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A 200-year ^{210}Pb record from Greenland

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Abstract. A continuous profile of ^{210}Pb activity extending back to 1766 has been developed for a firn/ice core collected at Site D in central Greenland in 1984. Unexpectedly high activities of ^{210}Pb were found at the base of this core (0.032 pCi kg^{-1} in samples more than 200 years old), calling into question the common assumption that supported ^{210}Pb can be neglected when constructing chronologies in glacial snow and ice. It is problematic to assert that all of the ^{210}Pb measured at depth should be attributed to the supported fraction, given previous estimates of dust loading in Greenland ice cores. However, even if an estimated constant value of 0.032 pCi supported ^{210}Pb kg^{-1} is subtracted from the measured values to estimate excess ^{210}Pb, the ^{210}Pb chronology for Site D yields ages that are significantly younger (mean accumulation rate too high) than an independent depth-age scale based on annual layer counting. It is apparent that the flux of excess and/or supported ^{210}Pb to this site must have decreased over the past 2 centuries, with decreasing trends in both fractions most likely. Previously published ^{210}Pb profiles for cores from Summit and Dye 3, Greenland, show similar trends, which had been interpreted as decreasing fluxes of excess ^{210}Pb only. For all three sites, it is not possible to separate variations in the fluxes of the excess and supported fractions of ^{210}Pb, but variations in the total ^{210}Pb flux will impact ^{210}Pb-based chronologies generally if these variations have not been restricted to the Greenland ice sheet.

Introduction

We have separately reported the results of detailed ^{210}Pb analyses conducted on ice cores collected at Dye 3 and Summit, Greenland [*Nijampurkar and Clausen*, 1990; *Dibb*, 1992]. In both cases it appears that the amount of ^{210}Pb delivered to high-elevation sites on the Greenland ice sheet has not been constant, rather, a significant decline is suggested through much of this century. Since applications of ^{210}Pb for geochronological studies generally assume either a constant activity at the surface or a constant flux over the time period being investigated, these findings may indicate a systematic bias in ^{210}Pb-derived ages determined in sediments as well as ice cores in northern high latitudes.

All previous investigators examining ^{210}Pb depth distributions in glacial ice [e.g., *Crozaz et al.*, 1964; *Crozaz and Fabri*, 1966; *Crozaz and Langway*, 1966; *Sanak and Lambert*, 1977; *Gäggeler et al.*, 1983] have interpreted the depth profiles in terms of the constant initial concentration (CIC) model. This model is also commonly applied in sediment studies [e.g., *Robbins*, 1978; *McCall et al.*, 1984]. The fundamental assumption in this model, that temporal variations in ^{210}Pb flux are exactly balanced by parallel changes in the flux of diluent (snow or sediment) to maintain a constant specific activity at the surface, is rarely, if ever, completely valid. However, general agreement between ^{210}Pb-derived and independently determined ages in many studies suggests that temporal averaging inherent in natural systems like sediments and firn accommodates short-term variations in ^{210}Pb activity at the top of the accumulating pile. At Dye 3 and Summit, stratigraphic depth-age scales had already been established with annual resolution. The CIC model yielded systematically young ages at both sites, which *Nijampurkar and Clausen* [1990] and *Dibb* [1992] suggested were due to decreasing fluxes of excess ^{210}Pb. (The independent depth-age scales allowed estimation of the ^{210}Pb flux on a sample by sample basis.)

These two studies also followed the lead of all previous investigations of ^{210}Pb in snow and ice by assuming that all of the ^{210}Pb measured was unsupported or "excess". (Supported ^{210}Pb is that which is in secular equilibrium with ^{226}Ra, such that radioactive decay of ^{210}Pb is balanced by production through the decay of the grandparent. In some cases the ^{226}Ra may also be supported by ^{238}U if the system in question has remained closed for long enough. Unsupported or excess ^{210}Pb has been separated from the decay chain, primarily by the escape of the short-lived, noble gas, intermediary, ^{222}Rn. Excess ^{210}Pb activity will decrease with the characteristic half-life of ^{210}Pb (22.3 years), thus it is the excess fraction which is of interest for dating purposes.) Consideration of the low dust loading of Greenland ice suggests that this should be a reasonable assumption. The mass of insoluble particulates in Greenland firn and ice varies seasonally, with low concentrations near 20 μg (kg snow)$^{-1}$ and spring peaks as high as 300 μg kg^{-1} [*Steffensen*, 1988; *Nijampurkar and Clausen*, 1990]. If this dust is compositionally similar to the North American shale composite [*Gromet et al.*, 1984], it should contain 1.7 - 3.8 μg U (g dust)$^{-1}$ or 0.6 - 1.3 pCi supported ^{210}Pb (g dust)$^{-1}$ (assuming secular equilibrium).

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Taking the product of the top of the ranges estimated for dust in Greenland snow and supported ^{210}Pb in that dust yields a maximum estimate of 4 x 10^{-4} pCi supported ^{210}Pb (kg snow)$^{-1}$. This represents approximately 0.06% of the measured ^{210}Pb activity at the surface at Dye 3 and Summit. Even in the oldest samples from these two cores, this high estimate of supported ^{210}Pb would only account for 0.6 and 2.7% of the measured ^{210}Pb (the bottom samples were 89 and 134 years old at Dye 3 and Summit, respectively).

Although the ^{210}Pb records from Dye 3 and Summit both suggested decreasing fluxes of excess ^{210}Pb from the late 1800s to the 1970s, it was not possible to determine whether these were due to variations in the ^{210}Pb burden throughout the northern hemisphere troposphere, a unique aspect of the accumulation of snow and impurities at high elevations on the ice sheet, or an artifact of assuming that there was no supported ^{210}Pb. We have now determined ^{210}Pb activities in a third Greenland ice core, analyzing samples as old as 216 years (nearly 10 half-lives of ^{210}Pb). This record allows assessment of the supported ^{210}Pb question at this core site as well as the assumptions used to estimate accumulation rates and ages from excess ^{210}Pb depth profiles. The Dye 3 and Summit ^{210}Pb results are also revisited in light of this new record and the insights gained from it.

Methods

The core was collected at Site D (70°38'23"N, 39°37'04"W) in 1984; some of the results of earlier analyses conducted on the core are discussed by *Clausen et al.* [1988] and *Steffensen* [1988]. For the present purposes it is important to note that the site is between Summit and Dye 3 (about 300 km south of Summit and 500 km north of Dye 3) at an elevation of 3018 m. Site D is about 100 km west of and 120 m lower than the ice sheet divide. For reference, the Summit core was drilled 30 km west of, but only 30 m lower than, the true summit of the ice sheet. Dye 3 is approximately 100 km east of the divide on the southern dome of the Greenland ice sheet (Figure 1).

Year by year dating and an accumulation time series (corrected for densification and strain) were developed from detailed δ^{18}O and electrical conductivity measurements on the Site D core [*Clausen et al.*, 1988]. This depth-age scale was used to cut annual increments (from quarter or half core sections remaining in the archives) for ^{210}Pb analyses. These annual samples were shipped frozen from Copenhagen to New Hampshire in 1993. At New Hampshire, each sample was manually scraped (in a cold room at -15 C) to remove the outer several millimeters, then 3-6 annual layers were combined to

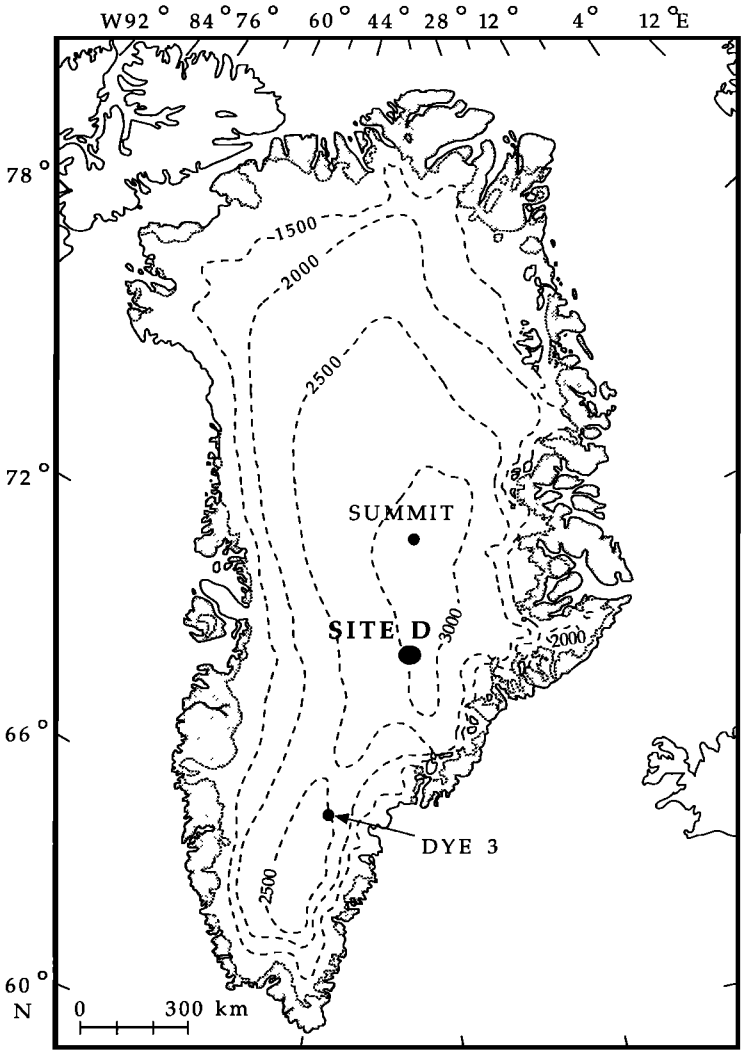

Figure 1. Map of Greenland showing the ice coring sites D, Summit and Dye 3. Contours are in meters above sealevel.

Table 1. Measured Values of ^{210}Pb Activity in the Firn and Ice at Site D, Greenland

Sample Interval	Mean Accumulation m ice yr^{-1}	Activity of ^{210}Pb pCi kg^{-1}
1984 - 1981	0.316	0.901 +/- 0.062
1981 - 1978	0.360	0.619 +/- 0.038
1978 - 1975	0.360	0.477 +/- 0.033
1975 - 1972	0.370	0.463 +/- 0.029
1972 - 1969	0.323	0.538 +/- 0.031
1969 - 1966	0.347	0.490 +/- 0.032
1966 - 1963	0.420	0.300 +/- 0.016
1963 - 1960	0.310	0.366 +/- 0.020
1960 - 1957	0.383	0.386 +/- 0.025
1957 - 1954	0.407	0.348 +/- 0.025
1954 - 1951	0.360	0.329 +/- 0.022
1951 - 1948	0.333	0.340 +/- 0.022
1948 - 1945	0.407	0.358 +/- 0.025
1945 - 1942	0.370	0.376 +/- 0.024
1942 - 1939	0.407	0.331 +/- 0.025
1939 - 1936	0.360	0.307 +/- 0.021
1936 - 1933	0.370	0.424 +/- 0.027
1933 - 1930	0.407	0.210 +/- 0.012
1930 - 1927	0.310	0.290 +/- 0.018
1927 - 1924	0.300	0.274 +/- 0.018
1924 - 1921	0.323	0.352 +/- 0.023
1921 - 1918	0.347	0.245 +/- 0.016
1918 - 1915	0.370	0.240 +/- 0.014
1915 - 1912	0.360	0.224 +/- 0.014
1912 - 1908	0.378	0.248 +/- 0.015
1908 - 1904	0.315	0.309 +/- 0.019
1904 - 1900	0.395	0.249 +/- 0.016
1900 - 1896	0.350	0.167 +/- 0.013
1896 - 1892	0.378	0.228 +/- 0.017
1892 - 1888	0.403	0.139 +/- 0.008
1888 - 1884	0.368	0.158 +/- 0.008
1884 - 1880	0.323	0.103 +/- 0.006
1880 - 1876	0.378	0.171 +/- 0.012
1876 - 1872	0.368	0.103 +/- 0.007
1872 - 1868	0.358	0.091 +/- 0.006
1868 - 1864	0.350	0.075 +/- 0.005
1864 - 1860	0.333	0.105 +/- 0.006
1860 - 1856	0.395	0.103 +/- 0.006
1856 - 1852	0.395	0.105 +/- 0.006
1852 - 1848	0.358	0.103 +/- 0.007
1848 - 1843	0.338	0.096 +/- 0.007
1843 - 1838	0.344	0.095 +/- 0.006
1838 - 1833	0.338	0.083 +/- 0.006
1833 - 1828	0.344	0.055 +/- 0.004
1828 - 1823	0.366	0.052 +/- 0.004
1823 - 1818	0.358	0.048 +/- 0.004
1818 - 1813	0.338	0.106 +/- 0.007
1813 - 1808	0.366	0.072 +/- 0.005
1808 - 1803	0.374	0.054 +/- 0.003
1803 - 1798	0.344	0.062 +/- 0.005
1798 - 1793	0.358	0.070 +/- 0.007
1793 - 1788	0.322	0.038 +/- 0.003
1788 - 1783	0.402	0.033 +/- 0.002
1783 - 1778	0.308	0.041 +/- 0.002
1778 - 1772	0.353	0.032 +/- 0.002
1772 - 1766	0.340	0.035 +/- 0.002

yield samples with masses between 1 and 3 kg (sample mass increased with age). Samples were spiked with ^{208}Po while still frozen. After melting, ^{208}Po and ^{210}Po were concentrated onto cation exchange resin, extracted in HCl, plated onto Ag planchets, and then analyzed in an alpha spectrometer (see *Dibb* [1990, 1992] for details). All of the samples were counted long enough to reduce the 1 σ uncertainty due to counting statistics to < 10% of the sample activity. Several distilled water blanks were carried through all steps of the procedure, these blanks produced < 1 count day^{-1} in the regions of the two Po peaks in the alpha spectrometer. Distilled water spiked with ^{208}Po gave responses for ^{208}Po similar to the melted ice core samples and contributed no significant counts in the ^{210}Po window (still < 1 count day^{-1} for counting periods as long as 12 days).

Results

Lead 210 was detected in all samples, including those more than 200 years old (Table 1). The activity profile of ^{210}Pb is well described by an exponential decrease with increasing depth (Figure 2). This type of curve fit forms the basis for estimating accumulation rates with the CIC model. The activity of excess ^{210}Pb at any depth ($A_{(d)}$, where depth must be expressed in terms that account for compaction) is assumed to be controlled by the activity at the surface (A_o, presumed constant), the ^{210}Pb decay constant (λ = 0.0311 yr^{-1}) and the accumulation rate (AR) (in meters ice equivalent yr^{-1} in this case) through the well-known relation

$$A_{(d)} = A_o \exp[-(\lambda/AR)\, d]$$

In Figure 2 we have assumed that all measured ^{210}Pb is excess. The estimated mean accumulation rate (0.83 m ice yr^{-1}) is more than twofold higher than the true mean accumulation rate of 0.36 m ice yr^{-1} [*Clausen et al.*, 1988]. The five deepest samples (1793 - 1766) have roughly constant activities a bit higher than 0.03 pCi kg^{-1} (Table 1 and Figure 2). In profiles of total ^{210}Pb activity in sediments (where there is always supported ^{210}Pb), approach to an asymptote at depth provides one means to estimate the supported ^{210}Pb activity. Thus,the measured activity in the five deepest samples may reflect supported ^{210}Pb activity in the ice at Site D.

We note that supported ^{210}Pb activities as high as 0.03 pCi kg^{-1} are not easily explained. No dust measurements were made on this section of the core, but if we assume the high estimate of dust loading used earlier (300 μg kg^{-1}), the ^{210}Pb activity of the dust would have to be 100 pCi g^{-1}. While we would expect some enhancement of the supported ^{210}Pb activity in fine aerosol particles transported through the free troposphere relative to the particles in average shale, enhancements approaching 2 orders of magnitude appear to be unrealistic. It is also possible that the measured activities in the bottom of the core include a significant

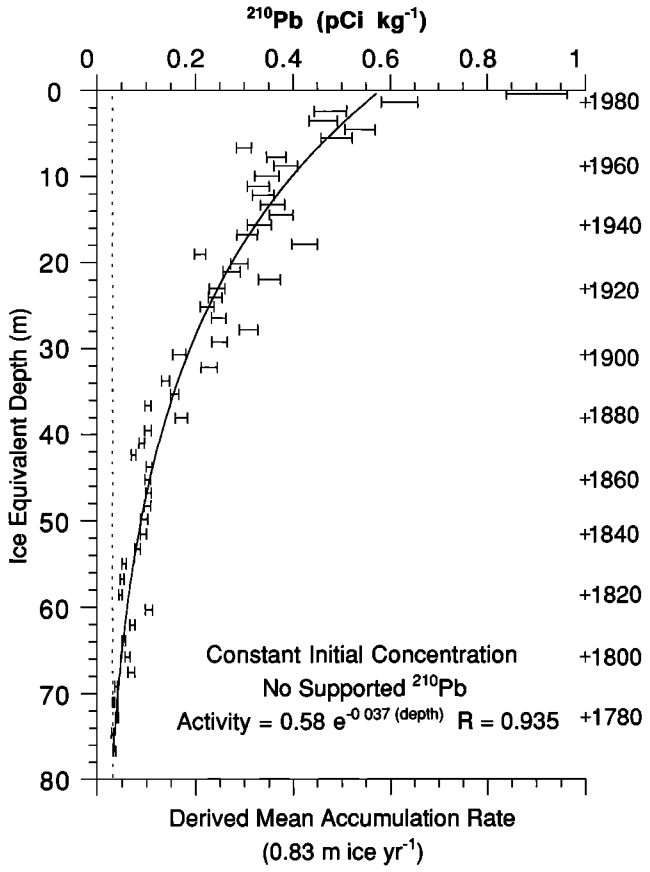

Figure 2. Measured ^{210}Pb activity in the Site D core. The curve is an exponential fit of activity against ice equivalent depth. If it is assumed that all of the ^{210}Pb is excess, this curve is an application of the constant initial concentration (CIC) model. The dashed line represents an estimate of the constant supported ^{210}Pb fraction suggested by the roughly constant activity in the five oldest samples.

fraction of excess ^{210}Pb if the initial activity at the surface was higher in the past. An initial activity of 16 pCi excess ^{210}Pb kg^{-1} 200 years ago would account for the measured activities with no contribution from supported ^{210}Pb. Of course, a 16-fold decrease in the initial excess ^{210}Pb activity over the period of record is probably as unlikely as a supported ^{210}Pb activity of 100 pCi g^{-1} in dust. It is more likely that the measured ^{210}Pb at the bottom of the core includes both excess and supported fractions, which cannot be separated using the available information (see discussion).

It is, nonetheless, still informative to assume that the lowest measured ^{210}Pb activity (0.032 pCi kg^{-1}) represents only supported ^{210}Pb. We have subtracted this apparent overestimate of supported ^{210}Pb from the measured value in each sample and fit an exponential curve to the resulting estimated excess ^{210}Pb depth profile. The CIC model yields an accumulation rate of 0.46 m ice yr^{-1} in this case, substantially lower than when all ^{210}Pb was assumed to be excess (0.83 m ice yr^{-1}, Figure 2) but still 28% higher than the mean accumulation rate determined by annual layer counting (0.36 m ice yr^{-1}).

Snow accumulation rate, averaged over the 3 to 6-year sample interval, ranges from 17% below to 17% above the long-term mean (Table 1). The CIC model assumptions require a corresponding variation in the flux of excess ^{210}Pb. An alternative conceptual model is that the flux of ^{210}Pb remains constant, implying that the initial activity would vary inversely to snow accumulation. This constant flux (CF) model [*Robbins*, 1978] may be more appropriate in some situations than the CIC model.

To facilitate comparison between the two models for the Site D ^{210}Pb record, we use each to calculate the model age of the layers defined by the sampling interval. For the CIC model the age at any given depth is given by;

$$t_{(d)} = -1/\lambda \ln (A_{(d)}/A_0).$$

The corresponding expression in the CF model is;

$$t_{(d)} = -1/\lambda \ln (1 - FI_{(d)}),$$

where $FI_{(d)}$ is the fractional inventory (the inventory (picocuries per square meter) of excess ^{210}Pb between the surface and depth d divided by the total inventory). To apply either model to the Site D record, the excess fraction of ^{210}Pb must be separated from the supported fraction; for lack of any other information, we again assume the supported ^{210}Pb activity is constant at 0.032 pCi kg^{-1} but reiterate that this is likely an overestimate. For the CIC model, A_0 is chosen to match the "observed" excess ^{210}Pb activity in the top sample.

Model-derived layer ages are compared to the stratigraphic age in Figure 3. The modeled ages vary widely at the bottom of the core as the log terms approach 0. For the first 30 years the CIC model overestimates ages, then systematically underestimates age by 30 - 50% for the bulk of the record. (Note that this model produces stratigraphic reversals when the activity of a deeper sample exceeds that of any samples above it.) The CF model ages are constrained to monotonically increase, but they systematically underestimate the age (Figure 3). After the first 30 years the errors in both models are similar. It should be noted that while we admit that our estimate of supported ^{210}Pb is problematically high, using a lower estimate would increase the discrepancies between modeled and stratigraphic age. Under the extreme (but hitherto commonly made) assumption that there is no supported ^{210}Pb, modeled ages > 100 years too young are calculated for 200-year-old firn at the bottom of the core.

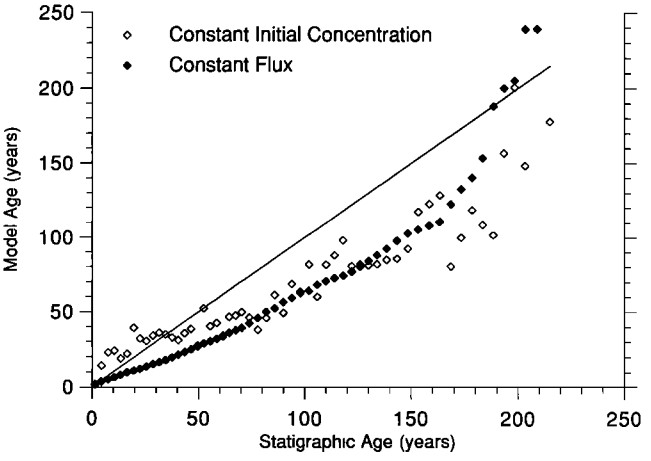

Figure 3. Ages calculated in the CIC and constant flux (CF) models from the estimated excess ^{210}Pb activity in the Site D core compared to the ages determined by annual layer counting [*Clausen et al.*, 1988]. The 1:1 line is shown for reference.

Discussion

It is apparent that the depth distribution of our estimate of excess ^{210}Pb activity in the firn at Site D is not consistent with either of these idealized (but widely used) models. In Figure 4 the "observed" values of excess ^{210}Pb (assuming supported ^{210}Pb activity = 0 or 0.032 pCi kg^{-1}) are compared to the profiles that would yield correct age determinations for all layers in the CIC and CF models. The differences between the two models are considerably less pronounced than those between either model and the observations. In the top 12 m (ice equivalent depth) there is "too little" excess ^{210}Pb in the firn, and for most of the rest of the record there is "too much" (Figure 4).

These discrepancies can be theoretically explained in two ways. The first is the explanation suggested by *Nijampurkar and Clausen* [1990] and *Dibb* [1992] for the Dye 3 and Summit records, a significant decrease in the flux of excess ^{210}Pb over the past 100 - 200 years. Alternatively, a decrease in the true supported ^{210}Pb flux over this time period would result in the same pattern if a constant value of supported ^{210}Pb was used to estimate the excess activity. In the absence of any independent measure of supported ^{210}Pb (i.e., ^{238}U or ^{226}Ra measurements), it is not possible to separate these two scenarios, but it is likely that the fluxes of both the supported and excess fractions of ^{210}Pb

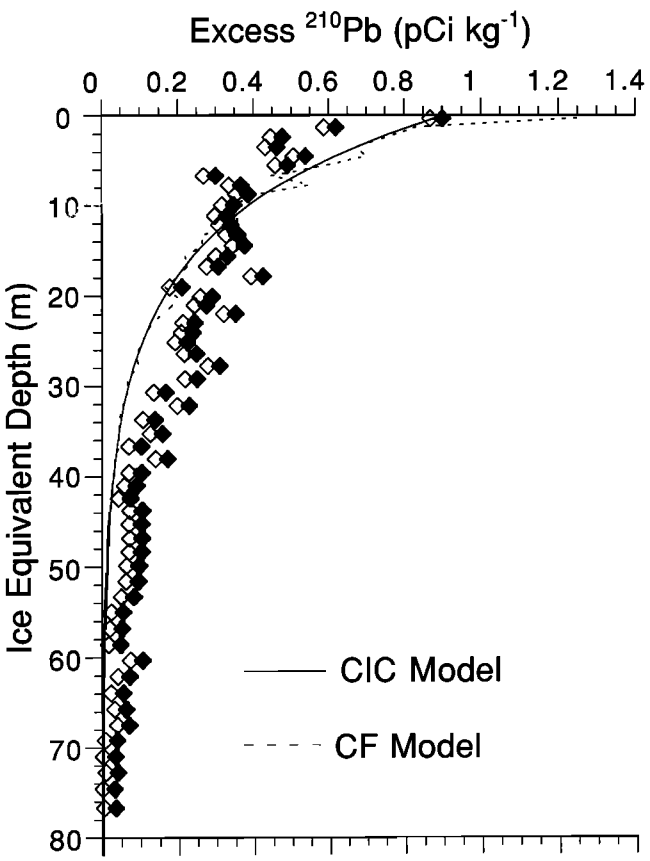

Figure 4. Estimated excess ^{210}Pb activities compared to the values that would yield correct ages in the CIC and CF models. (Solid diamonds assume constant supported ^{210}Pb activity = 0, open diamonds assume constant supported ^{210}Pb activity = 0.032 pCi kg^{-1}.) The flux of excess ^{210}Pb was calculated for each point in the CIC model by multiplying the decay-corrected activity by the independently determined accumulation rate of snow. The mean flux determined from this model was chosen as the constant flux value for the CF model.

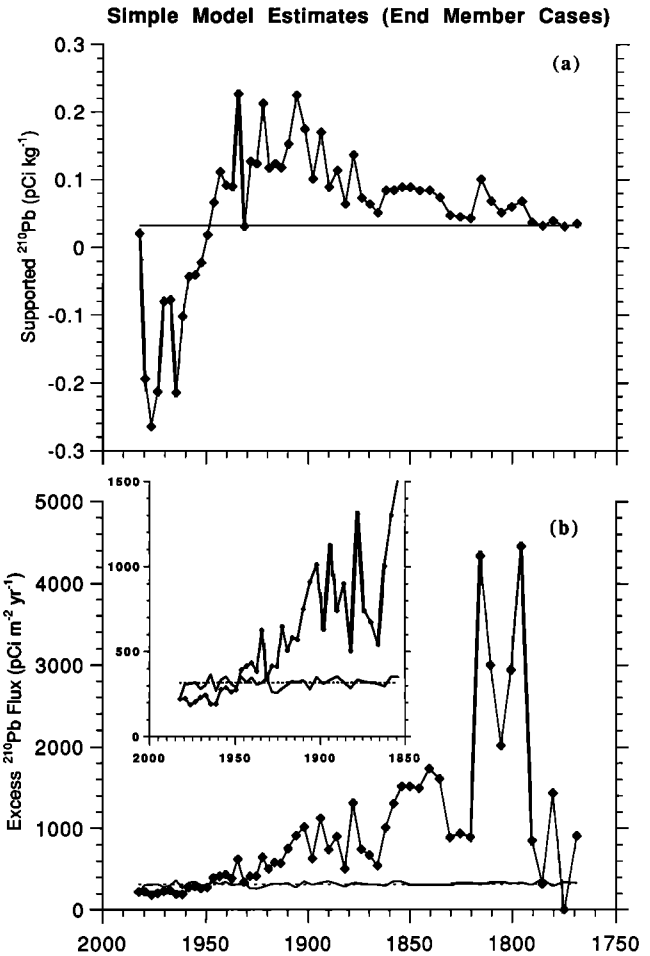

Figure 5. Variations in the concentration of (a) supported ^{210}Pb and (b) the flux of excess ^{210}Pb that would allow recovering the correct ages and accumulation rates from the ^{210}Pb profile at Site D (the inset in Figure 5b provides an expanded view of the time period since 1850). The horizontal line in Figure 5a reflects the constant level of supported ^{210}Pb estimated from the bottom of the core. The fluxes that correspond to the CIC and CF models are shown as the thin solid and dashed lines, respectively, in Figure 5b. It is likely that both factors have varied over the past 200 years (see text for discussion).

have decreased. Examining the magnitude of the change in either supported or excess ^{210}Pb alone that would be needed to resolve the underestimation of ages at Site D supports the hypothesis that decreases in both fractions must have occurred.

The activity of supported ^{210}Pb that would be required to place the observed excess ^{210}Pb on the CIC curve in Figure 4 is shown in Figure 5a. (This is simply the measured activity minus the modeled activity at each depth. Results are similar for the CF model (not shown).) For comparison, the estimated constant level of supported ^{210}Pb is also shown. Negative values of supported ^{210}Pb are not physically possible, and a change in the level of supported ^{210}Pb in excess of 0.5 pCi kg^{-1} in less than 50 years would also seem unlikely. As noted earlier, 4 x 10^{-4} pCi kg^{-1} was felt to be a conservatively high estimate of supported ^{210}Pb given dust loadings as high as 300 μg kg^{-1}. *Steffensen* [1988] reported a mean dust loading of 84 μg kg^{-1} for the 1919 -1891 interval of this core, a time period which would need to have among the highest levels of supported ^{210}Pb of the entire

record. In order for variations in supported ^{210}Pb alone to resolve the problems with the Site D ^{210}Pb dates, we would have to invoke a combination of enhanced dust loading and enrichment of ^{226}Ra in the aerosols that could create more than 1000-fold enhancements of supported ^{210}Pb. At the high end the dust reaching Greenland would have to resemble U ore if the ^{226}Ra was in turn supported by ^{238}U.

Figure 5b shows the excess ^{210}Pb flux derived by decay correcting the observed excess ^{210}Pb for the time since deposition and multiplying this by the annual average accumulation over each 3 to 6-year sampling interval. (This corresponds to the initial deposition rates calculated for the Dye 3 and Summit cores by *Nijampurkar and Clausen* [1990] and Dibb [1992], respectively, except that a constant level of supported ^{210}Pb was subtracted in the Site D case.) As expected, there is a pronounced downward trend as the present is approached. It is not likely that the excess ^{210}Pb flux was actually 10 - 20 times higher before 1850 than it was from 1960 - 1984, but only a small increase in supported ^{210}Pb in these older samples would result in more reasonable estimates of the excess flux. It is clear from the plots in Figure 5 that the failure of ^{210}Pb dating in the firn at Site D can be most plausibly explained by variations in the flux of both excess and supported ^{210}Pb, with a general decrease of both fractions over the past 200 years. Refining this statement will require a means to independently determine the temporal variation in the fluxes of supported and excess ^{210}Pb, perhaps through thermal ionization mass spectrometric (TIMS) determination of ^{238}U and/or ^{226}Ra, in parallel with ^{210}Pb measurements, in another Greenland ice core.

Dye 3, Summit, and Site D

We have calculated excess ^{210}Pb depth profiles that would yield the independently determined mean accumulation rates at Summit and Dye 3 with the CIC model. In Figure 6 the differences between the observed and modeled activities at all three sites are plotted against the year of deposition. (Note that we are not suggesting that the entire discrepancy between ^{210}Pb and stratigraphic ages should be attributed to variations in supported ^{210}Pb. Plots similar to Figure 5b, depicting the magnitude of change in the flux of excess ^{210}Pb alone that would reconcile the ages, have been previously presented for Dye 3 and Summit [*Nijampurkar and Clausen*, 1990; *Dibb*, 1992]). The decreasing trend at all three sites from about 1940 through the late 1970s must reflect reduced fluxes of ^{210}Pb, probably both the excess and supported fractions. Prior to 1940 the apparent ^{210}Pb fluxes were highest at Site D and lowest at Summit, perhaps largely due to differences in the activity of supported ^{210}Pb. (The measured ^{210}Pb activity in the 1766-1772 sample at Site D is more than twofold higher than that in the 1855 sample from Summit.) At all sites the apparent ^{210}Pb flux remains consistently higher during this earlier part of the record than it has been in the recent past (Figure 6). As discussed for the Site D results above, the general failure of the ^{210}Pb dating technique in Greenland firn must reflect decreasing fluxes over the time intervals sampled, a conclusion that is not negated by our inability to separately account for trends in the excess and supported fractions of ^{210}Pb.

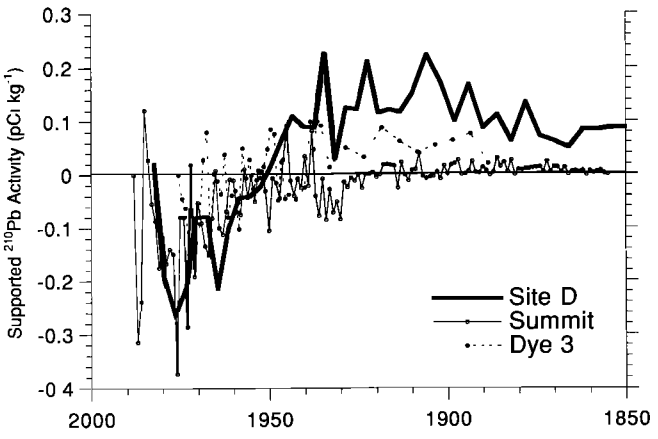

Figure 6. The difference between measured ^{210}Pb activity and the excess ^{210}Pb profile that would perfectly fit the known mean accumulation rate at Site D, Summit and Dye 3 under the assumptions of the CIC model. This is the same analysis presented in Figure 6a. The trends are felt to reflect variations in both excess and supported ^{210}Pb, rather than either alone.

Wider Implications of the Greenland Results

Similar discrepancies between ^{210}Pb-derived and stratigraphic ages (^{210}Pb ages significantly too young) have been found recently in three new firn cores collected in northern Greenland (D. Wagenbach, personal communication, March 1995). These findings extend the latitude range covered by the three cores discussed herein (65 - 72°N) to include most of the interior of the Greenland ice sheet. The large spatial extent of the observed decrease in ^{210}Pb deposition to Greenland suggests that an explanation must be sought in atmospheric processes of similar or larger spatial scales. A gradual decrease in ^{210}Pb activity in the northern high-latitude troposphere due to declining ^{222}Rn emissions is one possibility. Alternatively, increased scavenging of the aerosols carrying ^{210}Pb before continentally derived air masses reach Greenland could reduce the atmospheric ^{210}Pb burden over the ice sheet. A third possibility is a gradual increase in the influence of marine air masses (which carry relatively low burdens of ^{222}Rn and its progeny), at the expense of continental air masses, over the ice sheet. (*Gäggeler et al.*[1983] raised this same possibility to explain large interannual variations in ^{210}Pb activity in firn cores from Colle Gnifetti.) The most optimistic scenario for use of ^{210}Pb as a dating tool would be modification of air mass transport pathways to increase marine influence over Greenland.

A general decrease in tropospheric ^{210}Pb would introduce a bias toward young age estimates in all media where ^{210}Pb can be used to develop chronologies. Radon emission from high-latitude soils may vary in response to climate change (warming) that has been documented for Arctic land areas [e.g., *Taylor et al.*, 1982; *Lachenbruch et al.*, 1982]. Warmer temperatures over the past century might have reduced Rn flux to the atmosphere by decreasing the soil permeability in a deeper active (thawed) layer in summer (D. Wagenbach, personal communication, March 1995), though reduced length and thickness of snow cover during winter could have an opposite effect. At present, there does not appear to be any firm evidence that decreasing ^{210}Pb concentrations in the atmosphere can explain the trends seen in Greenland firn.

Large increases in scavenging by precipitation and/or dry fallout during transport to Greenland (with a constant Rn input) would imply that some regions must have experienced increasing ^{210}Pb fluxes while those in Greenland decreased. Thus ^{210}Pb chronologies could be biased in opposite directions, depending on the location being studied. We are not aware of any ^{210}Pb

records in the northern hemisphere that have shown significant biases toward ages that are too old, but this may just reflect the difficulty of obtaining independent chronologies to compare to ^{210}Pb-based age estimates in media other than polar ice cores. Application of ^{210}Pb dating to cold glaciers in the Alps appears to yield ages in better concordance with other dating techniques than we have found in Greenland [e.g., *von Gunten et al.*, 1982; *Gäggeler et al.*, 1983].

The ^{210}Pb data themselves do not allow discrimination between these possibilities, but other glaciochemical data from Greenland ice cores may be consistent with both an increasing marine influence over the ice sheet and increased aerosol removal rates from the northern free troposphere. The concentrations of soluble Ca^{2+} (considered to reflect inputs of continental dust) in the Greenland Ice Sheet Project 2 (GISP2) Summit ice core since 1940 are about 20 - 25% lower than the 1850 - 1940 mean concentration, while Cl^- concentrations steadily increase over the last 50 years of the record [*Mayewski et al.*, 1993]. No parallel increase is seen in Na^+ concentrations, suggesting that the Cl^- is not entirely from sea-salt aerosols. However, the excess Cl^- trend could reflect gaseous HCl released from sea-salt aerosols and mixed upward out of the marine boundary layer. Free tropospheric marine air enriched in HCl, but with very low levels of ^{222}Rn and ^{210}Pb, could be more readily transported to high elevations over Greenland than marine boundary layer air rich in sea-salt aerosols. On the other hand, microparticle analyses on the GISP2 core show no trend in either the number or mass of insoluble particles accumulating in the Summit region since 1850 (G. Zielinski, personal communication, December 1994). However, the Coulter counter technique used for ice core microparticle studies only detects particles > 0.63 μm diameter, which may be larger than those most important in transport and deposition of soluble Ca^{2+}.

Increased deposition of NO_3^- and SO_4^{2-} has been documented in several Greenland ice cores and has been attributed to anthropogenic emissions of NO_x and SO_2 [*Neftel et al.*, 1985; *Mayewski et al.*, 1986, 1990; *Finkel et al.*, 1986; *Laj et al.*, 1992]. The impact of elevated concentrations of acidic gases on aerosol microphysics is still not completely understood, but condensation of H_2SO_4 (from SO_2) on preexisting aerosols would increase their size and might enhance deposition. Such a process could cause earlier removal of ^{210}Pb-bearing aerosols along the transport paths from continental sources to Greenland. As noted above, if this mechanism is contributing to the decreasing ^{210}Pb flux in Greenland, there must also be an increasing trend in ^{210}Pb flux in some region that has not yet been identified.

Conclusions

The detection of ^{210}Pb in ice >200 years old at Site D has shown that the supported fraction of ^{210}Pb cannot be assumed negligible in Greenland ice cores. The ^{210}Pb activity at the bottom of the Site D core is about 2 orders of magnitude higher than published measurements of dust loading in Greenland ice would indicate, if all of the measured ^{210}Pb is supported and the dust resembles average shale in its U content. We suggest that part of the measured ^{210}Pb activity at these depths must still be excess, reflecting much higher fluxes of excess ^{210}Pb in past times. However, we doubt the excess ^{210}Pb flux was 16-fold higher 200 years ago than it is now, thus the supported ^{210}Pb fraction appears to be significantly higher than the estimate based on dust loading. This may indicate that the crustal aerosols reaching Greenland are enriched in U and its progeny relative to average soil or shale, perhaps reflecting the size selective processes acting on insoluble aerosols during long-range transport. Further investigation of this question could contribute to our understanding of the provenance of dust reaching high-elevation sites on the Greenland ice sheet.

Even after correcting the ^{210}Pb activities measured at Site D for an unexpectedly large (assumed constant) supported fraction, age estimates from the CIC and CF models tended to be much younger than the known ages. The failure of these models indicates that the flux of ^{210}Pb must have varied significantly over the past 1-2 centuries. It is not possible to partition ^{210}Pb flux variations between the excess and supported fractions from available data, but it is likely that both fractions have roughly covaried throughout this period. It might be possible to assess temporal variations of excess and supported ^{210}Pb separately by measuring ^{238}U and/or ^{226}Ra with ^{210}Pb in Greenland ice core samples, but the levels are so low that TIMS is probably the only technique that will be viable. The Site D record is similar to previously published results from Dye 3 and Summit, in that the ^{210}Pb flux appears to have steadily decreased throughout the present century. In all three cores the ^{210}Pb flux was particularly low in the period 1940 - 1980 relative to older portions of each record.

The findings from these Greenland cores may have implications for ^{210}Pb-based chronologies in coastal and lacustrine sediments as well as other ice core studies. Large variations in the flux of either excess or supported ^{210}Pb will bias age estimates from any of the standard models. It is possible that the ^{210}Pb decrease is unique to Greenland, reflecting an increasing marine influence on air masses advected over the ice sheet. However, other plausible explanations of the Greenland observations would perturb the atmospheric budget of ^{210}Pb over large regions of the northern hemisphere. Detailed investigations of ^{210}Pb in rapidly accumulating sediments that were independently dated could provide insight into this question since it would be possible to also determine supported ^{210}Pb activities (through ^{226}Ra measurements) relatively easily by direct gamma counting of each sample since the levels will be much higher than in glacial snow and ice.

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