

ANALYSIS OF THE SEASONAL VARIATION IN DUST, Cl^- , NO_3^- , AND SO_4^{2-} IN TWO CENTRAL GREENLAND FIRN CORES

by

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ABSTRACT

The concentrations of dust, Cl^- , NO_3^- and SO_4^{2-} in two firn cores covering the period A.D. 1891 to 1910 have been analyzed and compared. The cores were drilled 130 km apart — one west and one east of the north-south ice crest in central Greenland. The dust concentration is higher at the western site. This suggests a greater influence of continental aerosols in precipitation there, and that most of the dust comes from the west. The Cl^- concentration is higher at the eastern site in some years, probably because of greater marine influence on that site. Both sites have similar concentrations of NO_3^- and SO_4^{2-} . A rise in SO_4^{2-} concentration in the middle of the period is explained by volcanic activity and increasing anthropogenic influence. The seasonal variation in each component is discussed, and explained within the framework of an Arctic aerosol-reservoir model.

INTRODUCTION

Stratigraphic analysis of impurity concentrations in ice cores is an important source of information about past atmospheric conditions. The impurities are fractions of the

atmospheric aerosol. They arrive at the ice-sheet surface in precipitation, both by wash-out and by forming condensation nuclei for ice crystals, and also as dry fall-out (Junge 1977, Davidson and others 1985). The atmospheric circulation carries the impurities to central Greenland. Changes in impurity concentrations in precipitation reflect changes in the sources of the aerosol, in atmospheric circulation or in local atmospheric processes. Temporal changes in impurity concentrations over the past several thousand years can be monitored by stratigraphic analysis of dated ice cores (Oeschger 1985). Central Greenland cores are ideal for this kind of study, because the accumulation rate is high enough to allow sub-annual sampling and absolute dating by counting annual layers recorded by variations in $\delta^{18}\text{O}$. In addition, surface melting, which might destroy the stratigraphy, rarely occurs.

In this paper, results from an analysis of two central Greenland core segments, each covering precipitation from A.D. 1891 to 1910, are discussed. Firn from this period is currently being analyzed in detail in several of the central Greenland ice cores at our laboratory. Both cores were drilled with an electro-mechanical drill in a dry hole. One was drilled in 1984 at Site D ($70^\circ 38' \text{N}$, $39^\circ 47' \text{W}$; 3020 m

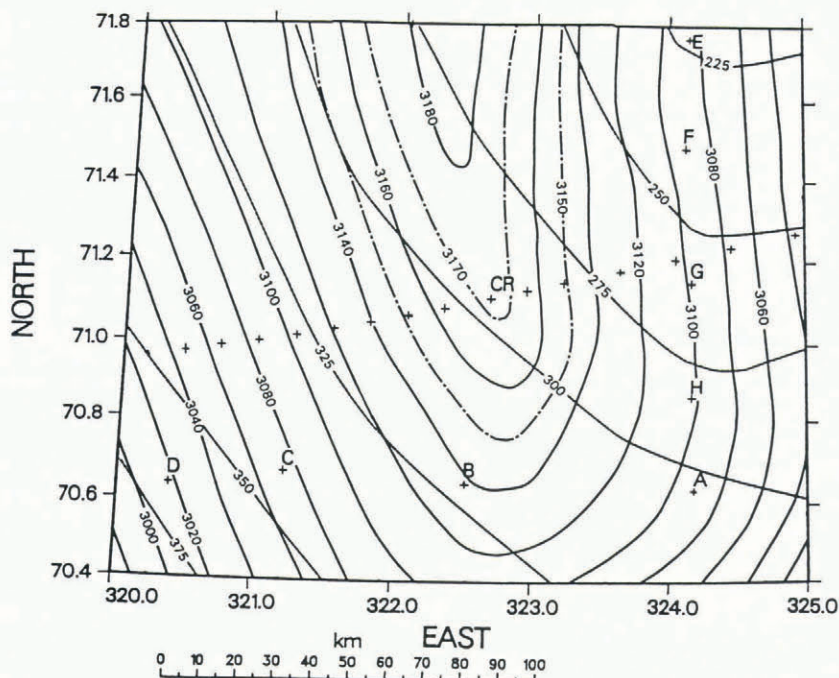
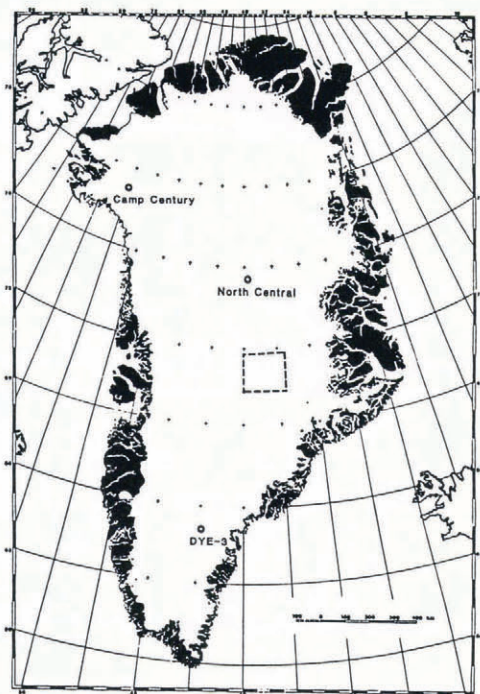


Fig.1. The location of the Crête area. On the detailed map (right), Crête (CR), sites A-H and the EGIG line (crosses) are marked. Shallow cores at sites A-H were recovered in 1984 and 1985. The 3000-3180 contours are

heights in meters over mean sea-level, and the 225-375 contours are accumulation rates in millimeter ice equivalents per year (Clausen and others 1988, this volume).

a.s.l.), about 90 km west of the north-south crest of the ice sheet, the other in 1985 at Site A ($70^\circ 38' N$, $39^\circ 49' W$; 3090 m a.s.l.), about 40 km east of the crest (Fig.1). I measured the levels and seasonal variations of dust, Cl^- , NO_3^- and SO_4^{2-} to investigate possible differences between the sites. Variations in $\delta^{18}O$ were also studied. The results are also compared with those from other Greenland sites.

EXPERIMENTAL TECHNIQUE

The two cores were brought to Copenhagen in a frozen condition and stored at $-17^\circ C$. They were of excellent quality, without breaks or internal cracks. $\delta^{18}O$ samples were cut according to a cutting scale and measured on the automatic mass spectrometer in our laboratory. Electrical conductivity measurements (Hammer 1980, Neftel and others

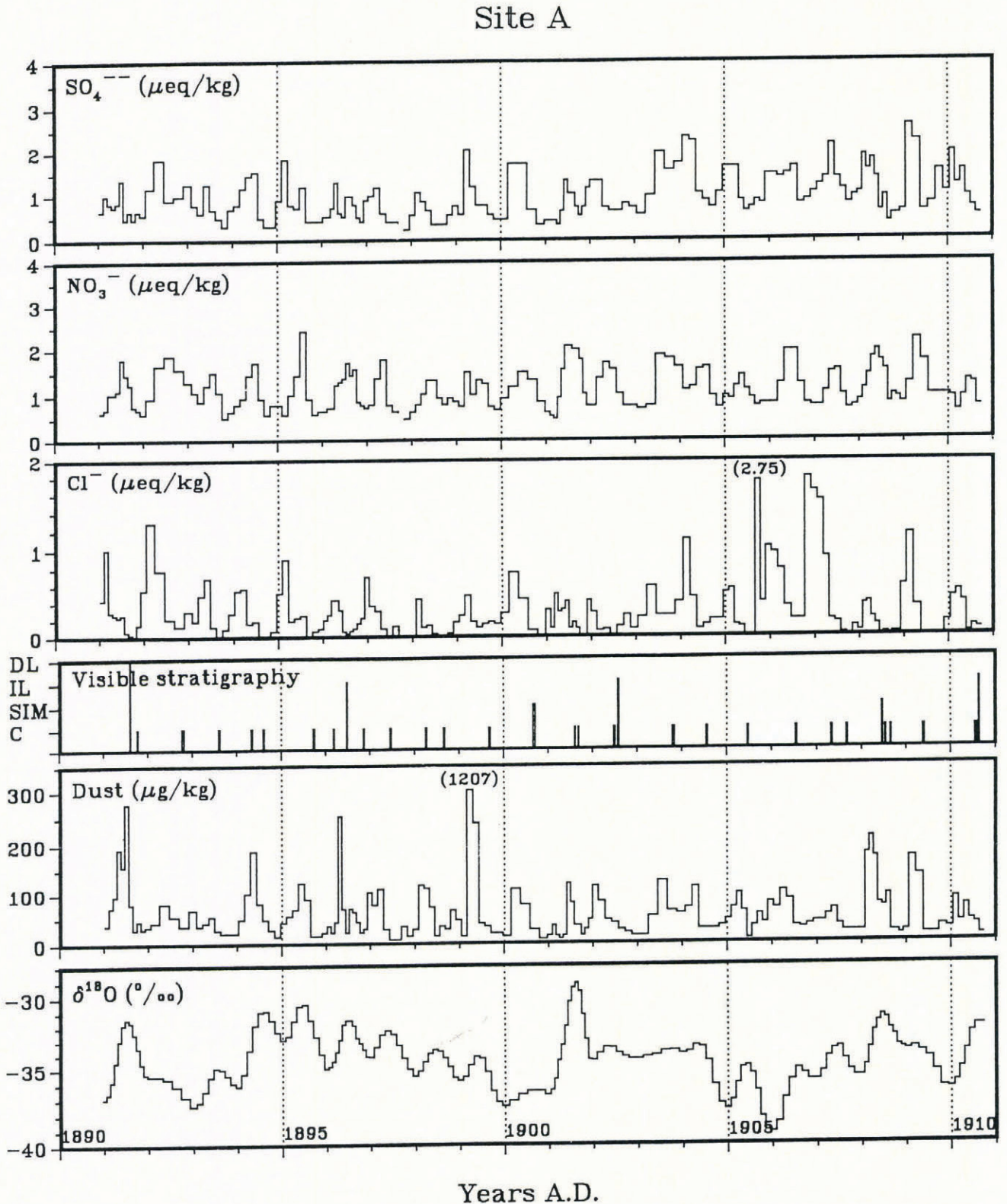


Fig.2a. Concentrations of dust and anions versus the time-scale obtained by $\delta^{18}O$ dating at Site A. Values out of range are given in parentheses. The visible stratigraphic features, in order of increasing changes by

melting, are: C (coarse layer), S (summer layer), SIM (slight indication of melt), DIM (definite indication of melt), and IL (ice layer). DL refers to "dark layer".

1985[a]) were made on both cores and visible stratigraphy was recorded. The electrical conductivity measurements served as a cross-check of dating based on the seasonal variations in $\delta^{18}O$ (Clausen and Hammer 1988, this volume). The Katmai 1912 eruption, which is clearly visible in the electrical conductivity record of cores from the Crête area (Fig.1), served as a marker (Hammer and others 1980,

personal communication from H.B. Clausen).

A total of 154 samples from Site A and 176 from Site D, each 5 cm long, was cut. They cover the depth intervals 37.40–45.10 m (Site A) and 42.35–51.15 m (Site D). The samples formed two continuous series covering the period A.D. 1891 to 1910, with a resolution of about eight samples per annual layer. The precipitation rates at sites A and D

Site D

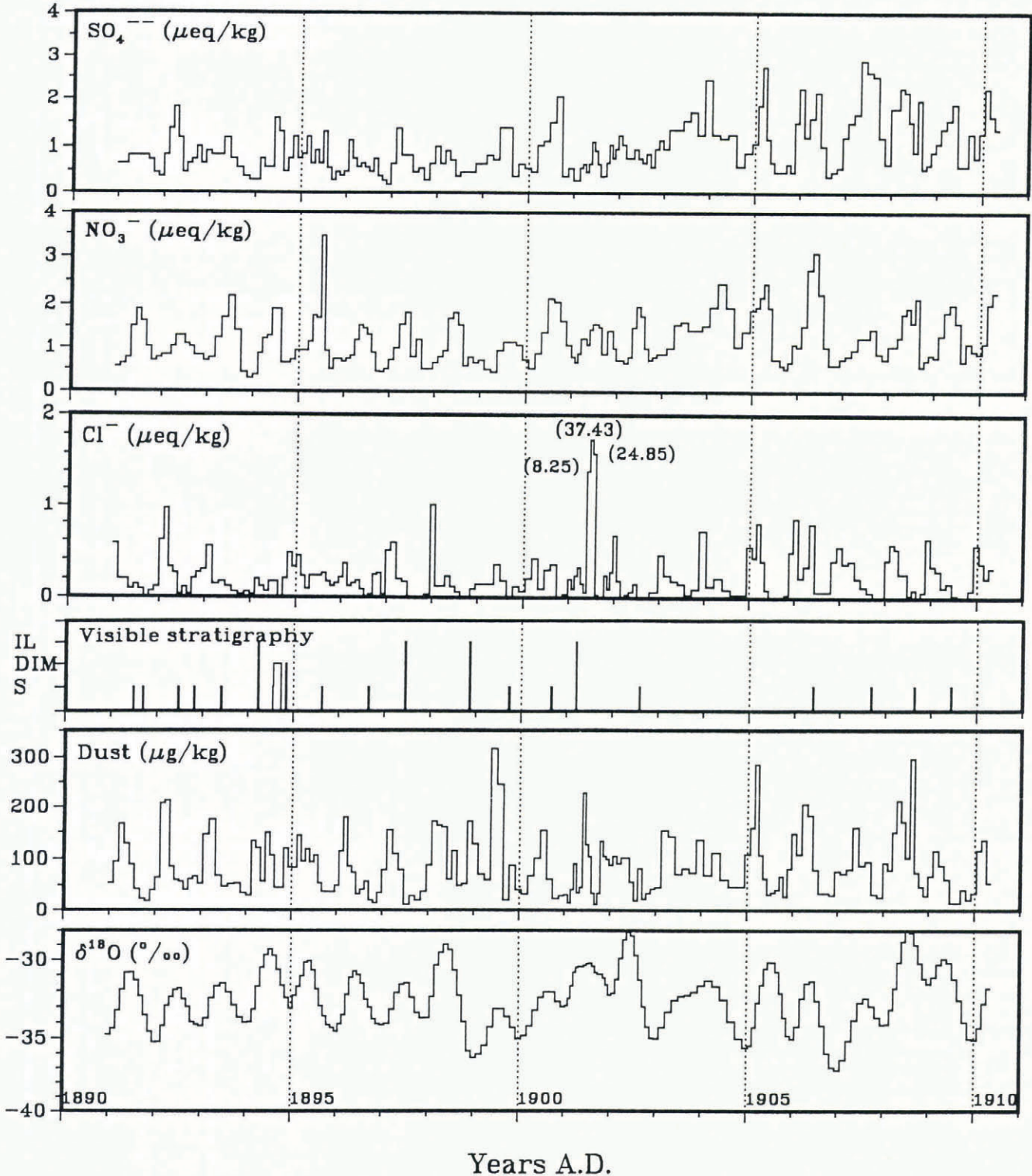


Fig.2b. Concentrations of dust and anions at Site D.

are 0.31 and 0.37 m ice per year respectively (Clausen and others 1988, this volume).

The samples were taken using a technique which is normally employed at our laboratory. The firn-core sections were cleaned by a stainless-steel microtome knife in the cold-room, under class 100 conditions. The outer 1–2 cm were cut away so that only firn from the interior of a core was used. This technique has been checked thoroughly for possible contamination, using artificial ice-core substitutes of frozen ultra-pure water. The samples were kept frozen in 25 ml Accuvettes until analysis. The samples were melted immediately before analysis in a clean-room (class 100), and a Dionex 2010i ion chromatograph was used for the chemical analysis. The bulk dust concentration was measured by laser light scattering (Hammer 1977[b]). The samples were taken in random order to eliminate possible "cross-talk". A number of blanks (ultra-pure water, >18 Mohm/cm) were treated in the same way as the samples, to provide a check on possible contamination during the chemical analysis.

RESULTS

Figure 2 shows the concentrations of dust and the three anions, together with the $\delta^{18}O$ profile and visible stratigraphic features. Each component shows the same seasonal variation at the two sites. Seasons are defined on the basis of the $\delta^{18}O$ profile, i.e. "summer" when $\delta^{18}O$ reaches a maximum, and "winter" when it reaches a minimum. The annual mean values of anion and dust concentrations and of $\delta^{18}O$ are presented in Figure 3.

Although surface melting seldom occurs in the Crête area, there are visible layers of changed firn structure and 1 mm thick ice layers in the cores associated with high summer-surface temperatures (see Fig.2). These layers are subjectively classified into four groups in order of increasing changes by melting: "C" (coarse layer), a barely visible layer of slightly enlarged crystals caused by recrystallization; "S" (summer layer), a layer of slightly enlarged crystals; "SIM" (slight indication of melting), a layer with a slight indication that regular melting-refreezing has taken place; "DIM" (definite indication of melting), a layer with definite indications of melting-refreezing; and "IL" (ice layer), a layer of refrozen water, where the firn structure has disappeared. The concentrations of dust, NO_3^- , Cl^- and SO_4^{2-} can be affected seriously by surface melting. Elevated NO_3^- and dust concentrations caused by melt processes are found in cores from the Dye 3 area (Finkel and others 1986, Steffensen 1986). The processes which affect the concentrations during melting and refreezing are still not known in detail.

In the Site A core, no concentration changes are observed in the visible layers. The dark layer observed in the summer layer of 1891 is not a melt feature; it is believed to be associated with the high dust concentration. Unfortunately the record of visible stratigraphy in the Site D core did not include the "coarse" layers as in the Site A core, therefore fewer features are marked in the Site D record (Fig.2). The firn structure in the visible layers at Site D indicates slightly more melting than at site A. Elevated concentrations of dust are associated with visible layers in 1898, 1901, 1902, 1906 and 1908. The NO_3^- record is affected in the 1895 and 1908 layers. The SO_4^{2-} and Cl^- records appear to be affected only in the 1908 layer. Although melting produces small concentration changes in the Site D core, their affect on the annual mean values is believed to be small.

The 20 year mean values of $\delta^{18}O$ are -34.34‰ at Site A and -32.48‰ at Site D. The annual mean temperatures are -29.8°C and -27.9°C respectively (Clausen and others 1988, this volume). The difference in $\delta^{18}O$ is thus about twice that expected if the normal conversion factor of $0.62\text{‰}/^\circ\text{C}$ (Dansgaard and others 1973) is used. This

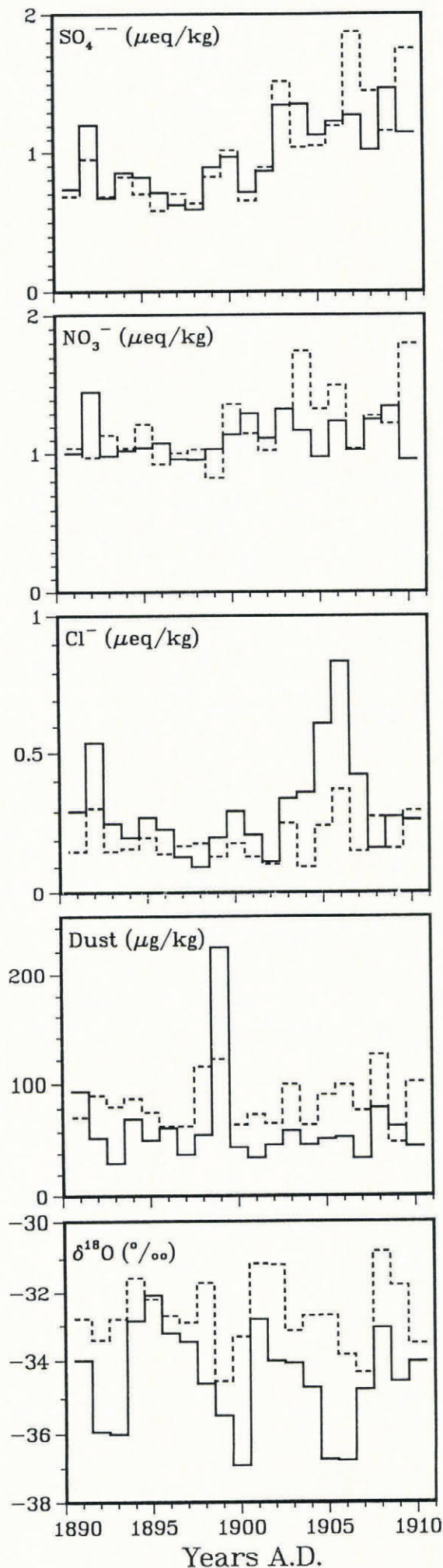


Fig.3. Annual mean values of the concentrations of dust, anions and $\delta^{18}O$ from Site A (solid line) and Site D (dashed line).

discrepancy is caused by $\delta^{18}\text{O}$ s which are significantly lower from Site A than from Site D in the years 1892–93, 1900, and 1905–06, and it is ascribed to local effects which will be discussed below.

Chloride and dust

The bulk of the dust (the insoluble fraction of the impurities) in the ice is of continental origin (Hammer 1977[c]). It has been transported to the Greenland ice sheet mainly in the mid-troposphere, as part of the tropospheric background aerosol (Hammer and others 1978, Junge 1977). The dust concentration is high in spring and summer, with a maximum in spring, and low in autumn and winter. This agrees with data from other Greenland and Canadian sites (Hamilton and Langway 1967, Murozumi and others 1969, Thompson 1977, Koerner 1977, Hammer 1977[a]). The 20 year mean concentrations are $64 \mu\text{g}/\text{kg}$ at Site A and $84 \mu\text{g}/\text{kg}$ at Site D. The high annual mean value in 1899 at Site A results from the very high spring peak.

Marine sea salt appears to account for most of the observed Cl^- in the Greenland ice cores. However, some major volcanic eruptions in which HCl is emitted may contribute a significant amount of Cl^- , but only for short periods (Herron 1982). Cl^- can therefore be used as an indicator of marine air masses. Cl^- is part of the marine aerosol. This stays mainly in the low troposphere, but small parts of it reach the high altitudes of central Greenland (Junge 1977).

The Cl^- concentration has seasonal variations, with high concentrations in winter–early spring and low concentrations in summer and autumn. This agrees with results from the Dye 3 area in south Greenland (Murozumi and others 1969, Finkel and others 1986, Mayewski and others 1986). In our study the Cl^- concentration reaches a maximum before the dust concentration; it starts to decrease in early summer and reaches a summer–autumn low which in several cases was below the detection limit of the ion chromatograph ($0.02 \mu\text{eq}/\text{kg}$). As for the summer Cl^- high reported by Mayewski and others (1986), only one is found in the Site D record. This value, which is exceptionally high ($37 \mu\text{eq}/\text{kg}$), occurred in 1901. The 20 year mean Cl^- concentration at Site A ($0.30 \mu\text{eq}/\text{kg}$) is higher than at Site D ($0.20 \mu\text{eq}/\text{kg}$). The annual mean values for 1905 and 1906 at Site A are significantly higher than at Site D. These high Cl^- concentrations are associated with low $\delta^{18}\text{O}$ values in these annual layers. This will be discussed below. The low $\delta^{18}\text{O}$ values of 1892 and 1900 also appear to be related to the elevated Cl^- concentrations.

Nitrate and sulfate

The origin of the sulfate and nitrate is still uncertain. Some of the SO_4^{2-} is marine and it is transported along with Cl^- . The ratio between marine SO_4^{2-} and Cl^- is 0.10 by $\mu\text{eq}/\text{kg}$. The remaining parts of SO_4^{2-} and NO_3^- come from sulfurous and nitrous gases (mainly SO_2 , NO and NO_2) that have been oxidized to form H_2SO_4 and HNO_3 . The main sources of these gases are believed to be continental (Logan 1983, Neftel and others 1985[b], Herron 1982). Both NO_3^- and SO_4^{2-} , which form part of the tropospheric background aerosol, and nitrous and sulfurous gases are normally transported from their sources to Greenland in the mid- and high troposphere (Junge 1977). However, transportation sometimes occurs in the stratosphere, e.g. SO_2 from large volcanic eruptions (Hammer and others 1980, Clausen and Hammer 1987).

The seasonal variation in the NO_3^- concentration is the same as that of $\delta^{18}\text{O}$, with mid-summer maxima and mid-winter minima. This agrees with results from Crête and Dye 3 (Finkel and others 1986, Risbo and others 1981). There is no significant difference in 20 year mean nitrate concentration at the two sites ($1.11 \mu\text{eq}/\text{kg}$ at Site A and $1.16 \mu\text{eq}/\text{kg}$ at Site D).

The SO_4^{2-} concentration is high in spring–early summer and low in autumn and winter, as at Dye 3 (Finkel and others 1986). Maximum concentrations of SO_4^{2-} and dust occur at the same time. The shifts between winter and summer concentration levels, and several minor variations in these concentrations, also appear to be simultaneous. The annual SO_4^{2-} records from the two sites are similar. In contrast to the concentrations of dust, Cl^- and NO_3^- , the annual mean concentrations of SO_4^{2-} change during the

period under consideration. Between 1891 and 1902 the annual mean SO_4^{2-} concentrations have a constant value of $0.78 \mu\text{eq}/\text{kg}$. They then rise to a mean level of $1.31 \mu\text{eq}/\text{kg}$ for the period 1903–10.

DISCUSSION

Continental dust

The annual cycle of dust concentration is believed to result from changes both in continental sources and in atmospheric circulation. An aerosol-reservoir model (Heidam 1984, Barrie 1986) can explain the observations. In summer the atmospheric polar front lies to the north of several of the major source areas and blocks the transport of dust to the Arctic. The mid- and high troposphere over the Arctic therefore becomes more and more depleted in dust because it is removed continuously by precipitation and sedimentation. In autumn the polar front moves south; this permits the influx of dust from the major continental sources. In winter the Arctic aerosol reservoir begins to fill up—slowly in mid-winter—because there is snow cover over the continents, but later, in spring, the influx of dust rises when the snow disappears. Vigorous circulation over the continents, associated with the northward movement of the polar front, causes both horizontal and vertical transport of dust from its sources into the mid- and high troposphere of the Arctic. This fills up the Arctic reservoir. The reservoir thus contains most dust in spring, just before the polar front moves to the north and isolates the reservoir from the major sources. The higher dust concentration at Site D shows that mid-tropospheric air masses of continental origin have more influence there than at Site A.

Marine Cl^-

The presence of mainly mid- and high tropospheric air masses over the central Greenland ice sheet in summer and autumn could account for the low Cl^- concentrations observed at both sites during these seasons. The tropospheric background aerosol is low in Cl^- because of its continental origin and because the marine aerosol is efficiently removed by precipitation before reaching the mid- and high troposphere (Junge 1977). In winter the North Atlantic air masses are transported, via the low troposphere, along the east coast of Greenland, mainly from the north-east (Heidam 1984). Increased cyclonic activity south-east of Greenland in late winter might inject marine aerosols from the low to the mid-troposphere, and marine Cl^- would then be able to enter the atmosphere over the ice sheet. This could explain the relatively high Cl^- concentrations at sites A and D in winter–early spring. During spring the atmospheric circulation changes and continental air masses, mainly from the west, begin to enter the central Greenland region; Cl^- concentrations at sites A and D begin to fall and dust concentrations rise. Sometimes the cyclonic activity in late winter might be so strong that some of the low-pressure systems south-east of Greenland are strong enough to transfer marine air masses into the mid-troposphere, and at the same time affect the weather over the central Greenland ice sheet, causing precipitating air masses carrying marine Cl^- to come in from the south-east. These low-pressure systems are more likely to affect the weather at Site A, some 300 km inland, than at Site D, which is on the west side of the crest and 100 km farther inland. This might explain the layers with high Cl^- concentrations at Site A in 1905 and 1906, but they are not observed at Site D. The thickness of these layers indicates a large amount of winter precipitation, and the high Cl^- concentrations suggest a more direct transport of Cl^- than under "normal" conditions. Relatively high winter precipitation would give lower annual $\delta^{18}\text{O}$ mean values in these layers, in agreement with what has been observed. Although the increases in Cl^- concentration in the layers of 1892 and 1900 are not as significant as in the 1905 and 1906 layers, they may result from the same mechanism.

The results suggest that the composition of precipitation at Site A is more likely to be influenced by eastern and more marine air masses than at Site D. This marine influence varies from year to year, causing larger variations in the annual mean values of Cl^- concentration and $\delta^{18}\text{O}$ at Site A than at Site D. This has to be taken into account when discussing the climatic information from the Site A $\delta^{18}\text{O}$ record.

Heidam (1984) presents results of aerosol measurements in air at sea-level at several coastal stations in Greenland. He found that the concentration of marine aerosol has a maximum in summer and a minimum in winter, and explained this variation by a coupling between the marine-aerosol concentration in coastal air and the extent of sea ice. The disagreement between these results and the central Greenland and Dye 3 observations strongly indicates that the coupling between the low troposphere at the coast and the mid-troposphere over the ice sheet is not simple. It also suggests that the concentration of marine aerosols in the mid-troposphere is determined not by their concentrations in the low troposphere, but by vertical atmospheric transport mechanisms.

The Cl⁻ peak of 1901 in the Site D core

The enormous Cl⁻ concentration peak observed at Site D is a mystery. It occurs in summer precipitation, where Cl⁻ concentrations are normally low. It is not caused by contamination in the sampling procedure, because it was confirmed by a second set of samples from the core. Since the samples in question were taken from the middle of a firn-core segment that was without any breaks or cracks and since only ice from the core interior was used, it is not believed to be caused by contamination of the core. The peak is unlikely to result from dry deposition because it is found in three neighbouring samples representing a total of 15 cm of ice, which is half the normal annual accumulation. It is not caused by deposition of HCl, since pH measurements show no elevated H⁺ concentrations. Nor is it likely to be marine Cl⁻, because the peak should then be accompanied by a rise in SO₄²⁻ concentration to about 3.7 $\mu\text{eq/kg}$. Because this Cl⁻ concentration peak is believed to be non-marine it is omitted in the annual mean concentration plot in Figure 3. Since the peak is not observed in the Site A core, it appears to be a local phenomenon, and it is therefore not used to draw any conclusions concerning the Cl⁻ load in the atmosphere over Greenland in general at the time. Measurement of the cation counterpart of the Cl⁻ should add further information about the origin of this Cl⁻. The slightly elevated dust concentrations in the same layer, which are atypical of summer precipitation, might be related to the Cl⁻ peak; this would indicate that the Cl⁻ has a continental origin.

Nitrate and sulfate

Parallel to dust, both nitrate and sulfate (and their gaseous precursors) are believed to be mainly of continental origin and to have been transported to Greenland via the mid- and high troposphere. I therefore tried to explain the seasonal variations in NO₃⁻ and SO₄²⁻ in the context of the Arctic aerosol-reservoir model. The concurrence between the seasonal variation in the NO₃⁻ concentration and the $\delta^{18}\text{O}$ record, combined with the regularity of the seasonal variations in NO₃⁻, suggests that the Arctic reservoir does not become depleted in NO₃⁻ during summer, probably because it contains enough nitrous gases to supply the NO₃⁻ by photochemical processes resulting from solar radiation, as suggested by Finkel and Langway (1985). The Arctic reservoir might be supplied from the stratosphere (Risbo and others 1981), or from photochemical reactions on N₂, to maintain a high enough content of nitrous gases during summer. The observed seasonal variation in NO₃⁻ might therefore be caused mainly by seasonal variations in the production rate of NO₃⁻ from nitrous gases. The similarities between the seasonal variations in SO₄²⁻ and dust indicate that these components might share the same atmospheric transportation mechanisms from a certain point in their transportation history. As can be seen from Figure 3, the concentration levels of SO₄²⁻ have shifted between 1902 and 1903. The 1891–1902 concentration level agrees with the findings of Herron (1982) in pre-industrial ice from Crête, but the SO₄²⁻ concentrations in the period 1903–10 are significantly higher. The sudden shift between 1902 and 1903 is believed to have been caused by the 1902 eruptions of the volcanoes Mont Pelée, Soufrière and Santa Maria. These injected enough SO₂ into the atmosphere to cause a rise in SO₄²⁻ concentrations in Greenland precipitation (personal communication from C.U. Hammer). The increasing anthropogenic SO₂ input to the atmosphere, possibly

combined with further volcanic injections, is believed to be the reason why the SO₄²⁻ concentrations remain high for the rest of the period. A similar shift in SO₄²⁻ concentration around 1901 has been observed by Mayewski and others (1986) in an ice core from the Dye 3 area.

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