The accumulation of ²¹⁰Pb at Summit, Greenland since 1855

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ABSTRACT

Detailed (202 samples) profiles of total beta and ²¹⁰Pb activity were determined from a 32 m firn core collected at the GISP2 site (72°20′ N, 38°45′ W) near Summit, Greenland in 1989. The beta radioactivity profile verifies year by year dating based on recognition of annual hoar layers back to 1955, and lends a high degree of confidence to the 74 year age assigned to the bottom of the core by this stratigraphic dating technique. The decay corrected activity of ²¹⁰Pb at the time of deposition shows considerable short term variability, but no clear seasonal or annual periodicity. ²¹⁰Pb activity in surface snow at this site has averaged 0.7 pCi kg⁻¹ since 1927, but the period 1915–1927 is characterized by a steady decline from higher levels. The average annual accumulation of ²¹⁰Pb has markedly declined since at least 1870. Similar observations at Dye 3 suggest that ²¹⁰Pb accumulation has decreased throughout this century over much of the Greenland Ice Sheet. If the records of ²¹⁰Pb in the firn on the Greenland Ice Sheet are mainly reflecting the northern hemisphere atmospheric burden of ²¹⁰Pb, these results will demand careful reassessment of ²¹⁰Pb-based radiochronologies. However, our current lack of understanding of the linkages between atmospheric and snow chemistry makes the widespread applicability of these findings an open question.

1. Introduction

The depth profiles of anthropogenic radionuclides from atomic bomb testing (e.g., Picciotto and Wilgain, 1963; Lorius et al., 1968) and the natural ²³⁸U series ²¹⁰Pb (e.g., Crozaz et al., 1964; Crozaz and Langway, 1966) have long been used to establish chronologies in polar firn and ice that have accumulated over the past several decades. In addition, these radionuclides may provide information about changes in atmospheric circulation and depositional processes over the same time scale (Lambert et al., 1966; Sanak and Lambert, 1977; Monaghan and Holdsworth, 1990; Nijampurkar and Clausen, 1990).

Very detailed profiles of total beta radioactivity (predominantly from the fallout isotopes 90 Sr and 137 Cs) and 210 Pb activity were determined from the 32 m A core recovered at the Greenland Ice Sheet Project Two (GISP2) site in 1989 (Fig. 1). These records provide an accurate chronology (± 1 year) for the past 74 years at this site, and help to

corroborate the independent dating techniques being applied to the upper part of the GISP2 deep core. Perhaps more importantly, the time series of annual ²¹⁰Pb accumulation at the GISP2 site since 1855 displays some interesting trends that may have important implications regarding the presumed constant flux of ²¹⁰Pb that underlies its use as a dating technique.

2. Methods

Core was collected by the Polar Ice Coring Office of the University of Alaska, Fairbanks, with a 5.2" electromechanical drill in a dry hole. One meter sections were slabbed longitudinally to prepare a surface for electroconductivity measurements in the field science trench. The small slab removed was sectioned for stable isotope studies. The remainder of the core was logged for visible stratigraphic features, placed in polyethylene sleeves and then into cardboard

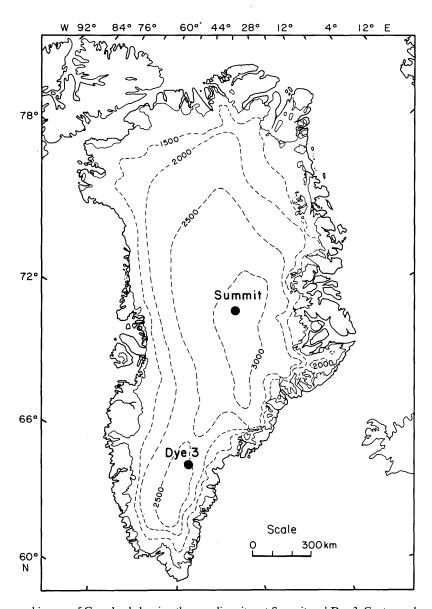


Fig. 1. Topographic map of Greenland showing the sampling sites at Summit and Dye 3. Contours show elevation (m) above sea level.

"core tubes". All of the core collected during the 1989 season was returned to UNH frozen for subsequent processing in dedicated freezers.

The A core was sectioned into 205 samples. In order to obtain a high resolution beta radioactivity record during the main fallout era (1953 to about 1975) a 10 cm interval was chosen for the 7–16 m

section of the core. The rest of the core was cut into 20 cm samples. After the analysis of the A core samples it was decided to extend the $^{210}{\rm Pb}$ record further back in time. Annual increments of accumulated snow, determined on the basis of stratigraphic observations supported by $\delta^{18}{\rm O}$ oscillations, were cut from the 30–50 m depth

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interval of the C core, also collected in 1989 within 30 m of the A core. (The 43–44 m section of the C core had previously been dedicated to other analyses.) Each section was double bagged in polyethylene and kept at or below -20° C until preparation for analysis.

The details of sample preparation are given in Dibb (1990). Briefly, the samples are spiked with ²⁰⁸Po and melted, then the radionuclides are concentrated onto cation exchange filters (Delmas and Pourchet, 1977) for determination of beta activity in a gas flow proportional counter. ²⁰⁸Po and ²¹⁰Po are subsequently leached from the filters with concentrated HCl and plated onto polished silver planchets for alpha spectrometric determination of ²¹⁰Pb activities (Crozaz and Fabri, 1966; Flynn, 1968). All of the planchets were counted long enough to reduce counting uncertainties below 15%: for most of the A core (175 out of 202 samples), and all of the C core samples, uncertainties range from 5-10%. Three of the samples were lost from leaking plastic bags.

3. Results

Unequivocal dating of 11 horizons is provided by the detailed beta activity profile (Fig. 2). The relatively small peak due to Chernobyl fallout in 1986 was confirmed by gamma spectrometric identification of ¹³⁷Cs and ¹³⁴Cs in this sample. The prominent peaks between 11.5 and 16.5 m reflect the well established history of radioactive fallout in the northern hemisphere and can be matched, peak for peak, with the independently dated beta profile from a core collected near Summit in 1974 (Claussen and Hammer, 1988). Annual layers identified by stratigraphic features are in perfect agreement with the beta horizon dating back to 1954. The stratigraphic dating indicates an age of 74 years at the bottom of the A core. (Note: annual oscillations of δ^{18} O in the B core, collected within 30 m of the A core, indicate an age of 74 or 75 years at 32 m depth.)

Average measured ²¹⁰Pb activities for 5-year intervals ranged from 0.02 pCi kg⁻¹ at 50 m to

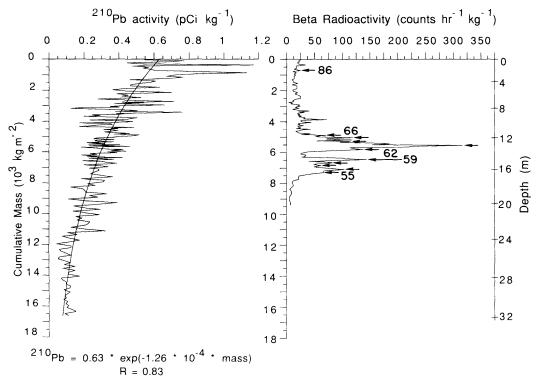


Fig. 2. Detailed profiles of ²¹⁰Pb and beta radioactivity against cumulative mass and depth. The curve on the ²¹⁰Pb profile is a least squares fit to an exponential decay function.

Table 1. Nominal 5-year averages of snow accumulation rate, measured, and initial, activity of Pb-210 at the GISP2 site near Summit, Greenland

Interval	Snow Accumulation (kg m ⁻² a ⁻¹)	Measured Activity (pCi ²¹⁰ Pb kg ⁻²)	Initial Activity (pCi ²¹⁰ Pb kg ⁻²)
1988-1986	192 ± 25	0.616 ± 0.168	0.633 ± 0.168
1985-1981	229 ± 37	0.672 ± 0.148	0.779 ± 0.134
1980-1976	187 ± 39	0.424 ± 0.124	0.574 ± 0.153
1971-1975	223 ± 24	0.408 ± 0.125	0.647 ± 0.192
1970-1966	201 ± 42	0.360 ± 0.052	0.668 ± 0.074
1965-1961	241 ± 52	0.334 ± 0.059	0.725 ± 0.118
1960-1956	219 ± 22	0.297 ± 0.040	0.754 ± 0.095
1955–1951	233 ± 37	0.274 ± 0.030	0.812 ± 0.076
1950–1946	213 ± 82	0.215 ± 0.042	0.750 ± 0.167
1945-1941	242 ± 74	0.211 ± 0.061	0.849 ± 0.207
1940-1936	225 ± 25	0.173 ± 0.069	0.811 ± 0.303
1935–1931	254 ± 68	0.103 ± 0.036	0.568 ± 0.176
1930-1926	211 ± 71	0.101 ± 0.028	0.659 ± 0.194
1925-1921	240 ± 51	0.105 ± 0.011	0.795 ± 0.091
1920–1916	202 ± 59	0.105 ± 0.008	0.923 ± 0.108
1915–1911	239 ± 36	0.082 ± 0.021	0.839 ± 0.200
1910–1906	211 ± 44	0.079 ± 0.020	0.943 ± 0.223
1905-1901	176 ± 39	0.062 ± 0.009	0.867 ± 0.150
1900-1896	229 ± 15	0.066 ± 0.012	1.084 ± 0.200
1895–1891	222 ± 41	0.053 ± 0.010	1.013 ± 0.181
1890-1886	244 ± 24	0.049 ± 0.012	1.106 ± 0.288
1885–1881	220 ± 58	0.047 ± 0.012	1.211 ± 0.305
1880-1879	200 ± 14	0.039 ± 0.001	1.123 ± 0.005
1874–1871	284 <u>+</u> 99	0.036 ± 0.010	1.311 ± 0.346
1870–1866	169 ± 56	0.030 ± 0.007	1.233 ± 0.256
1865-1861	176 ± 64	0.023 ± 0.003	1.101 ± 0.141
1860-1856	203 ± 63	0.021 ± 0.003	1.186 ± 0.130
1855	277	0.015 ± 0.001	0.906 ± 0.083

Pb-210 values back through 1919 are from subannual sampling of the 32 m long A core, those from 1855–1914 are annual samples from the C core, and the 1915–1918 period represents the average of duplicates from the two cores. Note there is no data for 1875–1878, as the 43–44 m interval of the C core had already been used for other analyses.

almost 0.7 pCi kg⁻¹ near the surface (Table 1). Subannual sampling in the 32 m long A core revealed an even greater range in ²¹⁰Pb activity, but the activity does decrease exponentially with increasing cumulative mass (Fig. 2). An exponential least squares fit of these data (Fig. 2) yields an average accumulation rate of 245 kg m⁻² a⁻¹. An age of 67 years at 32 m depth can then be estimated from the cumulative mass and accumulation rate.

4. Discussion

The 7-year discrepancy between the ²¹⁰Pb dating and that provided by beta radioactivity,

physical stratigraphy, and δ^{18} O, despite the exponential decay of 210 Pb with increasing cumulative mass, merits further consideration. It is apparent from Fig. 2 and other recent work on the Greenland ice sheet (Dibb, 1990) that the assumption of constant activity of 210 Pb in surface snow is not strictly justified for samples representing time spans less than a year. However, the success of the technique in previous investigations on both polar ice sheets (e.g., Crozaz and Langway, 1966; Crozaz et al., 1964) suggests that over longer time periods the high frequency variability of surficial 210 Pb is averaged out. Independent dating of this core allows back calculation of the surface 210 Pb activity for the past 74 years

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(Fig. 3). Although there is short term variability throughout the record, it seems to vary around a mean value of about 0.7 pCi kg⁻¹ to a depth of almost 28 m (1927). Below this depth the decay corrected ²¹⁰Pb activity increases steadily to the bottom of the core. An average accumulation rate of 225 kg m⁻² a⁻¹ can be calculated from the ²¹⁰Pb profile in the top 27.8 m of the core, in very good agreement with the rate of 223 kg m⁻² a⁻¹ based on an age of 74 years at 32 m.

The decay corrected ²¹⁰Pb profile shows no clear seasonal or annual signal, when compared to the annual signal recorded in the visible stratigraphy. The timing of the most prominent spikes in the profile of decay corrected ²¹⁰Pb activity (Fig. 3) all seem to coincide with documented volcanic eruptions that were recorded in the firn near Dye 3 (Lyons et al., 1990). It is tempting to interpret the ²¹⁰Pb spikes as additional signals of these eruptions. However, much of the variability in the decay corrected ²¹⁰Pb activity may also reflect: variable dilution of a constant flux of ²¹⁰Pb by changes in the flux of snow, some change in the timing of ²¹⁰Pb delivery to the site such that the snow containing elevated levels of ²¹⁰Pb is not retained in the snow pack (perhaps being removed by wind) in some years, a change in meso- or largescale atmospheric transport and depositional processes resulting in different amounts of ²¹⁰Pb being delivered to the Summit region, or more likely, some combination of these and other unknown factors. The complexity of this question certainly limits the amount of faith that can be placed in any simplistic interpretation of the apparently dynamic ²¹⁰Pb record preserved in this core.

In an effort to highlight trends while reducing the presently unexplainable "noise" contained in the ²¹⁰Pb concentration vs cumulative mass profile, annual values of ²¹⁰Pb accumulation were calculated. The bottom of the A core (1920-1915) showed a marked increase in the annual accumulation of ²¹⁰Pb. A recent study on a core collected at Dye 3 found the average annual accumulation of ²¹⁰Pb during the period 1886–1930 to be nearly two times higher than it was from 1931-1975 (Nijampurkar and Clausen, 1990). In order to confirm the trend hinted at in the bottom of the A core, and to determine whether the change in ²¹⁰Pb accumulation at Dye 3 was a local phenomenon, the ²¹⁰Pb record at Summit was extended back to 1855 by cutting annual increments from the 30-50 m interval of the adjacent C core.

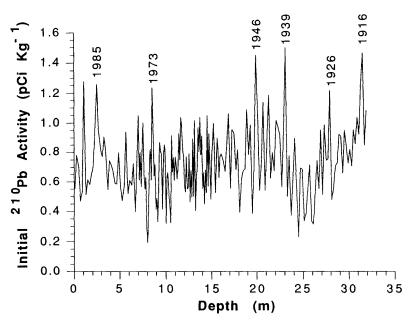


Fig. 3. ²¹⁰Pb concentrations backcalculated to the time of deposition using independent dating based on physical stratigraphy and beta activity reference horizons.

The two meters of overlap between the A and C cores contained four complete years (1915–1918) in common. Agreement between the annual accumulation of snow and ²¹⁰Pb in the two cores was quite poor for these four years (the difference between the cores averaged 70% of the mean for ²¹⁰Pb and 50% of the mean for snow), demonstrating the importance of small scale spatial heterogeneity in snow chemistry in the region. The average of the two cores was used for the four year period in all of the following, but the observed variability between the two cores suggests that little weight be given to any single year of the record, and even less to the "peaks" in the 210Pb concentration profiles representing only one or two samples (Fig. 3).

Spectral analysis of the detrended time series of ²¹⁰Pb and snow accumulation at Summit indicates weak periodicity near 2–3 years for both snow and ²¹⁰Pb, with perhaps an additional 20 year period present in the snow data and some periodicity near 6–7 years and 12–13 years for ²¹⁰Pb. However, the most striking feature of these time series is the coherence between the accumulation of ²¹⁰Pb and snow (Fig. 4). Cross correlation between the raw time series shows a dominant peak at zero lag (correlation = 0.60), indicating that the main factor determining short-term variability of ²¹⁰Pb accumulation near Summit is the variability in

snow accumulation. (When the linear trend in ²¹⁰Pb accumulation is removed the correlation increases to 0.70.)

The snow accumulation time series at Summit (smoothed by a 3 point moving average) varies between about 200-250 kg m⁻² a⁻¹, but shows no long-term trends (Fig. 4). In contrast, the smoothed annual accumulation of 210Pb has declined fairly steadily since about 1870 (Fig. 4). These results agree with the significant decrease in ²¹⁰Pb accumulation at Dye 3 (Nijampurkar and Clausen, 1990), and show that at Summit the decrease began at least 15 years earlier than the end of the Dye 3 record in 1886 (Table 2). The 2.3 times higher average annual accumulation of ²¹⁰Pb at Dye 3 from 1931-1975 might appear to simply reflect the 2.4 times higher rate of snow accumulation there during the period. However, the 1886-1930 period shows ²¹⁰Pb accumulating 3.3 times faster at Dye 3 than Summit while the ratio of snow accumulation rates stayed essentially constant at 2.5 (Table 2). Recent (1986–1987) accumulation of 210Pb in a snowpit 40 km southwest of Dye 3 was only about 1.5 times higher than at Summit for the same years, while the snow accumulation rate was 2.1 times higher at Dye 3 (Table 2). Similarly, the average annual accumulation of ²¹⁰Pb at North Central during 1961-1965 was identical to the average at the

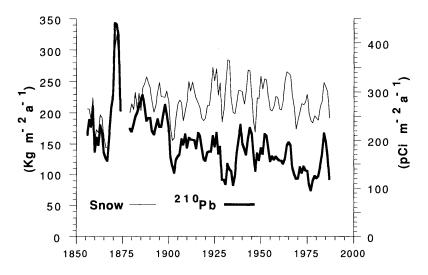


Fig. 4. Annual accumulation of snow and ²¹⁰Pb at the GISP2 site, smoothed by a 3 year moving average.

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Table 2. Average values of ²¹⁰Pb and snow accumulation at Summit and two other locations in Greenland

Interval	Snow (kg m ⁻² a ⁻¹)	²¹⁰ Pb (pCi m ⁻² a ⁻¹)	Snow $(kg m^{-2} a^{-1})$	²¹⁰ Pb (pCi m ⁻² a ⁻¹)	Notes
	Summit		Dye 3		
1988–1976	204	138			
1975-1931	228	165	553	387	1
1930-1886	219	200	544	669	1
1885-1855	209	250			
1987–1986	206	111	434	138-182	2
			North Central		
1965-1961	241	174	130	174	3

¹ Record for Dye 3 from Nijampurkar and Clausen, 1990.

GISP2 site, despite snow accumulating 1.9 times faster at Summit during these five years (Table 2).

The strong correlation between ²¹⁰Pb and snow accumulation, over the short-term at a single site, indicates the dominance of wet deposition, mainly as snow, in the delivery of ²¹⁰Pb to the Greenland Ice Sheet. Over longer time scales, and when more than one site is considered, the relationship between the accumulation of snow and ²¹⁰Pb is obviously quite complex, as demonstrated by the comparisons in Table 2. It is clear that considerably less ²¹⁰Pb has been accumulating over a large region of the Greenland Ice Sheet in the recent past than was the case around the turn of the century. However, the causes of this change can not yet be resolved. Possible contributors include; decreased ²²²Rn emanations from source regions, changes in transport pathways to the ice sheet resulting in different source regions or increased time in transport from continental areas, more efficient removal of 210Pb and its short-lived precursors during transport, decreased efficiency of ²¹⁰Pb scavenging over the ice sheet, or, likely, some combination of these and other factors. Much more study of the quantitative linkage between atmospheric and snow chemistry, like the ongoing atmospheric sampling component of GISP2, is required to allow sound interpretation of snow chemistry records in terms of atmospheric processes.

5. Conclusions

The combination of beta radioactivity horizons and stratigraphic features have allowed very accurate dating of the top 32 m of firn at the GISP2 site near Summit, Greenland. Agreement between these two dating techniques in the A core and the counting of δ^{18} O oscillations in the adjacent B core lends confidence to the interpretation of stratigraphic and stable isotope annual signals in at least the top several hundred meters of the deep GISP2 core.

²¹⁰Pb dating for the entire 32 m of the A core overestimates the average accumulation rate by 9% if constant surficial activity is assumed. Calculation of surficial activities at the time of deposition indicates that ²¹⁰Pb concentrations have varied around a constant value since about 1927, but from 1915 to 1927 they steadily declined from a higher level to the current average of 0.7 pCi kg⁻¹.

² Record for Dye 3 actually from a pit 40 km southwest of the station (Dibb, 1990).

³ North Central is at 74°37′N, 39°24′W, about 2.3 degrees north and almost 0.7 degrees west of the GISP2 site (Nijampurkar and Claussen, 1990).

The temporal intervals were selected to facilitate comparison to other results, and are not meant to suggest any natural breaks in the time series for Summit shown in Fig. 4.

The average annual accumulation of ²¹⁰Pb at the GISP2 site has declined fairly steadily since at least 1870. This corroborates the dramatic decrease in ²¹⁰Pb accumulation rate recently reported for a Dye 3 core (Nijampurkar and Claussen, 1990). The wide separation of these two sites argues against the possibility that this decrease is due to some local phenomenon. If it can be established that the ²¹⁰Pb records in the firn on the Greenland Ice Sheet are reflecting changes in the atmospheric burden of ²¹⁰Pb, rather than meso- to small-scale changes in atmospheric transport and/or depositional processes, the basic assumption of nearly constant ²¹⁰Pb flux, on which the radiochronologic technique is based, will have to be reassessed. However, our limited understanding of air-snow transfer functions do not yet allow us to unequivocally draw this conclusion.

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