SOOT SCAVENGING MEASUREMENTS IN ARCTIC SNOWFALL

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Abstract—Simultaneous measurements of elemental carbon (EC) concentrations in air and in fresh snowfall were made for six storms during the spring of 1984 in the Swedish Arctic. Air concentrations ranged from 0.8-3.05 μg m⁻³, while concentrations in the fresh snowfall ranged from 60-4 μg cm⁻² melted water with a marked decrease in both the air and snow concentrations over the sampling period. The EC scavenging ratio in the snowfall remained roughly constant at ca 100 and appeared to be independent of these concentration changes. In addition to the EC measurements, pH, conductivity, and some major ions were measured in the fresh snowfall. Excess sulfate varied from 0-12 μM with the lowest value at the end of the sampling period, while pH ranged from 4.6-5.2, with the highest value at the end of the sampling period.

Key word index: Elemental carbon, Swedish Arctic, snowfall, ion chromatography.

INTRODUCTION

Considerable new information has been gathered over the last decade concerning pollution in the Arctic atmosphere and the deposition of pollutant material to the snowpack. Most of the data regarding the composition and concentration of this pollutant material can be separated into atmospheric measurements (Rahn et al., 1977; Daisey et al., 1981; Heinzenberg et al., 1981; Raatz, 1983; Shaw, 1985) or measurement of material in ice cores or in the snowpack (Herren, 1982; Neftel et al., 1985; Clarke and Noone, 1985). Despite this large body of data, simultaneous or near simultaneous measurements of contaminants in air and fresh snowfall are relatively rare (Davidson et al., 1985, 1987). Because of this, the scavenging ratios for various atmospheric constituents present in the Arctic environment are poorly characterized. One contaminant of particular importance to the Arctic region because of its potential to alter the radiative balance of the region is elemental carbon (EC), or soot. Soot has been measured in the Arctic atmosphere (Rosen et al., 1981; Lunenfors et al., 1983) and in the Arctic snowpack (Clarke and Noone, 1985), and removal rates for the wet deposition of EC have been measured outside the Arctic (Ogren and Charlson, 1984). Lacking, however, are direct measurements of the scavenging of soot to the Arctic snowpack. In this paper, we present simultaneous measurements of EC in both air and in fresh snowfall collected near cloud level for six storms sampled in the Swedish Arctic.

EXPERIMENTAL APPROACH

The sampling campaign was conducted in March and April of 1984 at the Swedish Academy of Science Research Station in Abisko, Sweden. This was selected as an accessible remote site that we expected to reflect conditions characteristic of the Arctic. The station itself is located roughly 200 km north of the Arctic Circle on the shore of lake Torneträsk (68°32'N, 18°49'E) in the eastern foothills of the mountains range separating Sweden and Norway (Fig. 1). Samples were taken at four sites in the vicinity of the station. Two of the sampling sites, Vassjäure (Fig. 1), at 350 m above sea level and Snöfjället (Fig. 1), at 500 m above sea level, were located at higher elevations in the mountain range, and were generally above cloud base during sampling. The other two sites, Slikvatten (Fig. 1), at 350 m and Riksdalen (Fig. 1), at 500 m above sea level, were located at lower elevations in the mountain range, and were generally above cloud base during sampling. These remote sites were selected to minimize the possibility of contamination by local sources. Local sources include a two-lane road, a railway, and small communities along the roadway. Assuming the roadways and railroads to be line sources and using typical traffic conditions along the roadway and EC production estimates (Charlson and Ogren, 1982), an estimate of the potential EC air concentration due to local sources was calculated to be ca 0.03 μg m⁻³. This value is between 1-3% of the observed EC air concentration values for all storms, except the last, and thus local sources are not considered to have been important.

Portable air pumps (Bendix Microaer model 900-10) were used to pull air through quartz fiber filters (Pallflex, Gelman) for determination of the atmospheric EC concentrations. These pumps were battery powered, with flow rates between...
0.5 and 1 l/min. The filter assembly consisted of a plastic filter holder that attached directly to the pump, with a plastic tube as a shield upstream of the filter. The pumps ran at a steady flow rate for ca 12 h, at which time the batteries quickly discharged and the pumps would stop. Since during a sampling event the pumps were deployed for longer than 12 h, experiments were undertaken to measure the uncertainty on the total volume of air sampled by each pump. This uncertainty was found to be between 5-10% for the pumps used in the experiment.

Snow samples were taken using a plastic spatula to scrape fresh snow into 500 ml polyethylene jars. Plastic gloves and nylon outer clothing were worn, and samples were taken facing into the wind to minimize contamination of the snow. The jars were sealed and placed in plastic bags for transport back to the laboratory. The snow was kept frozen during transport so that soot could not be lost to the jar walls by contact with meltwater. Once in the laboratory, the snow was transferred to a filtration apparatus where it was melted and filtered through 25 mm diameter, 0.1 μm pore size Nuclepore filters. Melting and filtration were carried out simultaneously to minimize the contact time of the meltwater with the filtration vessel walls, thus minimizing possible particle loss. Particles large enough to fall through the meltwater were retained, and an aliquot was used for sulphate and nitrate measurements. The remainder of the meltwater was frozen in a polyethylene bottle for later analysis of major ions by ion chromatography (IC).

The amount of EC on the filters was determined by optical analysis. The quartz filters (air samples) were measured using the integrating miniature technique (Clarke, 1985a) and the Nuclepore filters (snow samples) were measured using a modification of the integrating plate technique (Lin et al., 1973) that employed a neutral density filter (Clarke, 1982b).

The techniques entail measuring the change in transmission of the filters before and after sampling. The change in transmission can be determined against the mass of soot on the filter, allowing an estimation of the amount of EC on the filter from absorption measurements. Clarke et al. (1987) describe an intercomparison of light absorption measurement techniques for estimating soot in atmospheric aerosol samples. Their results show good agreement between absorption measurements and measured amounts of EC on various filter materials for laboratory-generated aerosol samples. Since determination of the amount of EC in snowfall involves working with particles suspended in water rather than in air, a separate calibration was undertaken for this medium. The method of calibration was first to weigh a series of 0.4 μm pore size, 25 mm diameter Nuclepore filters using a CAHN 21 electrobalance. Next, the intensity of light (525 nm) transmitted through these uncoated filters (I0) was measured using the integrating plate method with a neutral density filter in the light path (Clarke, 1982b). A hydrosol was then prepared containing 100 mg of Monarch 71 (M71) calibration soot (Cabot Corporation) dissolved in 80 ml filtered water and 20 ml filtered isopropyl alcohol. The hydrosol was placed in an ultrasonic bath for several minutes to disperse the soot particles. This hydrosol was then pre-filtered through a 20 μm pore diameter Nuclepore filter, then again through a 0.8 μm pore diameter Nuclepore filter. The purpose of these pre-filtration steps was to produce soot particles in the same size range as are found in the atmospheric aerosol (Clarke et al., 1987). Starting with a 1:10 dilution of the stock hydrosol, each of 10 steps the solution was separated into three aliquots. One was saved as a reference, one diluted 1:2 for use in the subsequent steps, and the third was filtered through one of the prepared Nuclepore filters. The filters were left to dry overnight, and then measured on the same CAHN electrobalance. After weighing, the transmission of 525 nm light was then measured on the loaded filters (I1) and the fractional change ([I0 - I1]/I0) was calculated. This fractional transmission change could
then be calibrated against the measured amount of M1 soot on the filter. Since there are absorbing materials in the atmosphere other than soot, this optical technique does not allow an unambiguous determination of EC. The contribution of other absorbing materials can be estimated, however, by measuring the transmission change at several wavelengths. Materials other than EC in the atmosphere typically have a different absorption wavelength dependence for visible light than does EC (Bohren and Huffman, 1983). This multi-wavelength technique has been used to estimate the amount of EC in both fresh and aged snow (Clarke and Noone, 1985). Using this technique, the amount of the total absorption due to soot has been estimated for the air and snow samples in this study, and indicated that soot was the major absorber of consequence at the Aktoke site.

Major ion (NO$_3^-$, SO$_4^{2-}$, Na$^+$, K$^+$, NH$_4^+$) concentrations in the filtered meltwater from the snow samples were measured by ion chromatography. H$^+$ was calculated from pH measurements made using a glass electrode.

**RESULTS**

Table 1 presents the data set for the six storms. Each row corresponds to a simultaneous snow/air sample. Column 1 is the sample identification number. Column 2 identifies the location of the sampling site. Column 3 shows the date that the sample was collected expressed as Julian day (1 January = 1, 31 December = 366; note that 1984 was a leap year). Column 4 shows the volume (ml) of meltwater from the snow sample. Columns 5 and 6 show the conductivity ($\mu$S cm$^{-1}$) and pH of the samples. Column 7 is the maximum atmospheric EC concentration (mg m$^{-3}$), this assumes all of the absorption of the air filter sample is due to soot. Column 8 is the corrected atmospheric EC concentration (mg m$^{-3}$) using the multi-wavelength technique to account for absorption due to other materials. Comparing columns 7 and 8 for the atmospheric EC concentrations (also 9 and 10, respectively, for EC concentrations in meltwater) gives an estimate of the fraction of the total absorption due to soot. Columns 9 and 10 show the corresponding maximum and corrected EC concentrations in the fresh snowfall (mg g$^{-1}$ meltwater) column 11 is the dimensionless "scavenging ratio" for soot ([mg EC g$^{-1}$ meltwater]/[mg EC g$^{-1}$ air]) using the corrected EC concentrations (rather than the maximum values) and a value of 1.293 kg m$^{-3}$ for the density of air. The remaining columns show measurements made on the filtered meltwater. The accuracy of the major ion measurements is $\pm 12\%$ of the given value, while the accuracy of the pH measurements is 0.09 pH units. Excess, or non-seasalt sulfate (as SO$_4^{2-}$, $\mu$M, column 19) was determined by multiplying the measured sodium value (column 15, $\mu$M) by the ratio of SO$_4^{2-}$/Na$^+$ in seawater (Holland, 1978) and subtracting this amount from the total measured sulfate (SO$_4^{2-}$, column 14, $\mu$M). The IC instruments were run at high sensitivity in order to maximize the response for NO$_3^-$ and SO$_4^{2-}$. As a consequence, the Cl$^-$ peaks were always off scale, and the true Cl$^-$ concentration could not be determined. The Cl$^-$ values reported

<table>
<thead>
<tr>
<th>ID</th>
<th>Location</th>
<th>Julian Day</th>
<th>Volume (ml)</th>
<th>EC (air)</th>
<th>EC (snow)</th>
<th>EC (snow)</th>
<th>pH (air)</th>
<th>pH (snow)</th>
<th>K$^+$ (mg l$^{-1}$)</th>
<th>Na$^+$ (mg l$^{-1}$)</th>
<th>Cl$^-$ (mg l$^{-1}$)</th>
<th>NO$_3^-$ (mg l$^{-1}$)</th>
<th>SO$_4^{2-}$ (mg l$^{-1}$)</th>
<th>Excess SO$_4^{2-}$ (mg l$^{-1}$)</th>
<th>HCO$_3^-$ (mg l$^{-1}$)</th>
<th>NH$_4^+$ (mg l$^{-1}$)</th>
<th>Ca$^{2+}$ (mg l$^{-1}$)</th>
<th>Mg$^{2+}$ (mg l$^{-1}$)</th>
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<td>44.4</td>
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<td>44.4</td>
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here \( \text{Cl}^- \), column 12, \( \mu \text{M} \)) were obtained by multiplying the measured sodium values by the ratio of chloride to sodium in seawater, the inherent assumption being that all of the \( \text{Cl}^- \) in the sample was due to seawater. Computing a charge balance using the measured ions and this estimated \( \text{Cl}^- \) value \( (\Sigma_{\text{anions}} - \Sigma_{\text{cations}}) \) (column 20) gives negative values that are close to zero for nearly all of the samples, indicating that this assumption is reasonable. The \( H^+ \) values (column 18, \( \mu \text{M} \)) were calculated from the pH measurements.

Differences in both EC and major ion concentrations were observed between the two sampling sites during a given storm. Since, however, there were no systematic differences between the sampling sites in terms of the measured parameters (that is to say that no site always showed higher or lower concentrations relative to the second site in any given storm over the sampling period), the values for the separate sites were averaged to give a mean value for each storm. This subset of the data is presented as a time series in Figs 3a and 2b. Errors for EC concentrations (snow) were estimated for three storms using information from duplicate samples, and are shown as error bars in Fig. 2. The error bars reflect the 95% confidence level and indicate measurement uncertainties due to sampling and analytical errors, but do not reflect any site-to-site variability. To illustrate the magnitude of the site-to-site variability, samples having the maximum concentration difference between sites for a given storm are illustrated along with a site identifier.

The first samples were taken on 24 March 1984 (J.D. 84) and the last samples on 23 April (J.D. 115). An overall decrease in EC concentrations of roughly an order of magnitude can be seen over this time period, both in air and in snow (Fig. 2a). The magnitude and period of this decrease is the same as that reported at numerous other Arctic locations for other atmospheric constituents associated with Arctic haze (Joranger and Ottar, 1984, Barrie and Hoff, 1985).

This suggests that our data reflect regional Arctic haze behavior and its typical removal mechanisms. The variation in EC (air) concentrations of ca 0.8 to 0.05 \( \mu \text{g m}^{-3} \) is similar to the atmospheric values of 0.6 to 0.05 \( \mu \text{g m}^{-3} \) measured during aircraft flights out of Barrow, Alaska during April of 1982 (Clarke et al., 1984). The data in the Clarke et al. (1984) study were collected at varying altitudes and locations in the Arctic, and reflect the influence of different air mass sources. The data in this study reflect a temporal...
The comparison here is to illustrate that the range and trend of EC (air) values observed at the sites in this study reflect those observed for the extended Arctic region. The range of EC (snow) concentrations of ca 3–60 μg Cl\(^{-1}\) meltwater is comparable to the range of values found elsewhere in the Arctic (Clarke and Noone, 1985). The snow scavenging ratio for the six storms does not show a definite trend over the time period. The mean value and 95% confidence interval of the scavenging ratio for the six storms is 97 ± 34.

Since this scavenging ratio does not give information on any specific physical process incorporating material into precipitation, but rather the combined effect of the various removal processes, it is impossible to draw conclusions from this data regarding the removal mechanisms for soot. The absence of a clear trend in the scavenging ratio despite the large decrease in EC concentrations does indicate, however, that the total effect of the removal mechanisms was relatively constant during the time period the samples were taken.

In addition to the scavenging ratio for EC, another area of interest is the potential for the EC present in the Arctic snowpack to alter the radiative balance of the region. At the EC (snow) concentration levels reported here, Warren and Wiscombe (1985) estimate a possible decrease of several percent in the albedo of pure snow vs snow with incorporated EC.

Turning to the chemical parameters, the pH of the meltwater ranged from a minimum of 4.6 to a maximum of 5.2 over the sampling period. Ross and Granat (1986) report pH values of 4.8 for snow samples taken during February and March 1984 at sites in Sweden south of the Abisko area. In terms of hydrogen ion concentration (Fig. 2b), the lowest [H\(^{+}\)] value was at the end of the sampling period, and there was a general (though not monotonic) decrease in [H\(^{+}\)] over the same period as observed in the EC values. In contrast to the lowering of EC and hydrogen ion concentrations as a function of time, xs SO\(_{4}^{2-}\) does not show a clear trend over this period (Fig. 2a). However, NO\(_{3}^{-}\) and xs SO\(_{4}^{2-}\) are taken together, they parallel the [H\(^{+}\)] behavior in both sense and magnitude (Fig. 2b). Clearly, xs SO\(_{4}^{2-}\) and NO\(_{3}^{-}\) are controlling factors in determining the acidity of the snowfall at this site. It is interesting to note that the nitrate concentrations are usually similar to, and often slightly larger than the excess sulfate concentrations. There are two negative values given in Table 1 for xs SO\(_{4}^{2-}\) which are not physical, since there can be no negative concentrations. These negative values arise when the total SO\(_{4}^{2-}\) concentrations are low and the Na\(^{+}\) values are high. In these cases, small uncertainties in the Na\(^{+}\) measurements can induce large errors in the xs SO\(_{4}^{2-}\) values via the sea-salt correction.

Atmospheric xs SO\(_{4}^{2-}\) concentrations (expressed as xs SO\(_{4}^{2-}\)–S, sulfate as sulfur) for roughly the same time period as the samples reported here were taken have been reported for the Scandinavian area (Iversen and Jorasø, 1985). Interpolating from their results gives a value of roughly 1.2 μg xs SO\(_{4}^{2-}\)–S m\(^{-2}\). The average xs SO\(_{4}^{2-}\) concentration in snow for storms 2-6 is 0.16 μg xs SO\(_{4}^{2-}\)–S g\(^{-1}\). These values, along with the air density can be combined to give a scavenging ratio for excess sulfate of 175. Davidson et al. (1987) report scavenging ratios for sulfate in Greenland of between 100–200 during winter and 200-400 during summer. Our value for March–April in Abisko falls in their range of winter values.

A comparison of the ratio of excess sulfate to EC in fresh snow from Abisko can be made to measurements of this ratio in precipitation. Ogren and Charlson (1984) report a value of 8 for the wet deposition flux of excess sulfate (as sulfur) to EC in Sweden during April–August, 1981. For a given area and precipitation rate, this ratio is equivalent to the corresponding xs SO\(_{4}^{2-}\)–S/EC concentration ratio. The average ratio for storms 2-6 in Abisko is 3. These values are very close, especially considering the time difference between the two sampling campaigns and the potentially different scavenging mechanisms involved; precipitation fell both as rain and snow in the Ogren and Charlson study, while precipitation fell exclusively as snow here.

A more quantitative picture of how the various constituents are related to each other than that of the time series presented here can be obtained by examining the correlation matrix of the measured constituents. Table 2 is the correlation matrix for eight of the measured chemical constituents plus conductivity for storms 2-6. The matrix is symmetric, so only the lower half is shown. The values on the principal diagonal (all 1's) indicate perfect correlation and are not shown.

Table 2. Correlation matrix for selected chemical parameters

<table>
<thead>
<tr>
<th></th>
<th>EC (air)</th>
<th>H(^{+})</th>
<th>NO(_{3}^{-})</th>
<th>Na(^{+})</th>
<th>K(^{+})</th>
<th>NH(_{4}^{+})</th>
<th>EC (snow)</th>
<th>xs SO(_{4}^{2-})</th>
</tr>
</thead>
<tbody>
<tr>
<td>EC (air)</td>
<td>1</td>
<td>0.596</td>
<td>0.455</td>
<td>0.431</td>
<td>0.049</td>
<td>0.035</td>
<td>0.396</td>
<td>0.596</td>
</tr>
<tr>
<td>H(^{+})</td>
<td>0.596</td>
<td>1</td>
<td>0.77</td>
<td>0.694</td>
<td>0.672</td>
<td>0.406</td>
<td>0.77</td>
<td>1</td>
</tr>
<tr>
<td>NO(_{3}^{-})</td>
<td>0.455</td>
<td>0.77</td>
<td>1</td>
<td>0.567</td>
<td>0.492</td>
<td>0.492</td>
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<td>1</td>
</tr>
<tr>
<td>Na(^{+})</td>
<td>0.431</td>
<td>0.694</td>
<td>0.567</td>
<td>1</td>
<td>0.131</td>
<td>0.131</td>
<td>0.694</td>
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<tr>
<td>K(^{+})</td>
<td>0.049</td>
<td>0.672</td>
<td>0.492</td>
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<td>1</td>
<td>0.147</td>
<td>0.672</td>
<td>1</td>
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<tr>
<td>NH(_{4}^{+})</td>
<td>0.035</td>
<td>0.406</td>
<td>0.492</td>
<td>0.131</td>
<td>0.147</td>
<td>1</td>
<td>0.406</td>
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<tr>
<td>EC (snow)</td>
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<td>0.77</td>
<td>0.77</td>
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<td>0.147</td>
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<tr>
<td>xs SO(_{4}^{2-})</td>
<td>0.596</td>
<td>0.77</td>
<td>0.77</td>
<td>1</td>
<td>0.131</td>
<td>0.131</td>
<td>0.694</td>
<td>1</td>
</tr>
</tbody>
</table>

Correlation coefficients not significantly different from zero at the 95% level are in bold. The higher r values (0.89 and 0.77) show correlation between H\(^{+}\), NO\(_{3}^{-}\), and xs SO\(_{4}^{2-}\). High values are seen for EC (air) and EC (snow) (0.69) and between EC (snow) and xs SO\(_{4}^{2-}\) (0.653).
are the correlation of each variable with itself. The off-diagonal terms are correlations between pairs of variables defined by the row and column headings. The boldface entries in the table indicate correlation coefficients significantly different from zero at the 0.05 confidence level. The highest correlations are between \( H^+ \), \( NO_3^- \), and \( SO_4^{2-} \) (0.77 and 0.89, respectively).

An interpretation of these correlations would be that excess sulfate and nitrate are the important species defining the acidity of the snowfall at the site, as was illustrated in Fig. 2b. There is also a correlation between EC (snow) and \( SO_4^{2-} \) (0.65). Examining column 1 [EC (air)] Table 2, there are correlations between EC (air) and EC (snow) (0.69), \( H^+ \) (0.69), and \( SO_4^{2-} \) (0.60). The presence of EC and this grouping of species suggests an anthropogenic pollution component. Since the local contribution to the EC concentrations at the site is expected to be small, this likely reflects pollutant material that has been transported to the site over long distances.

CONCLUDING REMARKS

Simultaneous measurements of EC in air and in fresh springtime snowfall at a site in the Swedish Arctic show atmospheric concentrations decreasing from ca. 0.08 to 0.05 mg m\(^{-3}\), while the corresponding snow concentrations decreased from ca. 60 to 4 mg m\(^{-3}\). The scavenging ratios for EC obtained from these measurements average 97 ± 34 and are consistent with ratios determined for fine particle lead and sulfate found in Greenland.

Measurements of major ions in the melted snow samples show a correlation between EC and excess sulfate in the snowfall, as well as suggesting that excess sulfate and nitrate are the major factors in determining the acidity of the snowfall at the site.

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