 \text{ Bq kg}^{-1}$ in the low activity half. Since the low activity halves all had similar ^{137}Cs concentrations, the most likely explanation of these results is that the ^{137}Cs peak in the 6–7 cm sample was mainly due to the presence of a single hot ^{137}Cs particle. Excluding this particle,

^{137}Cs activity appeared to be uniformly distributed throughout the sediment matrix. The mean concentration in the matrix based on a weighted average of measured activities in the low activity splits was calculated to be $1441 \pm 81 \text{ Bq kg}^{-1}$. This is very similar to the mean of the ^{137}Cs activities in the adjacent 5–6 cm and 7–8 cm samples, $1270 \pm 14 \text{ Bq kg}^{-1}$ and $1442 \pm 16 \text{ Bq kg}^{-1}$, respectively.

For each high activity split the total ^{137}Cs activity can be divided into two components, one due to the sediment matrix and the other due to the presumed hot particle. The activity due to the matrix is calculated by multiplying the mass of the split by the ^{137}Cs concentration in the matrix. Excluding the value from the first split where the uncertainty was necessarily very large, the results, shown in Table 3, yield a consistent value for the activity of the hot particle of between 256 and 281 mBq with a weighted mean of $266 \pm 15 \text{ mBq}$. Higher weights were given to smaller splits where the uncertainties were necessarily smaller. A similar analysis of concentrations in the 11–12 cm section showed that in this case the

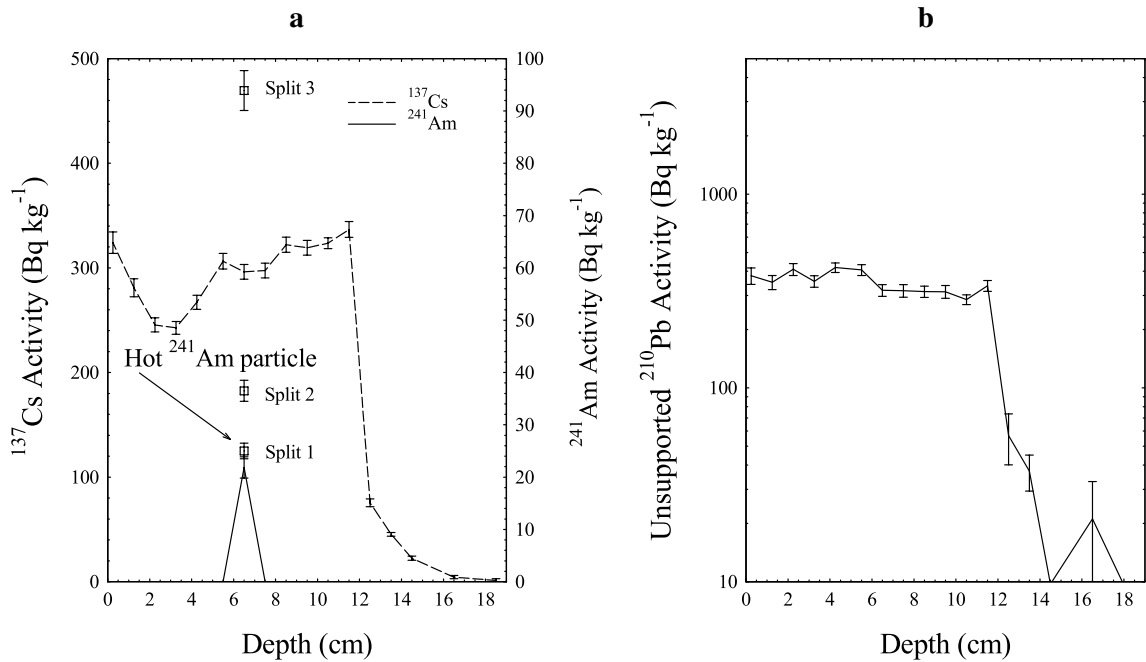


Fig. 4 Fallout radionuclides in the Kiantajärvi sediment core showing **a** ^{137}Cs and ^{241}Am and **b** unsupported ^{210}Pb concentrations versus depth. The ^{137}Cs and ^{210}Pb records both suggest a hiatus in the sediment record at 12 cm. An unusually high

^{241}Am concentration was recorded in the 6–7 cm sample. The markers show the ^{241}Am concentrations in the first three splits containing the hot particle

Table 2 Radiometric parameters for the three Finnish lake sediment cores

Core	Unsupported ^{210}Pb						^{137}Cs	
	Maximum activity		Inventory		Mean supply rate		Inventory	
	Bq kg^{-1}	\pm	Bq m^{-2}	\pm	$\text{Bq m}^{-2}\text{y}^{-1}$	\pm	Bq m^{-2}	\pm
Nuasjärvi	554	90	8255	186	257	6	19,979	291
Kiantajärvi	379	37	7598	220	237	7	14,611	236
Koirusjärvi	474	46	5496	133	171	6	41,934	573

Table 3 ^{210}Pb and ^{137}Cs concentrations in the matrix and successive high activity splits of the 6–7 cm slice from the Koirusjärvi core, and total ^{137}Cs activities in these splits attributable to the matrix and the presumed hot particle

Sub sample	Mass g	Specific activities				Total ^{137}Cs activity			
		^{210}Pb		^{137}Cs		Matrix		Hot particle	
		Bq kg^{-1}	\pm	Bq kg^{-1}	\pm	mBq	\pm	mBq	\pm
Matrix				1441	81				
Split 1	0.814	493	17	1635	11	1173	66	158	67
Split 2	0.463	470	24	2049	20	667	38	281	39
Split 3	0.223	498	28	2718	26	321	18	285	19
Split 4	0.109	597	68	3788	62	157	9	256	11
Split 5	0.058	626	145	5994	109	84	5	264	8
						Mean value		266	15

¹³⁷Cs activity was uniformly distributed throughout the sample and that the high concentration was due to elevated levels of fallout. Successive splits of this sample had relatively similar ²¹⁰Pb and ¹³⁷Cs concentrations.

The ¹³⁷Cs record in the Nuasjärvi core (Fig. 3a) also has two distinct peaks, a broad feature between 7 and 10 cm, and more narrowly defined feature within the 4–4.5 cm sample. There is also a small peak in ²⁴¹Am concentrations slightly deeper in the core, between 9 and 12 cm. High concentrations again suggest that both ¹³⁷Cs peaks originate in fallout from the 1986 Chernobyl accident, with the deeper and more substantial feature being a direct record of that event. This is confirmed by the ²¹⁰Pb results, which place 1986 at a depth of around 8 cm. Since the ²¹⁰Pb dates also place 1963 at a depth of around 10 cm, the ²⁴¹Am peak most probably records the early 1960s peak in fallout of its parent radionuclide ²⁴¹Pu from the atmospheric testing of nuclear weapons (Appleby et al. 1991). The relatively small gap between the 1963 and 1986 dates is attributed to a much lower sedimentation rate at that time. The CRS model calculations suggest a value of around 0.1 cm y⁻¹, compared to 0.24 cm y⁻¹ for the post-1986 period. The absence of a 1963 ¹³⁷Cs peak can again be attributed to downwards migration of Chernobyl ¹³⁷Cs, as can the presence of significant amounts of ¹³⁷Cs at depths predating the 1953 onset of global fallout from the atmospheric testing of nuclear weapons.

Sediments in the 4–4.5 cm section containing the more recent ¹³⁷Cs peak were dated to 2009 ± 2 years. An analysis carried out on a second subsample from this section suggested that the anomaly was again due to the presence of a single hot particle, and this was

confirmed by carrying out a sequential splitting analysis as described above. During this process ¹³⁷Cs concentrations in the hot splits increased from an initial value of 593 ± 8 Bq kg⁻¹ to 1952 ± 58 Bq kg⁻¹ in the 5th split (Table 4). The results again suggested that, excluding the hot particle, ¹³⁷Cs was relatively uniformly distributed throughout the section with a mean concentration of 487 ± 19 Bq kg⁻¹. Using this value, calculations of the ¹³⁷Cs activity in each hot split due to the presumed hot particle yielded a consistent value of between 57 and 64 mBq, with a weighted mean of 64 ± 4 mBq. A similar analysis of sediments in the 8–9 cm section showed that in this case the ¹³⁷Cs activity was uniformly distributed throughout the sample and that the high concentrations were due to elevated levels of fallout. Successive splits had relatively similar ²¹⁰Pb and ¹³⁷Cs concentrations.

Results from the Kiantajärvi core suggest that there is a hiatus in the sediment record at a depth of around 12 cm. At this depth there is an abrupt jump in unsupported ²¹⁰Pb concentrations from 57 ± 17 Bq kg⁻¹ in the 12–13 cm sample to 337 ± 22 Bq kg⁻¹ in the 11–12 cm sample, and a similar jump in ¹³⁷Cs concentrations from 76 ± 4 Bq kg⁻¹ below to 337 ± 8 Bq kg⁻¹ above. Although there is no clear feature identifying the 1986 Chernobyl event, high ¹³⁷Cs concentrations in all samples above 12 cm suggest that they all post-date 1986. Near constant values in the unsupported ²¹⁰Pb concentrations suggest that sediments above the hiatus span no more than a few years. One of the samples within this part of the core, at 6–7 cm, had an unusually high ²⁴¹Am activity of 25 ± 1 Bq kg⁻¹. At all other depths ²⁴¹Am concentrations were below detection limits. Since fallout from the Chernobyl accident was known to include hot

Table 4 ²¹⁰Pb and ¹³⁷Cs concentrations in the matrix and successive high activity splits of the 4–4.5 cm slice from the Nuasjärvi core, and total ¹³⁷Cs activities in these splits attributable to the matrix and the presumed hot particle

Sub sample	Mass g	Specific activities				Total ¹³⁷ Cs activity			
		²¹⁰ Pb		¹³⁷ Cs		Matrix		Hot particle	
		Bq kg ⁻¹	±	Bq kg ⁻¹	±	mBq	±	mBq	±
Matrix				487	19				
Split 1	0.536	763	20	593	8	261	10	57	11
Split 2	0.281	698	31	697	15	137	5	59	7
Split 3	0.151	739	43	909	20	73	3	64	4
Split 4	0.078	762	91	1313	46	38	2	64	4
Split 5	0.044	632	141	1952	58	21	1	64	3
						Mean value		64	4

^{241}Am particles (Falk et al. 1988), a sequential splitting analysis was carried out to determine whether that could account for the high activity in this sample. The results, shown in Table 5, were consistent with the presence of a single hot particle. At each stage of the analysis one of the splits was shown to contain all of the ^{241}Am activity, and the other negligible ^{241}Am activity. Calculated values of the activity in the hot split had a consistent value of between 15 and 21 mBq. The weighted mean was 17 ± 2 mBq. There was no evidence of significant ^{137}Cs activity on the hot particle, possibly indicating that it was a Group 1 particle as defined by Falk et al. (1988). All of the splits had essentially the same ^{137}Cs concentration. Although there are large uncertainties over the dating of this core, it is almost certain that sediments containing the hot particle were deposited at some time during the past decade.

Discussion and conclusions

Although their precise origin is to some extent immaterial, the hot particles reported here almost certainly originated in fallout from the Chernobyl cloud during its passage over Finland in late April and early May 1986. All three cores are from areas known to have been heavily impacted by fallout from Chernobyl. Further, measurements carried out in the immediate aftermath of the accident showed that hot particles from the cloud were deposited in Sweden (Persson et al. 1987) and Finland (Raunemaa et al. 1987). Releases from the Ignalina nuclear power plant in Lithuania (Marčiulionienė et al. 2015) appear to have been relatively localised. Atmospheric releases

from other nuclear installations in the Former Soviet Union (Suokko and Reicher 1993) were almost certainly either too localised or too remote. Although the Kyshtym accident in 1957 did release a large amount radioactive material into the atmosphere, the prevailing winds carried it mainly in a north–north east direction from that site (Norwegian Radiation Protection Agency 2007).

A study of hot particles collected one year after the Chernobyl accident (Falk et al. 1988) showed that they included fragments from the damaged reactor as well as highly radioactive inclusions formed within the fuel elements. Although initially the radioactivity would have been mainly associated with short-lived fission products, only longer-lived products such as ^{137}Cs and some transuranic elements including ^{241}Am are presently above detection limits. One such particle from Sweden (Falk et al. 1988) had a ^{137}Cs activity of 400 mBq (179 mBq decay corrected to 2021) but negligible ^{241}Am . Another from the Gotland Deep in the Baltic Sea (Pöllänen et al. 1999) contained both ^{137}Cs (59 mBq decay corrected to 2021) and ^{241}Am (100 mBq). Particles deposited in Finland appear to have been of a similar size, up to around 10- μm in diameter (Raunemaa et al. 1987). Assuming a similar composition to those collected in Sweden, the Nuasjärvi and Koirusjärvi ^{137}Cs particles are estimated to have diameters of between 6–11 μm and 10–18 μm respectively, and the Kiantajärvi ^{241}Am particle a diameter of around 6 μm .

Hot particles from the study sites account for only a small fraction of the ^{137}Cs inventory in the sediments. The sediment matrix accounted for 97% of the total ^{137}Cs activity in the core slices from Nuasjärvi checked for homogeneity. In the Koirusjärvi

Table 5 ^{210}Pb and ^{137}Cs concentrations in successive splits of the 6–7 cm slice from the Kiantajärvi core, and ^{241}Am concentrations in successive hot subsamples. Total ^{241}Am activities in the hot splits are presumed to be that of the hot particle

Sub sample	Mass g	Specific activities						Hot ^{241}Am particle	
		^{210}Pb		^{137}Cs		^{241}Am		mBq	±
		Bq kg ⁻¹	±	Bq kg ⁻¹	±	Bq kg ⁻¹	±		
Matrix									
Split 1	0.839	454	14	282	4	25	1	21	2
Split 2	0.404	466	16	333	5	36	2	15	1
Split 3	0.189	452	22	341	6	94	4	18	1
Split 4	0.100	403	52	343	13	183	9	18	1
Split 5	0.049	330	70	345	17	344	14	17	1
						Mean value		17	2

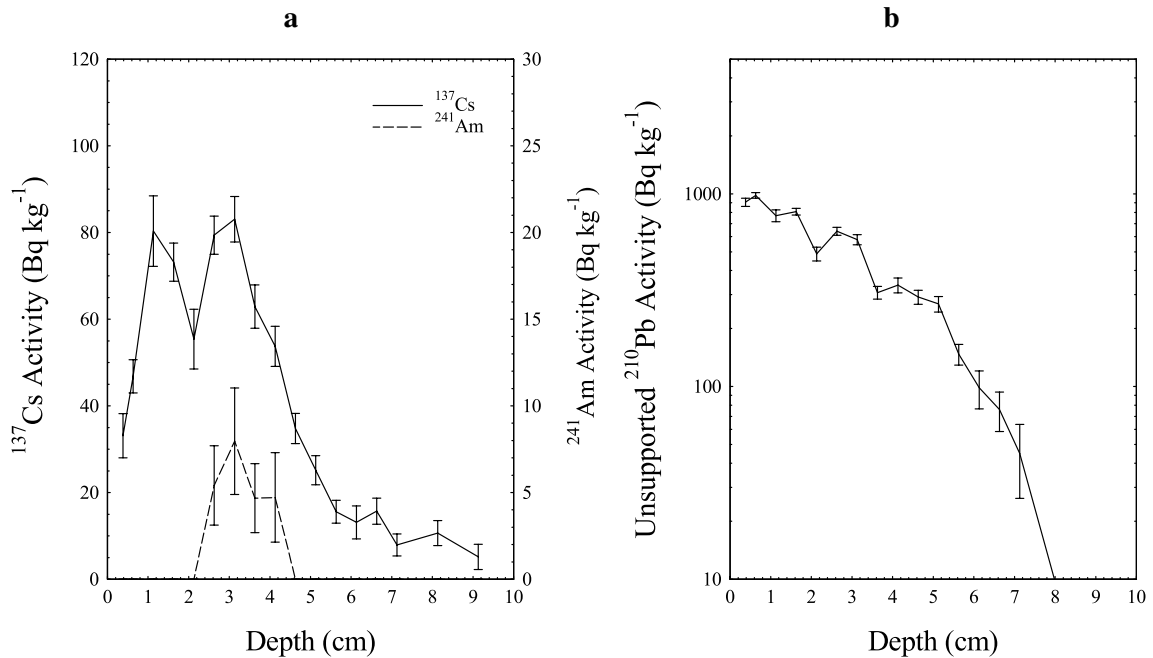


Fig. 5 Fallout radionuclides in the Karipääjärvi sediment core showing **a** ¹³⁷Cs and ²⁴¹Am and **b** unsupported ²¹⁰Pb concentrations versus depth. Substantially lower levels of Chernobyl fallout are reflected in the greatly reduced ¹³⁷Cs concentrations compared to the study sites. Although the ¹³⁷Cs record is superficially similar to that at Koirusjärvi (Fig. 2a), association of the deeper peak at 3–3.25 cm with a similar but smaller

²⁴¹Am peak suggests that this feature most probably records the 1963 fallout maximum from the atmospheric testing of nuclear weapons. The ²¹⁰Pb calculations, which take account of dilution due e.g. to episodes of more rapid sedimentation, suggest that the later ¹³⁷Cs peak almost certainly records fallout from the 1986 Chernobyl accident

core the corresponding figure was 96%. There was no evidence of the presence of hot particles in the remaining slices, in all probability they are likely to account for less than 1% of the total ¹³⁷Cs inventory. Since particles deposited directly onto the surface of the lake would have been either incorporated in the sediment record for 1986 or lost from the lake via the outflow, those detected in the present study were almost certainly deposited on the catchment and only transported to the lake some years later. Studies of catchment/lake transport (Appleby et al. 2019) have shown that significant quantities of ¹³⁷Cs deposited on the catchment may continue to be transported to the lake decades after the last fallout event. Regardless of contribution or pathway, these findings do, however, show that at sites significantly impacted by fallout from the Chernobyl accident, delayed inputs of hot particles originating in fallout from the 1986 Chernobyl accident can have a disproportionate impact on ¹³⁷Cs and ²⁴¹Am records and hence on their reliability as chronostratigraphic markers. This

has relevance not just for the ¹³⁷Cs/²⁴¹Am dates themselves but perhaps most importantly for their use in validating ²¹⁰Pb chronologies. It is widely recognized that neither of the standard ²¹⁰Pb models can be relied on absolutely and independent validation is essential, even where the ²¹⁰Pb dates appear to be unequivocal. The importance of reliable chronostratigraphic dates for validating ²¹⁰Pb dates is highlighted by data from Karipääjärvi, a small lake around 500 km further north in Finnish Lapland (Fig. 1). Figure 5 shows radiometric records from a core collected from this site in 2010. The results have been previously reported in Ruppel et al. (2013, 2015). The ¹³⁷Cs record has two distinct peaks, within the 1–1.25 cm and 3–3.25 cm sections, respectively. The presence of a small ²⁴¹Am peak within the 3–3.25 cm section showed that the earlier peak in this case recorded the 1963 weapons test fallout maximum. Confidence in the reliability of the ¹³⁷Cs date allowed it to be used as a reference point (Appleby 2001) to correct a small but significant discrepancy with the raw ²¹⁰Pb dates.

The superficial similarity of the Karipääjärvi records to those from Koirusjärvi shows that such confidence is not always justified and that ^{137}Cs dates can be misleading. Where there is any doubt, samples containing potentially anomalous features should be investigated more thoroughly for possible causes other than direct fallout.

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Declarations

Competing interests The authors declare no competing interests.

Conflicts of interest The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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References

- Anttila P, Kulmala M, Raunemaa R (1987) Dry and wet deposition of Chernobyl aerosols in southern Finland. *J Aerosol Sci* 18:939–942
- Appleby PG (2001) Chronostratigraphic techniques in recent sediments. In: Last WM, Smol JP (eds) *Tracking environmental change using lake sediments: basin analysis, coring, and chronological techniques*, vol 1. Kluwer Academic, Dordrecht, pp 171–203
- Appleby PG, Oldfield F (1978) The calculation of lead-210 dates assuming a constant rate of supply of unsupported ^{210}Pb to the sediment. *Catena* 5:1–8
- Appleby PG, Piliposyan G, Hess S (2022) Detection of a hot ^{137}Cs particle in marine sediments from Norway. *Geo-Mar Lett* 42(2):1–6. <https://doi.org/10.1007/s00367-021-00727-2>
- Appleby PG, Richardson N, Nolan PJ (1991) ^{241}Am dating of lake sediments. *Hydrobiologia* 214:35–42
- Appleby PG, Richardson N, Nolan PJ (1992) Self-absorption corrections for well-type germanium detectors. *Nucl Instrum Methods B* 71:228–233. [https://doi.org/10.1016/0168-583X\(92\)95328-O](https://doi.org/10.1016/0168-583X(92)95328-O)
- Appleby PG, Semertzidou P, Piliposian GT, Chiverrell RC, Schillereff DN, Warburton J (2019) The transport and mass balance of fallout radionuclides in Brotherswater, Cumbria (UK). *J Paleolimnol* 62:389–407. <https://doi.org/10.1007/s10933-019-00095-z>
- Appleby PG, Nolan PJ, Gifford DW, Godfrey MJ, Oldfield F, Anderson NJ, Battarbee RW (1986) ^{210}Pb dating by low background gamma counting. *Hydrobiol* 143:21–27
- Arvela H, Markkanen M, Lemmelä H (1990) Mobile survey of environmental gamma radiation and fallout levels in Finland after the Chernobyl accident. *Radiat Protect Dosi* 37:177–184
- Erlinger C, Lettner H, Hubmer A, Hofmann W, Steinhäusler F (2008) Determining the Chernobyl impact on sediments of a pre-Alpine lake with a very comprehensive set of data. *J Environ Radioact* 99:1294–1301
- Falk R, Suomela J, Kerekes A (1988) A study of “hot particles” collected in Sweden one year after the Chernobyl accident. *J Aerosol Sci* 19:1339–1342
- Foucher A, Chaboche P-A, Sabatier P, Evrard O (2021) A worldwide meta-analysis (1977–2020) of sediment core dating using fallout radionuclides including ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$. *Earth Syst Sci Data*. <https://doi.org/10.5194/essd-2021-168>
- Jokinen P, Pirinen P, Kaukoranta J-P, Kangas A, Alenius P, Eriksson P, Johansson M, Wilkman S (2021) Climatological and oceanographic statistics of Finland 1991–2020. Reports 2021:8. Finnish Meteorological Institute, 169 pp
- Kauppinen EI, Hillamo RE, Aaltonen SH, Sinkko KTS (1986) Radioactivity size distributions of ambient aerosols in Helsinki, Finland, during May 1986 after the Chernobyl accident: preliminary report. *Environ Sci Technol* 20:1257–1259
- Klaminder J, Appleby P, Crook P, Renberg I (2012) Post-deposition diffusion of ^{137}Cs in lake sediment: implications for radiocesium dating. *Sedimentology* 59:2259–2267. <https://doi.org/10.1111/j.1365-3091.2012.01343.x>
- Marčiulionienė D, Mažeika J, Lukšienė B, Jefanova O, Mikalauskienė R, Paškauskas R (2015) Anthropogenic radionuclide fluxes and distribution in bottom sediments of the cooling basin of the Ignalina Nuclear Power Plant. *J Environ Radioact* 145:48–57
- Norwegian Radiation Protection Authority (2007) The Kyshtym accident, 29th September 1957. NRP Bulletin p 4. ISSN 0806–895x

- Paatero J, Vaaramaa K, Buyukay M, Hatakka J, Lehto J (2015) Deposition of atmospheric ^{210}Pb and total beta activity in Finland. *J Radioanal Nucl Chem* 303:2413–2420. <https://doi.org/10.1007/s10967-014-3785-7>
- Pennington W (1981) Records of a lake's life in time: the sediments. *Hydrobiologia* 79:197–215
- Persson C, Rodhe H, De Geer L-E (1987) The Chernobyl accident—a meteorological analysis of how radionuclides reached and were deposited in Sweden. *Ambio* 16:20–31
- Pöllänen R, Ikäheimonen TK, Klemola S, Juhanoja J (1999) Identification and analysis of a radioactive particle in a marine sediment sample. *J Environ Radioact* 45:149–160
- Raunemaa T, Lehtinen S, Saari H, Kulmala M (1987) 2–10 μm sized hot particles in Chernobyl Fallout to Finland. *J Aerosol Sci* 18:693–696
- Renberg I, Hansson H (2008) The HTH sediment corer. *J Paleolimnol* 40:655–659
- Ruppel M, Gustafsson Ö, Rose N, Pesonen A, Yang H, Weckström J, Palonen V, Oinonen MJ, Korhola A (2015) Spatial and temporal patterns in black carbon (BC) deposition to dated Fennoscandian Arctic lake sediments from 1830 to 2010. *Environ Sci Technol* 49:13954–13963. <https://doi.org/10.1021/acs.est.5b01779>
- Ruppel M, Lund MT, Grythe H, Rose NL, Weckström J, Korhola A (2013) Comparison of spheroidal carbonaceous particles data with modelled atmospheric black carbon concentration and deposition and air mass sources in northern Europe, 1850–2010. *Adv Meteorol.* <https://doi.org/10.1155/2013/393926>
- Saari H, Luokkanen S, Kulmala M, Lehtinen S, Raunemaa T (1989) Isolation and characterization of Hot Particles from Chernobyl Fallout in Southwestern Finland. *Health Phys* 57(6):975–984
- Sandalls FJ, Segal MG, Victorova N (1993) Hot particles from Chernobyl: a review. *J Environ Radioact* 18:5–22
- Suokko KL, Reicher D (1993) Radioactive waste and contamination in the former Soviet Union. *Environ Sci Technol* 27:602–604
- Viksted H, Kivipelto J, Koivuhuhta A, Virtanen K, Tolonen KT, Weckström J, Luoto TP, Mykrä H, Riihimäki J, Hellsten S (2022) Project comparing mandatory monitoring and deep-bottom animal methods (Vepove). Report 28/2022, 214 pp (in Finnish). <https://urn.fi/URN:ISBN:978-952-398-024-2>

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