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Chemical species spatial distribution and relationship to elevation and snow accumulation rate over the Greenland Ice Sheet

Q. Yang,¹ P. A. Mayewski,¹ E. Linder,² S. Whitlow,¹ and M. Twickler¹

Abstract. Major chemical species (Cl^- , NO_3^- , SO_4^{2-} , Na^+ , K^+ , Mg^{2+} , Ca^{2+}) from 24 snowpits (sampled at a resolution of 3 cm, total 2995 samples) collected from northern, central, and southern Greenland were used for this investigation. The annual and seasonal (winter and summer) concentration of each chemical species was calculated and used to study the spatial distribution of chemical species over the central portion of the Greenland Ice Sheet. A two-sided t -distribution test ($\alpha=0.05$) suggests that concentrations of major chemical species in snow do not vary significantly over this portion of central Greenland. The relationship between chemical concentration and snow accumulation rate was investigated using annual data from two groups of snowpits: those from coastal sites (northern and southern Greenland); and those from high-altitude inland sites (central Greenland). The snowpit data from a single group, when examined independently of the other group, show that chemical concentrations do not vary with snow accumulation rate. However, when data from the two groups are integrated into a single data set, pseudorelationships appear, with NO_3^- concentration decreasing and Na^+ , K^+ , Mg^{2+} , and Cl^- increasing as snow accumulation rate increases. Therefore we suggest that it is improper to study the relationship between chemical concentration and snow accumulation rate by using data collected from different geographic sites. The relationship between elevation and chemical concentration was investigated using the same suite of annual data sets. We find that Cl^- , Na^+ , and Mg^{2+} concentrations decrease, while NO_3^- concentration increases, with increasing elevation on the Greenland Ice Sheet.

Introduction

Ice cores are an important tool for reconstructing past climatic and environmental change, and investigation of variations in chemical concentration snow and ice plays an important role in the interpretation of resultant records. In general, the concentration of a given chemical species in snow is determined by its sources, transportation pathways, and scavenging processes. For example, chemical species with marine sources tend to decrease with elevation in Greenland snow [Herron, 1982; Osada and Langway, 1993]. Gaseous species, such as HNO_3 and H_2O_2 , which are strongly influenced by atmospheric photochemistry, have significant postdepositional exchange between snow/firn and the atmosphere [Mayewski and Legrand, 1990; Bales and Wolff, 1995; Wolff, 1995; Yang *et al.*, 1995]. Many of these influen-

tial factors operate together; thus it is difficult to distinguish which is the controlling factor for the resultant chemical concentration in snow.

Previous studies of the spatial distribution of major cations and anions in Greenland snow have only been conducted on a regional basis [Mayewski *et al.*, 1987; 1990; Davidson *et al.*, 1989], while other studies have only focused on selected sections of ice cores from Greenland [Herron, 1982; Steffensen, 1988; Osada and Langway, 1993]. Notably, Herron [1982] and Osada and Langway [1993] used several ice cores of different ages from Greenland to suggest that concentrations of nitrate and non-sea-salt sulfate have a linear relationship to snow accumulation rate. However, Yang *et al.* [1995] reported that the chemical concentration of the major ions in central Greenland snow and ice demonstrate no predictable relationship to snow accumulation rate. The discrepancy may be because Herron [1982] and Osada and Langway [1993] used data from different ice core sites covering different time periods, while Yang *et al.* [1995] used continuous ice core data covering the last hundred years from a single location.

Understanding the spatial distribution and factors which affect chemical concentrations over the Greenland Ice Sheet can improve our ability to interpret the representativeness of detailed, single-site studies like the

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Greenland Ice Sheet Project 2 (GISP2) ice core. With snowpits at different locations but covering the same period, we are able in this study (1) to statistically examine chemical species spatial distribution on a year-by-year basis, (2) to investigate the relationship between chemical concentration and elevation, and (3) to reevaluate the relationship between snow accumulation rate and chemical concentrations over the Greenland Ice Sheet.

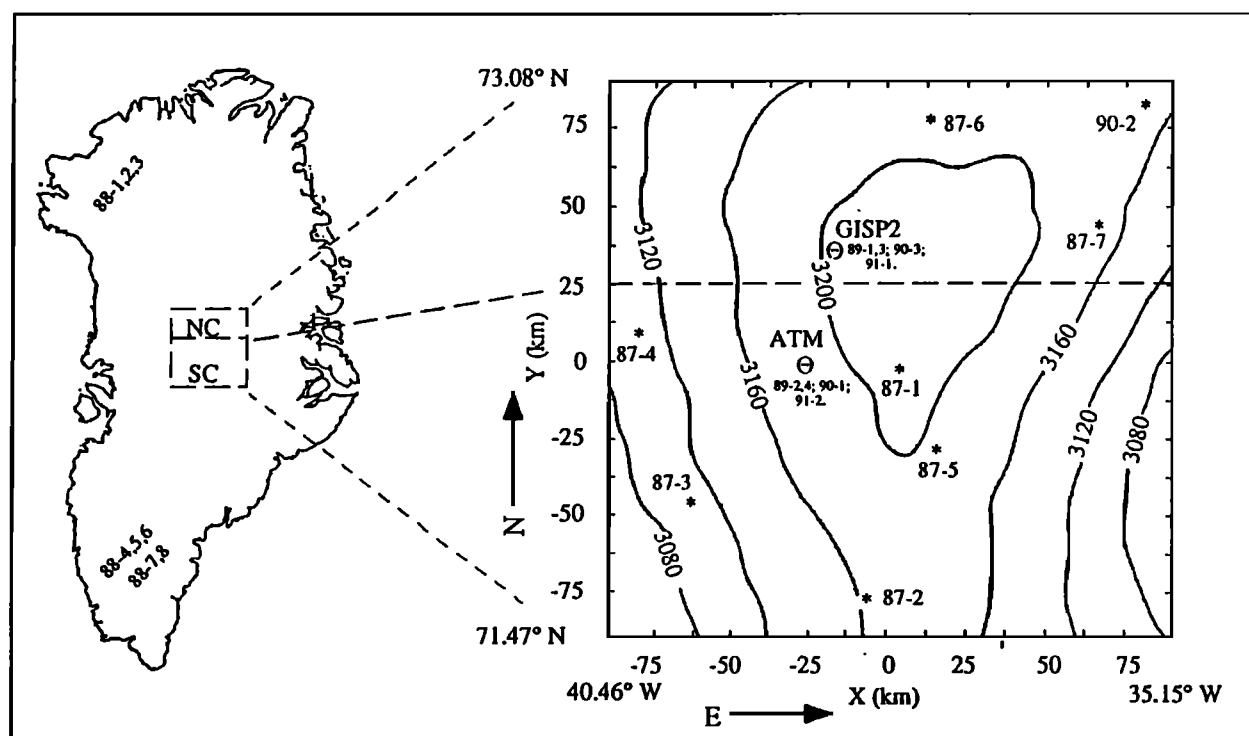
Methodology

Data retrieved from 24 snowpits were used in this investigation (Figure 1, Table 1). All snow pits in this study were sampled at a resolution of 3 cm (except for the 1990-ATM (see Figure 1) pit which was sampled at 5-cm intervals). To reduce significantly the possibility of sample contamination, all sampling tools and sample containers were precleaned using ultrapure water. During sampling, all personnel wore polyethylene gloves, nonparticulating clean suits, and masks. All samples were maintained frozen (in the field and during transportation to our laboratory) until analysis.

Measurements of the major anions and cations were performed by ion chromatography on a Dionex 4000 series instrument. Details of the technique are available in Whitlow *et al.* [1992].

The 3-cm sampling interval provides a high-resolution record (>12 samples per year) sufficient to document subseasonal inputs. Snowpit dating is based on oxygen isotope ($\delta^{18}\text{O}$) seasonal variations [Dansgaard *et al.*, 1973]. Oxygen isotope values for 1987 snowpits were provided by the Isotope Laboratory in Copenhagen [Mayewski *et al.*, 1990]. The rest of the oxygen isotope analyses were performed at the University of Washington. When the $\delta^{18}\text{O}$ data were not available or did not provide a clear seasonal cycle, seasonal signals in hydrogen peroxide (H_2O_2) [Sigg and Neftel, 1988] or the ratio between chloride and sodium (Cl/Na) [Whitlow *et al.*, 1992] were utilized.

Mean annual chemical concentration of each year was calculated by averaging all sample concentrations between two adjacent summer peaks. Because of the subseasonal sampling resolution we were able to calculate the seasonal concentrations of chemical species in each snowpit. For this purpose we identify the inflection points on the roughly sinusoidal annual oscillation of $\delta^{18}\text{O}$. Summer is taken to be all samples with $\delta^{18}\text{O}$ values higher (less negative) than the inflection points for each year. Winter is the other part of the $\delta^{18}\text{O}$ curve. Non-sea-salt chemical concentrations were calculated using Na^+ as the conservative marine source indicator [Legrand and Delmas, 1988; Mayewski *et al.*, 1990; Whitlow *et al.*, 1992]. Depth and period covered in each snowpit are presented in Table 1.



Legend

NC: north central Greenland; SC: south central Greenland; GISP2: Greenland Ice Core Project Two main camp (1989-1994); ATM: Atmospheric sampling site; *: snowpit site.

Figure 1. Location map of Greenland snowpits. Legend: NC, north central Greenland; SC, south central Greenland; GISP2, Greenland Ice Sheet Project 2 main camp (1989-1994); ATM, atmospheric sampling site; asterisk, snowpit site.

Table 1. Snowpit Information

Snowpit Year	Location, Lat., Long.	Depth, m	Number of Samples	Time Period Covered (yr A.D.)	Elevation, asl m	Mean Accuracy Rate, kg m ⁻² yr ⁻¹
<i>Northern Greenland</i>						
1988-1	77.30°N, 60.60°W	6.05	201	1981-1987	1850	293.1
1988-2	77.30°N, 59.20°W	3.74	124	1984-1987	1900	309.2
1988-3	77.75°N, 59.55°W	2.00	66	1986-1987	1900	254.3
<i>Central Greenland</i>						
1987-1	72.24°N, 37.67°W	6.00	200	1977-1986	3200	221.9
1987-2	71.57°N, 38.0°W	4.04	134	1982-1986	3160	249.4
1987-3	71.86°N, 39.66°W	2.00	66	1985-1986	3100	298.4
1987-4	72.35°N, 40.2°W	4.00	133	1982-1986	3100	267.1
1987-5	72.02°N, 37.28°W	4.04	134	1981-1986	3190	221.5
1987-6	72.95°N, 37.41°W	4.04	134	1979-1986	3180	168.9
1987-7	72.65°N, 35.28°W	4.04	134	1979-1986	3170	175.8
1989-1	72.6°N, 38.5°W	4.02	157	1983-1988	3200	219.1
1989-2	72.32°N, 38.8°W	4.02	157	1984-1988	3190	241.2
1989-3	72.6°N, 38.5°W	2.01	67	1987-1988	3200	189.2
1989-4	72.32°N, 38.8°W	2.04	68	1986-1988	3190	244.2
1990-1	72.32°N, 38.8°W	6.00	120	1981-1989	3190	235.3
1990-2	72.98°N, 35.28°W	4.05	135	1983-1989	3170	172.1
1990-3	72.6°N, 38.5°W	4.10	135	1984-1989	3200	213.8
1991-1	72.6°N, 38.5°W	2.25	75	1989-1990	3200	207.8
1991-2	72.32°N, 38.8°W	2.16	72	1988-1990	3190	252.4
<i>Southern Greenland</i>						
1988-4	66.72°N, 45.49°W	2.05	68	1986-1987	2200	300.1
1988-5	67.07°N, 45.57°W	4.00	134	1984-1987	2300	332.6
1988-6	67.34°N, 43.79°W	6.18	205	1982-1987	2400	361.5
1988-7	64.79°N, 44.29°W	4.20	140	1985-1987	2800	384.3
1988-8	65.64°N, 43.70°W	4.10	136	1986-1987	2600	566.8

Results and Discussion

Chemical Species Spatial Distribution

Chemical records from the 16 snowpits in central Greenland were examined to see if mean annual chemical concentrations are statistically homogeneous. Snowpits in central Greenland were divided into north central (NC) and south central (SC) (Figure 1). The mean annual concentrations and standard errors for the concentrations of Cl⁻, NO₃⁻, SO₄²⁻, Na⁺, K⁺, Mg²⁺, Ca²⁺ between NC and SC are presented in Table 2a. Since the variance within the two groups are not the same, a two-sided *t*-distribution test [Microsoft Corporation, 1992] with unequal variances was applied. A minimum of four snowpits are required in each group for this test. Hence it is clear from Table 1 that only three years (1984, 1985, and 1986) were suitable for testing. For all species and all years, a two-sided *t*-test ($\alpha=0.05$) shows no significant difference between the two groups.

The same statistical method was applied to the mean winter and summer concentrations of the species from NC and SC (Table 2b). Again, no significant difference between the groups was found. The two-sided *t*-distribution test was also conducted between concentra-

tions of chemical species from central and south Greenland for 1986 and 1987 (Table 2c). The *t*-test results suggest that concentrations of NO₃⁻, SO₄²⁻, Ca²⁺, and K⁺ are statistically similar between the regions in 1986 and 1987. Higher concentrations of Cl⁻, Na⁺, and Mg²⁺ in southern Greenland (Table 2c) than in central Greenland are statistically significant. We believe these differences are due to marine source proximity and elevation effect (Table 1).

However, it is apparent from Table 2 that variance in chemical concentrations within the groups is large. A recent surface snow chemistry spatial distribution survey conducted at the ATM station (Figure 1) shows that annual and seasonal chemical concentration under certain conditions can vary by as much as 30-40% on spatial scales of a few tens of centimeters [Dibb, 1996]. It is believed that such variations may be caused by physical or chemical processing while snow falls onto the glacier surface. The mechanisms of this process are not well understood yet. However, previous studies [Steffensen, 1988; Mayewski *et al.*, 1990] and our test results suggest that the GISP2 ice core chemical series record is representative of the annual and subannual glaciochemical signal for the central Greenland region. The small-scale temporal and spatial variation is averaged out and has

Table 2a. Mean Annual and Subannual Chemical Concentrations of Snowpit Group and Standard Errors

	Cl		NO ₃		SO ₄		Na		K		Mg		Ca		NC	SC
	SC	NC	SC	NC	SC	NC	SC	NC	SC	NC	SC	NC	SC	NC	n ₁	n ₂
86	16.2	20.3	6.0	127.4	78.1	106.7	5.7	6.7	0.9	1.2	1.5	1.5	10.5	11.8	4	4
Std. e.	1.9	4.1	5.4	18.5	5.5	24.9	1.0	1.6	0.2	0.4	0.2	0.5	4.1	5.0		
85	18.6	21.7	133.8	145.1	91.3	87.7	5.5	6.1	1.4	1.7	1.3	1.2	10.5	13.3	4	4
Std. e.	0.8	1.8	6.6	4.5	7.0	8.7	0.7	0.7	0.3	0.5	0.1	0.2	2.8	3.7		
84	16.9	18.2	157.0	156.7	106.6	106.3	4.9	5.7	1.9	1.3	2.1	1.6	14.7	13.9	4	4
Std. e.	1.2	0.8	10.2	8.8	8.3	5.6	0.8	0.9	0.4	0.3	0.1	0.4	2.6	0.8		

Std. e., standard error; SC, south central Greenland; NC, north central Greenland; SG, southern Greenland; CG, central Greenland; n₁ and n₂, number of samples used for calculating mean and *t*-test. (The difference between n₁ and n₂ is due to no measurements for Mg, K, Ca in snowpits 2, 3, 5 and 6, 1987.)

Table 2b. Mean Annual and Subannual Chemical Concentrations of Snowpit Group and Standard Errors

	Cl		NO ₃		SO ₄		Na		K		Mg		Ca		NC	SC
	SC	NC	SC	NC	SC	NC	SC	NC	SC	NC	SC	NC	SC	NC	n ₁	n ₂
89W	29.6	29.3	107.0	115.0	122.0	113.0	13.3	11.4	2.2	1.7	1.9	1.8	5.6	6.1	5	4
Std. e.	8.9	4.7	28.8	8.9	4.2	15.5	3.2	2.1	0.8	0.3	0.5	0.4	1.6	0.8		
87W	17.6	21.8	92.0	116.0	73.6	139.0	6.8	8.9	0.9	1.3	1.5	1.9	7.6	11.4	5	4
Std. e.	2.1	3.8	4.7	12.3	8.5	25.5	1.0	1.6	0.1	0.2	0.2	0.5	3.3	4.9		
86W	18.3	23.2	112.0	125.0	104.0	103.0	7.6	9.6	1.3	1.9	1.5	1.4	6.1	8.8	4	4
Std. e.	1.0	2.1	7.8	12.6	6.6	14.0	1.1	1.6	0.3	0.6	0.1	0.3	2.3	1.6		
85W	16.5	17.3	151.0	158.0	117.0	121.0	8.3	8.4	2.9	1.5	2.8	1.9	17.6	13.7	4	4
Std. e.	1.6	2.3	15.1	20.7	6.0	18.1	1.1	1.7	0.8	0.5	0.3	0.7	5.3	4.4		
86S	14.5	18.6	121.0	133.0	61.2	69.6	2.4	3.9	1.1	1.9	1.0	1.0	10.2	9.2	4	4
Std. e.	0.9	2.1	6.5	4.5	4.6	5.4	0.4	1.4	0.4	1.0	0.2	0.2	3.6	0.8		
85S	21.2	21.1	177.0	175.0	84.2	81.1	3.5	3.3	1.6	1.4	1.9	1.3	17.1	14.8	4	4
Std. e.	0.8	1.6	9.3	14.6	7.7	11.6	0.7	0.8	0.5	0.4	0.3	0.4	2.6	2.5		
84S	17.1	18.9	148.0	150.0	113.0	96.0	2.7	2.9	0.4	0.2	1.3	1.3	10.6	11.2	4	4
Std. e.	1.4	2.7	6.2	20.8	14.0	14.3	0.5	0.6	0.3	0.2	0.2	0.2	1.4	2.7		

W, winter; S, summer.

Table 2c. Mean Annual and Subannual Chemical Concentrations of Snowpit Group and Standard Errors

	Cl		NO ₃		SO ₄		Na		K		Mg		Ca		SG		CG	
	SG	CG	SG	CG	SG	CG	SG	CG	SG	CG	SG	CG	SG	CG	SG	CG	n2	n2
87	20.1	17.4	113.6	142.7	83.7	93.2	7.1	4.3	5	7	1.3	1.0	10.8	7.9	5	7		
Std. e.	1.7	0.9	15.6	8.4	13.0	6.0	0.4	0.5			0.1	0.1	1.7	0.7				
86	26.2	17.2	107.0	109.0	84.2	90.0	10.3	6.1	7	12	1.5	1.4	9.9	6.7	5	8		
Std. e.	2.9	2.0	8.7	9.1	10.2	10.9	1.1	0.8			0.2	0.3	1.7	0.8				

less impact on the mean annual and subannual chemical concentration in central Greenland.

Elevation Versus Chemical Species

Marine sea salt is believed to be a major source for Na⁺ and Cl⁻ observed in Greenland snow [Herron, 1982; Mayewski et al., 1984; 1990; Steffensen, 1988]. Changes in elevation can cause changes in sea-salt concentrations in snow because sea-salt aerosols are preferentially precipitated during air mass movement to higher altitudes. The locations of snowpits in this paper (Figure 1, Table 1) cover a range of 1850 to 3200 m above sea level. This allows investigation of the relationship between chemical species concentration and elevation using annual data sets.

Annual concentration of the major chemical species for the period 1984-1987 were generated from each snowpit and plotted against elevation (Figure 2). Since the snowpits cover different time periods (Table 1), the number of data points varies with elevation. Figure 2 shows that annual concentrations of Na⁺, Cl⁻, and Mg²⁺ decrease, while NO₃⁻ concentration increases, with increasing elevation.

The trends for Na⁺ and Cl⁻ in this study agree with those previously reported by Herron [1982] and Legrand and Delmas [1988]. To explain the similar trend of Mg²⁺, we calculated mean annual non-sea-salt concentrations of Cl⁻, Mg²⁺, Ca²⁺, K⁺, and SO₄²⁻ for all snowpits using Na⁺ as the conservative marine source indicator. The sea-salt component accounts for 61.9, 65.5, 3.5, 19.3, and 2% of Cl⁻, Mg²⁺, Ca²⁺, K⁺, and SO₄²⁻, respectively. It is apparent that Mg²⁺ is mainly (65.5%) derived from sea salt, while the sea-salt component of the other species is less than 20%. This finding is also supported by Keene et al. [1986], who considered Mg²⁺ to be a marine reference element in the study of the non-sea-salt chemical content of marine aerosols.

To explain the increasing nitrate concentration with increasing elevation over the Greenland Ice Sheet, it is necessary to understand the source and formation of nitrate in the atmosphere over Greenland. Although the relationship between measured nitrate concentration in snow and that of NO_y in the ambient atmosphere is far from well understood [Wolff, 1993; Dibb, 1994], it is clear that NO_x and OH radicals play important roles in the formation of HNO₃ [e.g., Logan, 1983; Warneck, 1988]. The major sources of NO_x in the northern hemisphere troposphere include combustion of fossil fuels, lightning at high altitude/low latitude, NO exhalation from soils, and stratospheric inputs [Logan, 1983; Mayewski et al., 1990]. The precursor of nitrate in Greenland snow is believed to be transported through the upper troposphere/lower stratosphere [Finkel et al., 1986; Yang et al., 1995], and the OH radical concentration shows an increase with altitude in high latitudes [Warneck, 1988]. Thus nitrate concentrations in snow would display a trend that increases with elevation over the Greenland Ice Sheet (Figure 2). This finding is also supported by Mulvaney and Wolff [1994], who demonstrated that nitrate concentration increases with increasing elevation over Antarctica.

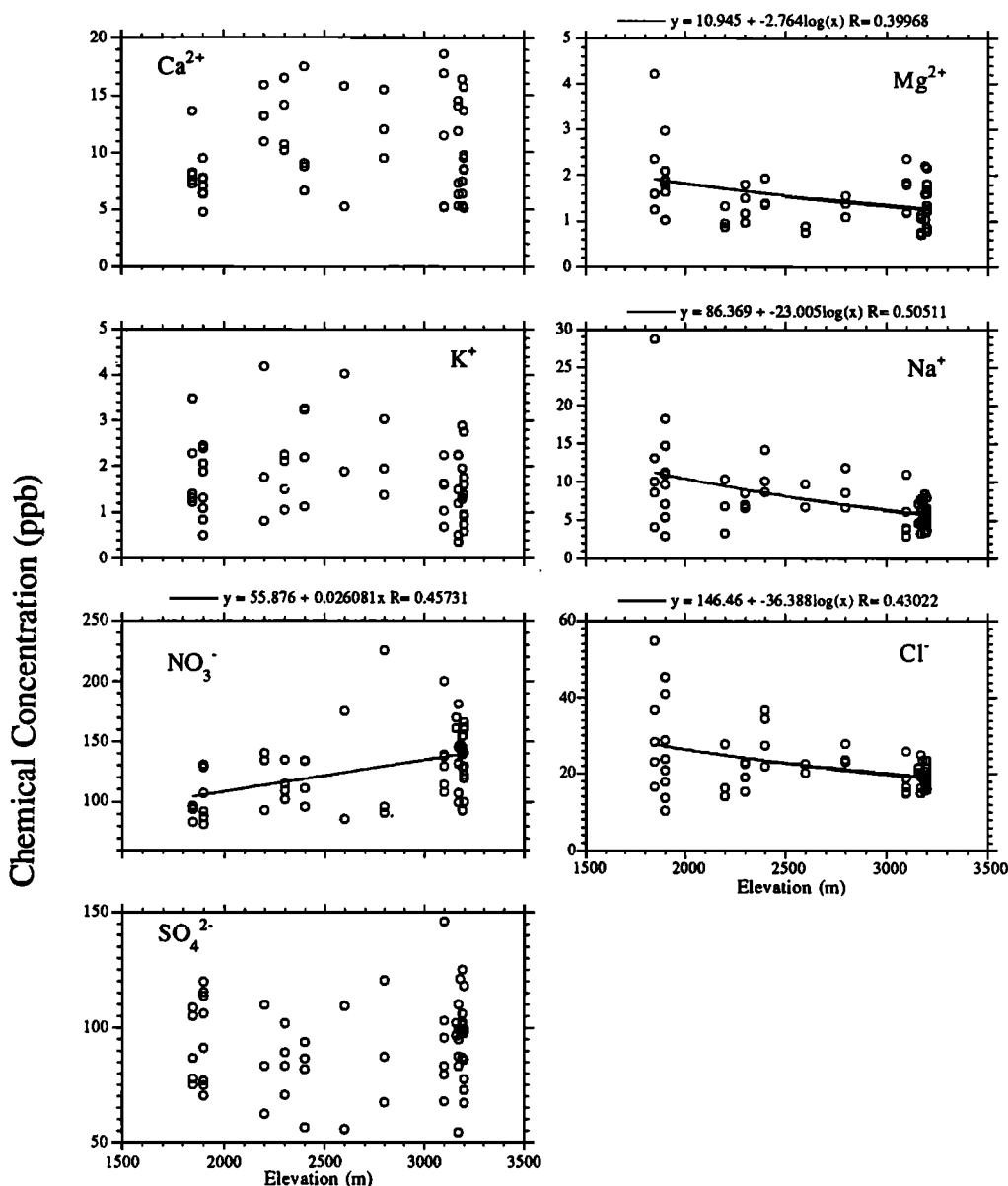


Figure 2. Annual chemical concentration (ppb) of major ions versus elevations above sea level (meters) from snowpits over the Greenland Ice Sheet for the period 1984-1987. (Each point represents individual annual chemical data and the elevation of the snowpit. Data sets from each snowpit vary in terms of the covered period.)

Concentration Versus Accumulation Rate

The relationship between the concentration of chemical species and snow accumulation rate is important since it can improve our ability to interpret deep ice core chemical data. For instance, changes in concentrations of chemical series from an ice core may be interpreted as (1) changes in atmospheric circulation and source regions, if chemical concentration is independent of snow accumulation rate; (2) changes in snow accumulation rate, if chemical concentration depends on snow accumulation rate. With annual concentration and annual snow accumulation data covering the Greenland Ice Sheet, we investigate the relationship between chemical concentration and snow accumulation rate in central Greenland and over the Greenland Ice Sheet for

the period 1984 to 1987. Using the data from snowpits located in central Greenland, scatterplots of concentration of Cl^- , NO_3^- , SO_4^{2-} , Na^+ , K^+ , Mg^{2+} , and Ca^{2+} against snow accumulation rate are presented in Figure 3 (open squares). As observed by Yang *et al.* [1995], there is no predictable relationship between chemical concentration and snow accumulation rate in central Greenland snow.

Chemical species concentration and snow accumulation data from snowpits located in southern and northern Greenland are also plotted on Figure 3 (solid triangles). No clear relationship is observed between the concentration of chemical species and snow accumulation rate for all chemical species studied if we examine only the data from southern and northern Greenland. However, by considering central, northern and southern

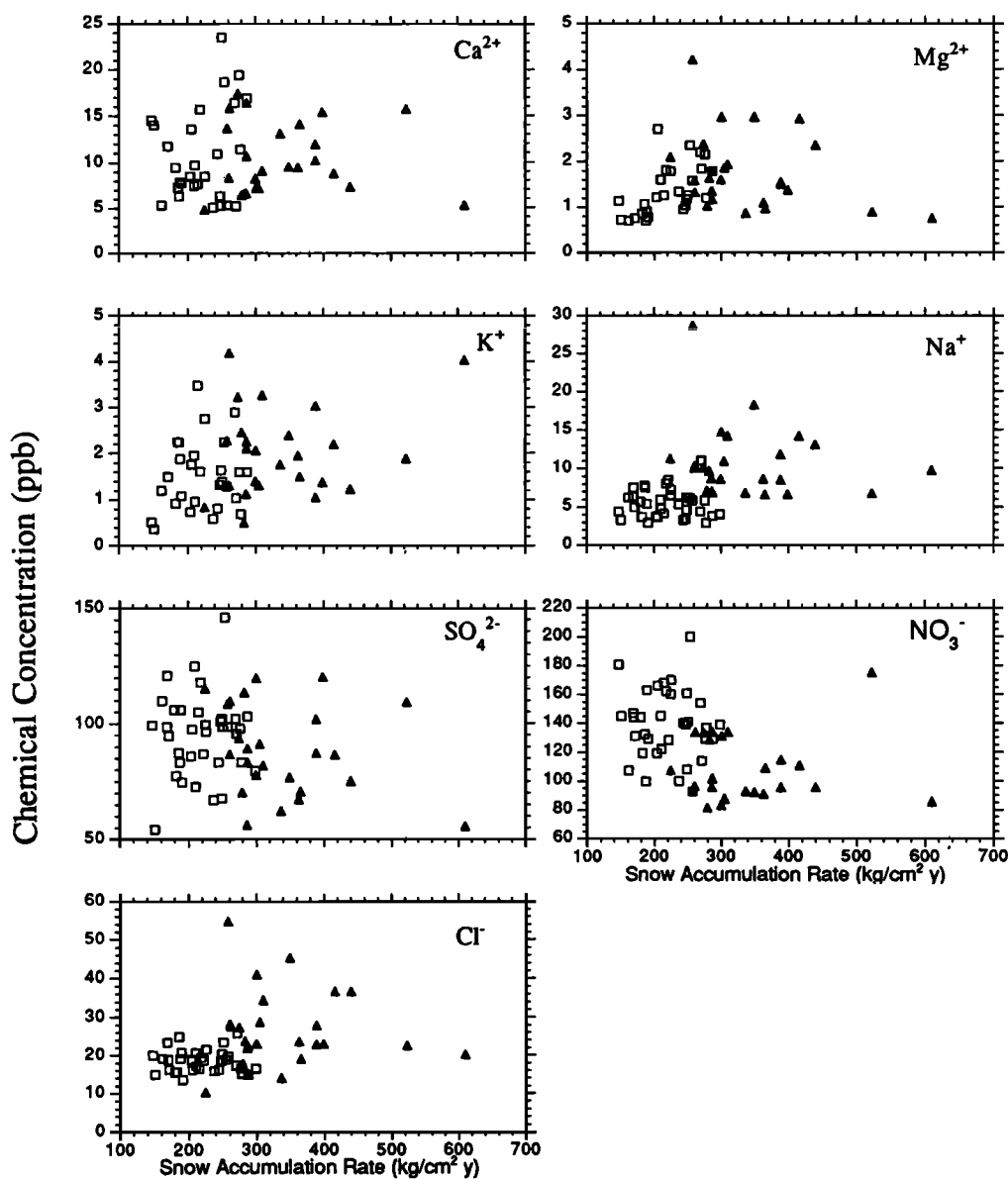


Figure 3. Annual chemical concentration (ppb) of major ions versus snow accumulation rate ($\text{kg m}^{-2} \text{yr}^{-1}$) from snowpits over the Greenland Ice Sheet for the period 1984–1987. (Each point represents individual annual chemical data and the accumulation rate of the snowpit. Open squares represent data collected from snowpits in central Greenland; solid triangles represent data collected from snowpits in southern and northern Greenland.)

data, a decrease in NO_3^{-} concentration with increasing snow accumulation rate is apparent as reported by *Herron* [1982] and *Osada and Langway* [1993] in Greenland and *Mulvaney and Wolff* [1994] in the Antarctic. Concentrations of Cl^{-} , Na^{+} and Mg^{2+} increase slightly with increasing snow accumulation rate considering all data in Figure 3.

We suggest that the major cause for the increase in NO_3^{-} concentration with decrease in snow accumulation rate in Figure 3 is not the accumulation dilution effect reported by *Herron* [1982], but source and transport pathways. Figure 3 reveals that the mean nitrate concentration in central Greenland (open squares) is higher than the mean nitrate concentration in southern and northern Greenland (solid triangles). As discussed

previously for Figure 2, NO_3^{-} concentration is higher in central Greenland than in southern and northern Greenland because the source of NO_3^{-} is from the upper troposphere/lower stratosphere. A pseudorelationship between chemical concentration and snow accumulation rate could appear (Figure 3) if mixed data, collected from different geographic sites, were put together without considering other factors (e.g., source, transport pathway, and elevation). These pseudorelationships would imply that Cl^{-} , Na^{+} , and Mg^{2+} concentrations increase, while NO_3^{-} concentrations decrease, with increasing snow accumulation rate over the Greenland Ice Sheet. However, accumulation rate on the Greenland Ice Sheet is characteristically inversely related to elevation. We also learned from Figure 2 that Cl^{-} ,

Na^+ , and Mg^{2+} concentration in snow collected from lower elevations of southern and northern Greenland are elevated due to proximity to marine sources. Thus the pseudotrends which show that Cl^- , Na^+ , K^+ , and Mg^{2+} concentrations increase, while NO_3^- concentrations decrease, with increasing snow accumulation rate over the Greenland Ice Sheet are actually a reflection of source, transportation pathway, and elevation, rather than accumulation rate. The influence of source, transport pathway, and elevation on chemical concentrations cannot be ignored when investigating the relationship between chemical concentration and snow accumulation rate using geographically mixed data.

Therefore we suggest that, when studying the relationship between chemical concentration and snow accumulation rate, it is important to constrain the investigation to an area over which geographic conditions and sources do not vary significantly. For example, a detailed ice core record covering a period with no major climate and environmental changes may be suited for such study [Legrand and Kirchner, 1990; Yang et al., 1995]. Otherwise, if other influencing factors are not considered, a pseudorelationship could appear between chemical concentration and snow accumulation rate.

Summary

In this paper we used 24 snowpits from northern, central, and southern Greenland, and covering the same time period, to reevaluate the spatial distribution of chemical species concentration, snow accumulation dependence, and the influence of elevation.

Instead of comparing the multiyear mean values of chemical species [Mayewski et al., 1990], we applied a two-sided *t*-distribution test to determine if annual and seasonal concentrations are statistically the same between northern and southern areas within central Greenland for the years 1984 through 1987.

The relationship between elevation and chemical species concentration was examined in detail using the same data sets. Cl^- , Na^+ , and Mg^{2+} decrease with increase in elevation, while nitrate concentration increases with increase in elevation.

The relationship between snow accumulation rate and concentration of chemical species was investigated using two groups of data: central Greenland data and southern and northern Greenland data. None of the chemical species show a predictable relationship to snow accumulation rate within a single group. An apparent relationship between chemical concentration and snow accumulation rate was observed by combining two groups of data. However, when factors such as source and transport pathways are introduced, we conclude that accumulation rate is not a major control on chemical concentrations over the Greenland Ice Sheet.

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