

BERYLLIUM-7 AND LEAD-210 IN AEROSOL AND SNOW IN THE DYE 3 GAS, AEROSOL AND SNOW SAMPLING- PROGRAM

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Abstract—Concentrations of cosmogenic ^7Be and the radon daughter ^{210}Pb were determined in surface-level aerosols and fresh and aging snow throughout DGASP. The distinct sources, yet common association with submicron aerosols, of these atmospheric radionuclides makes them valuable tracers of the transport, depositional and post-depositional processes leading to incorporation of submicron aerosols (and associated species) into the Greenland Ice Sheet.

The aerosol concentrations of atmospheric radionuclides tended to covary on the 1–7 day time scale of individual samples ($r=0.57$, $n=114$, $p=0.001$) and were also highly correlated when averaged by month ($r=0.80$, $p=0.002$). The year-long time series of both ^7Be and ^{210}Pb showed concentration peaks in spring (April) and fall (September–October) and minima in summer (July) and winter (November–February). This finding is in stark contrast to results from other arctic sites, where both radionuclides, but particularly ^{210}Pb , show pronounced concentration peaks in the winter. This indicates that the air masses over the Greenland Ice Sheet may be distinct from the air in the polar basin, or boundary layer processes strongly bias surface-level aerosol characteristics over the ice sheet, or, likely, some combination of these factors are operating.

The concentrations of ^7Be and ^{210}Pb in fresh snow at Dye 3 show wide variability between snow fall events. The lack of any obvious correlation between concentrations in aerosol and fresh snow support the contention that surface-level air at this site is often not reflecting air aloft. Aging snow studies did not reveal consistent trends in concentration, suggesting important heterogeneity in the initial concentrations of ^7Be and ^{210}Pb in surface snow over small spatial scales. Poor agreement between the ^{210}Pb profile in a 2 m snowpit sampled at the end of the project and the observed deposition of ^{210}Pb through the year indicate the potential importance of post-depositional processes in modifying the atmospheric “signals” preserved in the snowpack.

Key word index: Greenland, arctic aerosol, arctic precipitation, ^7Be , ^{210}Pb .

INTRODUCTION

The natural atmospheric radionuclides ^7Be and ^{210}Pb were included in the species measured during the Dye 3 Gas, Aerosol, and Snow Sampling Program (DGASP). (The objectives, design, and a summary of DGASP are discussed by Jaffrezo and Davidson (this issue)). Cosmogenic ^7Be is produced in the upper troposphere and stratosphere, with maximum production near 15 km (Bhandari *et al.*, 1970). Atmospheric ^{210}Pb results from the decay of ^{222}Rn in the atmosphere. Radon flux from the ocean surface to the atmosphere is negligible compared to that from continents (Turekian *et al.*, 1977), so the ultimate source of most atmospheric ^{210}Pb is the continental boundary layer. However, enough ^{222}Rn reaches the stratosphere before decaying to create a significant stratospheric ^{210}Pb reservoir (Bhandari *et al.*, 1966; Feely and Seitz, 1970; Lambert *et al.*, 1982). Once formed,

both of these radionuclides rapidly associate with atmospheric particles (Arnold and Al-Salih, 1955; Turekian *et al.*, 1977), predominantly those in the submicron fraction (Maenhaut *et al.*, 1979; Bondietti *et al.*, 1987; 1988). Hence, ^7Be and ^{210}Pb serve as tracers of submicron aerosol transport in the atmosphere. To a first order of approximation, ^7Be and ^{210}Pb reflect the fate of aerosols originating high in the free troposphere, and in the boundary layer, respectively. These radionuclides also allow examination of the processes removing the submicron fraction of the aerosol population from the atmosphere and the post-depositional processes modifying the concentrations of aerosol-associated species in snow before it is preserved as glacial ice.

^7Be and, to a lesser degree, ^{210}Pb , differ from most of the atmospheric constituents of more general interest in ice core and climate research in that their sources are diffuse and relatively constant. This tends to yield tropospheric distributions of these radionuclides that vary relatively smoothly with space and time. As a result, particularly for ^7Be , changes in tropospheric concentration over a site like Dye 3 will be more

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dependent on factors such as seasonal changes in stratosphere-troposphere and/or meridional exchange than discrete long-range transport events from a specific source region. Therefore, the concentrations of these radionuclides in surface-level air may be particularly useful in determining the extent of surface-layer/free troposphere communication on timescales ranging from seasonal down to daily changes.

Ground-based aerosol sampling at a number of low altitude Arctic sites has revealed pronounced seasonality in the concentrations of ^7Be and ^{210}Pb . Both radionuclides are characterized by broad winter peaks, though peak ^7Be concentrations occur slightly later in the year than those of ^{210}Pb (Rahn and McCaffrey, 1980; Daisey *et al.*, 1981; Larsen *et al.*, 1988; Feely *et al.*, 1988; Larsen and Sanderson, 1990; Dibb *et al.*, in review). Recent detailed snowpit studies near Dye 3 in south Greenland suggest either no seasonality in ^{210}Pb accumulation (Dibb, 1990a) or a slight tendency for higher concentrations to occur in snow from late summer to fall (Beer *et al.*, 1991). The depth distribution of ^{10}Be (a long-lived proxy for ^7Be) in the pit studied by Beer *et al.* (1991) did not show clearly seasonality, but there was a tendency for higher concentrations to occur in snow from spring and fall. The apparent lack of agreement between seasonality of these radionuclides observed in the Arctic Basin and in south Greenland snow indicates either distinct differences in the composition of air over the two regions, or serious fractionation between air and snow that varies over the year.

In this paper we examine this question by comparing aerosol concentrations of ^7Be and ^{210}Pb at Dye 3 to similar measurements at other locations in the Arctic, and to their concentrations in fresh and aging surface snow collected throughout DGASP. A depth profile of ^{210}Pb in a snowpit excavated at the end of the study is also compared to the time series of concentrations in both air and fresh snow. This ^{210}Pb profile is also used to assess the representativity of the DGASP year through comparison to the results of recent studies cited above.

EXPERIMENTAL

Aerosol sampling for atmospheric radionuclides commenced 5 August 1988 and terminated on 28 July 1989. For the 114 samples collected, sampling intervals varied from 1 to 7 days (depending entirely on vagaries of the weather and the enthusiasm of the operator on site at any given time). The sources of ^7Be and ^{210}Pb (unlike most of the other species sampled during DGASP) preclude the possibility of contamination by operations at Dye 3. As a result, sampling was continuous except for brief periods of pump failure, usually due to brushes wearing out. Aerosols were collected on Whatman GF/A glass fiber filters coupled to a high volume vacuum pump. Sample air volumes were determined by an in-line dry gas meter. Corrections for temperature and the reduced pressure in the sampling line were used to convert measured volumes to standard cubic meters (SCM). Four

47 mm diameter filters were exposed concurrently, at a sampling face velocity of about 0.5 m s^{-1} for a flow rate of approximately 13 SCM h^{-1} .

Samples of fresh snow were collected from 22 events during DGASP. Most sampling was conducted 1 km from the station, in the upwind direction of each event. On two occasions, samples were also collected at established sampling sites 10 and 23 km southwest of the station. Snow was sampled by carefully scraping about 1 kg of snow from the dominant, distinct, surface stratigraphic layer into polyethylene bags with clean Plexiglas scrapers. Sixteen samples of aged snow, that appeared to be related to specific fresh snow events, were also collected. Assignment of aged snow samples to specific snowfall events was based on careful observation of the snow surface on a daily basis and the absence of new snow or drifting due to wind during the intervening period. As will be shown below, it is not certain that this approach adequately identifies a given stratigraphic layer of the surface snow, even over time periods as short as a few days.

Accumulation of snow during DGASP was measured on stakes established at distances of 1, 10, 23 and 25 km from the station. At each stake, labeled polyethylene sheets were placed on the snow surface at intervals through the study to provide a series of chronologic horizons to relate to the snow pit samples collected at the end of DGASP. The snowpit at the 23 km site was excavated 7 August 1989. A block of snow, surface area approximately $20 \times 40 \text{ cm}^2$, extending from 1.5 to 117.5 cm depth, was cut from the wall of this pit for ^{210}Pb determinations. This block was returned frozen to UNH, where it was cleaned by scraping to a $15 \times 35 \text{ cm}^2$ column and sectioned at 4 cm depth intervals. (Cleaning and sectioning was conducted in a cold room at -20°C .) Two of the labeled sheets at this site were recovered within the depth interval encompassed by the ^{210}Pb samples.

Processing and analysis of the aerosol and fresh and aging snow samples for ^7Be and ^{210}Pb by gamma spectrometry is described elsewhere (Dibb, 1990b). The determination of ^{210}Pb in the snowpit samples by alpha spectrometry followed procedures outlined in Dibb (1990a), omitting the search for ^{137}Cs by gamma counting.

RESULTS

Aerosol

The concentration of ^7Be and ^{210}Pb in surface-level aerosols at Dye 3 varied markedly on the 1–7 day time scale defined by sampling intervals (Fig. 1). There was a strong tendency for covariance of these two radionuclides, despite their distinctly different sources ($r=0.57$, significant at $p=0.001$). Daily aerosol sampling at Summit, Greenland in the summers of 1989 and 1990 showed similar features (Dibb, 1990b; Dibb *et al.*, 1992a), as did high temporal resolution aerosol sampling on the Antarctic Ice Sheet (Sanak *et al.*, 1985; Wagenbach *et al.*, 1988). The high frequency and correlation of ^7Be and ^{210}Pb variability in surface-level air over both polar ice sheets implies a large influence of "local" atmospheric processes on boundary layer composition (Dibb, 1990b; Dibb *et al.*, 1992a). Below, we examine this hypothesis in detail.

Snow

Over the course of the year, ^7Be concentrations in fresh snow events around Dye 3 ranged over nearly an

order of magnitude, while ²¹⁰Pb concentrations showed greater than 25-fold variability (neglecting samples with concentrations below detection limit) (Table 1).

In contrast to their atmospheric boundary layer relationship, there is no clear indication that the concentrations of these radionuclides covary in fresh snow ($r=0.30$, significant at $p=0.3$).

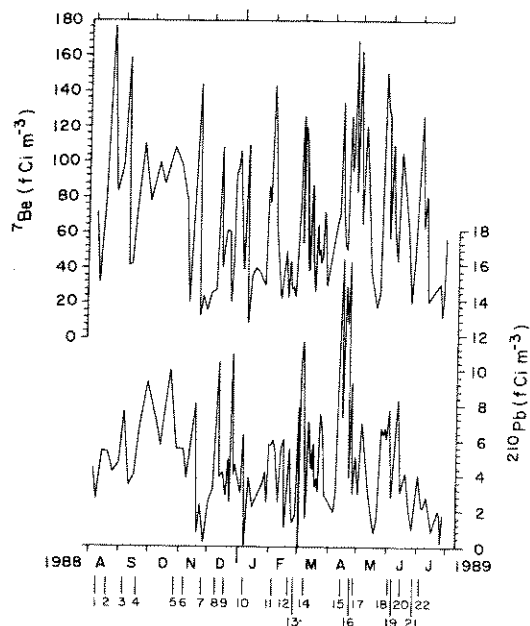


Fig. 1. Detailed time series of the concentrations of ⁷Be and ²¹⁰Pb in surface-level aerosol for the year of DGASP. The timing of fresh snow events that were sampled during the study (Table 1) is indicated by the numbered vertical lines beneath the time axis.

Beryllium-7 concentrations showed three-fold variability in September and a two-fold range in April when fresh snow was sampled along a 23 km traverse (Table 2). In the same samples ²¹⁰Pb concentrations varied six-fold and two-fold, respectively. Such variability is actually encouragingly low, given our recent observations of 50-fold ⁷Be and 20-fold ²¹⁰Pb concentration variations along a 60 km traverse near Summit (Dibb, 1990b).

Nine fresh snowfall events were resampled after aging for periods up to 35 days (Table 3). The motive for this sampling was to examine changes in surface snow chemistry during aging, and perhaps the opportunity to obtain an estimate of the rate and importance of dry deposition processes for incorporating atmospheric constituents into the snowpack (Davidson *et al.*, 1987). For example, if dry deposition and/or mass loss of snow by sublimation were consistently important processes, concentrations of aerosol-associated species should increase in snow exposed at the surface.

An example of an apparently successful application of this technique was described for our 1989 sampling at Summit (Dibb, 1990b). However, the foregoing discussion of spatial variability in surface snow chemistry suggest that this is an overly simplistic approach,

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Table 1. Concentrations of ⁷Be and ²¹⁰Pb in fresh snow collected 1 km from Dye 3 throughout DGASP. Mass-basis scavenging ratios calculated from these and the aerosol data are also included

No.	Date	Activity (pCi Kg ⁻¹)		Scavenging ratio		S _{Be} /S _{Pb} *
		⁷ Be	²¹⁰ Pb	⁷ Be	²¹⁰ Pb	
1	8/11/88	7.88 ± 0.52	0.97 ± 0.17	244	362	0.67
2	8/21/88	8.29 ± 0.43	0.58 ± 0.15	47	109	0.43
3	9/9/88	58.51 ± 1.89	2.13 ± 0.36	1386	405	3.42
4	9/21/88	7.70 ± 0.58	1.22 ± 0.17	70	175	0.40
5	10/30/88	6.57 ± 0.93	0.34 ± 0.13	70	61	1.17
6	11/6/88	6.36 ± 0.45	1.27 ± 0.15	81	232	0.35
7	11/28/88	18.64 ± 1.08	0.23 ± 0.28	1135	1045	1.09
8	12/11/88	6.92 ± 0.67	n.d.†	63	—	—
9	12/20/88	7.10 ± 0.60	1.63 ± 0.22	117	565	0.21
10	1/7/89	n.d.	1.40 ± 0.22	—	223	—
11	2/6/89	18.01 ± 1.07	0.11 ± 0.09	377	20	18.85
12	2/22/89	6.21 ± 0.70	1.51 ± 0.23	214	271	0.79
13	2/25/89	9.95 ± 0.73	n.d.	400	—	—
14	3/10/89	11.96 ± 0.79	1.69 ± 0.22	308	1012	0.30
15	4/15/89	8.19 ± 0.67	0.51 ± 0.13	156	69	2.26
16	4/22/89	7.70 ± 0.67	n.d.	76	—	—
17	4/27/89	9.17 ± 0.47	1.53 ± 0.16	109	508	0.21
18	6/5/89	26.30 ± 1.40	2.76 ± 0.35	413	968	0.43
19	6/6/89	14.66 ± 1.48	n.d.	329	—	—
20	6/15/89	13.68 ± 0.70	1.10 ± 0.18	159	356	0.45
21	6/26/89	10.83 ± 0.82	2.95 ± 0.34	245	2875	0.08
22	7/2/89	28.03 ± 1.06	1.13 ± 0.25	222	276	0.80
Mean		13.48	1.08	315	547	
S.D.		11.89	0.83	374	683	

*Ratio of the scavenging ratios.
 †n.d., not detected.

Table 2. Concentrations of ^7Be and ^{210}Pb in fresh snow collected at three sites along a 23 km traverse from Dye 3

Site (km)	Date	Activity (pCi kg ⁻¹)	
		^7Be	^{210}Pb
1	9/21	7.70 ± 0.58	1.22 ± 0.17
10	9/23	2.79 ± 0.36	0.19 ± 0.11
23	9/24	6.16 ± 0.46	1.10 ± 0.14
1	4/27	9.17 ± 0.47	1.53 ± 0.16
10	4/29	8.63 ± 0.61	2.81 ± 0.24
23	4/29	16.53 ± 0.82	1.22 ± 0.18

as is borne out by examination of Table 3. Out of 23 cases (including successive samples from the "same layer" of aging snow), the concentrations of ^7Be and ^{210}Pb both increased on only 9 occasions (Table 3). Only one out of five cases where a given layer was resampled on at least two later dates showed monotonic increases in the concentrations of both radionuclides (the September–October series). (In the other four cases, the only time either species steadily increased was in the July–August series for ^{210}Pb , but in this instance the concentration apparently triples from one day to the next after experiencing an insignificant increase during the preceding 18 d.) The present results indicate that extreme care must be taken to ensure that sampling is restricted to the same stratigraphic layer, and small-scale spatial heterogeneity must be assessed, if studies of fresh and aging snow at the surface are to provide insight into the question of post-depositional modification of snow chemistry.

The depth distribution of ^{210}Pb in the snowpack at the end of DGASP was quite comparable to results from recent snowpit studies near Dye 3 (Dibb, 1990a; Beer *et al.*, 1991) (Fig. 2, left frame). Temporal control for this pit was provided by the well-defined annual oscillation of ^{18}O (not shown, but the position of the first maximum below the surface is indicated as "summer 1988") and the recovery of two dated polyethylene sheets. There does appear to be a subtle ^{210}Pb concentration maximum in late summer–fall snow, as was noted by Beer *et al.* (1991). The total accumulation of ^{210}Pb between the surface and the "summer 1988" horizon was 153 pCi m⁻², in very good agreement with the annual accumulation of 138–182 pCi m⁻² for the period May 1986–May 1988 in a snowpit sampled 40 km southwest of Dye 3 (Dibb, 1990a). The depth profile of ^{210}Pb at the end of DGASP will be compared to the time series of both fresh snow and aerosol concentrations in the discussion below.

DISCUSSION

It must be reiterated here that we are discussing a single year of observations at Dye 3, with no *a priori* assurances that this was a representative year. However, the depth profiles of ^{210}Pb and sulfate (Jaffrezo

Table 3. Comparison of the concentrations (pCi kg⁻¹) of ^7Be and ^{210}Pb in fresh and aged snow around Dye 3

	Date	^7Be	^{210}Pb
<i>1 km site</i>			
Fresh	9/21	7.70 ± 0.58	1.22 ± 0.17
Aged	10/19	15.15 ± 0.69	2.00 ± 0.24
Aged	10/26	20.66 ± 0.84	2.01 ± 0.31
Fresh	12/20	7.10 ± 1.63	1.63 ± 0.22
Aged	1/3	3.37 ± 1.02	1.02 ± 0.18
Fresh	1/7	n.d.*	1.40 ± 0.22
Aged	1/23	8.10 ± 0.39	1.56 ± 0.12
Aged	1/28	5.63 ± 0.64	0.59 ± 0.14
Fresh	2/6	18.01 ± 1.07	0.11 ± 0.09
Aged	2/19	5.02 ± 0.46	1.40 ± 0.16
Fresh	2/25	9.95 ± 0.73	n.d.
Aged	3/5	6.33 ± 0.62	n.d.
Fresh	3/10	11.96 ± 0.79	1.69 ± 0.22
Aged	3/22	7.49 ± 0.95	1.65 ± 0.25
Aged	4/5	n.d.	n.d.
Aged	4/8	2.93 ± 0.51	0.06 ± 0.24
Fresh	4/22	7.70 ± 0.67	n.d.
Aged	4/25	7.53 ± 0.61	0.58 ± 0.16
Fresh	6/15	13.68 ± 0.70	1.10 ± 0.18
Aged	6/25	33.65 ± 1.10	0.44 ± 0.19
Aged	7/2	14.71 ± 0.74	1.61 ± 0.21
<i>10 km site</i>			
Fresh	9/23	2.79 ± 0.36	0.19 ± 0.11
Aged	10/4	6.33 ± 0.47	2.37 ± 0.19
<i>23 km site</i>			
Fresh	7/18	9.00 ± 0.62	0.50 ± 0.11
Aged	8/5	27.06 ± 1.13	0.58 ± 0.13
Aged	8/6	22.11 ± 1.02	1.85 ± 0.24

* n.d., not detected.

and Davidson, 1992) in the snowpack that accumulated during the year of DGASP are very similar to the annual patterns reported for the past 10–15 yr on the basis of numerous snowpits sampled near Dye 3. This suggests, but does not prove, that the DGASP aerosol record may be representative of the recent seasonal pattern of atmospheric composition over Dye 3.

Aerosol

Aerosol concentrations of ^7Be and ^{210}Pb in surface-level air over the Greenland Ice Sheet are characterized by high variability on time scales as short as one day (likely even shorter if sampling frequency was high enough to observe it) (Fig. 1; Dibb, 1990b; Dibb *et al.*, 1992a). The covariation of ^7Be and ^{210}Pb concentrations at a given site but lack of coherence in their concentration time series between Summit and Dye 3 during June–July 1989, led to the hypothesis that "local" atmospheric processes strongly influence the composition of surface-level air over the ice sheet (Dibb, 1990b). Further investigation during the 1990 summer season at the Summit showed that ^7Be and ^{210}Pb concentrations (as well as those of other aerosol-associated species) dropped sharply when strong,

Concentrations (pCi/kg) aged snow

	²¹⁰ Pb
1.58	1.22 ± 0.17
1.69	2.00 ± 0.24
1.84	2.01 ± 0.31
.63	1.63 ± 0.22
.02	1.02 ± 0.18
	1.40 ± 0.22
.39	1.56 ± 0.12
.64	0.59 ± 0.14
.07	0.11 ± 0.09
.46	1.40 ± 0.16
73	n.d.
62	n.d.
79	1.69 ± 0.22
95	1.65 ± 0.25
51	n.d.
51	0.06 ± 0.24
57	n.d.
51	0.58 ± 0.16
70	1.10 ± 0.18
0	0.44 ± 0.19
74	1.61 ± 0.21
6	0.19 ± 0.11
7	0.37 ± 0.19
2	0.50 ± 0.11
3	0.58 ± 0.13
2	1.85 ± 0.24

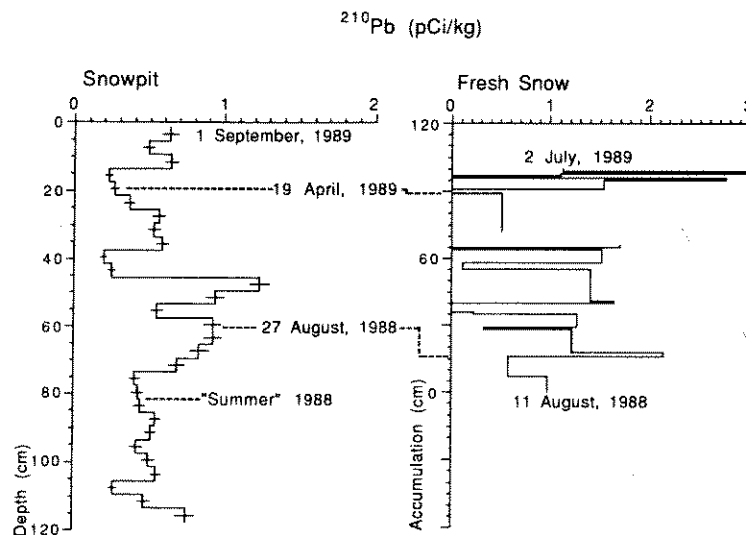


Fig. 2. Depth profile of ²¹⁰Pb concentration in a snowpit sampled at the end of DGASP. For comparison, a "deposition series" has been constructed by plotting the concentration of ²¹⁰Pb in each fresh snow sample over the accumulation measured at the 1 km site for the given event.

Table 4. Comparison of the mean concentrations of aerosol-associated atmospheric constituents between periods characterized by high and low ⁷Be concentrations. See text for discussion of how these intervals were defined. Trace element data from Mosher and Jaffrezo (1993).

Higher when ⁷ Be is high	Lower when ⁷ Be is high
(Significant at 99% confidence level) ²¹⁰ Pb, Al, Mn, Ca, Sc, Br, As, Mg, Fe	Na, Cl, V
(Significant at 95% confidence level) I, Sb, Se, Zn	

pack that accumulates very similar to the past 10–15 yr on the ice near Dye 3. This is the DGASP aerosol from the recent seasonal cycle over Dye 3.

The ²¹⁰Pb in surface-level aerosols are characterized as short as one week when the frequency was high in 1990b; Dibb *et al.*, 1990b. ²¹⁰Pb concentrations in the snow at Summit and Dye 3 support the hypothesis that the high ⁷Be influence the ²¹⁰Pb concentrations over the ice sheet during the 1990 winter. The ⁷Be and ²¹⁰Pb concentrations of other aerosols when strong,

low-level (below 1 km) inversions "sealed" the surface-level air from the aloft (Dibb *et al.*, 1992a). As an extension of this hypothesis, we would now like to suggest that, in general, intervals when the surface-level concentrations of ⁷Be and ²¹⁰Pb are both low reflect isolation of surface air from higher levels, and, conversely, high concentrations indicate periods of more efficient mixing of free tropospheric air to the surface. The DGASP data set does not prove this hypothesis, as we lack any information about atmospheric structure over Dye 3, but it does provide additional support.

To test this hypothesis regarding the influence of troposphere/boundary layer exchange on the composition of surface-level air at Dye 3, we must define characteristic high and low concentrations of the radionuclides. The average concentration of ⁷Be in the Arctic free troposphere is in the neighborhood of 200 fCi SCM⁻¹ (Dibb *et al.*, 1992b; Rama, 1963). As expected, surface concentrations at Dye 3 are lower than this (average during DGASP was 69 ± 39 fCi ⁷Be SCM⁻¹), but frequently approach free tropospheric levels (Fig. 1). Twenty-five sampling periods, totaling 61 days out of the 358 day duration of DGASP, had

⁷Be concentrations exceeding 100 fCi SCM⁻¹. On the other hand, there were 47 samples (covering 128 days) with ⁷Be concentrations less than 50 fCi SCM⁻¹.

We have used these somewhat arbitrarily chosen levels of high and low ⁷Be concentration to filter the ²¹⁰Pb and trace element (Mosher and Jaffrezo, 1993) aerosol data, then compared their mean concentrations during the two different periods (Table 4). Among the species showing significant increase during intervals with high ⁷Be concentrations, Al, Mn, Ca, Sc and Fe appear to reflect a dominant crustal source (Barrie *et al.*, 1989; Landsberger *et al.*, 1990; Mosher and Jaffrezo, 1993). Previous work has indicated that subsidence of air masses under the influence of a high pressure ridge over Greenland is responsible for marked increases in the dust content of the surface-level aerosol at Dye 3, suggesting that most of this material is transported to South Greenland through the free troposphere (Hogan *et al.*, 1984; Davidson *et al.*, 1985).

The concentrations of the sea-salt components Na and Cl tend to be higher when ⁷Be levels are low, suggesting that sea salt aerosols mainly travel to Dye 3 at lower levels in the atmosphere, perhaps largely

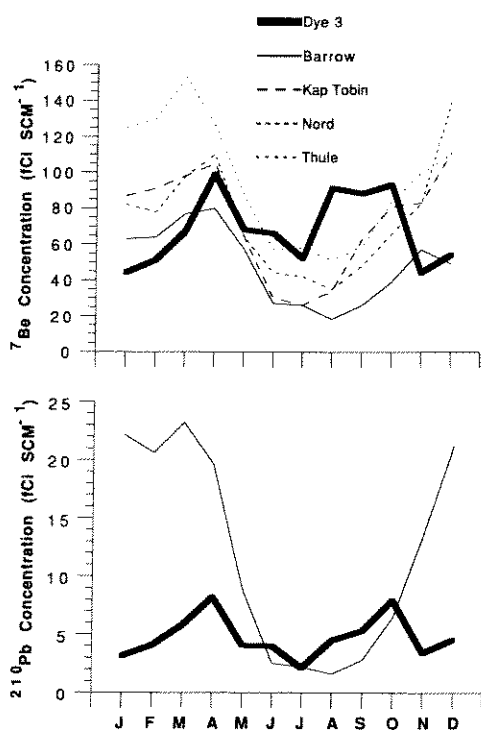


Fig. 3. Monthly average concentrations of ^7Be and ^{210}Pb in surface-level aerosol at a number of arctic sites. The Dye 3 data has been weighted by the sampled volumes for the 114 samples of varying exposure time presented in Fig. 1. Data for the other stations is from EML data reports and represents a mix of monthly values and averaged weekly samples, depending on the station.

within the boundary layer, and originate from source regions within several hundreds of km of Dye 3. Vanadium concentrations are also elevated when ^7Be is low. Mosher and Jaffrezo (1993) suggest that Dye 3 station operations constitute an important local source of V, thus it is not surprising to find V enriched in air masses that apparently reached the sampling sites via low-level transport.

Davidson *et al.* (1985) observed maximum Na concentrations at the end of their month-long sampling campaign (17 April–15 May 1983) when Dye 3 was influenced by “moist upslope flow” from the south east. Detailed examination of April 1989, at Dye 3 (Davidson *et al.*, 1993; Mosher and Jaffrezo, 1993) also shows a dramatic “sea salt” episode occurring during a similar meteorologic situation. It may be that this specific meteorologic situation characterizes most transport events bringing sea salt to the ice sheet in southern Greenland.

Our working hypothesis is that all of the species increasing with ^7Be (Table 4) are generally transported to South Greenland at levels above the planetary boundary layer. With the exceptions of Br and I, the major sources of these elements in the Arctic free troposphere are crustal and/or anthropogenic, and are

thousands of km from Dye 3 (Heidam, 1984; 1985; Barrie *et al.*, 1989; Landsberger *et al.*, 1990). It might be expected that Br and I would be associated with Na and Cl, given their common dominant marine source (Berg *et al.*, 1984; Barrie *et al.*, 1989; Li and Winchester, 1989a; 1989b). However, like ^{210}Pb , aerosol Br and I can be derived from gaseous precursors, which may be more readily mixed upward into the free troposphere than sea salt aerosols (Mosher and Jaffrezo, 1993).

Short episodes of ^7Be concentrations >100 fCi SCM^{-1} occurred throughout DGASP, but were more frequent in the spring and late summer–fall (Fig. 1). These two seasons also contained the fewest intervals of low (<50 fCi SCM^{-1}) ^7Be concentration. This pattern is clearly reflected in the volume-weighted monthly average concentrations of both ^7Be and ^{210}Pb at Dye 3 (Fig. 3). It is interesting to note that, on a monthly basis, ^7Be and ^{210}Pb aerosol concentrations are highly correlated at Dye 3, and Barrow, ($r=0.80$, significant at $p=0.002$, and $r=0.89$, significant at $p=0.001$, respectively). However, the seasonal pattern at Dye 3 is distinct among the Arctic sites for which data are available. The sharp April peak for ^7Be and ^{210}Pb at Dye 3 comes at the end of the broad winter maximum seen at the other sites, and the broad peak from August–October is not observed at any of the other stations. Similar spring peaks at Dye 3, compared to extended winter–spring maxima at Arctic Basin sites, were also observed for SO_4^{2-} (Jaffrezo and Davidson, 1992) and crustally derived species (Mosher and Jaffrezo, 1993).

The observation that aerosol concentrations of ^7Be and continentally derived (natural and anthropogenic) species sharply increased together in spring suggests they are all reflecting increased exchange between the free troposphere and near surface air. On the other hand, Beer *et al.* (1991) invoke increased stratosphere/troposphere exchange in spring and fall to explain the observed seasonality of ^{10}Be in the snowpack at Dye 3.

Results of the NASA GTE ABLE 3A aircraft-based sampling campaign in the North American Arctic troposphere indicate important stratospheric influence on tropospheric chemistry during the summer (Browell *et al.*, 1992; Dibb *et al.*, 1992b; Gregory *et al.*, 1992; Jacob *et al.*, 1992; Talbot *et al.*, 1992; Wofsy *et al.*, 1992); and several studies have documented potentially significant injections of stratospheric air into the Arctic troposphere during the winter (e.g. Shapiro *et al.*, 1984; Raatz *et al.*, 1985; Oltmans *et al.*, 1989). Measurements of ^7Be and ^{10}Be in surface-level air at Alert, NWT, indicate that a stratospheric source is significant for ^7Be and dominates ^{10}Be concentrations in the Arctic troposphere throughout the year. During March and April, a 3-fold increase of stratospheric influence on the composition of surface air was estimated on the basis of the Be isotope concentrations and ratio (Dibb *et al.*, in review). Therefore, we feel that stratospheric influences on the Arctic troposphere are

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rations >100 fCi .SP, but were more nmer-fall (Fig. 1). he fewest intervals ntration. This pat- e-weighted month- ⁷Be and ²¹⁰Pb at o note that, on a sol concentrations | Barrow, (r=0.80, 9, significant at p e seasonal pattern tic sites for which peak for ⁷Be and f the broad winter d the broad peak ved at any of the is Dye 3, com- ma at Arctic .O₄²⁻ (Jaffrezo and d species (Mosher

centrations of ⁷Be nd anthropogenic) in spring suggests ange between the air. On the other sed stratosphere/- fall to explain the snowpack at Dye

3A aircraft-based American Arctic ratospheric influ- ring the summer 2b; Gregory *et al.*, il., 1992; Wofsy *et* ocumented poten- pheric air into the er (e.g. Shapiro *et* ans *et al.*, 1989). urface-level air at spheric source is Be concentrations t the year. During e ratospheric fac air was esti- pe concentrations refore, we feel that c troposphere are

most important in the spring, but are then relatively constant throughout the rest of the year.

The ground-level aerosol ⁷Be and ²¹⁰Pb seasonality at Dye 3 (Figs 1 and 3) strongly suggests that April–May and August–October are characterized by more efficient mixing of free tropospheric air to the surface than at other times (at least during the year of DGASP). The spring peaks in many of the trace elements measured by Mosher and Jaffrezo (1993) (Table 5) are also apparently at least partly due to these atmospheric processes. The fact that most of the trace elements with spring concentration peaks have reduced, or no, secondary peaks in the fall suggests important seasonality in the free tropospheric burden of these elements over Greenland, presumably due to complex interactions between variations in sources and large-scale atmospheric circulation patterns.

Snow

The concentrations of ⁷Be and ²¹⁰Pb in fresh snow during DGASP show considerable variability between events, but there is no clear indication of spring and late summer–fall peaks corresponding to the aerosol time series (Table 1 and Figs 1 and 3). It also does not seem that snowfall events consistently cause depression in the concentrations of the radionuclides in surface level air (Fig. 1), as would be expected if precipitation scavenging was a very efficient removal process. (It may be that the length of aerosol sampling intervals was generally too long to show such a relationship, if it was, in fact, operating.) The lack of coherence between snow and air chemistry suggests that the snow is generally “sampling” a different air mass than that at the surface. Uncertainties about the relationship between the chemistry of boundary layer and overlying air plague any surface-based atmospheric sampling campaign, particularly when attempts are made to directly compare the chemistry of surface air to precipitation. This may be particularly troublesome for studies on the Greenland Ice Sheet due to nearly complete lack of information about the structure and chemistry of the atmosphere directly above the ice sheet.

These considerations indicate that scavenging ratios (calculated after Davidson *et al.*, 1989) for individual snowfall events at Dye 3 are highly suspect, but the apparent tendency for ²¹⁰Pb scavenging ratios to be higher than those for ⁷Be is surprising (Table 1). The common association of these radionuclides with submicron aerosols suggests that they ought to be scavenged by similar processes. Todd *et al.* (1989) calculated mass-basis scavenging ratios for nearly 3 yr of precipitation data in Virginia that were, on average, two-times higher for ⁷Be than ²¹⁰Pb. They argued that the difference was largely due to using surface aerosol concentrations for the calculations, i.e. that ⁷Be probably increased more rapidly with height than ²¹⁰Pb, so that scavenging ratios calculated using aerosol concentrations at cloud height (if they were known) would show little or no difference between the two radionuc-

Table 5. Seasonality in the surface-level aerosol concentrations of the trace elements at Dye 3 (Mosher and Jaffrezo, 1993). The elements in Table 4 that are not included in this table had too many samples below detection limit to calculate meaningful monthly averages for the entire DGASP record

April–May peak	December–February peak
Al*, Mn*, Ca*, Sc*, Zn, Br, I	Na, Cl

* These elements had secondary concentration peaks in October.

lides. For Dye 3 there is no compelling reason to assume that ⁷Be vertical profiles differ greatly from those of ²¹⁰Pb, at least upto cloud height. However, any attempt to reconcile the calculated scavenging ratios would require invoking some combination of vertical profiles that resulted in the relative concentration of ²¹⁰Pb increasing more quickly with height than ⁷Be (or making the assumption that the air sampled by newly forming snow, whether above or upwind of Dye 3, is generally enriched in ²¹⁰Pb (or depleted in ⁷Be) relative to the air at the surface). A plausible mechanism to accomplish this is not readily apparent, implying that, at least for the year of DGASP, ²¹⁰Pb was more efficiently removed from the atmosphere by snowfalling at Dye 3 than was ⁷Be.

Given the foregoing discussion, it should not be surprising that neither the ²¹⁰Pb depth profile from the snowpit or the “deposition series” from fresh snow sampling (Fig. 2) reflects the seasonality in surface aerosol ²¹⁰Pb concentrations (Figs 1 and 3) very closely. The similar lack of correspondence between the aerosol and fresh snow concentrations of ⁷Be is probably also the result of significant differences between surface-level air and that sampled by snow. We must point out that the timing of annual peak concentrations in the air, fresh snow and snowpit records for SO₄²⁻ and dust indicators do tend to agree (Jaffrezo and Davidson, 1992; Mosher and Jaffrezo, 1993). However, the amplitude of the spring aerosol peaks for these species is much greater than their corresponding peaks in the snow, as well as being approximately an order of magnitude greater than the seasonal peaks in aerosol-associated radionuclide concentrations. Thus, part of the reason the snow does not reflect the seasonality in ⁷Be and ²¹⁰Pb aerosol concentrations may be a tendency for the snow to record a muted version of any atmospheric signal.

Another fundamental concern for the interpretation of chemical records from snow and ice is the question of how accurately the snowpack preserves any atmospheric signals delivered to it in the form of new precipitation. In this regard, the poor correspondence between the accumulated snow and what fell during the course of the study is disturbing (Fig. 2). The 4 cm sampling interval in the pit certainly often averaged layers from more than one depositional event and the fresh snow sampling may suffer from artifacts similar

to those discussed in relation to the fresh and aging snow study, but it is not obvious how the "deposition series" could be smoothed to make it resemble the pit profile. The snowpack may also have received dry deposition inputs that we would not expect to be reflected in the fresh snow samples, but it seems that the snowpack has less ^{210}Pb than was delivered in fresh snow. (Unfortunately, it is not possible to accurately estimate ^{210}Pb flux from the fresh snow samples because the area and volume of snow collected were not routinely recorded, but for the two months of June and July a rough estimate ranges 27–53 pCi m^{-2} (Dibb, 1990b), compared to total accumulation of 153 pCi m^{-2} for the entire year.) The snowpack at Summit, Greenland, also appears to retain considerably less ^{210}Pb than would be expected from observed deposition (Dibb, 1990b). Erosion by wind has been suggested as a possible explanation for the apparent deficit in ^{210}Pb accumulation compared to inputs (Dibb, 1990b). However, it is unclear if this is a viable mechanism or, more importantly, whether whatever combination of processes modifies snowpack chemistry before preservation is repeatable and predictable enough to deconvolve snow and ice records to recover the original signals in the fresh snow. These questions form one focus of our ongoing studies of air–snow transfer functions in the Summit region of the Greenland Ice Sheet.

SUMMARY AND CONCLUSIONS

Continuous aerosol sampling at Dye 3 for a one-year period revealed marked spring and fall peaks in the concentrations of ^7Be and ^{210}Pb in surface-level air. The pattern is quite different from multi-year averages at a number of Arctic sites in the EML network, all of which show broad concentration peaks for both radionuclides in the winter. At Dye 3 it appears that variation, over temporal scales ranging from days to seasonal, in the effectiveness with which free tropospheric air is mixed down to the surface of the ice sheet exerts a key influence on the concentrations of the atmospheric radionuclides in surface-level air. A suite of trace elements, mainly crustally and/or anthropogenically derived, tended to covary with the atmospheric radionuclides. This indicates that their surface-level concentrations are also strongly influenced by vertical mixing over the ice sheet, and suggests that the free troposphere may be the dominant pathway for their long-range transport to South Greenland.

The concentrations of ^7Be and ^{210}Pb in fresh snow showed no obvious spring and fall peaks, in contrast to their ground-level aerosol concentrations. This suggests that assessing fractionation of aerosol-associated species between the atmosphere and fresh snow on the basis of aerosol samples collected at the surface must be attempted with caution. Careful examination

of high frequency atmospheric samples, representative fresh snow samples, information about the height of snow formation, and the specific meteorologic situation during a given snow event should allow better quantitative comparison of air and snow chemistry. However, general comparisons between surface-level air and fresh snow on the Greenland Ice Sheet will often be qualitative at best. Given these caveats, it appears that ^{210}Pb may be removed from the atmosphere over Greenland, by both precipitation and dry deposition, more rapidly than ^7Be . This preliminary finding will certainly be examined in our ongoing studies in the Summit region of Greenland.

This study also indicated that the accumulation of ^{210}Pb in the snowpack near Dye 3 did not closely reflect a time series of ^{210}Pb concentrations in fresh snow samples collected through the year. However, the snowpit depth profile was quite similar to several profiles for pits recently sampled in the area. The snowpit findings provide some reassurance that the DGASP year was not anomalous, but the discrepancy with the fresh snow sampling raises the possibility that post-depositional modification of snow chemistry may significantly alter atmospheric signals contained in fresh snow sampling raises the possibility that post-depositional modification of snow chemistry. Verification, and quantification, of such processes is essential in order to fully exploit the wealth of paleoclimatic and past atmospheric chemistry information preserved in the snow and ice of the Greenland Ice Sheet.

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