

## SEASONAL VARIATIONS IN SULFATE, NITRATE AND CHLORIDE IN THE GREENLAND ICE SHEET: RELATION TO ATMOSPHERIC CONCENTRATIONS

C. I. DAVIDSON, J. R. HARRINGTON, M. J. STEPHENSON, M. J. SMALL, F. P. BOSCOE and R. E. GANDLEY

Departments of Civil Engineering and Engineering and Public Policy, Carnegie Mellon University, Pittsburgh, PA 15213, U.S.A.

(First received 16 May 1988 and in final form 6 February 1989)

**Abstract**—Samples from three snowpits near Dye 3 in South Greenland have been used to study seasonal variations in contaminant transport from the atmosphere to the Ice Sheet. The snowpits cover the years 1982–1987. The samples have been dated by comparing  $\delta^{18}\text{O}$  values with meteorological data from Dye 3. Airborne concentrations of  $\text{SO}_4^{2-}$  over the Ice Sheet have been estimated for the dates corresponding to each snowpit sample by statistically analyzing data from several air monitoring stations throughout the Arctic, and computing average values from the appropriate stations. Seasonal variations in concentrations in air, concentrations in snow, and mass-basis scavenging ratios (concentration in snow divided by concentration in air) have been identified. Results indicate that concentrations of  $\text{SO}_4^{2-}$  in the air show a strong peak in late February, resulting from long-range transport of mid-latitude anthropogenic emissions, while those in the snow show a broad peak in January, February and March with smaller seasonal variation overall. The smaller variation in the snow is attributed in part to the effect of riming, which results in more efficient scavenging during warm weather when airborne concentrations are low. The importance of riming is also supported by the annual cycle in scavenging ratio which peaks in mid-summer coincident with maximum temperatures. In agreement with previous estimates, dry deposition appears to account for 10–30% of the total  $\text{SO}_4^{2-}$  in the snow. Concentrations of  $\text{NO}_3^-$  in the snow show a strong peak in summer; natural material from the stratosphere as well as anthropogenic emissions transported from the mid-latitudes may be responsible. Concentrations of  $\text{Cl}^-$  in the snow are maximum in January, with relatively high concentrations during October through March and a smaller peak in July. The winter peak is believed to reflect long-range transport (LRT) of marine aerosol from north Atlantic storms, while the summer peak is attributed to seaspray from nearby coastal Greenland. Riming also may influence the seasonal variations in  $\text{NO}_3^-$  and  $\text{Cl}^-$  in the snow.

**Key word index:** Sulfate, nitrate, chloride, precipitation scavenging, scavenging ratio, dry deposition, ice, snow, Arctic, Greenland.

### 1. INTRODUCTION

Several investigators have examined concentrations of chemical species in glacial snow and ice as a means of studying changes in atmospheric concentrations. Although a large number of contaminants have been examined, recent efforts have focused on acid  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$ . For example, Neftel *et al.* (1985) and Mayewski *et al.* (1986) have reported that concentrations of both of these species in Greenland snow have increased over the past several decades. The trends are attributed to increasing  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  emissions from anthropogenic activities in the mid-latitudes.

While gross changes in atmospheric levels are known to be reflected in ice cores, a quantitative link between airborne concentrations and the glacial record has not yet been established. This is because the mechanisms of transport from the atmosphere to the snow surface are extremely complex. Wet deposition may dominate during certain times of the year at some locations. This category includes nucleation of particles during the formation of cloud droplets or ice

crystals, and scavenging of gases and particles by hydrometeors within and below clouds. Dry deposition may dominate at other times and locations. This involves wind eddy transport from the free atmosphere to the boundary layer above the snow surface, followed by Brownian diffusion, interception, or inertial transport across the boundary layer. Sedimentation by gravity may influence particles that are sufficiently large. Other effects such as electrostatic or phoretic forces may also be important. The overall rates of transport by wet and dry deposition are dependent upon the mechanisms involved and are likely to be highly variable with space and time. A further complication is that the upper layers of snow may be redistributed by the wind, and contaminant migration may occur within the snowpack. The ways in which wet and dry deposition influence the glacial record have been discussed in detail elsewhere (Davidson, 1989).

In order to improve our understanding of air-to-snow transport, we previously conducted simultaneous air and snow sampling at Dye 3 on the south-