Relationships between synoptic-scale transport and interannual variability of inorganic cations in surface snow at Summit, Greenland: 1992-1996

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Abstract. To fully utilize the long-term chemical records retrieved from central Greenland ice cores, specific relationships between atmospheric circulation and the variability of chemical species in the records need to be better understood. This research examines associations between the variability of surface snow inorganic cation chemistry at Summit, Greenland (collected during 1992-1996 summer field seasons) and changes in air mass transport pathways and source regions, as well as variations in aerosol source strength. Transport patterns and source regions are determined through 10-day isentropic backward air mass trajectories during a 1 month (late May to late June) common season over the 5 years. Changes in the extent of exposed continental surfaces in source regions are evaluated to estimate aerosol-associated calcium and magnesium ion source strength, while forest fire activity in the circumpolar north is investigated to estimate aerosol ammonium ion source strength. During the 1995 common season, 3 times more calcium and magnesium accumulated in the snowpack than the other study years. Also, an increasing trend of ammonium concentration was noted throughout the 5 years. Anomalous transport pathways and velocities were observed during 1995, which likely contributed to the high levels of calcium and magnesium. Increased forest fire activity in North America was concurrent with increased levels of ammonium and potassium, except for 1996, when ion levels were above average and forest fire activity was below average. Because of the ubiquitous nature of soluble ions, we conclude that it is very difficult to establish a quantitative link between the ion content of snow and firn at Summit and changes in aerosol source regions and source strength.

1. Introduction

High-resolution chemical and physical measurements from polar ice cores are frequently used to reconstruct past atmospheric composition, temperature, and circulation patterns [e.g., *Mayewski et al.*, 1997; *White et al.*, 1997; *Legrand*, 1995; *Alley et al.*, 1993; *Taylor et al.*, 1993]. Glaciochemical records retrieved from Summit, Greenland (72.58 °N, 38.48 °W; 3207 m elevation) during the Greenland Ice Sheet Project 2 (GISP2) and the European Greenland Ice Core Program (GRIP) serve as proxy records of atmospheric composition and climate variability over the past ~110,000 years [e.g., *Meeker et al.*, 1997; *Laj et al.*, 1997]. Variations in ionic species measured in the Summit ice cores have been attributed to changes in the size and shape of the circumpolar vortex, the relative intensity of atmospheric circulation, and changes in aerosol source type and source region (e.g., continental, marine, and biogenic) [*De Angelis et al.*, 1997; *Taylor et al.*, 1996; *O'Brien et al.*, 1995; *Mayewski et al.*, 1994].

While a great deal of insight into past atmospheric conditions has been gained from the GISP2 and GRIP chemical records, specific relationships between the composition of the overlying air and the ice are quite complex, making quantitative estimates of past conditions difficult. In order to fully utilize the long chemical records retrieved from the ice cores, the transfer function between air and snow, snow and firn (the transitional material between snow and glacial ice), then firn and ice needs to be more clearly defined [*Dibb and Jaffrezo*, 1997]. The transfer function of chemical constituents between air and snow involves many physical and chemical processes, some of which can cause significant fractionation: the relative importance of dry, wet, and vapor deposition will differ between the chemical species present in the atmosphere over Summit. Variations in the chemical composition of overlying air masses result from horizontal and vertical transport from a variety source regions and source types [*Bales and Wolff*, 1995]. In an effort to improve the understanding of how variations in aerosol sources and transport may impact snow chemistry, this study focuses on linking interannual variability of certain inorganic cations in surface snow to changes in air mass transport pathways and source regions, as well as changes in aerosol source strength. We are therefore attempting to determine if changes in transport and sources dominate the variability of surface snow chemical signals at Summit.

2. Background and Objectives

2.1. Summit Ice, Firn, and Snow Records

The GISP2 and GRIP ionic records show that, during the last glacial maximum, continental dust (as reflected by water-soluble

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calcium (Ca^{2+})) increased approximately tenfold during stadial (cold) compared to interstadial (milder) periods [*Mayewski et al.*, 1997; *Fuhrer et al.*, 1993]. *Mayewski et al.* [1994] suggest that the increases in dust during stadials reflect periods during which the circumpolar vortex expanded southward to include new source regions not covered by ice, aridity increased, and/or circulation intensified over source regions. Holocene (~11,600 years before present (ybp) to present) levels of Ca^{2+}, on the other hand, vary by a factor of 2-3 interannually, which has been attributed to general changes in source regions and/or transport pathways [*O'Brien et al.*, 1995; *Fuhrer et al.*, 1993]. In comparison, subannual records of Ca^{2+} from Summit snowpits and firn cores (5-100 year records), sampled at high resolution (10-20 samples per year), display strong seasonal patterns consisting of springtime peaks that are as much as 50 times greater than baseline values and vary interannually by 2-3 times within most records [e.g., *De Angelis and Legrand*, 1994; *Silvente and Legrand*, 1993; *Whitlow et al.*, 1992]. It is important to note that even though the Holocene portion of the GISP2 and GRIP ice cores and the more recent snowpit/firn core Ca^{2+} records indicate the same magnitude of interannual variability, concentration variability could have been partly smoothed in the ice core records due to a coarser sample resolution (e.g., bi-annually for GISP2 [*O'Brien et al.*, 1995]).

In contrast to the two-stage (stadial and interstadial) Ca^{2+} record through the last glaciation, GISP2 and GRIP ice core ammonium ion (NH_4^+) concentrations display low levels throughout the glacial portion of the core (~15,000 ybp – 110,000 ybp). The low levels have been attributed to decreased biological activity resulting from ice coverage extending over most source regions [*Meeker et al.*, 1997; *Fuhrer et al.*, 1996]. During the glacial-to-interglacial transition, the frequency of large sporadic enhancements (spikes), and background NH_4^+ levels began to increase, reflecting the retreat of the North American and Eurasian ice sheets [*Fuhrer et al.*, 1996; *Legrand and De Angelis*, 1996]. Maximum NH_4^+ values were reached at ~12,000 ybp, decreasing threefold to fourfold from around the beginning of the Holocene to the present [*Meeker et al.*, 1997; *Fuhrer et al.*, 1996].

Spring, summer, and fall background levels of NH_4^+ at Summit are primarily derived from mid-latitude to high-latitude continental biological production and diffuse forest fire emissions, while spikes of NH_4^+ normally indicate short-term forest fire inputs from the circumpolar north [*Savarino and Legrand*, 1998; *Legrand et al.*, 1995; *Legrand and De Angelis*, 1996; *Whitlow et al.*, 1994]. Gaseous NH_3 from both biomass burning and biological production is normally incorporated into submicron acid aerosols and cloud water droplets in the boundary layer and lower troposphere and can subsequently be transported long distances to remote regions (like Summit) [*Seinfeld and Pandis*, 1998]. Recent airborne sampling campaigns in the Arctic and subarctic regions of Alaska, Canada, and Greenland suggest that the summer Arctic troposphere contains occasional elevated levels of biomass burning byproducts (e.g., carboxylic acids and NH_4^+), providing additional support for biomass burning being a major source of NH_4^+ at Summit [*Gorzelska et al.*, 1994; *Talbot et al.*, 1992; *Wofsy et al.*, 1992].

Subannually resolved records of NH_4^+ at Summit from snowpits and firn cores show concentration spikes superimposed on broad summer maxima [*Legrand et al.*, 1992, 1995; *Whitlow et al.*, 1994]. These spikes normally occur in summer and fall when the boreal forest fire season is at its peak [*Dibb et al.*, 1996]. *Legrand and De Angelis* [1995] and *Legrand et al.* [1992]

have suggested that a chemical fingerprint of boreal biomass burning inputs at Summit consists of the presence of a mixture of NH_4^+ and formate close to a molar ratio of 1, implying that the NH_4^+ is mainly present as ammonium formate. These authors also proposed that biomass burning aerosols are present in Greenland snow and ice layers when NH_4^+ concentrations are higher than 1 nmol g^{-1}. However, *Savarino and Legrand* [1998] demonstrate that seasonally resolved records often have summertime NH_4^+ concentrations above 1 nmol g^{-1}, even without direct inputs from forest fires. Consequently, *Savarino and Legrand* [1998] suggest that the "double condition" of both formate and NH_4^+ is needed to recognize biomass burning inputs in Summit snow, firn, and ice.

Along with changes in Ca^{2+} and NH_4^+, this study examines the interannual variability of magnesium ion (Mg^{2+}) and non-sea-salt (nss) potassium ion (K^+) in the surface snow samples. *O'Brien et al.* [1995] and *De Angelis et al.* [1997] estimate that 95%-99% of the Ca^{2+} and 50%-70% of the Mg^{2+} in Summit snow, firn, and ice are of continental origin, with the remaining amount attributed to sea-salt aerosols. During spring it is thought that the continental dust source for both species is very dominant [e.g., *Steffensen*, 1988].

Late spring, summer, and fall surface snow NH_4^+ and nss-K^+ at Summit are thought to have corresponding origins [*Dibb et al.*, 1996]. Spikes of nss-K^+ in the GISP2 record normally occur at the same time as NH_4^+ spikes, indicating that biomass burning is a predominant source of this ion at Summit [*Whitlow et al.*, 1994]. In addition, NH_4^+ and K^+ were among the major soluble ionic components of aerosols and surface snow that show large enhancements during a documented Canadian forest fire plume that was transported to and sampled at Summit in August 1994 [*Dibb et al.*, 1996]. Along with biomass burning sources, there is also a crustal component of nss-K^+ in Summit aerosol, snow, and ice [e.g., *Laj et al.*, 1997].

2.2. Study Objectives

Among the many measured parameters in ice cores used to reconstruct past climatic and atmospheric conditions, aerosol-associated species such as Ca^{2+}, Mg^{2+}, K^+, and NH_4^+ are thought to undergo little postdepositional modification [*Dibb*, 1996; *Bergin et al.*, 1994]. Knowledge of what controls the variability of the species in surface snow at Summit can therefore help interpret changes observed in ice cores. Consequently, the present study's primary objectives are to (1) link changes in surface snow Ca^{2+}, Mg^{2+}, K^+, and NH_4^+ content to changes in transport pathways and source regions as indicated by 10-day isentropic backward trajectories during a 5 year period (1992-1996) and (2) examine interannual variability of aerosol source strength to determine if tropospheric background levels of the ions of interest may have increased or decreased during the study period.

3. Methods

3.1. Surface Snow Collection and Analysis

Three replicates of surface snow were collected every day during five summer field seasons (season length in days: 1992 = 95; 1993 = 53; 1994 = 93; 1995 = 76; 1996 = 30). Field personnel wearing polyethylene gloves and nonparticulating clean suits gathered the samples into precleaned polyethylene containers. Careful attention was paid to collecting the entire thickness of the uppermost layer without including any of the

underlying layers. The "uppermost" layer consisted of the entire thickness of fresh snow on snowfall event days (typically 1.0 – 3.0 cm), and the top ~1 cm on other days. Detailed field notes on local meteorological conditions and physical characteristics of the surface snow layer sampled each day allowed discrimination between new layers and those that aged at the surface. The snow samples were weighed to provide the snow inventory (g cm^{-2}), which was then used to calculate the inventories of various ionic species (nmol cm^{-2}). Seasonal cumulative ion inventory is calculated by summing the daily inventories.

After being weighed, the samples were melted in the field and 7 mL aliquots were immediately refrozen and shipped to the University of New Hampshire for ion chromatographic analysis of eight major inorganic species (Ca^{2+}, Na^+, Mg^{2+}, K^+, NH_4^+, Cl^-, NO_3^-, and SO_4^{2-}) [Mayewski et al., 1987]. The concentrations of Ca^{2+}, Mg^{2+}, and K^+ were partitioned into sea-salt and non-sea-salt fractions using Na^+ as the sea-salt indicator and the seawater Ca^{2+}, Mg^{2+}, and K^+ to Na^+ bulk ratios [Whitlow et al., 1992]. We have neglected the continental component of Na^+ based on ionic analyses of the Holocene portion of the GISP2 and GRIP ice cores, which indicated that 95%-99% of total Na^+ was of marine origin [De Angelis et al., 1997; O'Brien et al., 1995].

3.2. Surface Snow Classification

Because both fresh and aged snow samples were collected, the snow layers had to be classified after collection. The daily samples were segregated into one of three categories: (1) fresh snow, (2) aged snow that was not buried as a result of drifting or substantial snowfall, or (3) buried layer (described below). To qualify as a fresh snowfall event, at least 1 cm of new snow had to be present at the sampling site. A slight dusting of snow was not considered a snowfall event, nor was clear-sky precipitation (diamond dust) or fog deposition.

Surface snow was classified postcollection as a buried layer if (1) snow had drifted substantially between daily sampling sessions, or (2) a snowfall event occurred between daily sampling. In either case, the snow sample from the day before the snowfall or the drifting was considered a buried layer. As a result, not all samples were used to calculate the cumulative ion inventory of buried layers, even though the surface snow was sampled every day. Buried layer ion inventory can therefore include wet deposition from large snowfall events, light dustings of snow, and diamond dust, along with any post-depositional additions (e.g., fog and dry fallout), as well as any postdepositional losses (e.g., sublimation and degassing) that may have occurred. By determining the cumulative ion inventory of buried layers, as opposed to the cumulative ion inventory of each daily sample, we are assured that the same layer of snow is not accounted for more than once when calculating the seasonal cumulative ion inventory for that year. While fresh snow events should bear a more direct relation to the overlying air, buried layers represent what would likely be incorporated into the glaciochemical record.

Interannual variations in surface snow ion content are established by comparing average soluble ion concentration (nmol g^{-1} H$_2$O) of fresh snow events and cumulative ion inventory (nmol cm^{-2}) of buried layers during the total sampling season, and a common sampling season of each year. The "common" season extends from May 22 to June 22 and is delineated based on the time period of the shortest sampling season (1996). Because of the contrast in both field season length and the relative time of year the seasons occurred, it would

be misleading to determine interannual variability of average fresh snow concentration and buried layer cumulative inventory based solely on samples collected during each entire field season. Therefore we decided that the common time period within the 5 years would better represent changes in spring to early summer snow chemistry at Summit from year to year.

3.3. Air Mass Transport Patterns and Source Regions

Ten-day isentropic backward trajectories were calculated twice daily (0000 and 1200 UT) using a model developed at the Climate Monitoring and Diagnostics Laboratory of the National Oceanic and Atmospheric Administration [Harris and Kahl, 1994]. The model estimates the three-dimensional transport pathways of air masses arriving at Summit at an altitude of 400 m above ground. Trajectory precision and spatial homogeneity of the flow are evaluated by producing five trajectories at each time interval (0000 and 1200 UT) within a 50 km radius centered at the Summit site [cf. Kahl et al., 1989]. A 10-day trajectory was chosen because shorter duration trajectories normally do not track the air mass history far enough back to include potentially important source regions, particularly in summer [Kahl et al., 1997; Davidson et al., 1993].

It is important to note that the accuracy of air mass trajectories is limited by the spatial and temporal resolution of the meteorological data on which they are based [Rolph and Draxler, 1990; Kahl and Samson, 1986]. In regions such as the Arctic, observational meteorological data are sparse, thus limiting the trajectory model considerably [Kahl, 1993]. In a comparison of three trajectory models ran in the Arctic basin, Kahl et al. [1989] report that median horizontal displacements between 5-day trajectory endpoints were primarily in the 800-1000 km range. It is also important to note that a modeled trajectory is an estimate of the transport pathway of an infinitesimally small air parcel and therefore does not take into account vertical and horizontal dispersion. Thus a trajectory should not be interpreted as a direct connection between a receptor and a source, but rather as the general transport pathway of an air mass [Kahl, 1993]. Given the known uncertainties and limitations of backward trajectories, they can nonetheless provide a useful qualitative estimate of the source regions and transport pathways of the air masses advected to Summit.

The trajectories produced for this study are manually classified into four regions in the circumpolar countries and the United States (Figure 1). Whenever three or more of the five trajectories had 10-day origins in a particular sector, a "hit" is recorded. By compiling hits throughout the 1-month common time period of each year, a seasonal total for each region is established and is reported as a percentage of the 60 sets of trajectories calculated. The vertical component of the trajectories is evaluated by noting when three or more of the five trajectories entered the planetary boundary layer ($\leq$ 1 km elevation) during their 10-day history. This information can help determine the likelihood of aerosol entrainment from potential source regions that the air mass passed over en route to Summit. The dominant horizontal and vertical patterns in the trajectories observed are then related to interannual variability of the surface snow chemistry. In addition, transport pathways and sources regions estimated by the trajectories produced for this study are compared to a 28-year record (1962-1989) of 700 mbar 10-day isobaric backward trajectories produced and analyzed by Kahl et al. [1997].
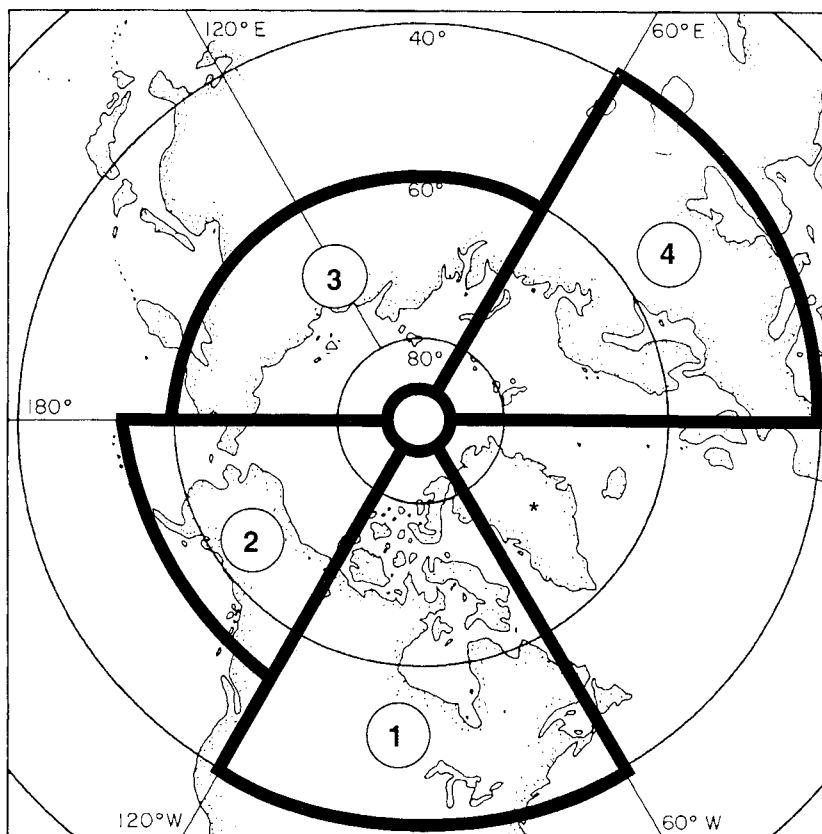
Figure 1. Study source regions for delineating origins of 10-day isentropic backward trajectories and snow cover extent during the common season 1992-1996. Numerals refer to 1, Canada/northern United States; 2, Yukon region; 3, Siberia; 4, western Eurasia.

4. Results

4.1. Variations in Total Season Surface Snow Ion Chemistry

Springtime maximum inputs of nss-Ca^{2+} and nss-Mg^{2+} are apparent in the time series of daily average surface snow chemistry for each year, with the values nearing baseline as sampling progressed into summer (Figure 2). This diagram also indicates the relative timing of snowfall events and buried layers throughout the 5 years of the study. It is likely that the total sampling season did not include the entire spring dust inputs during certain years. For example, during 1996 the nss-Ca^{2+} and nss-Mg^{2+} record shows that the spring maxima likely occurred earlier than the start of the field seasons (May 22). Because the 1995 field season started earlier in the year (April 24) than the 1996 season, a larger portion of spring inputs of the two species were included in the total sampling season time interval (Figure 2). This is also reflected in the nss-Ca^{2+} and nss-Mg^{2+} total season average concentration of snowfall events and cumulative inventory of buried layers during 1995, which are the highest values throughout the 5 years (Table 1).

The time series of daily average NH_4^+ and nss-K^+ concentrations of surface snow illustrates that these species are at or near baseline values during most of the sampling seasons (Figure 3). The total season mean ion concentration of snowfall events shows an increasing pattern from 1992 to 1996 in both species; in contrast, the cumulative ion inventory illustrates that 1992 and 1994 accumulated the highest amount of NH_4^+ and nss-K^+ during the 5 years (Table 1). However, 1992 and 1994

contain the greatest number of buried layers (31), leading to a higher amount of ions accumulating. In the daily average time series, occasional spikes occur during portions of the records. For example, the highest NH_4^+ value recorded (10.5 nmol g^{-1}) throughout the five years was observed in mid-summer of 1995 (Figure 3). Also during 1995, elevated levels of nss-K^+ occur throughout the season. Because the timing of some of these events coincides with nss-Ca^{2+} and nss-Mg^{2+} inputs (Figure 2), and the fact that NH_4^+ is near baseline, these nss-K^+ spikes are likely associated with crustal sources. In comparison, during the summer of 1994 both NH_4^+ and nss-K^+ display elevated levels that are likely due to biomass burning inputs. In fact, the spike in early August 1994 coincides with an event documented by *Dibb et al.* [1996] as being associated with Canadian boreal forest fire emissions that were efficiently transported to Summit. The authors observed anomalously high levels of NH_4^+, nss-K^+, and carboxylic acids in aerosols and water-soluble gases during this time period. These authors also observed enrichment in nss-Ca^{2+} and nss-Mg^{2+} in surface snow, which is also seen in Figure 2. Other investigations at Summit have revealed that elevated levels of nss-Ca^{2+} and nss-Mg^{2+} are occasionally present in ice layers impacted by biomass burning [e.g., *Savarino and Legrand*, 1998]. Trace element analysis of biomass burning emissions supports this observation by illustrating enrichment in Ca [*Echalar et al.*, 1995; *Susott et al.*, 1991]. The association of nss-Ca^{2+} and nss-Mg^{2+} with biomass burning emissions can be attributed to the remobilization of dust particles deposited on the vegetation, entrainment of soil dust in updrafts associated with
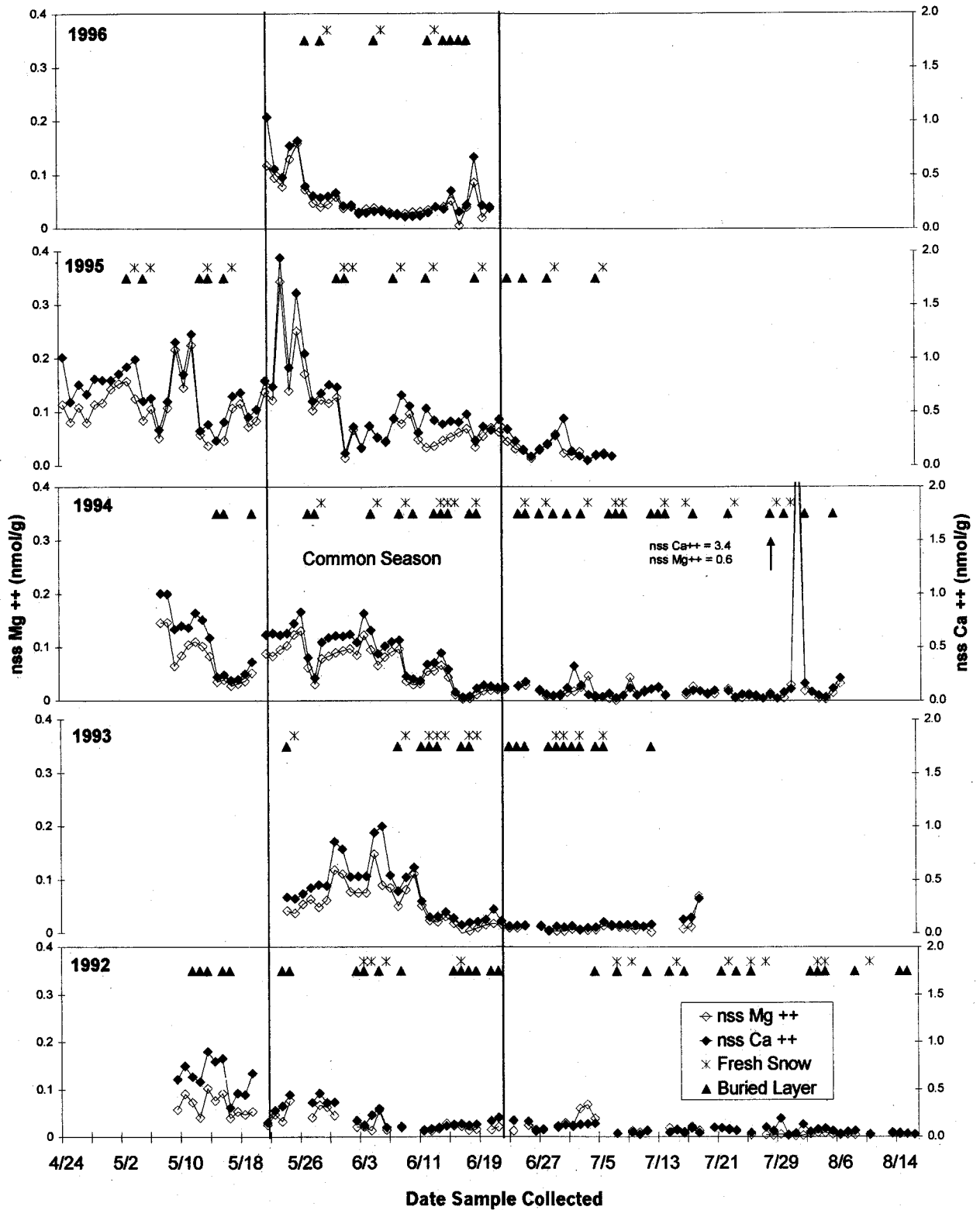
Figure 2. Total sampling season daily average concentration (≥ 3 replicates d^{-1}) of surface snow non-sea-salt (nss) calcium (Ca^{2+}) and nss-magnesium ion (Mg^{2+}). Also shown is the relative timing of the common season (May 22 to June 22) and the dates when fresh snow events and buried layers were recorded.

Table 1. Total Sampling Season Mean Ion Concentration of Snowfall Events and Cumulative Ion Inventory of Buried Layers

| Year | Number of Events | Fresh Snow, nmol g⁻¹ H₂O | | | | Number of Layers | Buried Layers, nmol cm⁻² | | | |
		Ca^{2+}	Mg^{2+}	NH_4^+	K^+		Ca^{2+}	Mg^{2+}	NH_4^+	K^+
1992	13	0.10 (0.02)[a]	0.020 (0.002)	0.22 (0.03)	0.024 (0.004)	31	2.17 (0.22)[b]	0.33 (0.04)	3.25 (0.50)	0.31 (0.05)
1993	11	0.13 (0.03)	0.021 (0.005)	0.26 (0.04)	0.043 (0.009)	18	0.88 (0.05)	0.18 (0.01)	1.30 (0.08)	0.17 (0.03)
1994	17	0.16 (0.04)	0.032 (0.007)	0.61 (0.09)	0.054 (0.009)	31	1.43 (0.08)	0.28 (0.01)	6.26 (0.36)	0.62 (0.09)
1995	11	0.45 (0.08)	0.07 (0.01)	0.68 (0.10)	0.053 (0.009)	14	3.25 (0.34)	0.61 (0.07)	2.04 (0.43)	0.20 (0.03)
1996	3	0.23 (0.04)	0.052 (0.003)	0.91 (0.26)	0.06 (0.01)	8	0.62 (0.03)	0.13 (0.01)	2.52 (0.11)	0.25 (0.02)

[a]Standard error.
[b]Standard deviation.

the fires, and because Ca and Mg are components of biomass [*Baker*, 1983].

4.2. Common Season Interannual Variability of Surface Snow Chemistry

The common season mean concentration of nss-Ca^{2+} and nss-Mg^{2+} in fresh snow displays a twofold increase from 1992 to 1995 (Figure 4a). Because ionic concentration in snow is influenced by the amount of water contained in the snow, it is necessary to consider whether the water content of the fresh snow has varied from year to year. If, for example, seasonal snowfall during a particular year contained an above average amount of water, this would cause the concentration of ionic species to be below average. There is, however, no statistical difference in the cumulative water mass of snowfall events between 1992-1996 (Table 2). Therefore a difference in ion concentration of fresh snow events between years likely reflects the atmospheric loading of the chemical species.

Similar to fresh snow concentration, the buried layer cumulative inventory of nss-Ca^{2+} and nss-Mg^{2+} shows the largest influx during 1995, despite 1992 recording the highest cumulative water mass and the highest number of buried layers during the 5 years (Figure 4b and Table 3). If atmospheric loading was roughly constant throughout the 5 years, one might expect that the greater water flux in 1992 would cause a depressed mean concentration in fresh snow, while enhancing the cumulative inventory, yet neither effect is evident. Because of the large contrast between 1992 and 1995 of nss-Ca^{2+} and nss-Mg^{2+} in both fresh snow and buried layers, we elected to study differences between these 2 years in detail in an attempt to link variations in air mass transport and source regions to the different surface snow chemical signatures.

The common season snowfall mean concentration of NH_4^+ displays an increase throughout the 5 years (Figure 5a), similar to what was calculated for the total sampling season (Table 1). The buried layer cumulative inventory of NH_4^+ and nss-K^+ shows an increase from 1994 to 1996 for NH_4^+ and an increase from 1995 to 1996 for nss-K^+. The different pattern in the nss-K^+ record likely reflects inputs from sources other than those contributing NH_4^+, such as continental dust aerosols. In 1992 the above average number of buried layers (12) likely contributed to an increase in NH_4^+ and nss-K^+ cumulative inventories (Figure 5b and Table 3), which does not appear in mean snowfall concentrations during that year (Figure 5a). From 1993 to 1995 there is no significant difference in the buried layer cumulative inventory of nss-K^+ (Figure 5b).

4.3. Air Mass Transport and Source Regions

Isentropic back trajectories indicate that at least 50% of the air masses transported to Summit during the common season had 10-day origins in North America, with the dominant region being Canada/northern United States (Table 4). In comparison, 88% of the 10-day isobaric (700 mbar) trajectories produced by *Kahl et al.* [1997] for spring and summer originated in North America. Transport from western Eurasia accounted for the bulk of the remaining trajectories during our study (~25% of all trajectories), compared to approximately 10% from Eurasia in the *Kahl et al.* [1997] study. Additionally, there were no trajectories originating in eastern Asia in our study, compared to an average of 7% of the spring/summer 700 mbar trajectories in the Kahl et al. study. Also, during the common seasons of 1992-1994 and 1996, greater than 20% of the trajectories had 10-day upwind origins in
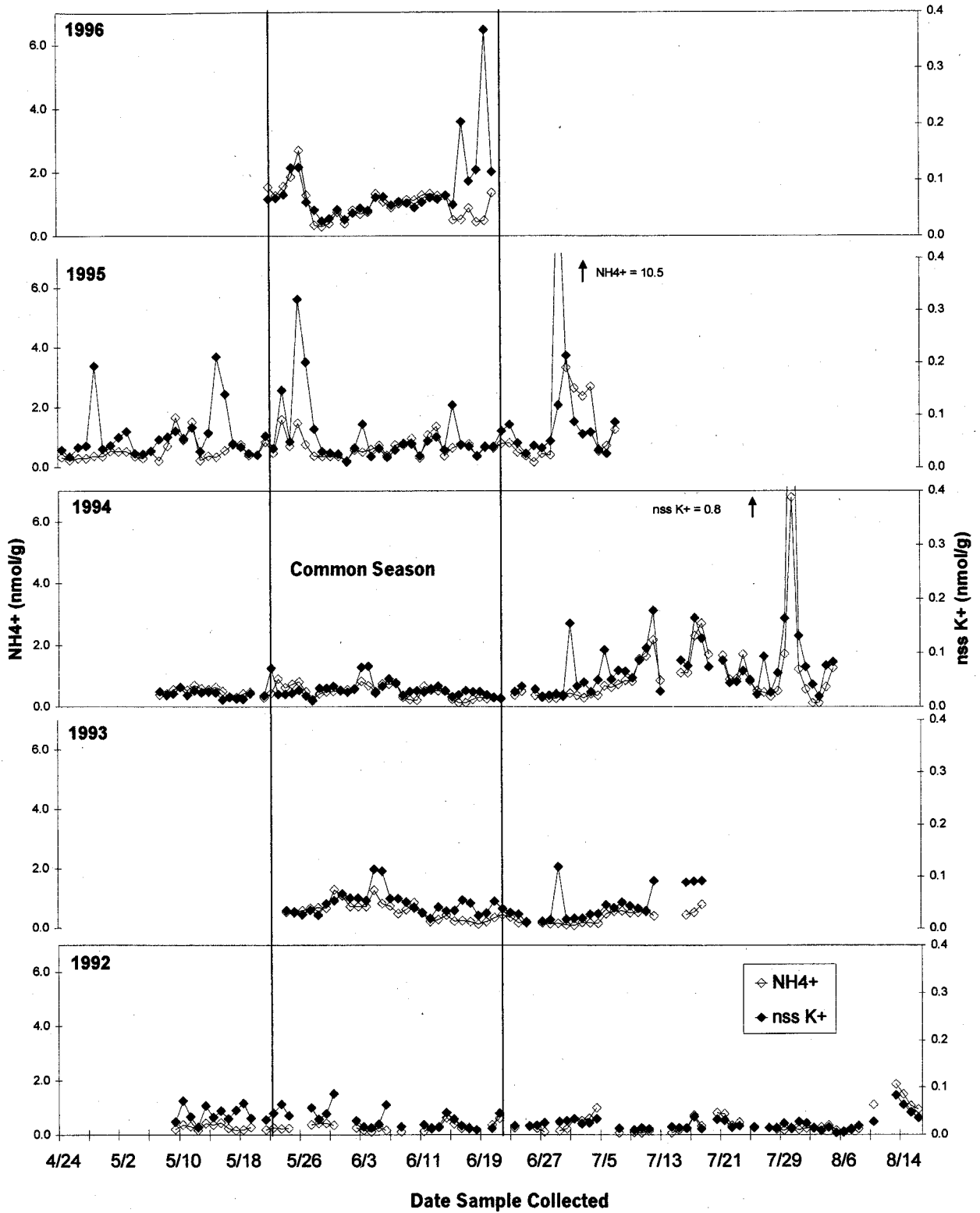
Figure 3. Total sampling season daily average concentration ($\geq$ 3 replicates d^{-1}) of surface snow ammonium (NH$_4^+$) and non-sea-salt potassium ion (nss-K$^+$). The common season (May 22 to June 22) is also shown.
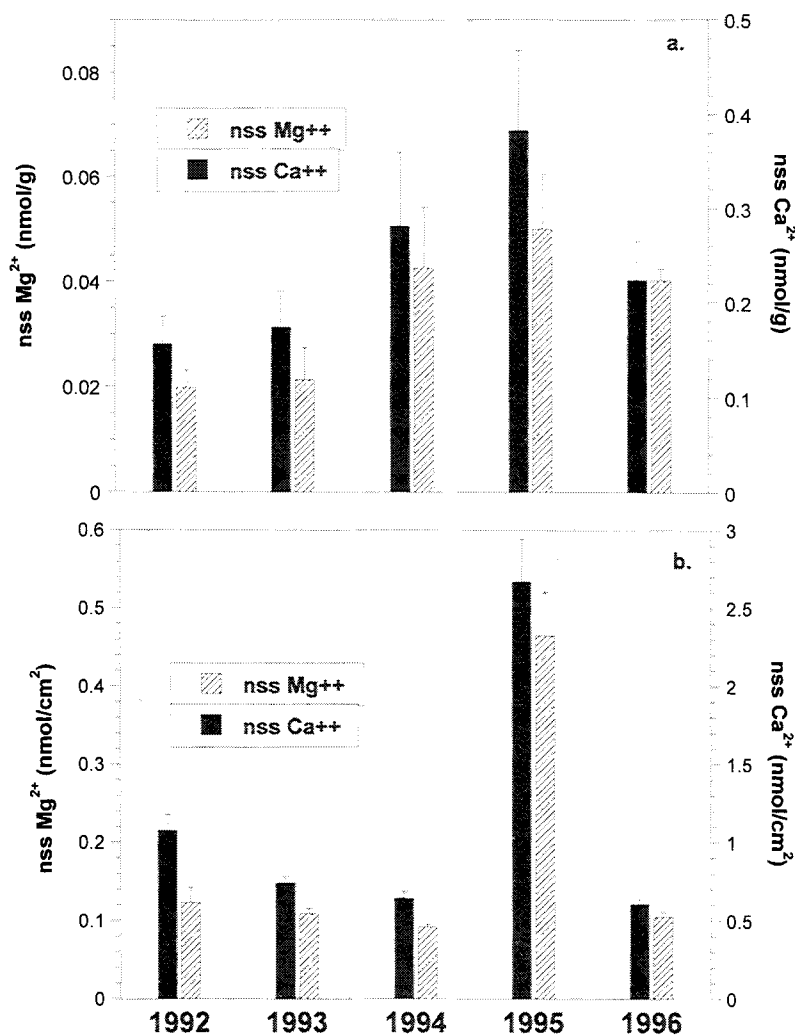
Figure 4. (a) The common season snowfall average nss-calcium (Ca^{2+}) and nss-magnesium ion (Mg^{2+}) concentrations (with standard error) and (b) the common season cumulative buried layer nss-Ca^{2+} and nss-Mg^{2+} cumulative inventory (with standard deviation). Buried layers are determined through explicit field notes (see text); cumulative inventory is calculated by multiplying the ion concentration by the water equivalent of each sample, then summing the inventories for the entire season.

the Yukon region and Siberia and generally had more northerly transport pathways than those observed in 1995. Notably, there were no trajectories that originated in these regions during 1995 (Table 4).

The synoptic-scale meteorological conditions that impacted the different transport patterns and source regions influencing Summit in 1992 and 1995 were investigated by examining 700 mbar (~3 km aloft) atmospheric pressure charts. Although planetary boundary layer conditions influence air mass flow above the ice sheet, geostrophic flow at the 700 mbar height will give a qualitative indication of the flow direction of air masses reaching Summit on a synoptic scale.

Source regions during 1995 were either Canada/northern United States (59%) or western Eurasia (41%). Dominant transport paths consisted of over-the-pole transport from western Eurasia and long-range rapid transport from North America (Figure 6). The over-the-pole transport pattern was caused by a cyclone system that tracked up the eastern coast of Greenland and persisted near the northeast corner of the landmass for several days (Figure 7a). *Serreze et al.* [1997] points out that the leeward

side of Greenland is an area of cyclogenesis under conditions of westerly flow over the southern part of the ice sheet. However, the systems normally form in winter and then move north and northeast while intensifying [*Serreze et al.*, 1997]; thus their presence in May-June is unusual.

Synoptic meteorological evidence indicates that the long-range transport from North America observed in 1995 (Pattern B,

Table 2. Common Season Water Equivalent of Snowfall Events 1992-1996

Year	Number of Events	Average Inventory, g H_2O cm^{-2}	Cumulative Mass, g H_2O cm^{-2}
1992	4	0.245 (0.070)[a]	0.982 (0.140)[a]
1993	7	0.114 (0.064)	0.795 (0.169)
1994	7	0.137 (0.069)	0.961 (0.183)
1995	5	0.138 (0.069)	0.689 (0.154)
1996	3	0.270 (0.042)	0.811 (0.073)

[a] Standard deviation.

Table 3. Common Season Water Equivalent of Buried Layers 1992-1996

Year	Number of Buried Layers	Average Inventory, g H_2O cm^{-2}	Cumulative Mass, g H_2O cm^{-2}
1992	4	0.245 (0.070)[a]	0.982 (0.140)[a]
1993	7	0.114 (0.064)	0.795 (0.169)
1994	7	0.137 (0.069)	0.961 (0.183)
1995	5	0.138 (0.069)	0.689 (0.154)
1996	3	0.270 (0.042)	0.811 (0.073)

[a] Standard deviation.

Figure 6) was caused by a strong low-pressure system at 700 mbar west of Great Britain (Figure 7b). This pattern dominated the air mass flow from late May to early June and was not present in the common season of the other four years of this study. Development of these midlevel low-pressure areas frequently occurs in association with surface midlatitude cyclone systems; baroclinicity is common on the eastern side of an upper air low, hence supporting the surface cyclone [*Barry and Chorley*, 1992].

During 1992 the major portion of backward trajectories originated in either Canada/northern United States (43%) or western Eurasia (34%, Table 4). Zonal westerly transport was generally at higher latitudes in 1992 than during 1995, resulting in a portion of the 1992 trajectories originating in the Yukon region and Siberia (12% and 9 %, respectively). A comparison of the vertical distribution of the trajectories between 1992 and 1995 reveals that 60% of the trajectories during 1992 entered the boundary layer during transport to Summit, compared to just 10% during 1995. This indicates that transport was generally more rapid in 1995, as a result of more flow patterns within the free troposphere. Also during 1992, an alternating pattern in trajectory origin was observed. In mid to late May, source regions alternated between Canada and western Eurasia (Figure 8) as cyclones migrated from west to east across the North Atlantic Ocean. This pattern repeated itself at a frequency of 5-7 days throughout the common season as a result of the formation, intensification, and movement of these low-pressure systems. Atmospheric pressure distributions (700 mbar) during late May illustrate (1) the formation of a cutoff low in the Baffin Bay region (Figure 9a), (2) deepening of the system as it migrates across the North Atlantic Ocean (Figure 9b), and (3) continued
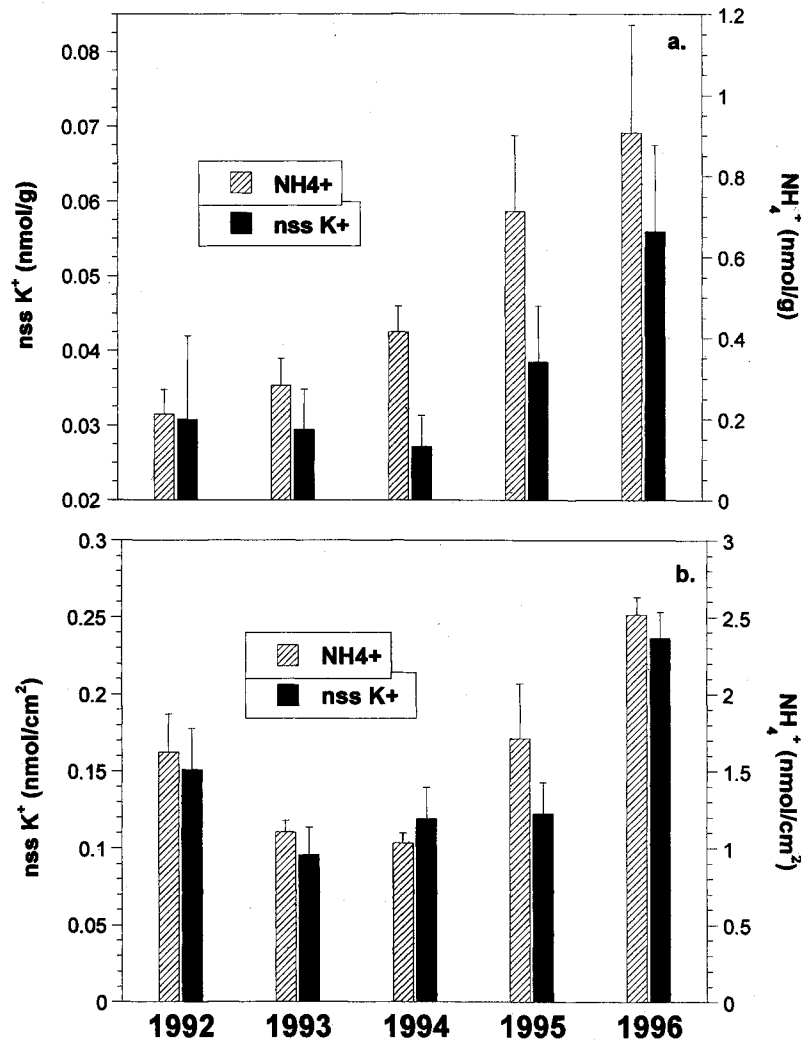

Figure 5. (a) Common season snowfall average ammonium (NH_4^+) and nss-potassium ion (K^+) concentration (with standard error) and (b) the common season cumulative buried layer NH_4^+ and nss-K^+ cumulative inventory (with standard deviation).

Table 4. Summary of the Origin of 10-Day Isentropic Backward Trajectories During the Common Season 1992-1996[a]

Year	Study Regions From Figure 1 (With Coordinates)			
	1. Canada and Northern United States, 40°-85°N x 60°-120°W	2. Yukon Region, 55°-85°N x 120°-180°W	3. Siberia, 60°-85°N x 60°-180°E	4. Western Eurasia, 40°-85°N x 0°-60°E
1992	43[b]	12	9	34
1993	50	-	31	19
1994	44	13	19	16
1995	59	-	-	41
1996	50	28	9	12

[a]Units in percent.
[b]Percentage of 60 sets of trajectories calculated per common season.

migration of the system and the formation of a new cutoff low south of Baffin Bay (Figure 9c).

5. Discussion

5.1. Continental Dust and Atmospheric Circulation

Nearly 3 times more nss-Ca^{2+} and nss-Mg^{2+} accumulated in the surface layers of the snowpack at Summit during the 1995 common season than any other year (Figure 4b). In addition, the average concentration of nss-Ca^{2+} and nss-Mg^{2+} in fresh snow was more than 2 times higher in 1995 than in 1992 (Figure 4a). As previously stated, there was no statistical difference in the cumulative mass of water contained in fresh snow between the 5 years of our investigation, allowing us to assume that the difference in average concentration in fresh snow of these species

between 1992 and 1995 is caused mainly by differences in atmospheric loading. When comparing the transport pathways between the 2 years, trajectories influenced by westerly flow during 1992 were largely north of the 60th parallel, and those in 1995 were observed to have a more southerly route (Figures 6 and 8). The vertical profile of the trajectories during the common season in 1995 indicates that transport was mainly above the boundary layer; 90% of all trajectories calculated maintained a height of 2-4 km for the duration of their modeled lifetime (10-days). Weekly snow cover charts for 1992 indicate that 70-80% of the area above the 60th parallel was covered by snow or ice in mid May to late May, with the snow-covered area declining through June [*National Oceanic and Atmospheric Administration, National Snow and Ice Data Center*, 1995]. Thus during 1995 air masses originated in, or passed over,
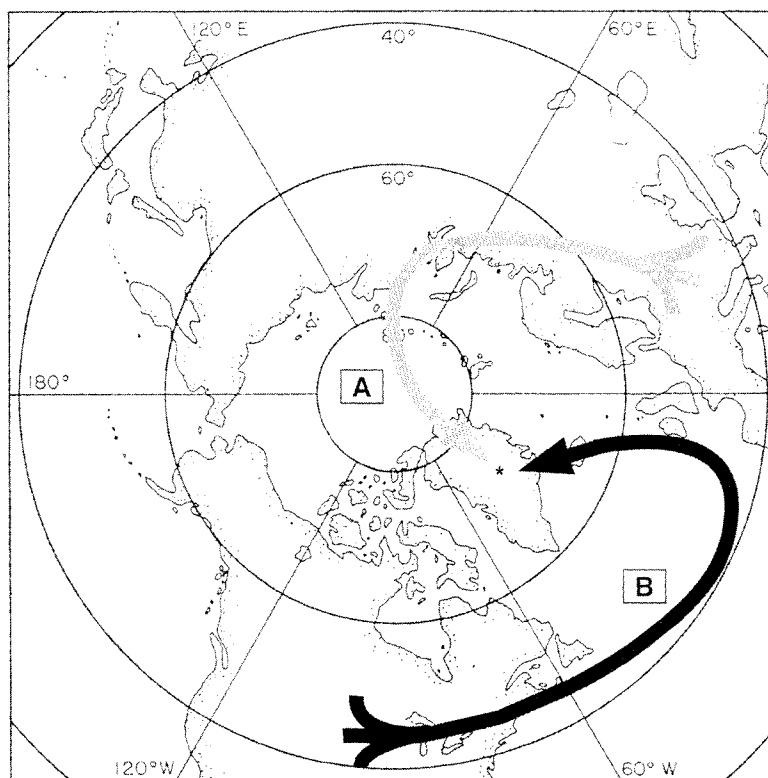

Figure 6. Main features of the transport pathways during the common season 1995 based on 10-day isentropic backward trajectories. Transport Pattern A occurred during mid to late May; Transport Pattern B occurred during late May to early June.
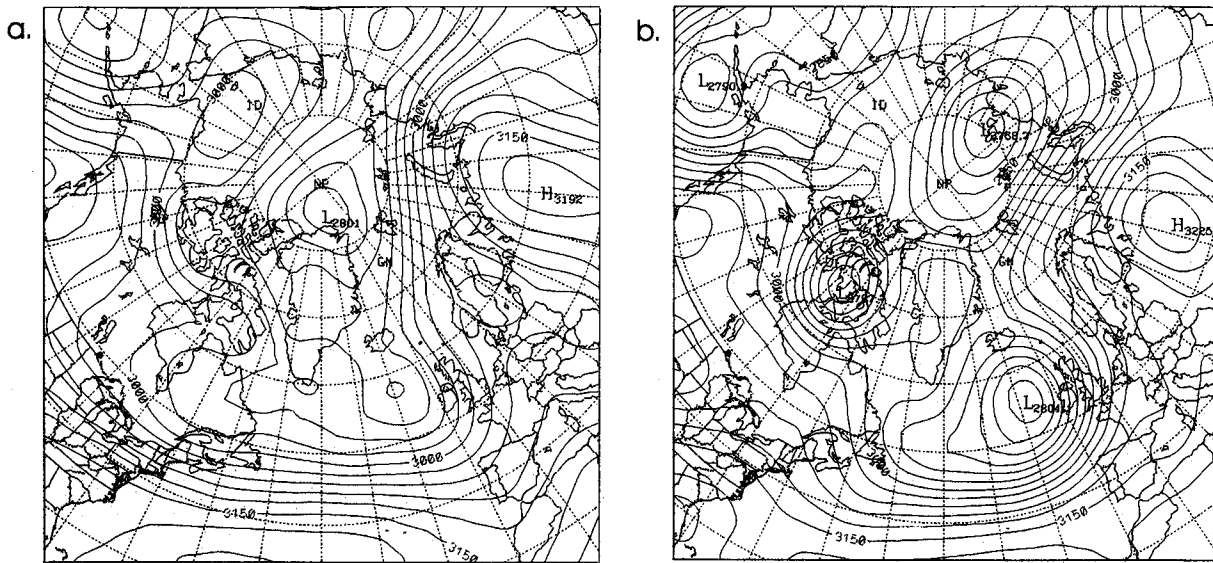
Figure 7. Mean 700-mbar atmospheric pressure surfaces (meters) showing features of the synoptic conditions during the periods of interest in 1995. Plot (a) (May 22-25) shows a low system in the northeast corner of Greenland. Plot (b) (May 26-28) illustrates a strong low system that persisted off the coast of Great Britain for several days. (NASA's GTE Project Office, 2000: http://www-gte.larc.nasa.gov).

regions where more continental surfaces were exposed, which can cause an increase in aeolian dust in the free troposphere, thereby increasing the likelihood of crustal species being transported to Summit.

The long-range transport pathways observed in late May to early June in 1995 (Pattern B, Figure 6), reflecting high wind velocities aloft, likely facilitated the transport of the dust to

Summit. *Kahl et al.* [1997] detected a similar transport pattern in spring and summer 700 mbar isobaric trajectories; however, the pattern only accounted for a seasonal average of 3% of the trajectories calculated during their 28-year study. Long-range transport of continental material to remote regions is dominated by transport in the free troposphere, where wind velocities are high enough to prevent the settling of larger particles [*Merrill et*
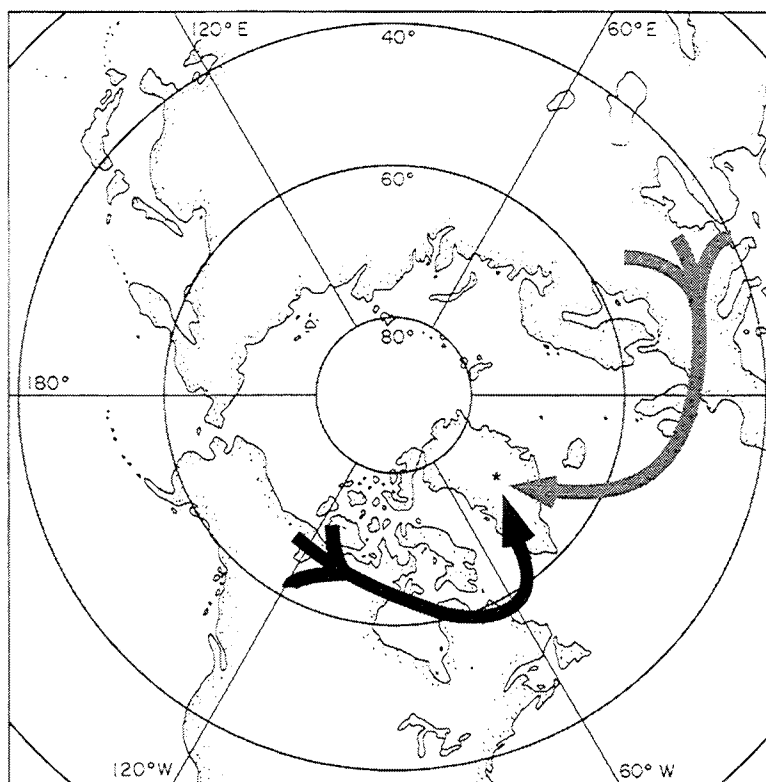

Figure 8. Main features of the transport pathways during the common season of 1992 based on 10-day isentropic backward trajectories. The two transport pathways alternated throughout the season with a frequency of 5-7 days.
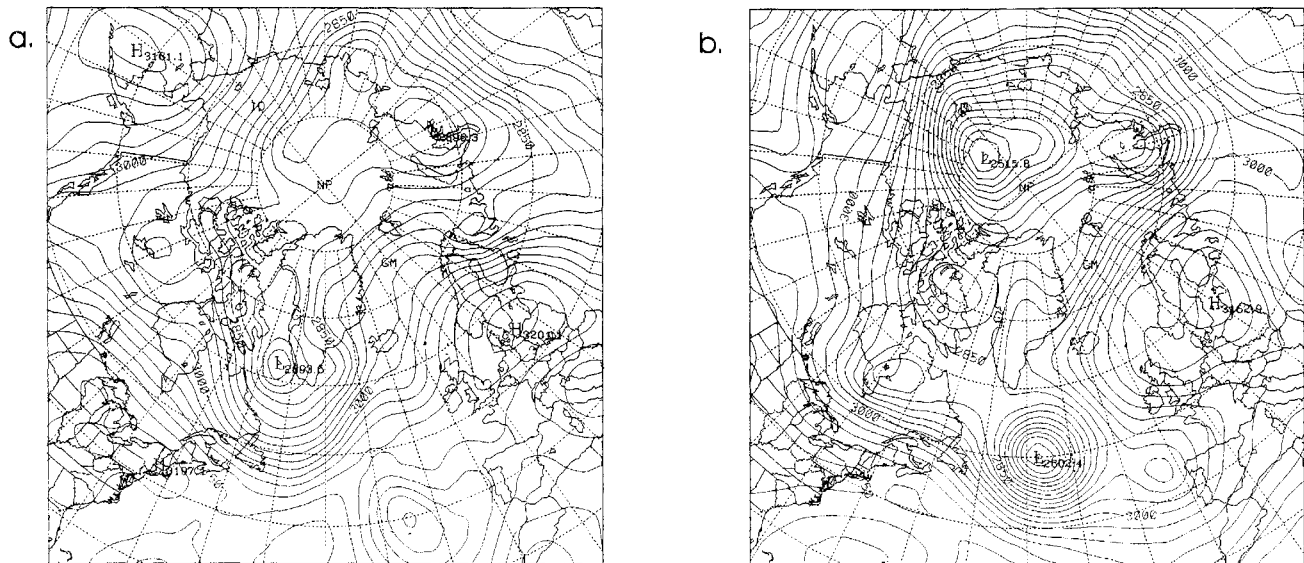
Figure 9. Daily 700-mbar atmospheric pressure surfaces (meters) showing the features of synoptic conditions during late May 1992: (a) A low system forms in the Baffin Bay region (May 20, 1992, 0000 UT) and (b, c) then proceeds to move across the North Atlantic Ocean, while a new system is formed in Baffin Bay (Figure 9b, May 23, 1992 1200 UT; Figure 9c, May 25, 1992 0000 UT) (NASA's GTE Project Office, 2000: http://www-gte.larc.nasa.gov).

al., 1994]. Thus the air masses transported to Summit during late May to early June 1995 likely experienced a reduction in the deposition of large crustal aerosols, as compared to the shorter trajectories (lower wind velocity) observed in the remaining years of this study (particularly 1992).

To investigate if changes in mid-latitude to high-latitude atmospheric background source strength could also be contributing to the above average nss-Ca^{2+} and nss-Mg^{2+} observed during 1995, monthly Northern Hemisphere snow cover data were examined (D. Robinson, Rutgers University, personal communication, 1998). Weekly snow cover charts were not available during this study's common season for all years. Instead, monthly averaged coverage was calculated from satellite imagery for each of our study regions [cf. Robinson et al., 1995]. A possible contribution to enhanced free troposphere loading of dust would be a decrease in Northern Hemisphere snow cover, exposing more continental surfaces. There is, however, no substantial variability in the snow cover extent for any of the regions throughout the 5 years of this study (Figure 10). For example, during May and June 1995 monthly snow cover extent (32% and 21%, respectively) for Canada/northern United States (Region 1) is within the range of the 5-year mean of each month for this region (May = 34 ± 4%; June = 20 ± 2%). Therefore the large enhancement in nss-Ca^{2+} and nss-Mg^{2+} observed in 1995 (as compared to 1992) is probably not caused by an increase in background source strength but is likely caused by a combination of anomalous transport velocities above the planetary boundary layer (resulting in reduced settling of large dust particles), as well as more southerly flow pathways over source regions not covered by snow.

5.2. Surface Snow Ammonium Concentration and Inventory

The increase of common season NH_4^+ concentration in snowfall events during 1992-1996 and inventory of buried layers during 1994-1996 (Figures 5a and 5b) suggests (1) an increase in

circumpolar north forest fires or that transport is changing to include regions where fires are more prevalent, and/or (2) spring to early summer background levels of NH_4^+ in the Arctic troposphere are increasing. Because formate was not measured in the surface snow, we cannot use the "double condition" of the presence both formate and NH_4^+ to recognize biomass burning inputs in Summit snow suggested by Savarino and Legrand [1998]. It is therefore difficult to separate the NH_4^+ resulting from NH_3 emitted to the atmosphere by soil exhalation and that produced by biomass burning. However, it is likely that a forcing such as a warming trend in circumpolar countries will result in an increase in both NH_3 emissions from undisturbed soils, as well as an increase in the frequency and extent of forest fires [Kasischke et al., 1995; Wotton and Flannigan, 1993]. We concluded that it

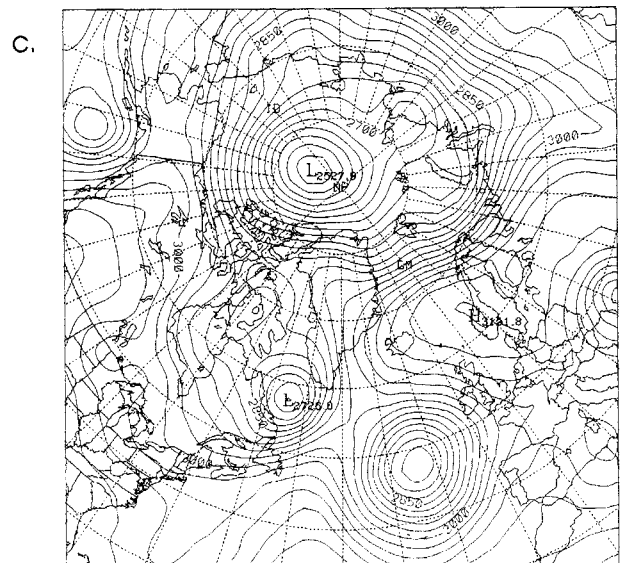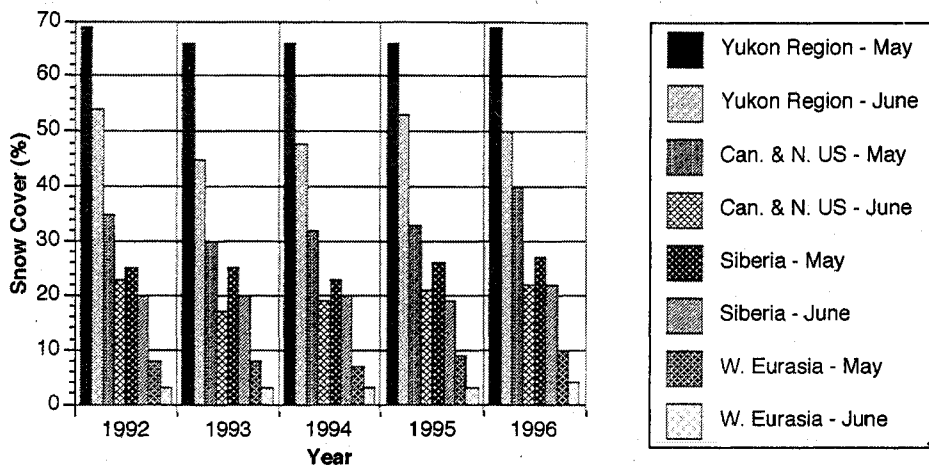**Figure 9.** (continued)

Figure 10. Monthly average circumpolar snow coverage of study regions derived from satellite imagery for May and June 1992-1996 (David Robinson, Rutgers University, personal communication, 1998).

would be more relevant to determine whether these two sources may have changed during our study period, rather than to try to separate them.   If a change in high-latitude NH_3 source strength could be demonstrated, it would help to explain the observed increase of NH_4^+ in snowfall during 1992-1996 and buried layers during 1994-1996. The different pattern in nss-K^+ during 1992-1995 likely reflects mixed sources and cannot be adequately explained.

5.3. Variability of Ammonium Source Strength

Forest fire statistics during 1992-1996 are readily available for the northern United States, Canada, and Alaska; however, there is no accurate record of fires in Russia during this time period. Given that the 10-day origin of $\geq$ 50% of the air masses reaching Summit throughout the 5 years of this study was in North America (Regions 1 and 2, Table 4), fire activity during 1992-
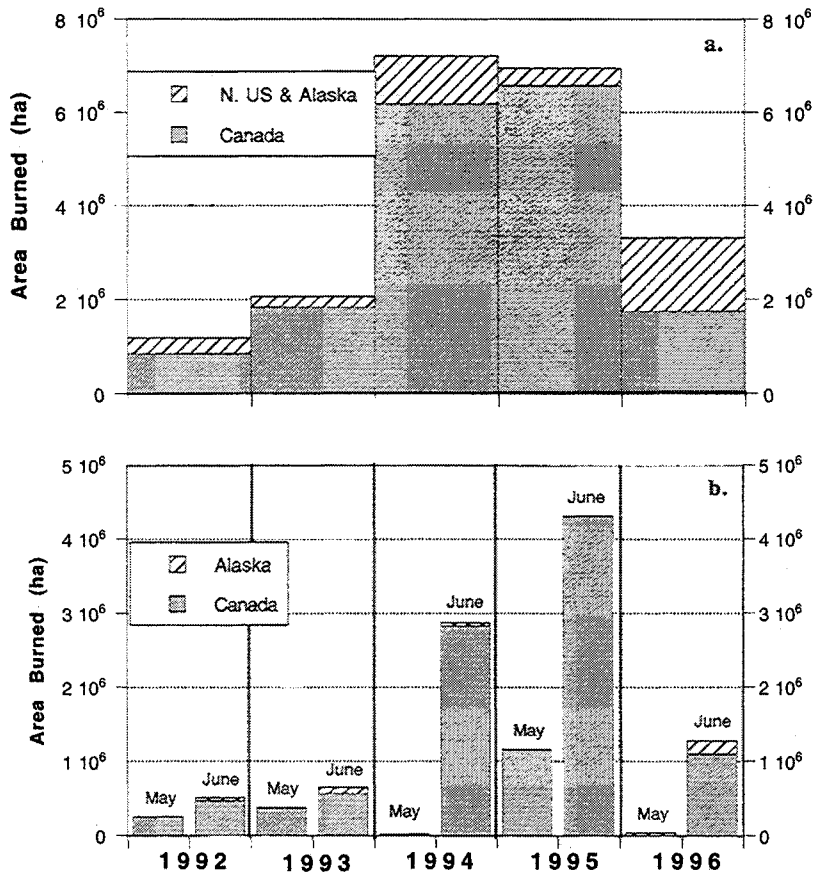

Figure 11. North American forest fire activity from 1992 to 1996: (a) Annual average area burned in North America (in hectares (ha)) and (b) monthly average area burned in Canada and Alaska for May and June (CCFM, 1997; BLM, 1998).

1996 for North America (Figure 11a) can be used to estimate changes in NH_3 source strength that may affect NH_4^+ in surface snow at Summit.

Our common season coincides with the start of the temperate and boreal forest burning season; Canadian and Alaskan fire statistics indicate that from 1992 to 1996 greater than 60% of the forest fires occurred during May and June (Figure 11b) [*Canadian Council on Forest Ministries*, 1997; *U.S. Bureau of Land Management*, 1998]. In addition, *Stocks and Lynham* [1996] report that extreme fire-weather conditions occur in southern Siberia in early May and move northward as spring arrives at higher latitudes, while *Valendik* [1996] presents evidence that greater than 50% of forest fires in Siberia from 1970 to 1983 burned in May and June. Any interannual variability in forest area burned in the circumpolar region can therefore be reflected in spring to early summer surface snow samples at Summit.

An increasing pattern similar to that observed in our NH_4^+ snowfall concentration (Figure 5a) is seen in the monthly average of area burned for Canada and Alaska during the years 1992-1995 (Figure 11b). However, the decrease in area burned during 1996 is not reflected in the ion pattern. Several authors have pointed out that not all fires in source regions will be preserved in snow and ice at Summit [e.g., *Fuhrer et al.*, 1996; *Whitlow et al.*, 1994]. If a biomass burning event is going to be recorded in the snow, meteorological factors must be favorable for transport from the source to the site, precipitation scavenging en route must be minimal, and snowfall must transfer the atmospheric signal to the surface snow. It is possible that more efficient transport and deposition processes were in place during 1996 compared to the other study years, accounting for the observed higher values of NH_4^+.

Along with forest fires, another major contributor to tropospheric NH_4^+ in the Arctic is NH_3 fluxes from undisturbed soils in temperate and boreal forests. An increase in temperature can cause soil trace gas emissions to increase as a result of more biological activity [e.g., *Apps et al.*, 1993]. Increased temperatures can also cause the soil freeze/thaw transition period to occur earlier in spring [e.g., *Running et al.*, 1999]. Determining if there was any interannual variability in NH_3 fluxes from soils in source regions during the 5 years of our study period proved to be difficult. Although recent investigations point toward a general warming trend and an increase in the growing season in the Arctic over the past ~20 years [*Rigor et al.*, 2000; *Running et al.*, 1999], we cannot unequivocally link Arctic temperature trends, NH_3 emissions, and the NH_4^+ content of surface snow at Summit during 1992-1996.

6. Summary and Conclusions

The Holocene portion of the GISP2 and GRIP records of nss-Ca^{2+} and nss-Mg^{2+} concentration displays twofold to threefold interannual variations that have been attributed to increased complexity of atmospheric circulation. We have observed a similar increase in nss-Ca^{2+} and nss-Mg^{2+} of about a factor of 3 in the snowpack during May-June 1995, compared to the other 4 years of this study. To determine if an increase in exposed continental surface area as the northern snow line retreated contributed to an increase in background levels of atmospheric dust, Northern Hemisphere snow cover extent was studied. There was, however, no substantial change in snow cover extent in any of our delineated source regions during the 5 years of this study.

Although there was no significant change in dust source strength, anomalous high-velocity transport mainly in the free troposphere during May and June 1995 likely facilitated transport of crustal aerosols to Summit (compared to the other years in this study).

We also observed a twofold to threefold increase in NH_4^+ and nss-K^+ accumulation in surface snow from 1993 to 1996, which is on a similar order of magnitude as changes in summertime background levels of NH_4^+ that were observed throughout the Holocene [*Fuhrer et al.*, 1996]. Backward trajectories indicated that source regions throughout this study were mostly ($\geq 50\%$) in North America. The bulk of the remaining trajectories (~25%) had 10-day origins in western Eurasia. An increase in the area of forest burned in North America from 1992 to 1995 was concurrent with the increasing pattern in the snowfall concentration of NH_4^+ during those years, leading to the conclusion that changes in atmospheric levels of these species can be recorded in surface snow at Summit. However, we were unable to connect recent Arctic temperature trends (and associated NH_3 soil emissions) and the observed NH_4^+ pattern in the snow.

Because of the ubiquitous nature of soluble ionic species in the troposphere, establishing quantitative relationships between the variability in atmospheric transport pathways, changes in source regions, and changes in surface snow chemistry at Summit is difficult. Even though we focused on ionic species that are thought to have independent sources, we observed some similarity in concentration fluctuations among the four ions (e.g., during the early August 1994 biomass burning event). We recommend that future studies aimed at connecting the interannual variability of Summit surface and subsurface snow chemistry and changes in atmospheric circulation and source strength on a seasonal basis should include more source specific tracers to track combustion aerosol inputs (such as polycyclic aromatic hydrocarbons and carbon isotopes) and crustal aerosols (such as rare earth elements).

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