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A century old record of lead-210 fallout on the Greenland ice sheet

By V. N. NIJAMPURKAR, *Physical Research Laboratory, Navrangpura, Ahmedabad 380 009, India* and H. B. CLAUSEN, *Geophysical Institute, Department of Glaciology, Haraldsgade 6, DK 2200 Copenhagen N, Denmark*

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ABSTRACT

Systematic measurements of the stable oxygen isotope ratio ($\delta^{18}O$) and artifical radioactivity (total β) on a 77 m ice core from the DYE 3 location in S.E. Greenland provided precise age indices to study the fallout of lead-210 during the last century. The results obtained on the Dye 3 core and those of similar studies conducted on two other ice cores from the Greenland locations, Camp Century and North Central, are discussed in relation to the average fallout of ^{210}Pb and the probable contributions made by natural and artificial sources in the atmosphere. The results indicate that: (i) the fallout of ^{210}Pb observed over a period of 100 years has not remained constant, an assumption usually made in geochronological studies; (ii) the fallout of ^{210}Pb over the period 1886–1930 is observed to be higher at least by a factor of two than that observed in the period 1930–1975; (iii) the higher ^{210}Pb fallout observed over the period 1886–1930 coincides with high volcanic activity in this period; (iv) there is no evidence for significant production of ^{210}Pb due to testing of nuclear weapons in the three decades: 1950s to 1970s.

1. Introduction

Polar glaciers and ice sheets preserve undisturbed records of past climatic changes, atmospheric and nuclear fallouts, volcanic debris, wind-borne materials (dust particles, sea salts etc.) and a wealth of other information. Precipitation falling on these ice sheets is extremely pure and the snow is slowly converted into ice due to the pressure of the succeeding years' snow fall. Polar ice from high altitude regions preserves these records more systematically and efficiently than any other reservoirs on earth because little melting and run-off take place in these regions. The Greenland ice sheet is, compared to Antarctica, fed by relatively frequent and heavy snow falls and analytical study of a well-dated ice core from Greenland offers a clue to reveal the past climatic and volcanic records, fallouts of several radioisotopes and other matter.

Lead-210, owing to its appropriate half-life (22.3 years) for chronological studies, has found several applications in diverse fields like glaciology (Crozaz and Langway, 1966; Sanak and Lambert, 1977; Nijampurkar et al., 1982; Gäggeler et al., 1983), limnology (Krishnaswamy and Lal, 1978), oceanography (Craig et al., 1973) and atmospheric studies (Jacobi and André, 1963; Bhandari, 1965; Joshi et al., 1969). The major natural source of ^{210}Pb is in the troposphere, where ^{222}Rn emanates from the Earth's crust as a result of decay of ^{226}Ra. The production of ^{210}Pb from the oceans is small (Table 1) compared to that produced from the land, since ^{226}Ra concentration in seawater is much less than that in the Earth's crust (Broecker et al., 1967).

Lead-210 may also be produced during volcanic eruptions, which inject large amounts of gases and other impurities into the atmosphere. These gases are rich in acidic gases like SO_2, long-lived daughters of ^{222}Rn and volatile elements from the magma. High concentrations of ^{210}Pb have been measured in the plumes of

Table 1. *Sources of ^{210}Pb in the atmosphere*

Source	Amount (kCi yr^{-1})	Ref.
from radon-222 exhalation from lands	690	Jaworowski et al. (1978a)
from radon-222 exhalation from oceans	25	Jaworowski et al. (1978a)
from tetraethyl lead	2.3	Jaworowski et al. (1978a)
from radium-rich phosphate fertilizers	0.1–0.6	Krishnaswami and Lal (1978)
from burned coal	0.0004	Jaworowski et al. (1978a)
from volcanoes	insignificant	This paper
from atmospheric thermonuclear bomb tests	insignificant	This paper

some volcanoes, but are not comparable to its production from the continental source (Lambert et al., 1979). ^{210}Pb production from other artificial sources like tetraethyl lead, burning of fossil fuels and radium-rich phosphate fertilizers is small compared to its production from the Earth's crust (Table 1). In addition to these sources, ^{210}Pb could also be injected into the atmosphere from the testing of nuclear weapons by the nuclear reaction $^{208}Pb(2n,\gamma)^{210}Pb$ (Peirson et al., 1966) with lead as a target material. On the basis of calculations of ^{210}Pb in thermonuclear conditions, (Grotowoski et al., 1977) and of the observed correlation between fission products (^{137}Cs) and increased level of ^{210}Pb in temperate glacier ice during the period of nuclear test series, Jaworowski et al. (1978b) suggested production of ^{210}Pb in the atmospheric thermonuclear bomb tests. Arguments for and against bomb-produced ^{210}Pb have been made by several researchers (Bhandari et al., 1966; Robbins, 1978; Turekian et al., 1977; Jaworowski et al., 1978a; Nijampurkar et al., 1982). The problem of ^{210}Pb production from different natural and artificial sources is not well understood, but rough estimates have been made in the past (Table 1), (Jaworowski et al., 1978a).

The present work, performed on well-dated ice cores from Greenland, provides an unique opportunity to verify the basic assumption, which is usually made in geochronological studies, that the fallout of ^{210}Pb has remained constant over the past few centuries, and to study the natural and artificial sources of ^{210}Pb in the atmosphere.

2. Sampling details

Three Greenland ice cores from Dye 3 (65°11′N, 43°29′W, 2480 m a.s.l.), North Central (74°37′N, 39°24′W, 2930 m a.s.l.) and Camp Century (77°11′N, 61°53′W, 1880 m a.s.l.) were raised (Fig. 1) in 1976 and 1977 during the American–Swiss–Danish Greenland Ice Sheet Program (GISP) using a light-weight core drill (Johnsen et al., 1980). The ice cores were stored in frozen conditions at −25°C at the Geophysical Institute in Copenhagen, Denmark. The entire length of the three ice cores were analysed in details for $\delta^{18}O$. Figs. 2, 3a, 3b and 3c show the $\delta^{18}O$ records for part of the ice cores in this

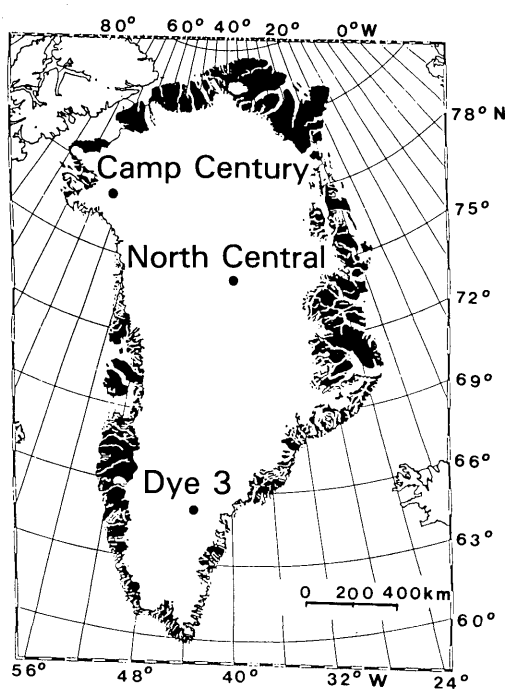

Fig. 1. Ice core locations showing Dye 3, North Central and Camp Century in Greenland.

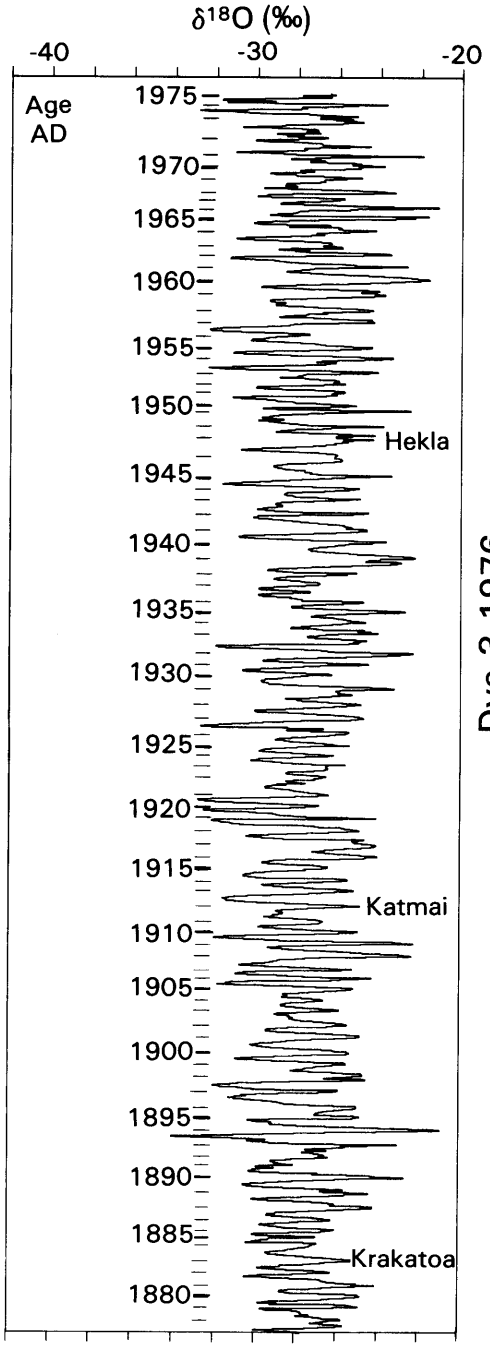

Fig. 2. Variation of $\delta^{18}O$ (‰) along a 77 m deep ice core from the Dye 3 location for the period 1880–1975. Each $\delta^{18}O$ value corresponds to $\frac{1}{16}$ of an average yearly accumulation rate of 56.1 cm ice equivalent. The horizontal bars indicate the position of summer precipitation in the record. Volcanic eruptions are marked as reference points.

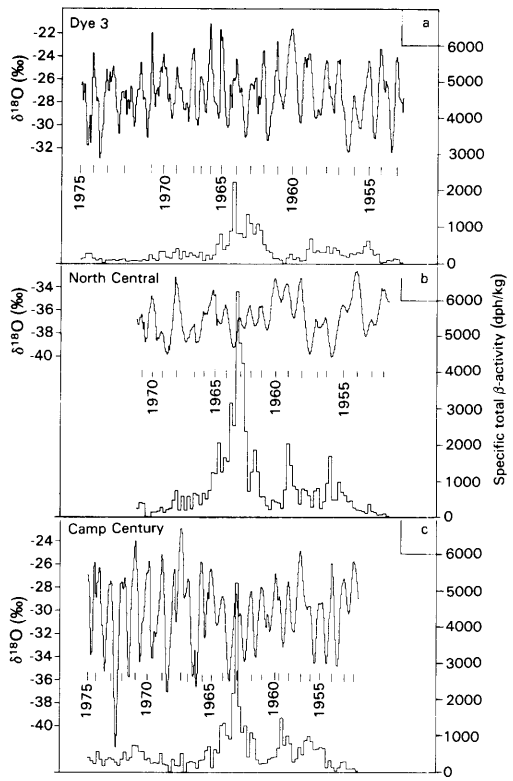

Fig. 3. $\delta^{18}O$ oscillations in per mille (scale to the left) and specific total β activity (dph kg^{-1} l of ice) (scale to the right) for three Greenland ice cores from Dye 3 (a), North Central (b) and Camp Century (c) during the periods 1953–1975, 1952–1971 and 1952–1971, respectively. The records show the characteristic maximum during 1963–64 due to the Russian nuclear tests in the 1960s.

study. These records are the basis for the selection of the ice core samples, which correspond to the accumulated amount of ice for a given year; the samples were analysed for total β- and ^{210}Pb-activity.

Dye 3 is located in S.E. Greenland and receives an average of 60 cm of ice equivalent as annual precipitation. The ice core raised at Dye 3 was cut into different annual sections for the period 1886–1975. The samples, representing from 1–5 years of accumulation (Tables 2 and 3), were analysed for ^{210}Pb activity to study possible variations in the natural and artificial fallout levels of ^{210}Pb. For samples from 1886–1930,

Table 2. *Observed annual specific activity and calculated initial deposition rate of Pb-210 for the period 1931–1975 in the 1976 Dye 3 ice core*

Year in sample AD	Weight of sample (kg)	Specific activity (dph kg^{-1})	Accumulated rate (cm of ice equivalent yr^{-1})*	Initial rate of deposition (dph cm^{-2} yr^{-1})
1975	0.98	79.3 ± 3.3	31.6	2.68 ± 0.11
1974	1.18	71.7 ± 2.0	49.1	3.89 ± 0.11
1973	2.15	66.8 ± 1.8	80.6	6.15 ± 0.17
1972	1.69	51.4 ± 2.0	119.2	7.21 ± 0.28
1971–70	4.33	44.7 ± 2.0	101.7	2.80 ± 0.13
1969	2.06	52.2 ± 2.0	70.1	4.73 ± 0.18
1968	1.01	66.4 ± 3.6	38.6	3.41 ± 0.19
1967	0.91	72.1 ± 5.6	35.1	3.48 ± 0.27
1966	1.46	46.2 ± 2.1	52.6	3.45 ± 0.16
1965	1.13	58.9 ± 2.6	38.6	3.32 ± 0.15
1964	2.05	55.4 ± 3.1	73.6	6.16 ± 0.34
1963	1.63	50.3 ± 3.4	52.6	4.12 ± 0.28
1962	1.24	58.9 ± 3.0	42.1	3.98 ± 0.20
1961	1.92	42.0 ± 2.9	42.1	2.93 ± 0.20
1960	2.28	45.9 ± 2.4	73.6	5.78 ± 0.30
1959	1.96	45.3 ± 2.9	63.1	5.04 ± 0.32
1958	2.45	33.9 ± 2.4	77.1	4.76 ± 0.34
1957	1.86	52.5 ± 2.3	59.6	5.87 ± 0.26
1956	1.58	43.7 ± 2.1	52.6	4.45 ± 0.21
1955	1.87	47.1 ± 1.8	59.6	5.61 ± 0.21
1954	2.26	38.3 ± 2.2	70.1	5.53 ± 0.32
1953	1.44	39.3 ± 2.3	45.6	3.81 ± 0.22
1952	1.61	38.9 ± 2.5	42.1	3.59 ± 0.23
1951	1.45	40.9 ± 2.3	45.6	4.22 ± 0.24
1950	1.53	44.2 ± 1.8	52.6	5.42 ± 0.22
1949	1.74	48.1 ± 2.4	56.1	6.49 ± 0.32
1948	1.56	46.0 ± 2.4	52.6	6.00 ± 0.31
1947	2.62	28.1 ± 1.1	87.7	6.31 ± 0.25
1946	2.29	36.2 ± 1.4	77.1	7.38 ± 0.29
1945	2.45	25.7 ± 1.2	84.2	5.89 ± 0.28
1944	1.40	25.6 ± 1.9	49.1	3.53 ± 0.26
1943–41	2.60	22.5 ± 1.2	199.9	4.48 ± 0.24
1940–39	4.54	22.9 ± 0.7	161.3	5.97 ± 0.18
1938	0.79	38.0 ± 2.8	42.1	5.42 ± 0.40
1937	1.32	30.4 ± 2.2	73.6	7.82 ± 0.57
1936	1.04	36.5 ± 3.4	56.1	7.38 ± 0.69
1935	0.87	34.9 ± 4.7	45.6	5.91 ± 0.80
1934–31	3.79	22.8 ± 0.7	256.0	5.86 ± 0.18

* The rate of accumulation corresponds to the years mentioned in the first column of this table.

successive samples of 5 years accumulation (Table 3) were combined to increase the signal level of ^{210}Pb. To obtain information on the spatial ^{210}Pb distribution in Greenland, we analysed two other ice cores from Camp Century (CC) and North Central (NC), (Fig. 1) for ^{210}Pb, total β and $\delta^{18}O$.

3. Analytical procedures

3.1. Oxygen isotope ratio ($\delta^{18}O$)

Mass-spectrometrical $\delta^{18}O$ measurements were performed on ice samples, which represented about one month's precipitation, with a precision of $\pm 0.05‰$ (Dansgaard et al., 1973).

Table 3. *Observed 5-year means of specific activity and initial deposition rate of Pb-210 for the period 1886–1975 in the 1976 Dye 3 ice core*

Year in sample AD	Weight of sample (kg)	Specific activity (dph kg^{-1})	Accumulated rate (cm of ice equivalent yr^{-1})	Initial rate of deposition (dph cm^{-2} yr^{-1})
1975–71	8.16	62.8 ± 1.0*	66.3	4.75 ± 0.08
1970–66	7.60	56.3 ± 1.5*	49.4	3.71 ± 0.10
1965–61	7.97	53.1 ± 1.3*	49.8	4.12 ± 0.10
1960–56	10.13	44.3 ± 1.1*	65.2	5.25 ± 0.13
1955–51	8.63	40.9 ± 1.0*	52.6	4.57 ± 0.11
1950–46	9.74	40.5 ± 0.8*	65.2	6.56 ± 0.14
1945–41	6.45	23.8 ± 0.6*	66.6	4.59 ± 0.12
1940–36	7.69	30.1 ± 1.0*	66.6	6.80 ± 0.23
1935–31	4.66	25.2 ± 1.0*	60.3	6.02 ± 0.23
1930–26	4.78	24.8 ± 0.8	65.2	7.47 ± 0.24
1925–21	4.03	19.8 ± 0.7	56.8	6.07 ± 0.21
1920–16	3.07	25.0 ± 1.1	59.6	9.40 ± 0.41
1915–11	3.27	19.7 ± 0.7	61.0	8.85 ± 0.31
1910–06	3.25	15.4 ± 1.8	54.7	7.25 ± 0.85
1905–01	3.17	15.8 ± 1.1	56.8	9.02 ± 0.63
1900–96	3.50	15.8 ± 0.8	66.6	12.36 ± 0.63
1895–91	2.13	16.4 ± 1.0	53.3	11.99 ± 0.73
1890–86	3.18	8.1 ± 0.7	59.6	7.74 ± 0.67

* Calculated values, based on the figures in Table 2.

3.2. Specific total β activity

Total β activity was measured by 2π flow-type β counters with low background ($\approx$3 cph), (Clausen, 1973; Nijampurkar, 1974).

3.3. Specific lead-210 activity

^{210}Pb activity of the samples was extracted from melted ice samples at pH 4–5 acidified by nitric acid and with lead and bismuth carriers added in the form of lead nitrate ($\approx$80 mg) and bismuth nitrate ($\approx$40 mg). The samples were evaporated to near dryness, and bismuth was separated from lead by complexing Bi with pure Dalzine reagent in chloroform (Navratnam, 1974) according to the procedure given by Nijampurkar et al. (1982). The ^{210}Bi activity (half life 5.013 days) was measured on $BiPO_4$ in the flow-type β counters mentioned above. Several blank samples were run through the complete procedure and they did not show any ^{210}Pb activity. Duplicate measurements carried out on a few ice samples were reproducible within 5%.

4. Results and discussion

Dating of the ice layers along an ice core is an important parameter. The continuous measurements of δ^{18}O on the three ice cores demonstrated the typical seasonal δ^{18}O variation and provided a time scale for the ice cores. The total β records provide the time scale with fixed points, like 1955 and 1963 due to the atmospheric thermonuclear test series in 1950s and 1960s, respectively. Furthermore, continuous acidity measurements provide the time scale with fixed points, because major volcanic eruptions like Hekla 1947, Katmai 1912 and Krakatoa 1883 mark the Greenland ice with an elevated level of acidity due to the acid gases released to the atmosphere by the volcanic eruptions. Combination of δ^{18}O seasonal variations with the fixed points from total β and acidity measurements provide the ice cores with an absolute time scale (±1 year) for the years in discussion (Clausen et al., 1988; Clausen and Hammer, 1988). A hundred-year-old δ^{18}O record from Dye 3 is

shown in Fig. 2, whereas a 45-year $\delta^{18}O$ record along with a 23-year record of total β activity are shown in the period 1931–1975 in Fig. 3a. The observed ^{210}Pb activities for the Dye 3 core for the past 100 years vary from 8–80 dph kg^{-1} of ice (Tables 2, 3). The uncertainties of the specific activities are predominantly determined by the statistical uncertainties of the counting procedure; therefore the stated uncertainties in Tables 2, 3 correspond to $\pm 1\sigma$ of the counted values. The ^{210}Pb activities have been calculated using the net activities of ^{210}Pb (i.e., by subtracting the systematic blank contributions and background from the total observed activity of ^{210}Bi at $t = 0$), and the probable influence of blank contributions have been carefully considered. Secondly, the typical concentration of dust in the Dye 3 core ranges from 20–200 $\mu g\ kg^{-1}$ of ice and the ^{226}Ra contributions from the trapped dust is less than 0.5%, particularly in the older samples (assuming the U concentration in the Earth crust to be 2 dpm g^{-1}, the concentration of ^{226}Ra is 2.4×10^{-2} dph kg^{-1} of ice). The yearly, bi-yearly and five-yearly means of the observed ^{210}Pb activities have been plotted against time in Fig. 4. The extrapolated regression line drawn through the observed scattered ^{210}Pb data shown in Fig. 4, shows an initial ^{210}Pb activity of 74 dph kg^{-1} of ice for the year 1980. The theoretical decay curve of ^{210}Pb based on this activity, shown by the dotted line, has a slope different from that of the observed natural decay curve (solid line). This suggests that the specific activity of ^{210}Pb in Dye 3 precipitation has not remained constant back in time for a period of 100 years. Using these activities, corrected for radioactive decay since deposition, and respective rates of accumulation (in cm of ice equivalent per year, ice density 918 kg m^3, Fig. 5a) for these years, the calculated initial fallout of ^{210}Pb (dph cm^{-2} yr^{-1}) has been plotted as a function of time (Fig. 5b). We have used the half life of ^{210}Pb to be 22.3 years and the net ^{210}Bi activity for calculating the initial fallout of ^{210}Pb. The errors in the calculation of the initial specific activity measurements (dph kg^{-1} of ice) are generally less than 7% and the uncertainty of the ^{210}Pb fallout estimates even in the oldest samples are insignificant.

Though one expects seasonal changes in the relative amounts of winter and summer precipi-

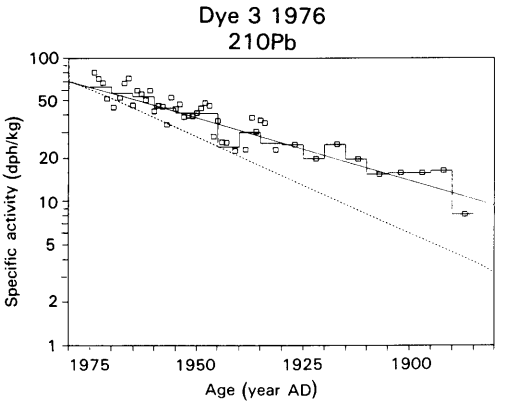

Fig. 4. Specific ^{210}Pb activity (dph kg^{-1} of ice) in the Dye 3 ice core for the period 1886–1975. The horizontal step curve shows 5-year mean values. The regression line (solid line) is based on the 5-year means. The slope of this line deviates from that of the ^{210}Pb decay (dotted line), see text.

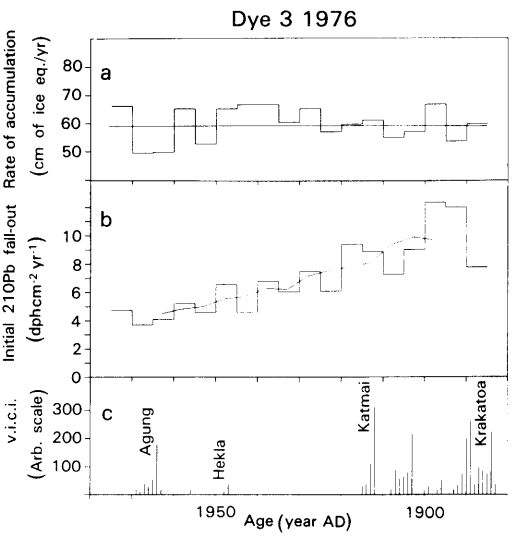

Fig. 5. (a) 5-year means of the annual rate of accumulation at Dye 3 (cm of ice equivalent per year) for the period 1886–1975. (b) The step curve represents 5-year means of the annual initial ^{210}Pb fallout (dph cm^{-2} yr^{-1}) for the period 1886–1975. The smooth curve represents 25-year running means of the initial ^{210}Pb fallout. (c) The volcanic impurity concentration index (v.i.c.i.) has been plotted on an arbitrary scale for the period 1886–1975. The names of volcanic eruptions like Katmai, Hekla etc. are placed to the left of the v.i.c.i. signal.

tation as well as an annual cycle of local atmospheric ^{210}Pb activity, our discussions in the present work are restricted only to the total fallout in the annual deposition of ice. It may be worthwhile to study in detail the seasonal variations of ^{210}Pb deposition rates over a period of a few decades to see whether cyclic variations in the annual deposition exist in comparison with the seasonally trapped dust and $\delta^{18}O$; however, this has not been a part of this study. Fig. 5c presents the volcanic impurity concentration index (v.i.c.i.) on an arbitrary scale, (Hammer et al., 1981). This index is derived from Lamb's volcanic dust production index (v.d.p.i., Lamb, 1970) by correction for different latitudes of individual eruptions and the residence time of aerosols in high latitude stratosphere. Fig. 6 and Tables 2, 4a and 4b present the distribution of

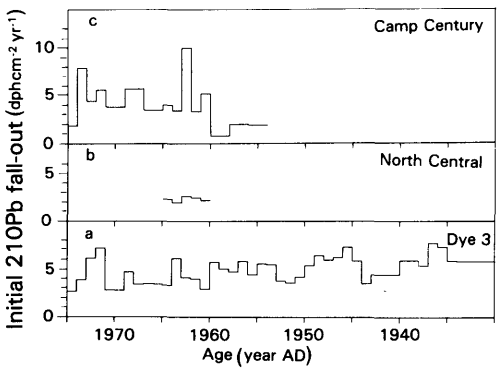

Fig. 6. The yearly initial ^{210}Pb fallout (dph cm^{-2} yr^{-1}) is shown for the ice cores: Dye 3 (1931–1975) (a); North Central (1961–1965) (b); Camp Century (1955–1975) (c).

Table 4a. *Observed annual specific activity and calculated initial deposition rate of Pb-210 for the period 1955–1975 in the 1977 Camp Century ice core*

Year in sample AD	Weight of sample (kg)	Specific activity (dph kg^{-1})	Accumulated rate (cm of ice equivalent yr^{-1})	Initial rate of deposition (dph cm^{-2} yr^{-1})
1975	0.05	160 ± 33	10.9	1.9 ± 0.4
1974	0.22	182 ± 8	39.4	7.9 ± 0.3
1973	0.20	118 ± 10	32.8	4.4 ± 0.4
1972	0.23	121 ± 14	39.4	5.6 ± 0.6
1971–70	0.42	91 ± 6	33.9	3.8 ± 0.3
1969–68	0.57	95 ± 4	45.9	5.7 ± 0.2
1967–66	0.39	84 ± 3	29.5	3.5 ± 0.1
1965	0.20	79 ± 11	35.0	4.0 ± 0.6
1964	0.23	61 ± 8	37.2	3.4 ± 0.4
1963	0.30	147 ± 7	43.8	10.0 ± 0.5
1962	0.32	63 ± 6	32.8	3.3 ± 0.3
1961	0.38	80 ± 7	39.4	5.2 ± 0.5
1960–59	0.37	23 ± 3	19.7	0.8 ± 0.1
1958–57	0.69	31 ± 3	35.0	2.0 ± 0.2
1956–55	0.49	25 ± 3	38.3	1.9 ± 0.2

Table 4b. *Observed annual specific activity and calculated initial deposition rate of Pb-210 for the period 1961–1965 in the 1977 North Central ice core*

Year in sample AD	Weight of sample (kg)	Specific activity (dph kg^{-1})	Accumulated rate (cm of ice equivalent yr^{-1})	Initial rate of deposition (dph cm^{-2} yr^{-1})
1965	0.20	98 ± 9	16.2	2.3 ± 0.2
1964	0.21	85 ± 10	15.3	2.0 ± 0.2
1963	0.22	126 ± 9	13.5	2.6 ± 0.2
1962	0.18	132 ± 13	11.7	2.5 ± 0.2
1961	0.23	92 ± 9	14.4	2.2 ± 0.2

initial ^{210}Pb fallout for 3 locations in Greenland during the time period of atmospheric thermonuclear bomb tests. The spaced sites and time periods in the records are Dye 3 (1931–1975), North Central (1961–1965) and Camp Century (1955–1975). The values of the mean fallout at Dye 3, North Central and Camp Century are 4.5, 2.3 and 3.9 dph cm^{-2} yr^{-1}, respectively, with a mean of 3.6 dph cm^{-2} yr^{-1} for Greenland. The mean initial ^{210}Pb activity for the period 1961–1965 in the surface samples of three ice cores from Camp Century, North Central and Dye 3, in the present work (2.1, 2.7 and 1.4 dpm kg^{-1} of ice, respectively) is very similar to that observed by Crozaz et al. (1966) (3 dpm kg^{-1} of ice from Camp Century) and Goldberg (1963) (1.3 dpm kg^{-1} of ice from Crete in Central Greenland). Sanak and Lambert (1977) conducted similar studies at the South Pole and observed that the fallout of ^{210}Pb for a period about 100 years varied from 1.0 to 2.5 dph kg^{-1} of ice with an observed value of 1.8 dpm kg^{-1} for the period 1961–1965. Their findings indicated that changes (either change in accumulation rate or ^{210}Pb fallout or both) occurred near the South Pole around 1920 and 1954. Surprisingly, the ^{210}Pb fallout on Greenland (from the present work) also shows changes in the fallout of ^{210}Pb around the same time. Gäggeler et al. (1978) have estimated a uniform accumulation rate of 35 cm w. eq. yr^{-1} using the ^{210}Pb (^{210}Po) method for dating of cold alpine ice cores, which cover a time span of 90 years. The surface activity of ^{210}Pb concentration for the year 1976–77 was observed to be 4 dpm kg^{-1} of ice. On Himalayan glaciers, Nijampurkar et al. (1982) observed the ^{210}Pb fallout, which varied from 3 to 8 dpm kg^{-1} of ice. The higher fallout in the lower latitude compared to polar regions is expected on the basis of more exposure of the Earth's crust in the lower latitudes than the polar regions.

The observations which emerge from our figures are now summarized. (i) The rate of accumulation at Dye 3 has remained fairly constant over the past century with a mean value of 59.5 cm of ice equivalent per year. (ii) The fallout of ^{210}Pb has not remained constant during the last century, and the observed fallout over the period 1886–1920 is significantly higher, at least by a factor of 1.8, than that observed in the period 1920–1975. The observed higher ^{210}Pb fallout during 1886–1920 coincides with the higher volcanic activity in this period compared to the volcano quiet period 1920–1975 with only Hekla, 1947 and Agung-Agung 1963 as main contributors to volcanism in the northern hemisphere. In geochronological studies, it is usually assumed that the fallout of ^{210}Pb has remained constant during the period of investigation. This assumption does not hold for the last 100 years in South Greenland, where a steady long-term decreasing trend exists since the end of the last century. This trend can hardly be explained by a changing rate of annual precipitation in South Greenland, as this rate shows no systematic trend in the past 100 years. The coincidence of high ^{210}Pb deposition and increased volcanic activity in the oldest part of the records is hardly explained by volcanism being a source for the observed "^{210}Pb excess", because the slowly decreasing trend of the ^{210}Pb deposition in the volcano quiet period 1920–1975 is in contradiction to the residence time of about one year for ^{210}Pb in the stratosphere, even though we consider ^{210}Pb and/or its mother nuclei brought into the stratosphere by violent volcanic eruptions. We find it more reasonable to connect the observed decreasing ^{210}Pb fallout trend with changes in the main source of ^{210}Pb (^{222}Rn exhalation from lands), which depends primarily on the nature of the soil (porosity, grain size, moisture content etc.) and environmental conditions (wind pattern, pressure, temperature etc.). ^{222}Rn from oceans is two order of magnitude lower than from land due to the low concentration of ^{226}Ra in seawater. (iii) The ^{210}Pb activities do not correlate with the high total β activity in the periods of atmospheric thermonuclear bomb tests and there is no elevated level in the ^{210}Pb fallout in 1962–1965 (Fig. 6).

The three records do not intercorrelate and especially in the Dye 3 and Camp Century records, the year-to-year values often vary more than a factor of two. Thus, the 1963 value in the Camp Century record appears to be just an oscillation in the natural ^{210}Pb fallout pattern. Therefore, the marginal excess observed in 1963 in the Camp Century record can hardly be explained by a contribution from the atmospheric thermonuclear bomb tests in the 1960s, and we exclude artificially produced ^{210}Pb from being present in Greenland ice cores.

5. Conclusion

A continuous record of ^{210}Pb fallout in Greenland reveals a steady decreasing trend in the last 100 years with a factor of 2 more deposited yearly at the start of our century than today. If this trend is valid on a global scale, it has an important impact on geochronological studies based on ^{210}Pb. In Greenland, we do not find evidence for any "excess" contribution of ^{210}Pb production either by violent volcanic eruptions or by atmospheric thermonuclear bomb tests.

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