

Sediment profile dating and reconstructing nuclear events from annually laminated lake sediments in northern Finland

Eeva Haltia^a, Ari-Pekka Leppänen^{b,*}, Antti Kallio^c, Timo Saarinen^a

^a Department of Geography and Geology, University of Turku, 20014, Turku, Finland

^b Monitoring and Situation Awareness, Radiation and Nuclear Safety Authority - STUK, Lähteentie 2, 96400 Rovaniemi, Finland

^c Natural Radiation Regulation and Health, Radiation and Nuclear Safety Authority - STUK, Lähteentie 2, 96400 Rovaniemi, Finland

ARTICLE INFO

Keywords:

lake sediment
Deposition
¹³⁷Cs
²⁴¹Am
²¹⁰Pb

ABSTRACT

The sediments deposited in Lake Kevojärvi (69°45'N, 27°00'E) in northernmost Finland were investigated for radioactivity. Freeze cores recovered from the 35-m deep basin has preserved a distinct succession of annual laminations deposited between 1909 and 2015. The basic varve structure is composed of a detrital snowmelt layer and an organic-rich post-snowmelt layer lying on top of the snowmelt layer. The past nuclear events have been preserved in the lake sediment. In order to study these, sediment freeze with annual sediment laminations were recovered from the lake bottom and measured for gamma emitting radionuclides. A total of 53 sediment subsamples, each incorporating sediment from one to two varves i.e. sediment deposited during one to two years, were taken for gamma spectroscopy measurements using low-background HPGe detector system. The measurements showed large variations in ¹³⁷Cs and ²⁴¹Am concentrations between different laminae marking different nuclear events in history. This highly resolved profile revealed a detailed record of anthropogenic radioactive fallout from atmospheric nuclear weapons testing during 1950s and 1960s and the Chernobyl accident in 1986. The measured ¹³⁷Cs concentrations varied between 0.6 and 229 Bq/kg dry weight (dw) while the measured ²⁴¹Am concentrations varied between 0.6 and 6.4 Bq/kg dw. The ¹³⁷Cs showed downward migration in the sediment column since ¹³⁷Cs was also found in varves dating before 1945. The first clear increase in ¹³⁷Cs concentration was observed in 1956 varve and the ¹³⁷Cs concentrations were found to peak in the 1964, 1970 and 1986 varves. The ²⁴¹Am was first observed in 1957 varve and the concentrations peaked in 1960–1962, 1964 and 1970 varves. This demonstrated that ²⁴¹Am can be a complementary chronostratigraphic marker to ¹³⁷Cs in sediment. A time delay of one to two years was observed between the years of intensive atmospheric nuclear weapons testing and the increased ¹³⁷Cs and ²⁴¹Am concentrations in the sediment varves. In the ²¹⁰Pb concentrations long-time periods of low (~1930–1950 and ~1990–2000) and high concentrations (~1950–1980) were observed. The reason for large variation was the amount of sediment input. The Constant rate of supply (CRS) model results showed good agreement with the varve counting. The model results showed that the ²¹⁰Pb deposition has been constant over the last 80 years and no effect of climate change to the ²¹⁰Pb deposition was observed.

1. Introduction

Long and continuous varve records are preserved in some lakes providing possibilities for high-resolution climatic and environmental reconstructions. Varved sequences have been found in lacustrine, marine and estuarine depositional environments (Jokinen et al., 2015; Wohlfarth et al., 1998). Because of their potentially highly resolved independent sediment chronologies, varved sediment records have been shown to provide reliable records of spring discharges and variability

therein (Sander et al., 2002; Kämpf et al., 2014). The varve type most often described in Fennoscandian lakes is characterized by a basal detrital layer which is deposited as the result of the annual spring flooding following snowmelt. The thickness of the snowmelt layer in these varve records is commonly inferred in terms of winter climate, where coldness and length of winter are assumed to influence the net accumulation of snow in the catchment (Snowball et al., 2002; Tiljander et al., 2003; Ojala et al., 2008; Saarni et al., 2016). However, interpretation of a varve record and information preserved in its snowmelt layers

* Corresponding author.

E-mail address: ari.leppanen@stuk.fi (A.-P. Leppänen).

<https://doi.org/10.1016/j.jenvrad.2021.106611>

Received 27 January 2021; Received in revised form 26 March 2021; Accepted 28 March 2021

Available online 12 April 2021

0265-931X/© 2021 The Author(s). Published by Elsevier Ltd. This is an open access article under the CC BY license (<http://creativecommons.org/licenses/by/4.0/>).

may remain tentative in case gauged meteorological and hydrological observations are scarce near the study sites.

We investigated a 107-year long varve record from an estuarine depositional site in Lake Kevojärvi located in Northern Finland. This study was part of a larger project studying regional changes in environment using varved sediment layers as the record of the past. The aim was to detect the nuclear events that occurred during the last 60 years from the annually laminated sediment varves. This would validate the varve chronology with a highly resolved profile where this information can be used in further e.g. in climatic studies. Here we show a detailed record of nuclear events conducted during 1950s and 1960s and the Chernobyl accident that occurred in 1986.

1.1. Site description

The Utsjoki River (60 km in length) flows in a valley bordered by steep slopes in northernmost Finland. It can be described as a chain of interconnected elongated lakes, most of them roughly in north-south direction. Lake Kevojärvi ($69^{\circ}45'34''\text{N}$, $27^{\circ}00'23''\text{E}$) (Fig. 1) is one of these lakes, lying between Lake Puksalanjärvi in the South and Lake Jomppalanjärvi in the North. Lake Kevojärvi is a clear-watered and oligotrophic fluvial lake. The Utsjoki River is unregulated and one of the

tributaries of the Tenojoki River that discharges into the Barents Sea through Tana fjord in Finnmark in Norway. Two tributaries of the Utsjoki River, i.e., the Kevojoki River (33 km long) and the Tsarsjoki River (26 km long), drain directly into Lake Kevojärvi from the west (Fig. 1). Besides these, a few smaller streams drain into the lake. Lake Kevojärvi has a small surface area (1.02 km^2) in comparison with its catchment area (1520 km^2). Detrital sedimentation in the western part of Lake Kevojärvi is assumed to be mainly influenced by the Kevojoki and Tsarsjoki rivers, whose combined catchment area is ca. 730 km^2 . The lake's outlet in the North is rather narrow, and the largest annual changes in water level (447 cm, data from Finnish Environment Institute (SYKE) in Finland) have been measured in Lake Kevojärvi.

The drainage basin is characterized by variable topography where hilly landscapes and fell areas are dissected by river valleys. Vegetation in higher altitude areas are dominated by brush vegetation with mountain birch (*Betula pubescens* subsp. *pumila*) and dwarf birch (*B. nana*). In the sheltered river valleys the forest line of Scots pine (*Pinus sylvestris*) reaches 200 m above sea level.

Because of the proximity to the North Atlantic and the Arctic Ocean and their warming oceanic influence, the climate in the study area is mild considering its position in the high Northern latitudes. Climate in the Lake Kevojärvi area is characterized by a fully humid snow climate

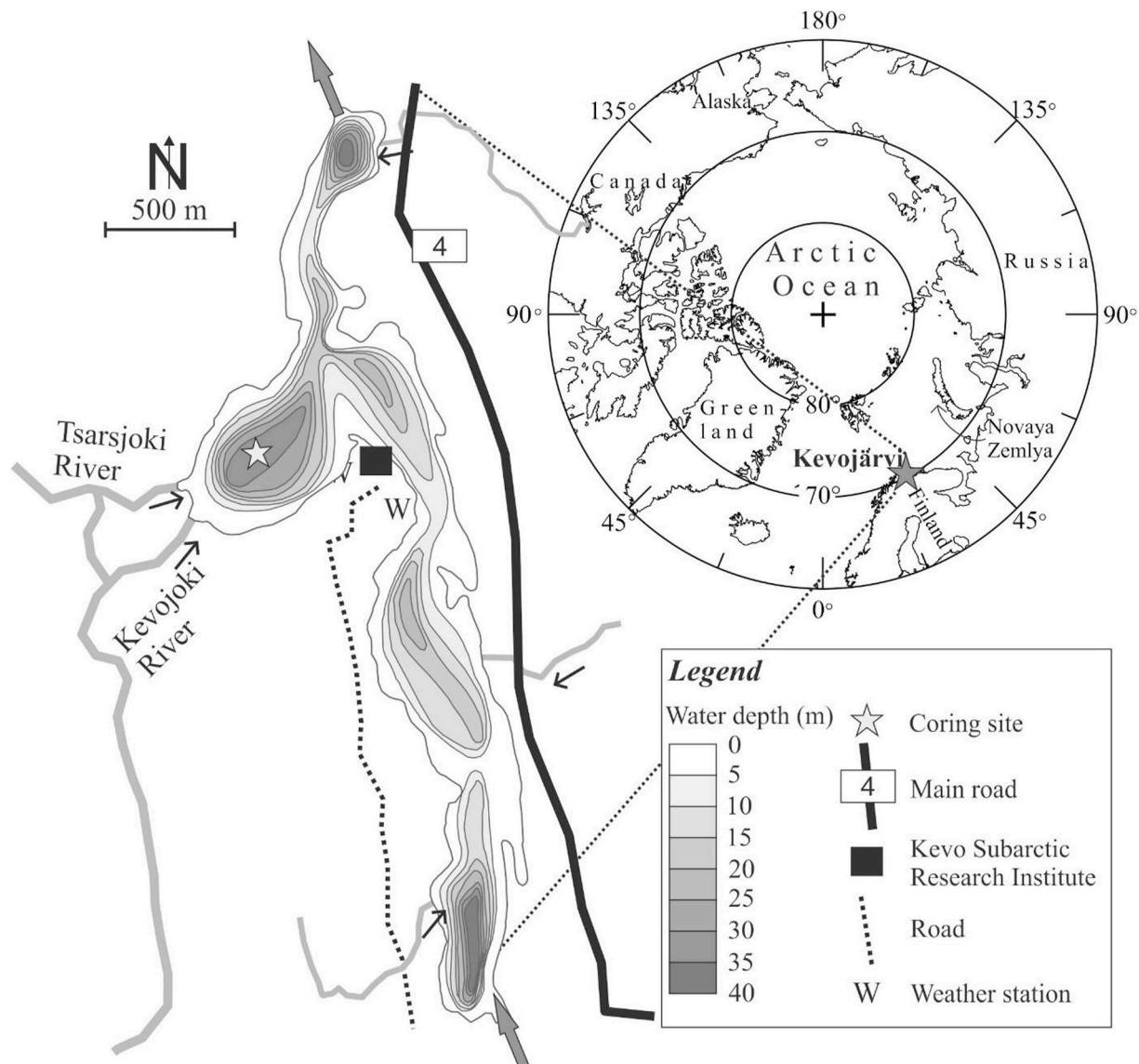


Fig. 1. Location of Lake Kevojärvi. Arrows in the southern and northern ends of the basin show the direction of flow of the Utsjoki River.

with cool summers. The annual mean temperature is $-1.3\text{ }^{\circ}\text{C}$ and the mean precipitation is 430 mm (all climate data from Finnish Meteorological Institute; reference period 1981–2010). There is snow on the ground for 8 months per year (Fig. 2). In general, winters are long and cold, and summers are short and cool, but also exceptionally high summer temperatures have been recorded at the Kevo weather station. Climate data relevant to Lake Kevojärvi and its surroundings are gathered in Fig. 2 (see Fig. 3).

2. Material and methods

2.1. Lake sediment coring

The 35m deep western basin of Lake Kevojärvi lies in the mouth of the Kevojoki River and Tsarsjoki River. The bottom sediments in the western 35 m deep basin, that has steep flanks and a flat bottom, have been sampled annually in April between 2007 and 2018 using the lake ice as a stable coring platform. The lake sediment was sampled using freeze coring technique (e.g. Shapiro 1958; Renberg, 1981) and a 100 cm long wedge-shaped freeze corer filled with carbon dioxide ice and denaturated alcohol. Test coring showed that sediment is laminated only in the western part of Lake Kevojärvi in a deep basin. For this project University of Turku, Finland has recovered three freeze cores from Lake Kevojärvi in 2011, 2013 and 2016, were named as KEVO-1 (62 cm in

length), KEVO-3 (75 cm in length) and KEVO-6_1m, respectively. The varve thickness is somewhat variable in freeze cores taken in different years. There were also cases where the boundaries between laminations were undulating and therefore more indistinct. Some cores contain abundantly plant macrofossils, apparently *Betula* leaves and pieces of *Pinus* bark. The KEVO-1 core was chosen for radioactivity studies because of its high sediment quality where water-sediment interface appears to be undisturbed and distinctly laminated sediment structure.

2.2. Core documentation, varve counting procedure and sediment stabilization by epoxy resin

The surfaces of the freeze cores were carefully scraped clean in the laboratory. The sediment stratigraphy was described macroscopically, and the freeze cores were photographed. Sediment was continuously laminated, and the thickness of laminations decreases from over 2 cm to some millimeters going down-core from the top of the core. The laminations have a basic internal structure in which a basal clastic layer is overlain by a darker layer composed of organic matter. This type of dichotomous structure resembles closely clastic-organic annual laminations, i.e. varves found in few Fennoscandian lakes where varved structure echo the seasonality of climate inducing changes in the sediment source during the course of the year. At this point, the laminations in lake Kevojärvi sediment were tentatively classified as varves. Varve counting was carried out by a single analyst in nine segments, where each segment consisted of a set of 5–12 varves sandwiched between two marker laminae. The counting was repeated three times, which serves as a basis for an error estimate of the varve chronology. Uncertainty of varve counting was derived from the standard deviation of the independent varve chronologies. The sediment chronology was reconstructed by counting of laminations directly from the surface of the freeze cores KEVO-1, KEVO-3 and KEVO-6_1m and photographs taken from them. The composite record created from KEVO-1, KEVO-3 and KEVO-6_1m cores ensured that the highest possible quality of the varve counting by avoiding sediment sections where the varve quality was locally compromised. Additional support for the initial assumption that laminations represent varves was provided by visual observation of a new pair of layers (detrital and organic) in the topmost part of the sediment sequence and comparing the freeze cores taken in the consecutive years.

2.3. Varve chronology

The varve counts from the composite varve record from cores KEVO-1, KEVO-3 and KEVO-6_1m indicate the investigated 71 cm of the sediment profile contains 107 varves that have been deposited between 1909 and 2015. The cumulative varve counting error is estimated as $\pm 0.5\%$, which can be considered closely comparable with varve chronologies from other sites globally (Ojala et al., 2012). Between 2010 and 1929 the number of varves is identical in all three counts. The preciseness of the varve counting reflects the clarity of the varve structure. In the two lowermost segments varve counting error increases because the sediment is more minerogenic and the varve boundaries are not as clear as in the overlying sediment. Between 1915 and 1928 the counting error reaches $\pm 2.0\%$ and in the lowermost segment from 1914 to 1906 it is $\pm 3.2\%$. Most lamination couplets are distinct and easily identifiable by an unaided eye, but a few of them, especially those where the spring snow melt layer (SML) is particularly thin, such as those deposited the years in 1928 and 1990, appeared less distinct. These varves could have become erroneously interpreted to belong to the preceding couplet. After it was noted that the SML properties correspond rather well with the annual peak discharge value of the Utsjoki and Tenjoki rivers, small errors in the varve chronology could be detected and corrected by identifying the prominent flood layers in the sediment record.

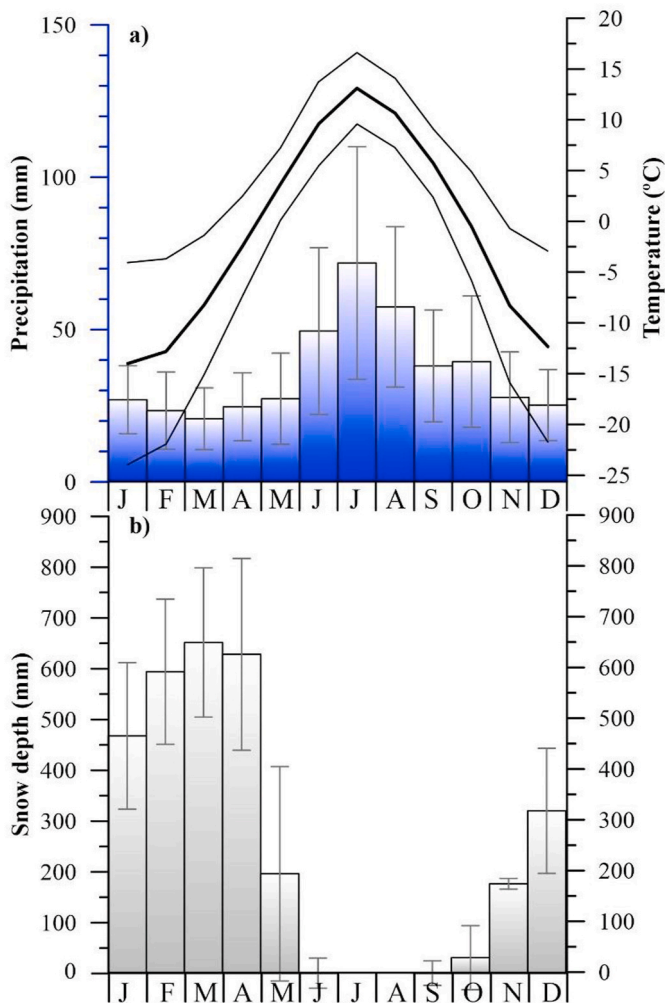


Fig. 2. a) Monthly mean precipitation (columns with standard deviation) and mean temperature (bold line; mean, thin lines: standard deviation) at the Utsjoki Kevo weather station between 1981 and 2010, b) Monthly mean snow depth (with standard deviation) at the Utsjoki Kevo between 1981 and 2010.

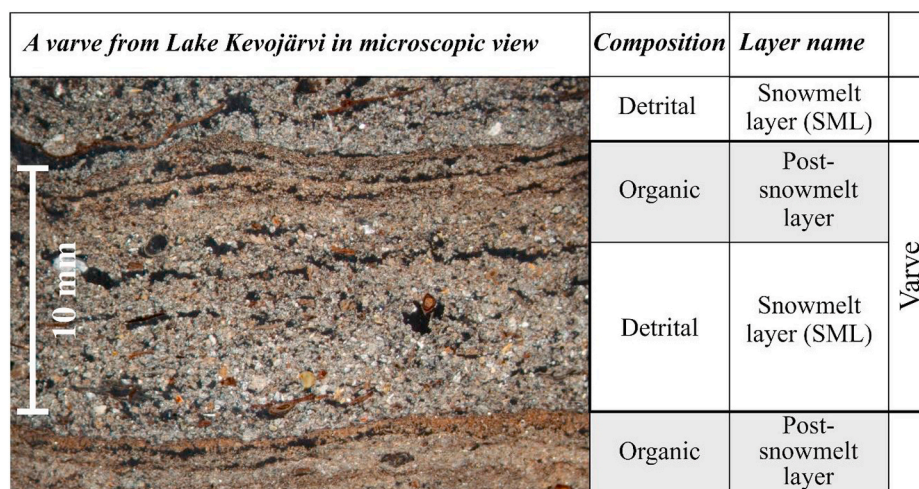


Fig. 3. Microscopic image of the clastic-organic varve of the KEVO-3 freeze core. The composition describes the main material in each layer. The panels on the right side describe the basic varve structure and their naming.

3. Measurements and the data analysis

The KEVO-1 freeze core was taken for gamma spectrometric analyses. A total of 53 subsamples were selected from the KEVO-1 freeze core for radioactivity measurements following the boundaries between the laminations. The varve chronology reconstructed by varve counting was used in targeting subsampling. The aim for time periods of high interest (times of atmospheric nuclear weapons testing and Chernobyl accident) was to take subsamples from every varve while outside of these time periods subsamples were taken with 2–5 years time steps depending on the varve quality. For water content and dry bulk density, the sediment subsamples were weighed and then oven-dried in 60 °C until dry. The dried subsamples were weighed and gently ground using an agate mortar and a pestle. To have enough material for measurements, five subsamples with a dry weight under 0.5 g were combined with an adjacent subsample (1995/1994, 1993/1992, 1984/1983, 1977/1976, 1952/1951). The subsamples were measured at the Radiation and Nuclear Safety Authority – STUK, Regional Laboratory in Northern Finland located in Rovaniemi, Finland. The laboratory was accredited according to the ISO17025:2005 laboratory standard.

The sediment subsamples were measured with an electrically cooled High Purity Germanium (HPGe) detector equipped with a carbon fibre window also allowing the detection of low energy gamma rays. The detector models were either Canberra BE5030 P/s or Ortec GEM-FX8530P4-PLB-S. The HPGe detector was placed inside a 6-inch thick Ultra-low background (ULB) grade lead shielding for reduction of background radiation. The detectors were calibrated using semi-empirical methods where the initial calibration measurement was done with a standard sample containing known activities of known isotopes between the energy range of 46–1836 keV. The measured subsamples had varying weights between 0.5 and 3.8 g in dry weight (d. w.). The large variation in subsample weight was due to the variations in varve thickness.

For gamma spectrometry measurements the sediment subsamples were placed into a plastic beaker of 42 mm in diameter. Thus, the actual subsample formed a thin layer, typically 1–2 mm in thickness, at the bottom of the beaker. The sample beakers were covered with a lid and taped tight. Due to the partly organic nature of the subsample and the thin thickness it formed at the bottom of the beaker, the self-absorption was considered to be low and hence reliable determination of low energy gamma rays. The individual subsample measurement time varied between three to seven days.

The measured radionuclides were ^{137}Cs , ^{241}Am , ^{210}Pb , ^{214}Bi , ^{214}Pb and ^{40}K . The activity concentrations were calculated using the full

energy peaks of 661.7, 59.5, 46.5, 609, 295/352 and 1460.8 keV, respectively. The activity concentrations were calculated using the corresponding full energy peaks, coincident summing correction factors and sample density and thickness corrections. ^{137}Cs and ^{241}Am are anthropogenic nuclear fission products with half-lives of 30.2 years and 432 years, respectively while the half-life of ^{210}Pb is 22.23 years. The gamma spectrometry measurement method employed in this work has been described in more detail in (Kremp et al., 2018).

^{210}Pb is a naturally occurring radionuclide of the ^{238}U decay series. ^{210}Pb activity in surface sediments and soil is in disequilibrium with the long-lived parent ^{226}Ra because the intermediate gaseous isotope ^{222}Rn can partially escape from solid matter into the atmosphere. ^{222}Rn decays through short-lived progeny into ^{210}Pb and these ^{222}Rn daughters attach to aerosols. Wet and dry deposition removes aerosols and ^{210}Pb from the atmosphere into land and water surfaces (Paatero et al., 2015). The most dominant method of ^{210}Pb deposition is wet scavenging and in Kevo and in Northern Finland the annual ^{210}Pb deposition has been found to be 19–55 Bq/m²/a (Paatero et al., 2015; Leppänen, 2019). A part of the deposition gets incorporated into sediments either by falling directly into the water body and getting scavenged, or by transport into the sedimentation area from further away in the catchment. In sediments there are thus two fractions of ^{210}Pb , supported and unsupported. The supported ^{210}Pb is formed from the decay of ^{226}Ra originating in transported solid matter. ^{226}Ra is determined from the decay products ^{214}Pb and/or ^{214}Bi . The unsupported ^{210}Pb is the fraction which originates from atmospheric deposition. The unsupported fraction decays with the ^{210}Pb half-life of 22.23 years while the supported fraction is in radioactive equilibrium with ^{226}Ra , decaying with the half-life of 1600 years. The unsupported ^{210}Pb was calculated by subtracting the mean ^{226}Ra activity from each measured ^{210}Pb total activity. The mean value for ^{226}Ra was used instead of layer-wise subtraction of ^{226}Ra because in 11 out of 53 measured subsamples the ^{226}Ra concentration was below minimum detectable concentration due to small size of the subsamples. The standard deviation of mean ^{226}Ra was propagated to the uncertainty of unsupported ^{210}Pb . The results for each observed radioisotope with the corresponding varve year are shown in Table 1 Fig. 4.

3.1. ^{137}Cs profile

The measured ^{137}Cs activity concentrations in Lake Kevojärvi sediment subsamples are presented in Fig. 5. The results have been decay-corrected to the KEVO-1 freeze core coring date (April 9, 2011). ^{137}Cs was detected almost throughout the Lake Kevojärvi sediment column from the uppermost to the 1922 varve (Fig. 5 and Table 1). Between the

Table 1

The measured radionuclide concentrations in Bq/kg in dw (dry weight) for each varve. In the table (Unc) refers to 1σ uncertainty and $^{210}\text{Pb}(\text{ex})$ to the calculated excess fraction of ^{210}Pb . The supported fraction of ^{210}Pb and its uncertainty used in the calculation of $^{210}\text{Pb}(\text{ex})$ were the mean and standard deviation of ^{226}Ra -results.

varve year	^{137}Cs	Unc [%]	^{210}Pb	Unc [%]	^{226}Ra	Unc [%]	^4K	Unc [%]	^{241}Am	Unc [%]	$^{210}\text{Pb}(\text{ex})$	Unc [%]
2009	30.5	9	274.6	8			281.2	13			259	9
2007	36.2	11	324.9	10			184.1	28			309	11
2005	36.1	11	340.9	10	24.1	22	172.1	28			325	11
2002	36.9	8	196.9	9	19.0	27	320.1	10			181	11
1999	60.3	10	400	11	37.1	37	170.0	40			385	12
1997	40.0	9	223.5	10	22.8	36	125.1	31			208	12
1994–1995	55.7	9	331.77	9	5.0	71	133.1	36			316	10
1992–1993	71.0	7	405.9	7	36.0	18	205.7	19			390	8
1991	60.8	9	273.03	13	20.0	46	60.8	9			258	14
1989	70.6	10	305.08	12	14.6	60	146.2	46			290	13
1987	91.8	6	138.54	12	11.0	32	269.8	13			123	15
1986	228.6	5	143.97	10	11.0	19	293.5	10			129	13
1985	59.9	10	231.3	16			241.8	32			216	18
1983–1984	39.9	7	111.6	10	12.5	15	246.0	10			96	15
1982	56.3	7	187.8	10	24.2	40	176.4	20			172	12
1981	33.1	7	88.16	11	9.0	23	233.1	10			73	18
1979	34.3	8	84.99	17	7.8	32	254.4	12			69	24
1976–1977	31.2	7	62.91	15	5.2	73	258.1	10			47	27
1975	34.4	7	82.45	15	7.1	51	267.0	11	1.6	33	67	23
1974	44.5	10	198	12			177.7	49			183	14
1973	57.9	6	109.1	10	11.3	24	308.4	10	2.2	32	94	15
1971	79.8	6	123.2	18	21.1	40	315.5	16	2.7	48	108	22
1970	80.5	7	138.6	17			276.5	20	3.0	38	123	20
1968–1969	48.9	6	69.7	14	8.7	18	265.3	9	1.4	26	54	24
1967	62.7	6	75.8	21			339.6	12			60	30
1966	76.81	5	80.1	12	9.0	21	285.4	9	2.7	27	65	20
1965	94.03	6	91.9	15	14.3	31	232.5	12	3.5	27	76	21
1964	127.1	7	93.6	30			183.1	33	5.4	30	78	38
1963	87.3	5	98.5	13	10.2	30	295.3	10	3.6	23	83	19
1962	82.9	6	85.8	21			339.0	15	5.6	22	70	29
1961	64.7	7	163.3	18	29.3	22	303.1	20	6.2	29	148	21
1960	64.3	8	100.1	21			394.5	15	6.4	30	85	27
1959	47.5	7	77.5	18	9.5	40	337.6	10	2.6	35	62	27
1958	39.3	8	80.17	18	17.8	16	347.0	12	2.4	35	65	26
1957	43.8	9	96.1	34	37.0	29	401.6	20	3.1	44	81	42
1956	46.3	7	128.7	12	30.7	23	243.6	24			113	16
1955	17.9	7	39.0	17	6.0	48	341.6	7	1.6	31	23	47
1954	11.4	7	58.6	7	11.7	20	307.4	7	0.8	35	43	23
1953	13.3	28	98.2	44			289.3	34			83	53
1952	10.2	14	75.7	16			185.0	15			60	25
1950–1951	18.6	14	119.7	16	23.2	17	105.0	34			104	20
1948	7.4	20	45.6	24	14.0	20	196.8	16			30	47
1946	8.8	17	64.4	22	21.4	17	184.3	19			49	34
1943–1944	9.7	16	120.4	12	6.4	48	186.9	17			105	16
1939	11.2	15	68.9	27	15.0	26	208.1	20			53	39
1935	5.1	23	62.4	25	14.4	22	265.1	13			47	38
1932	4.8	17	53.7	17	9.5	21	275.3	9			38	33
1927–1928	2.6	22	52.4	20	9.2	48	241.4	9			37	37
1920	1.8	15	16.4	22	10.0	9	312.8	5			1	1091
1918–1919	0.6	44	37.7	12	10.5	17	354.9	9			22	45
1917	<2.1		39.5	29	7.5	38	404.4	9			24	60
1915	<1.2		24.9	11	6.4	15	356.8	6			9	99
1913	<2.5		29.4	28	20.5	12	234.0	10			14	87

years 1922 and 1950, i.e., the period prior to large atmospheric nuclear weapons tests, ^{137}Cs concentrations varied from 0.60 to 11 Bq/kg in dry weight (dw), indicating downward diffusion of ^{137}Cs . The atmospheric nuclear weapons testing started in late 1940's following an intensive testing in 1954–1958. Between the United States and the Soviet Union there was a moratorium between 1959–1960 after which intensive testing resumed. Generally, 1954 was the year when the first high yield thermonuclear weapon was exploded and this also is considered as the year when measurable amounts of ^{137}Cs are found in soils (Longmore, 1982). Due to the downward diffusion and radioactive decay of ^{137}Cs it is not possible to say in which sediment varve ^{137}Cs was first deposited in but the first varve with clearly increased ^{137}Cs concentration was 1956 (46 Bq/kg dw). After 1956 the ^{137}Cs concentrations increased clearly until 1964 when the concentration reached the maximum of 127 ± 9 Bq/kg dw. After this peak value, ^{137}Cs concentrations steeply decreased until 1968. In 1970 and 1971 increased ^{137}Cs concentrations of 81 and

80 Bq/kg dw were observed. This is visible as a subsidiary peak is in the profile in Fig. 5. The lamination identified as 1986, based on the lamination counting record, the highest concentration of the whole profile was observed (229 ± 11 Bq/kg dw). In the years following the Chernobyl accident, there is a decreasing trend in ^{137}Cs concentration when moving towards top of the sediment record due to delayed input of ^{137}Cs into the sediment.

Finnish Meteorological Institute (FMI) has performed airborne radioactivity measurements since 1965 in Rovaniemi, Finland. Rovaniemi is located about 430 km south of Lake Kevojärvi. Since 1965 the annual airborne ^{137}Cs concentrations in Rovaniemi show a decreasing trend until 1968 when the annual airborne ^{137}Cs concentration began to rise, reaching the peak value in 1971 after which the concentrations began to drop again (Salminen-Paatero et al., 2019). Bergan (2002) reported annual average ^{137}Cs air concentrations measured in Norway from 1956 to 1981. The air concentrations measured at Rovaniemi and

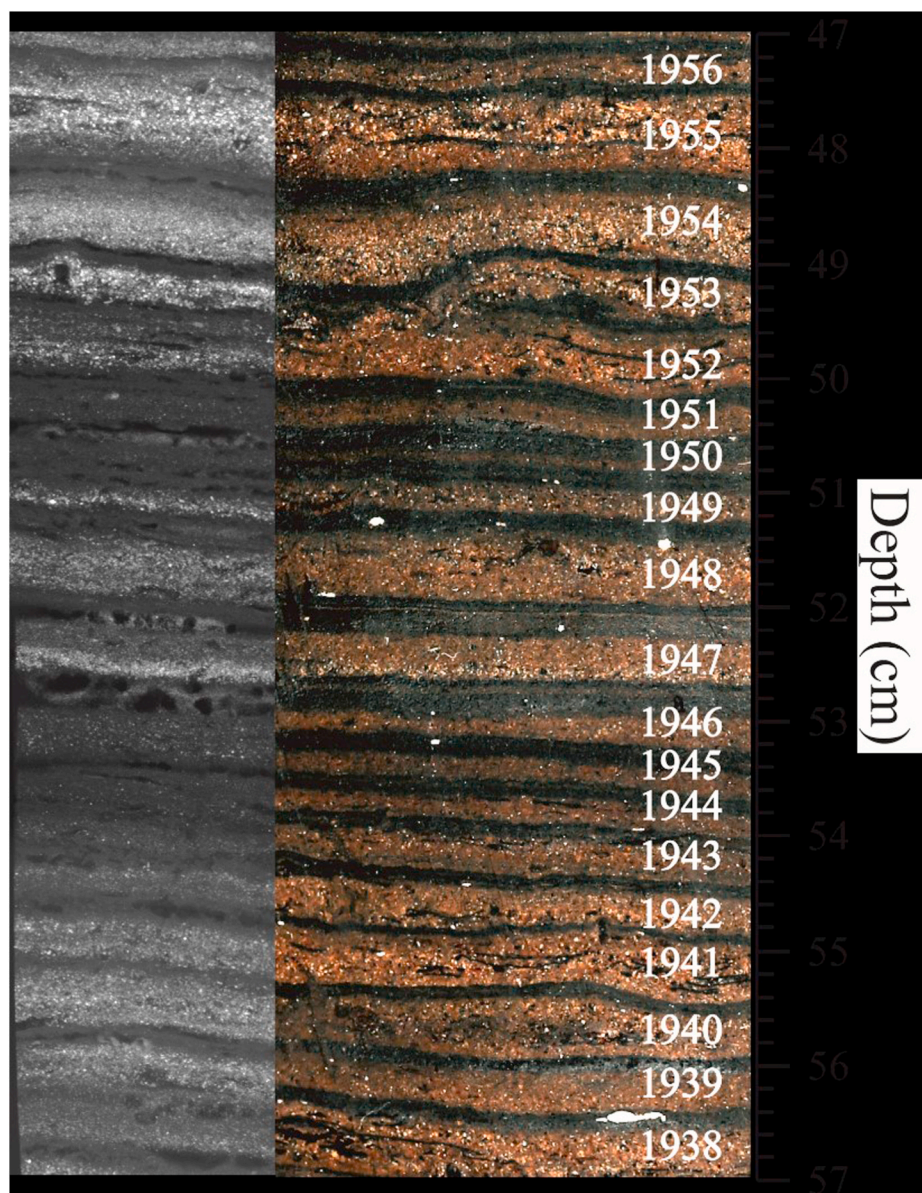


Fig. 4. An X-ray image of the KEVO-1 core and an image taken from the KEVO-3 freeze core surface from Lake Kevojärvi. White numbers label the varve years. The porous structure in the x-ray image in the varves deposited in 1946 and 1947 are assumed to be produced by escaping gases from the sediment during melting of the freeze core.

in Norway are similar to the sediment concentrations in varves shown in this study. The very last atmospheric nuclear weapons test to date was executed in October 16th, 1980 in Lop Nur, China. Interestingly, the ^{137}Cs concentration shows a minor increase from the value of 33 Bq/kg dw in 1981 to 56 Bq/kg dw, occurring in 1982.

The concentration profile in Fig. 5 represents the measured values when decay corrected to the freeze core drilling date. However, it should be noted that the ^{137}Cs in the deeper layers have had more time to decay. There is a 22-year time difference between the 1964 varve where the signal from intensive atmospheric nuclear weapons tests and the 1986 varve where the Chernobyl fallout is present. In order to get a better estimation on the ^{137}Cs deposition that occurred in a particular year, the results shown in Fig. 5 were recalculated using the varve year as the reference point for decay correction. Fig. 6 panel A shows the ^{137}Cs concentrations after using the varve year for decay correction. With this correction, it is thought to better reflect the level of ^{137}Cs fallout in that each year. This is not an accurate measure but a crude estimation. In the figure, the annual cumulative nuclear weapons test yields are indicated

with bars in panels A, B and C (United Nations Scientific Committee on the Effects of Atomic Radiation UNSCEAR, 2000). From Fig. 6 panel A it can be observed that, most likely, Chernobyl accident caused the highest ^{137}Cs fallout in the Lake Kevojärvi region, but the difference compared to the 1963 fallout is not large, only about 8% higher. From the Chernobyl accident Northern Finland received 1000 Bq/m² of ^{137}Cs . (Lepänen et al., 2011; Solatie et al., 2008). If the cumulative fallout through the whole nuclear weapons testing era (1956–1980) is being considered, then the ^{137}Cs fallout from nuclear weapons testing was significantly higher than the one from the Chernobyl accident. As other studies have shown, the ^{137}Cs concentrations in various environmental samples in Northern Scandinavia, e.g. in fish, milk, reindeer and in whole body-counting of humans, were higher in mid- and late 1960s than in 1986–1987 after the Chernobyl accident (see e.g. (AMAP Assessment, 2009)).

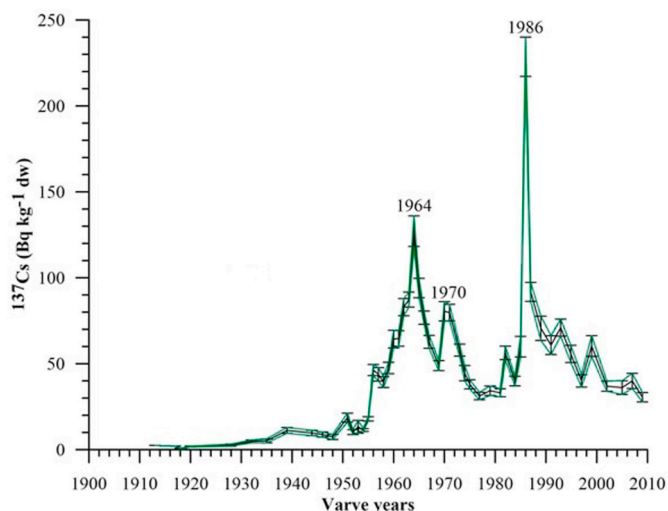


Fig. 5. ^{137}Cs concentration (Bq/kg dw [= dry weight]) measured from Lake Kevojärvi sediment (green line, with standard deviation for each subsample). The selected concentration maxima are labelled with the corresponding varve year. The subsamples for gamma spectrometry measurements were taken from KEVO-1 freeze core. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

3.2. ^{241}Am profile

^{241}Am is a manmade radioisotope with a half-life of $T_{1/2} = 432$ years and it is produced in the neutron capture reaction of uranium. The sources of ^{241}Am in nature are the atmospheric nuclear weapons tests and the Chernobyl accident where the ^{241}Am precursor, ^{241}Pu , was released in large quantities. Similarly to ^{137}Cs , ^{241}Am can be found in environmental samples, e.g., sediment profiles and can be used for dating purposes. About 98% of the ^{241}Am found in the Finnish nature originate from the nuclear weapons tests and only 2% from the Chernobyl accident (Lehto, 2009). However, it should be noted that the concentration of ^{241}Am in a fresh fallout from nuclear weapons tests is nearly zero and the observed ^{241}Am today in the nature is mainly due to the decay of ^{241}Pu . Unfortunately, more accurate radiochemical analyses of the sediment subsamples were not possible since the analyzed subsamples could not go through a destructive analysis. Hence, only gamma spectrometry analyses were performed. It was expected that ^{241}Am could only be found in the 1950s–1970s varves, while in the 1986 layer the ^{241}Am concentration would be too low to be observed. The ^{241}Am deposition from the nuclear weapons tests was rather uniform throughout Finland and the total deposition was estimated to be around 20 Bq/m^2 (Lehto, 2009). The ^{241}Am deposition from Chernobyl accident varied strongly between different region varying between 0.059 and 9.3 Bq/m^2 where Lake Kevojärvi region belongs to the region with low ^{241}Am deposition (Salminen et al., 2005). In nature, ^{241}Am is in the form of dust and the concentrations in nature are increasing due to the decay of ^{241}Pu , the mother of ^{241}Am . In the forest environment ^{241}Am is most abundantly found in soils in organic layer. In the lake environment ^{241}Am is enriched in sediments (Lehto, 2009). Since ^{241}Pu has a relatively short half-life ($T_{1/2} = 14.35$ years), the ^{241}Am concentrations will continue to increase until 73 years after the deposition when the in-grown activity equals that of the activity of ^{241}Pu . After this point the activity of ^{241}Am begins to gradually decrease.

Fig. 6 panel B shows the observed ^{241}Am concentration found in the Lake Kevojärvi sediment varves where the concentrations varied between 0.8 and 6.4 Bq/kg dw (Table 1). Three periods of increased concentrations were observed: first from 1955 to 1957, second period during 1960–1965 and the third during 1970–1973. Cambray et al. (1989) reported on the ^{241}Am fallout in Northern hemisphere. Significant ^{241}Am fallout was first observed in 1955 and higher concentrations

were found in fallout in 1959, 1962–1964 but not in 1970. In the Lake Kevojärvi highest concentration of 6.4 Bq/kg dw was observed in the 1960 varve. The high concentration years of 1960–1965 correspond to the years of high cumulative yields of 1961–1962 added with a two-year time delay from the explosion to the deposition into sediments. Interestingly, the ^{241}Am concentrations appear to be higher in 1960–1962 varves than in the 1964 varve where the ^{137}Cs concentration peaked. It should be noted that the ^{241}Am measurement uncertainties are relatively large and the variations during 1960–1965 are within the measurement uncertainties, hence, drawing definite conclusions is difficult. If the ^{241}Am concentrations truly peak in 1960–1961, the ^{241}Pu originated from the tests conducted in 1958. Bergan (2002) estimated the age of fallout observed in Norway and the fallout in 1960 was estimated to be 21 months old pointing to the tests conducted in 1958. The fallout in 1963–1964 was estimated to be 4–7 months old pointing to the tests conducted in 1962. The dashed lines in panels A, B and C indicate time delay of two years added to the years of high yields. One possible explanation maybe that the yields of nuclear tests in the late 1950s were lower compared to that of e.g. the largest nuclear weapon ever exploded (Tsar Bomba) which released large quantities of radioactivity into the stratosphere. Lower yield weapons could have released larger amounts of plutonium into troposphere producing a larger regional ^{241}Pu fallout. Wendel et al. (2013) studied the isotopic ratios of plutonium and uranium from air filter collected in Norway in early 1960's. They concluded that the fallout from Semipalatinsk and from Novaya Zemlya test sites via troposphere caused a significantly larger regional fallout that has been previously understood. Lake Kevojärvi is some 1000 km southwest from the Novaya Zemlya test site and in the same region as the Norwegian sampling sites used in the study by Wendel et al. (2013).

The third increase in ^{241}Am concentrations were observed during 1970–1973 which is after the United States, Soviet Union and United Kingdom had stopped the atmospheric nuclear weapons tests. In late 1960s and early 1970s China and France were still conducting atmospheric nuclear weapons tests. Salminen-Paatero et al. (2020) reported the annual concentrations of plutonium isotopes found in aerosol filters at Rovaniemi in Finnish Lapland during 1965–2011. The observed ^{241}Pu concentrations were the highest in 1965 after which the concentrations decreased until 1968–1971 when again higher concentrations of ^{241}Pu in the surface air were observed. The increasing concentrations of ^{241}Am found in Lake Kevojärvi sediment varves during 1968–1973 are in good agreement with the observations of Salminen-Paatero et al. (2020) after considering some time delay between air filter observation and sedimentation.

3.3. ^{210}Pb profile

The measured ^{210}Pb results show large variations in adjacent sediment varves (Fig. 7 panel A) and they do not follow the simple exponential curve that would be expected if both sedimentation rate and flux of atmospheric ^{210}Pb were close to constant. Therefore, the constant initial concentration (CIC) family of ^{210}Pb -dating models cannot be used for radiometric dating of this sediment, although these curves are shown in Fig. 7 in panels A and B for visual reference (Appleby 2001). Instead, the constant rate of supply (CRS) model was used, which assumes a constant rate of supply (flux) of atmospheric ^{210}Pb to the sediment surface but allows variable sediment mass accumulation rates (Appleby et al., 1978; Appleby 2001). The unsupported or excess ^{210}Pb (Table 1) is shown in Fig. 7 panel B as a function of mass depth. The porosity and dry bulk density were calculated as in Lima et al. (2005), using a sediment dry density of 2.3 g/cm^3 for the solid matter. The midpoint-procedure (Appleby, 2001) was used to calculate the mass depth and cumulative-excess- ^{210}Pb for the CRS-model because all varves in the sediment core were not measured and some measured subsamples were combinations of two varves to increase the sample size for gamma spectrometry measurement. The calculated values relevant for the CRS model results are shown in Table 2. It is evident, from the results of the deepest

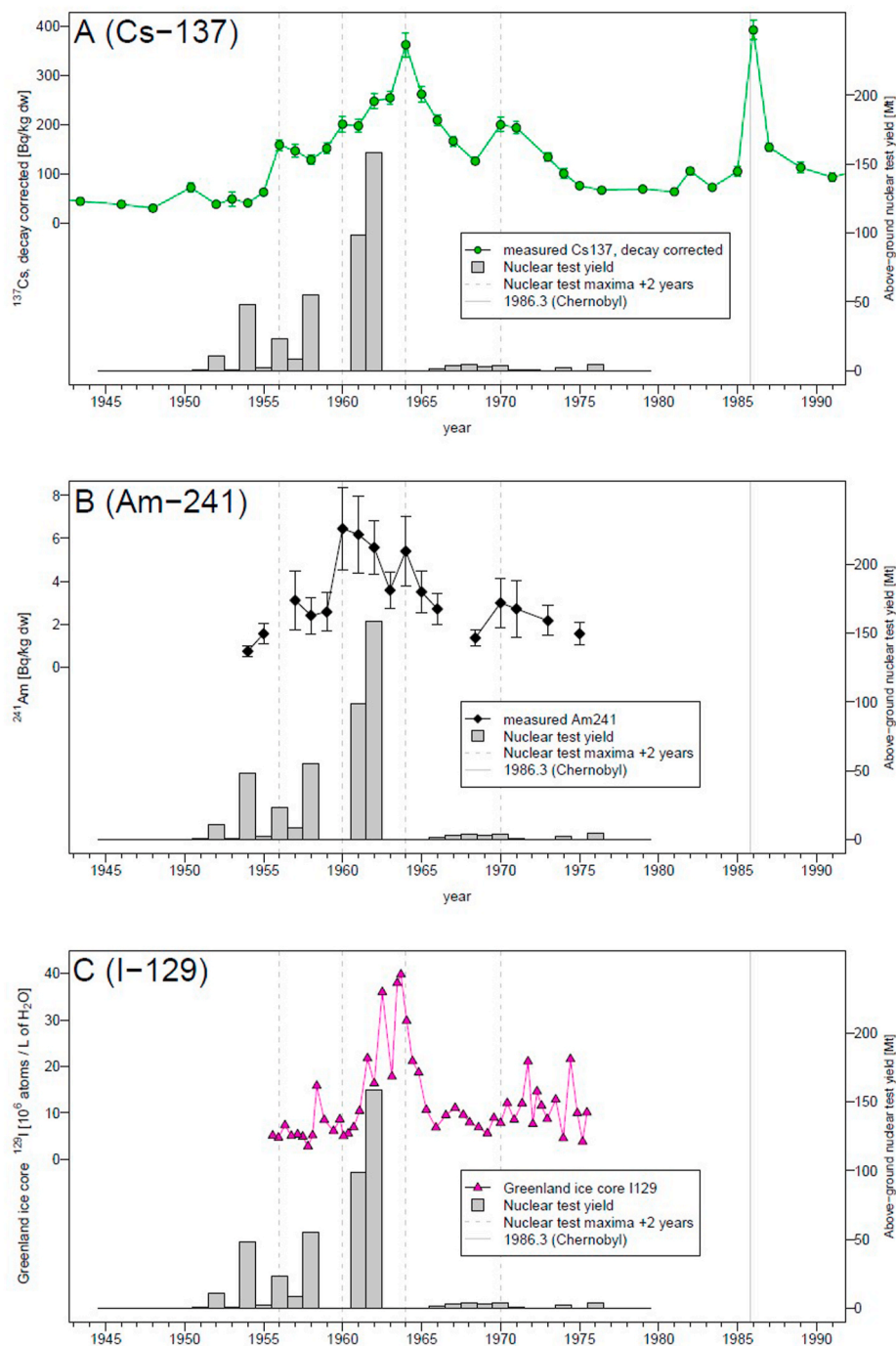


Fig. 6. Time series of different radioisotope concentrations. Panel A shows the ^{137}Cs concentrations founded in the Kevojärvi sediment subsamples where activity is corrected to the varve year. Panel B shows the ^{241}Am concentrations found in the Lake Kevojärvi sediment subsamples ranging from 0.8 to 6.4 Bq/kg dw Panel C shows the ^{129}I concentration found in ice core in SE Dome in Greenland (Bautista et al., 2018). The bars in all graphs indicate the annual yield from atmospheric nuclear weapons tests and the dashed lines shows two years after a year with high annual yield from the nuclear weapons tests (United Nations Scientific Committee on the Effects of Atomic Radiation UNSCEAR, 2000).

measured subsamples, that the excess ^{210}Pb has not yet reached zero at the bottom of the measured profile at 61 cm depth. The variability of excess ^{210}Pb concentration with depth is such that no clearly defined basal accumulation rate can be calculated by fitting of a curve. Therefore, the calculation of excess ^{210}Pb for the basal section (i.e., the excess ^{210}Pb missing from the measured profile) was done using the method of known independent reference date (Appleby 2001). In this case the ^{137}Cs peak of 1964 was used to estimate the excess ^{210}Pb remaining in the unmeasured basal section. The results of these calculations give a total inventory of unsupported excess ^{210}Pb of $11910 \pm 450 \text{ Bq/m}^2$ with the calculated contribution from the unmeasured basal section being $440 \pm 20 \text{ Bq/m}^2$. The rate of supply of atmospheric ^{210}Pb to the sediment surface is calculated to be $370 \text{ Bq/m}^2/\text{a}$, using the CRS model equations (Appleby 2001).

The calculated ages from the CRS-model are in the range of ± 2 years compared to varve-ages until the age of 80 years (47 out of 53 subsamples), and the age-residuals for this time period are normally distributed (Shapiro-Wilk test for normality gives p-value of 0.62) with a mean close to zero (Fig. 7 panel D). During the age interval of 80–100 years the residuals are one-sided ranging from +4 to +9 years, and the deviation from zero residual increases towards the older subsamples. The uncertainties for CRS model ages in Table 3 were calculated with the first order method of (Binford 1990).

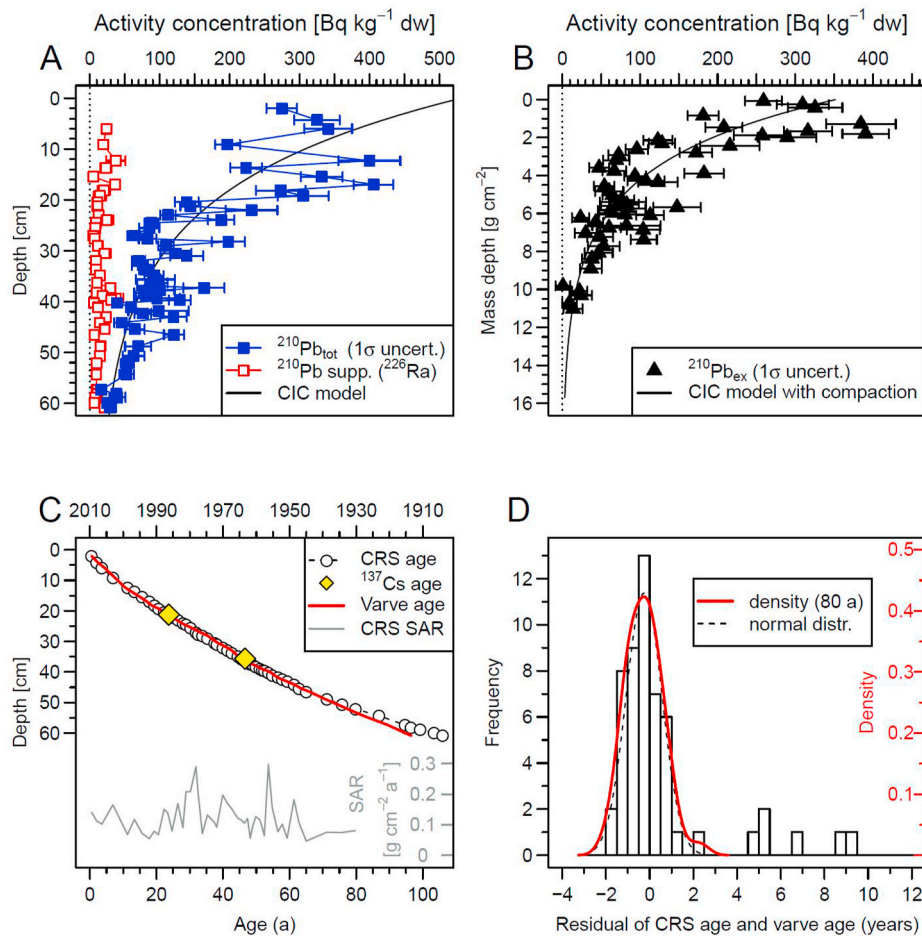


Fig. 7. A) Activity concentrations of total measured ²¹⁰Pb and supported ²¹⁰Pb (represented by ²²⁶Ra) with depth; B) unsupported excess ²¹⁰Pb (²¹⁰Pb_{ex}) with cumulative mass depth; C) CRS model ages with ¹³⁷Cs and varve ages, calculated at midpoints of varve-subsamples. CRS sediment accumulation rate (SAR) is shown for the time period (0–80 a) when CRS ages agree closely with varve ages; D) The residuals of CRS model ages when compared to varve ages (histogram); kernel density plot of residuals until 80 years age; a theoretical normal distribution using the mean and standard deviation from CRS model ages.

4. Discussion

4.1. Anthropogenic ¹³⁷Cs and ²⁴¹Am

The United States, the Soviet Union and the United Kingdom executed atmospheric nuclear testing increasingly in the 1950s. The 1950s culmination in the number of nuclear testing took place in 1958 when over one hundred atmospheric nuclear tests were executed (Norris and Arkin, 1995). By 1954, the atmospheric levels of ¹³⁷Cs had risen sufficiently high in the Northern Hemisphere to produce a measurable signal to be detected in sedimentary records deposited in lakes. A particular increase in the ¹³⁷Cs concentration is visible in 1956 in the Lake Kevojärvi sediments. About one thousand kilometers northeast from Lake Kevojärvi, the former Soviet nuclear test site is located in the archipelago of Novaya Zemlya in the Barents Sea, where a total of 75 atmospheric nuclear tests were conducted between the 1954 and 1962 including the largest thermonuclear weapon ever detonated (Tsar Bomba). (Khalturin et al., 2005). With 110 detonations, the global number of atmospheric nuclear tests reached the maximum in 1962 (Norris and Arkin, 1995). The peak concentration of atmospheric radioactivity resulting from atmospheric nuclear testing was recorded in 1963 (Bergqvist and Ferm, 2000). In spring and summer of 1963, the ¹³⁷Cs maximum activity of ca. 14 000 μBq/m³ was measured in ground-level air in Sodankylä in northern Finland (Salminen et al., 2009).

There were distinct maxima visible in both the ¹³⁷Cs and ²⁴¹Am concentrations in the sediment profile. The maximum concentration of ¹³⁷Cs in Lake Kevojärvi occurs in 1964, while the maximum ²⁴¹Am concentration can be seen in the varves of 1960–1961. However, as

noted in section 4.2, the variations in ²⁴¹Am concentrations are all within the measurement uncertainties and drawing definite conclusions is difficult. The 1960s peak in ¹³⁷Cs found in sediments is usually interpreted as a chronostratigraphic marker denoting the year 1963. In Finland, the maximum deposition of ¹³⁷Cs occurred in 1963 when the average was 422 Bq/m² (Outola and Saxén, 2012). In the studies where the resolution of subsampling allowed pinpointing of the chronostratigraphic placement of the 1960s fallout peak, it is placed between the years 1962 and 1964 (Widerlund and Roos, 1994; Crusius and Anderson, 1995; Klaminder et al., 2012). Direct transportation of radioactivity from Novaya Zemlya was not detected in Finland, despite its relative proximity and intensive use of the test site in 1961 (Salminen and Paatero, 2009). In 1963, the Partial Test-Ban Treaty (PTBT) between the Soviet Union, Great Britain and the United States banned the atmospheric nuclear weapons tests, however, France and China were conducting atmospheric nuclear weapon testing until 1974 and 1980, respectively. The sediment of the Lake Kevojärvi records an increase in ¹³⁷Cs and ²⁴¹Am concentrations in between 1968 and 1973 where peak value was observed in 1970. Similarly, the ¹³⁷Cs profile from Lake Nylandssjön in central Sweden showed an increase centered in 1970 (Klaminder et al., 2012). The ²⁴¹Am profile shows a distinct structure during the atmospheric nuclear weapons test era and serves as a complementary radioisotope marker to ¹³⁷Cs. Due to the longer half-life than ¹³⁷Cs, ²⁴¹Am will serve as a chronostratigraphic marker of the nuclear weapons testing era in years to come after all ¹³⁷Cs has decayed away.

The fallout from the Chernobyl accident was highly uneven and governed by the local weather conditions and occurred mainly as a wet deposition (Arvela et al., 1989; Paatero et al., 2010). The estimated surface activity resulting from the ¹³⁷Cs from the Chernobyl accident in

Table 2
The individual sediment varve information.

Assigned varve date	mid-varve date	mid-varve age [a]	wet weight [g]	dry weight [g]	varve mid-depth [cm]	varve bottom depth [cm]	thickness [cm]	water content	porosity	dry bulk density [g/cm ³]	mass depth [g/cm ²]
2009	2009.5	0.5	19.13	1.30	2	3	2	0.9318	0.9692	0.07091	0.07091
2007	2007.5	2.5	9.96	0.92	4.28	4.702	0.84	0.9079	0.9578	0.09711	0.2625
2005	2005.5	4.5	14.64	1.36	6.016	6.548	1.06	0.9075	0.9576	0.09763	0.4315
2002	2002.5	7.5	17.96	2.77	9.115	9.764	1.30	0.8458	0.9266	0.1689	0.8445
1999	1999.5	10.5	7.61	0.75	12.3	12.59	0.59	0.9015	0.9546	0.1043	1.279
1997	1997.5	12.5	8.68	1.17	13.71	14.08	0.74	0.8649	0.9364	0.1463	1.456
1995	1994.9	15.1	12.57	1.08	15.45	16.21	1.52	0.9142	0.9608	0.09019	1.661
1993	1992.9	17.1	16.63	1.38	16.98	17.74	1.54	0.9169	0.9621	0.08722	1.797
1991	1991.5	18.5	10.00	0.76	18.14	18.54	0.80	0.9243	0.9656	0.07907	1.894
1989	1989.5	20.5	7.56	0.65	19.21	19.47	0.52	0.9141	0.9608	0.09024	1.984
1987	1987.5	22.5	10.98	1.76	20.4	20.74	0.69	0.8398	0.9234	0.1761	2.143
1986	1986.5	23.5	14.72	2.6	21.24	21.74	0.99	0.8234	0.9147	0.1961	2.299
1985	1985.5	24.5	4.76	0.64	22.02	22.3	0.57	0.8664	0.9372	0.1445	2.432
1984	1983.9	26.1	15.01	3.19	22.99	23.68	1.38	0.7878	0.8952	0.2411	2.619
1982	1982.5	27.5	8.82	1.09	23.95	24.22	0.54	0.8767	0.9424	0.1325	2.799
1981	1981.5	28.5	10.37	2.41	24.52	24.83	0.60	0.7679	0.8839	0.2671	2.913
1979	1979.5	30.5	9.87	2.43	25.53	25.78	0.50	0.7538	0.8757	0.286	3.191
1977	1976.9	33.1	14.4	3.15	27.03	27.33	0.61	0.7812	0.8914	0.2497	3.593
1975	1975.5	34.5	7.67	1.91	27.63	27.93	0.60	0.7514	0.8742	0.2893	3.756
1974	1974.5	35.5	3.74	0.53	28.23	28.53	0.60	0.8593	0.9335	0.1529	3.888
1973	1973.5	36.5	10.75	1.88	29.03	29.53	1.00	0.8254	0.9158	0.1937	4.026
1971	1971.5	38.5	5.29	0.71	30.53	30.71	0.37	0.8653	0.9366	0.1458	4.281
1970	1970.5	39.5	6.65	0.74	31.02	31.33	0.61	0.8883	0.9482	0.1192	4.346
1969	1968.9	41.1	13.06	3.83	31.98	32.62	1.30	0.7064	0.8469	0.352	4.571
1967	1967.5	42.5	6.369	1.31	32.86	33.09	0.46	0.794	0.8986	0.2331	4.829
1966	1966.5	43.5	14.6	3.01	33.74	34.38	1.30	0.7938	0.8985	0.2334	5.034
1965	1965.5	44.5	13.18	2.32	34.89	35.39	1.01	0.8238	0.9149	0.1957	5.281
1964	1964.5	45.5	6.82	0.85	35.65	35.92	0.52	0.8755	0.9418	0.1339	5.407
1963	1963.5	46.5	1.69	0.69	36.32	36.72	0.80	0.8309	0.9187	0.187	5.514
1962	1962.5	47.5	6.58	1	36.89	37.06	0.35	0.8481	0.9278	0.1661	5.615
1961	1961.5	48.5	5.39	0.58	37.33	37.59	0.53	0.8928	0.9504	0.1141	5.676
1960	1960.5	49.5	6.16	0.79	37.75	37.92	0.33	0.8723	0.9402	0.1376	5.73
1959	1959.5	50.5	7.88	1.69	38.27	38.62	0.71	0.7858	0.894	0.2438	5.828
1958	1958.5	51.5	7.76	1.40	38.92	39.22	0.59	0.8194	0.9125	0.2011	5.973
1957	1957.5	52.5	2.70	0.35	39.36	39.5	0.29	0.8687	0.9384	0.1418	6.048
1956	1956.5	53.5	4.33	0.62	39.7	39.89	0.38	0.8567	0.9322	0.156	6.098
1955	1955.5	54.5	13.68	3.42	40.26	40.64	0.75	0.7504	0.8736	0.2906	6.225
1954	1954.5	55.5	12.55	2.74	41.14	41.65	1.01	0.7814	0.8916	0.2494	6.462
1953	1953.5	56.5	1.52	0.35	41.84	42.02	0.38	0.7708	0.8855	0.2634	6.64
1952	1952.5	57.5	7.19	1.24	42.33	42.64	0.62	0.8272	0.9168	0.1915	6.753
1951	1950.9	59.1	10.31	1.15	43.03	43.42	0.78	0.8883	0.9481	0.1193	6.862
1948	1948.5	61.5	10.58	1.81	44.15	44.57	0.84	0.8291	0.9177	0.1892	7.034
1946	1946.5	63.5	9.28	1.11	45.42	45.75	0.65	0.88	0.944	0.1287	7.237
1944	1943.9	66.1	9.23	1.16	46.52	46.91	0.78	0.8744	0.9412	0.1352	7.381
1939	1939.5	70.5	5.09	0.79	48.8	49.04	0.48	0.8457	0.9265	0.169	7.728
1935	1935.5	74.5	6.27	1.08	50.7	50.94	0.48	0.8275	0.9169	0.1912	8.07
1932	1932.5	77.5	10.47	2.23	52.14	52.55	0.82	0.7867	0.8945	0.2426	8.382
1928	1927.9	82.1	7.71	1.62	54.32	54.72	0.81	0.7904	0.8966	0.2378	8.906
1920	1920.5	89.5	13.22	3.93	57.37	57.77	0.81	0.7024	0.8445	0.3577	9.814
1919	1918.9	91.1	5.21	1.56	58.15	58.54	0.77	0.7004	0.8432	0.3607	10.1
1917	1917.5	92.5	4.89	1.17	58.83	59.12	0.58	0.7598	0.8792	0.2779	10.31
1915	1915.5	94.5	9.31	3.25	59.93	60.18	0.49	0.6507	0.8107	0.4353	10.71
1913	1913.5	96.5	5.46	1.35	60.8	61.06	0.52	0.7518	0.8745	0.2887	11.02

northern half of Finland was relatively low (≤ 2 kBq m⁻²) compared to more contaminated areas in the southern-central part of Finland (from 45 to 78 kBq m⁻²) (Arvela et al., 1989; Lehto et al., 2013). The accurate position of the ¹³⁷Cs peak in the 1986 varve validates the completeness of the sediment record. Down-core mobilization of ¹³⁷Cs is evident because even the oldest measured subsample from year 1913 contains trace amounts of this artificial radionuclide. Measurement of ¹³⁷Cs from sediment profiles can reveal mobility of this radionuclide to a degree which precludes its use as a chronological marker (Davis et al., 1984). It is also clear that the total inventory of ¹³⁷Cs northern Finland received from global fallout in the 1950s and 1960s was greater in comparison to that from the Chernobyl accident. After 1986, sediments kept receiving ¹³⁷Cs from the catchment or as a re-deposition of sediment from the shallower areas of the lake.

In the Lake Nylandsjön region in Sweden Klaminder et al. (2012)

investigated post-depositional diffusion of ¹³⁷Cs in varved sediment cores taken both prior and after the Chernobyl accident. They noted rapid remobilization of ¹³⁷Cs in the varves of Lake Nylandsjön. The cesium from Chernobyl accident had diffused only two years after the accident to underlying sediments layers deposited over 20 years earlier and nearly concealed the bomb-testing peak. Using varved sediments from Lake Nurmijärvi, Finland, Ojala et al. (2017) could establish a well-resolved ¹³⁷Cs peak from the Chernobyl accident, but the sedimentary record of bomb testing in the 1950s and 1960s was nearly completely obliterated by post-depositional downward diffusion of ¹³⁷Cs. Owing to the fact that Lake Kevojärvi is located in an area with low Chernobyl fallout, the ¹³⁷Cs record from the nuclear testing appears bold. The absence of vertical mixing, which is also a prerequisite for the preservation of varves, enables the detailed preservation of the ¹³⁷Cs record with several useful marker horizons to confirm the fidelity of the

(Bautista et al., 2018). It is possible that the Lake Kevojärvi varve counting contains an error of one year, hence, the time lag between nuclear weapons test and the ^{137}Cs and ^{241}Am deposition into the sediment was estimated to be 1–2 years. However, there was no time lag was observed between the Chernobyl accident and ^{137}Cs deposition into the sediment since the ^{137}Cs peak was found in the 1986 varve. In 1986 the spring flooding in Lake Kevojärvi started in mid-May and the bulk of the deposition from the Chernobyl accident occurred in Northern Lapland before that.

4.2. ^{210}Pb profile, radiometric dating and ^{210}Pb supply rate

The presence of large variations in the concentration of ^{210}Pb between the annual laminae show that the ^{210}Pb record has not been obliterated by, for example, diffusion through pore water. Significant bioturbation or other types of vertical mixing of sediment can already be ruled out on the basis that the varved structures are beautifully preserved in the sediment. The predicted ages from the CRS model with an accuracy of ± 2 years (until 80 years) is comparable to other studies of varved sediments with the CRS model (Appleby et al., 1979; Lima et al., 2005). For most of the measured sediment subsamples the CRS ages are accurate to ± 1 year. This is evidence of the integrity of the sediment, robustness of the varve chronology, preservation of the ^{210}Pb profile against chemical, physical or biological alteration and constancy of the annual atmospheric flux of ^{210}Pb at least for the last 80 years. The deep part of the age-profile at 80+ years behaves in a way that is described for situations where the limit of ^{210}Pb detection is relatively high and some unaccounted excess ^{210}Pb still remains in the basal section (MacKenzie et al., 2011). In this study the detection efficiency of ^{210}Pb was limited by the small subsample sizes (sometimes < 1 g), despite relatively long count times. Overall, the estimation of the unmeasured excess ^{210}Pb in the basal section using the 1964 ^{137}Cs marker can still be considered successful.

The accuracy of the ^{210}Pb -dating for the older ages could be improved further by the measurement of the deepest subsamples with a more sensitive methods (e.g., radiochemical analysis or possibly Compton-suppression or well-type detectors for gamma spectrometry, Lima et al., 2005; MacKenzie et al., 2011). The use of sediment isotope tomography (SIT) model for age calculations could allow variable atmospheric flux of ^{210}Pb (Tylmann et al., 2013). Also getting a longer sediment sample and making additional measurements of the deeper basal section of the sediment would produce a more representative total inventory of excess ^{210}Pb and possibly more accurate ages for the deepest parts of the profile. Care must be taken when interpreting data from sediments when there is no knowledge of the preservation of the sediment record against, e.g., pore water diffusion, bioturbation, mixing, erosion, gas bubbles or other disturbances occurring at a particular site. The use of multiple radionuclides (^{210}Pb , ^{137}Cs , ^{241}Am) is important, and at least one other source of information, such as ^{137}Cs , chemical data on heavy metals or another independent time marker should be used to check the validity of information obtained from excess ^{210}Pb (Appleby 2001; Baskaran et al., 2014). If the different radiometric parameters and other markers are in complete disagreement, then there could be disturbances in the sediment that also affect other environmental parameters. In this case the ^{210}Pb and ^{137}Cs -chronologies are consistent. The ^{210}Pb profile was corrected with the ^{137}Cs peak of 1964 and it correctly reproduces the age of the varve containing the ^{137}Cs peak of 1986. Due to the fluctuations in sedimentation accumulation rate, the ^{137}Cs data by itself could not be used to date accurately the varves outside the distinct markers of 1986 and 1964.

The good agreement of ^{210}Pb -ages with varve-ages show that the variations in the ^{210}Pb concentration profile can be explained by the changes in sediment accumulation rate according to the CRS model. This also means that the annual average flux of ^{210}Pb to the sediment surface at Lake Kevojärvi has been close to constant for the last 80 years, as this is an assumption in the CRS model and a prerequisite for its successful

use. If this assumption was not correct, the CRS model ages would not be comparable to ages from varve counting. The calculated mean annual rate of supply of atmospheric ^{210}Pb into the sedimentation area is 370 $\text{Bq}/\text{m}^2/\text{a}$. This is significantly higher than the typical atmospheric fallout of ^{210}Pb in northern Finland of 19–55 $\text{Bq}/\text{m}^2/\text{a}$ (Paatero et al., 2015; Leppänen, 2019), indicating a drainage area to sedimentation area ratio of approximately 10–20. A more typical ratio of ^{210}Pb supply rate to ^{210}Pb fallout would be closer 1–2 (Appleby 2001), at least in more temperate climates. This order of magnitude difference is not surprising, due to the very large ratio of catchment to lake area (factor of 700, see Site Description) and further sediment focusing into a smaller sedimentation area inside the lake area. Additionally, the long eight-month snowy winter causes a spring thaw and flooding which deposits unsupported ^{210}Pb into the lake from a much larger area than would occur from direct atmospheric deposition (see Appleby et al., 1995, for comparison). There has been discussion that the changes in precipitation caused by climate change could cause problems for the ^{210}Pb -dating in the future (e.g. Paatero et al., 2015), but at least until 2010 this effect is not visible in the Lake Kevojärvi sediments. The question whether the annual flux of atmospheric ^{210}Pb to Lake Kevojärvi has been invariable even before 1930, cannot be answered decisively from the data in this study. In the future, varved sediment sites in Northern latitudes, such as Lake Kevojärvi, will be important for the study of environmental records and the effects of climate change on atmospheric deposition.

5. Conclusions

The clastic-organic varves deposited in Lake Kevojärvi represent the northernmost site on Eurasian mainland with ongoing varve deposition. Owing to varve deposition, the concentrations of anthropogenic ^{137}Cs and ^{241}Am in the Lake Kevojärvi sediment varves showed, with remarkable accuracy, the different nuclear events in history. The observed events were the atmospheric nuclear testing conducted in the late 1950s and the intensive testing in the early 1960s and also the tests conducted far away from Northern Finland in the late 1960s and 1970 and the Chernobyl nuclear power plant accident in 1986. The sediment records showed a time lag of 1–2 years between the intensive testing years and the increase in ^{137}Cs and ^{241}Am concentrations in the sediment varves. It was shown that the ^{241}Am can serve as a complementary radioisotope marker to ^{137}Cs and due to the longer half-life of ^{241}Am , it will serve as a chronostratigraphic marker of the nuclear weapons testing era in years to come when all ^{137}Cs has decayed away. The ^{210}Pb concentrations were also determined and used for calculating the age of the varves by using the CRS model which showed good agreement with the varve counting hence confirming that the varve counting was correct. The ^{210}Pb measurements together with sediment accumulation rates showed that the ^{210}Pb deposition in the area has been constant over the last 80 years and no effect of climate change on ^{210}Pb deposition could be observed.

Declaration of competing 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.

Acknowledgments

This study has been funded by Academy of Finland (grant n:o 269834) and Emil Aaltonen Foundation. Hannu Wenho, Saija Saarni and Arto Peltola took part in lake sediment coring. The authors would also like to thank the two anonymous reviewers who helped to improve this manuscript.

References

- Amap Assessment 2009: Radioactivity in the Arctic. Arctic Monitoring and Assessment Program (AMAP), Oslo, Norway, Xii + 92pp. ISBN 13978-82-7971-059-2.
- Appleby, P.G., 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, P.G., Oldfield, F., Thompson, R., Huttunen, P., 1979. ^{210}Pb dating of annually laminated lake sediments from Finland. *Nature* 280, 53–55.
- Appleby, P.G., Jones, V.J., Ellis-Evans, J.C., 1995. Radiometric dating of lake sediments from Signy Island (maritime Antarctic): evidence of recent climatic change. *J. Paleolimnol.* 13, 179–191.
- Appleby, P.G., 2001. Chronostratigraphic techniques in recent sediments. In: Last, W.M., Smol, J.P. (eds.), *Tracking Environmental Change Using Lake Sediments, Volume I: Basin Analysis, Coring, and Chronological Techniques*. 171–203. Kluwer, Dordrecht.
- Arvela, H., Markkanen, M., Lemmelä, H., Blomqvist, L., 1989. Environmental Gamma Radiation and Fallout Measurements in Finland, 1986–1987. STUK-A76.
- Baskaran, M., Nix, J., Kuyper, C., Karunakara, N., 2014. Problems with the dating of sediment core using excess ^{210}Pb in a freshwater system impacted by large scale watershed changes. *J. Environ. Radioact.* 138, 355–363.
- Bautista, A.T., Miyake, Y., Matsuzaki, H., Iizuka, Y., Horiuchi, K., 2018. High-resolution ^{129}I bomb peak profile in an ice core from SE-Dome site, Greenland. *J. Environ. Radioact.* 184–185, 14–21.
- Bergan, T.D., 2002. Radioactive fallout in Norway from atmospheric nuclear weapons tests. *J. Environ. Radioact.* 60, 189–208.
- Bergqvist, N.-O., Ferm, R., 2000. Nuclear Explosions 1945–1998. FOA-R-00-01572-180, Stockholm, Sweden.
- Binford, M.W., 1990. Calculation and uncertainty analysis of ^{210}Pb dates for PIRLA project lake sediment cores. *J. Paleolimnol.* 3, 253–267.
- Cambray, R.S., Playford, K., Leweis, G.N.J., Carpenter, R.C., 1989. report Radioactive Fallout in Air and Rain: Results to the End of 1987, AERE Report R-13226, Harwell.
- Crusius, J., Anderson, R.F., 1995. Evaluating the mobility of ^{137}Cs , $^{239+240}\text{Pu}$ and ^{210}Pb from their distributions in laminated lake sediments. *J. Paleolimnol.* 13, 119–141.
- Davis, R.B., Hess, C.T., Norton, S.A., Hanson, D.W., Hoagland, K.D., Anderson, D.S., 1984. ^{137}Cs and ^{210}Pb dating of sediments from soft-water lakes in New England (U.S.A.) and Scandinavia, a failure of ^{137}Cs dating. *Chem. Geol., Geochronology of recent deposits* 44, 151–185.
- Jokinen A., Sami, Virtasalo J., Joonas, Kotilainen T., Aarno, Saarinen, Timo, 2015. Varve microfabric record of seasonal sedimentation and bottom flow-modulated mud deposition in the coastal northern Baltic Sea. *Marine Geology* 366, 79–96. <https://doi.org/10.1016/j.margeo.2015.05.003>.
- Kämpf, L., Brauer, A., Swierczynski, T., Czmyzik, M., Mueller, P., Dulski, P., 2014. Processes of flood-triggered detrital layer deposition in the varved Lake Mondsee sediment record revealed by a dual calibration approach. *J. Quat. Sci.* 29, 475–486.
- Khalturin, V.I., Rautian, T.G., Richards, P.G., Leith, W.S., 2005. A Review of Nuclear Testing by the Soviet Union at Novaya Zemlya, 1955–1990. *Sci. Glob. Secur.* 13, 1–42.
- Klaminder, J., Appleby, P., Crook, P., Renberg, I., 2012. Post-deposition diffusion of ^{137}Cs in lake sediment: Implications for radiocaesium dating. *Sedimentology* 59 (7), 2259–2267. <https://doi.org/10.1111/j.1365-3091.2012.01343.x>.
- Kremp, A., Hinners, J., Klais, R., Leppänen, A.-P., Kallio, A., 2018. Patterns of vertical cyst distribution and survival in 100-year-old sediment archives of three spring dinoflagellate species from the Northern Baltic Sea. *Eur. J. Phycol.* 53 (2), 135–145.
- Lehto, J., 2009. Americium in the Finnish environment. *Boreal Env. Res.* 14, 427–437.
- Lehto, J., Vaaramaa, K., Leskinen, A., 2013. ^{137}Cs , $^{239,240}\text{Pu}$ and ^{241}Am in boreal forest soil and their transfer into wild mushrooms and berries. *J. Environ. Radioact.* 116, 124–132.
- Leppänen, A.-P., Muikku, M., Jaakkola, T., Lehto, J., Rahola, T., Rissanen, K., Tillander, M., 2011. Effective half-lives of ^{134}Cs and ^{137}Cs in reindeer meat and in reindeer herders in Finland after the Chernobyl accident and the ensuing effective radiation doses to humans. *Health Phys.* 100 (5), 468–481.
- Leppänen, A.-P., 2019. Deposition of naturally occurring ^7Be and ^{210}Pb in Northern Finland. *J. Environ. Radioact.* 105995, 208–209.
- Lima, A.L., Hubeny, J.B., Reddy, C.M., King, J.W., Huguen, K.A., Eglinton, T.I., 2005. High-resolution historical records from Pettaquamscutt River basin sediments: 1. ^{210}Pb and varve chronologies validate record of ^{137}Cs released by the Chernobyl accident. *Geochim. Cosmochim. Acta* 69, 1803–1812.
- Longmore, M.E., 1982. The caesium-137 dating technique and associated applications in Australasia review. In: Ambrose, W., Duerden, P.(Eds.), *Archaeometry: an Australasian Perspective*. Australian National University Press, Canberra, Australia, pp. 310–321.
- MacKenzie, A.B., Hardie, S.M.L., Farmer, J.G., Eades, L.J., Pulford, I.D., 2011. Analytical and sampling constraints in ^{210}Pb dating. *Sci. Total Environ.* 409 (7), 1298–1304. <https://doi.org/10.1016/j.scitotenv.2010.11.040>.
- Norris, R.S., Arkin, W.M., 1995. Known Nuclear Tests Worldwide, 1945–1994. *Bull. At. Sci.* 51, 70–71.
- Ojala, A.E.K., Alenius, T., Seppä, H., Giesecke, T., 2008. Integrated varve and pollen-based temperature reconstruction from Finland: evidence for Holocene seasonal temperature patterns at high latitudes. *Holocene* 18, 529–538.
- Outola, I., Saxén, R., 2012. Radionuclide Deposition in Finland 1961–2006. STUK-A, 253. Helsinki, Finland.
- Ojala E.K., A., Francus, P., Zolitschka, B., Besonen, M., Lamoureux F., S., 2012. Characteristics of sedimentary varve chronologies – A review. *Quat. Sci. rev.* 43, 45–60. <https://doi.org/10.1016/j.quascirev.2012.04.006>.
- Ojala, A.E.K., Luoto, T.P., Virtasalo, J.J., 2017. Establishing a high-resolution surface sediment chronology with multiple dating methods – Testing ^{137}Cs determination with Nurmijärvi clastic-biogenic varves. *Quart. Geochron.* 37, 32–41. <https://doi.org/10.1016/j.quageo.2016.10.005>.
- Paatero, J., Hämeri, K., Jaakkola, T., Jantunen, M., Koivukoski, J., Saxén, R., 2010. Airborne and deposited radioactivity from the Chernobyl accident — a review of investigations in Finland. *Boreal Environ. Res.* 15, 19–33.
- 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.
- Renberg, I., 1981. Improved methods for sampling, photographing and varve-counting of varved lake sediments. *Boreas* 10, 255–258.
- Saarni, S., Muschiettiello, F., Weege, S., Brauer, A., Saarinen, T., 2016. A late Holocene record of solar-forced atmospheric blocking variability over Northern Europe inferred from varved lake sediments of Lake Kuninkaislampi. *Quat. Sci. Rev.* 154, 100–110.
- Salminen, S., Jaakkola, T., Lehto, J., Paatero, J., 2005. Americium and curium deposition in Finland from the Chernobyl accident. *Radiochim. Acta* 93, 771–779.
- Salminen, S., Paatero, J., 2009. Concentrations of ^{238}Pu , $^{239+240}\text{Pu}$ and ^{241}Pu in the surface air in Finnish Lapland in 1963. *Boreal Env. Res.* 14, 827–836.
- Salminen, S., Paatero, J., Hatakka, J., Makkonen, U., 2009. Airborne ^{137}Cs , ^{90}Sr and total beta activity in northern Finland in the 1960s. *NBC 2009 7th Symp. CBRNE Threats Meet. Future Chall* 205–209.
- Salminen-Paatero, S., Tölix, L., Kivi, R., Paatero, J., 2019. Nuclear contamination sources of surface air in Finnish Lapland in 1965–2011 studied by means of ^{137}Cs , ^{90}Sr and total beta activity. *Environmental Science and Pollution Research* 28 (21), 21511–21523.
- Salminen-Paatero, S., Vira, J., Paatero, J., 2020. Measurements and modeling of airborne plutonium in Subarctic Finland between 1965 and 2011. *Atmos. Chem. Phys.* 20, 5759–5769.
- Sander, M., Bengtsson, L., Holmquist, B., Wohlfarth, B., Cato, I., 2002. The relationship between annual varve thickness and maximum annual discharge (1909–1971). *J. Hydrol.* 263, 23–35.
- Shapiro, J., 1958. The core-freezer—a new sampler for lake sediments. *Ecology* 39, 748.
- Snowball, I., Zillen, L., Gaillard, M.-J., 2002. Rapid early-Holocene environmental changes in northern Sweden based on studies of two varved lake-sediment sequences. *Holocene* 12, 7–16.
- Solatie, D., Leppänen, A.-P., Niskala, P., Ylipietä, J., 2008. ^{90}Sr and ^{137}Cs in deposition, milk and grass in Northern Finland. In: Strand P, Brown J, Jolle T (eds). *Proceedings: Posters – Part 2. International Conference on Radioecology & Environmental Radioactivity, 2008 Jun 15–20; Bergen, Norway*. Østerås: Norwegian Radiation Protection Authority; 2008. p. 296–299.
- Tiljander, M., Saarnisto, M., Ojala, A.E.K., Saarinen, T., 2003. A 3000-year palaeoenvironmental record from annually laminated sediment of Lake Korttajärvi, central Finland. *Boreas* 32, 566–577.
- Tylmann, W., Enters, D., Kinder, M., Moska, P., Ohlendorf, C., Poreba, G., Zolitschka, B., 2013. Multiple dating of varved sediments from Lake qazduny, northern Poland: Toward an improved chronology for the last 150 years. *Quat. Geochronol.* 15, 98–107.
- United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), 2000. In: Annex, C (Ed.), Report to the General Assembly, with scientific annexes Vol.
- Wendel, C.C., Fifield, L.K., Oughton, D.H., Lind, O.C., Skipperud, L., Bartnicki, J., Tims, S.G., Hoibraten, S., Salbu, B., 2013. Long-range tropospheric transport of uranium and plutonium weapons fallout from Semipalatinsk nuclear test site to Norway. *Environ. Int.* 59, 92–102.
- Widerlund, A., Roos, P., 1994. Varved sediments in the Kalix River estuary, Northern Sweden. *Aqua Fenn.* 24, 163–170.
- Wohlfarth, B., Holmquist, B., Cato, I., Linderson, H., 1998. The climatic significance of clastic varves in the Angermanalven Estuary, northern Sweden, AD 1860 to 1950. *Holocene* 8, 521–534.