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Wet deposition of elemental carbon and sulfate in Sweden

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ABSTRACT

Elemental carbon and sulfate were measured in monthly precipitation samples at 12 rural sites in Sweden from April to August, 1981. Wet deposition of excess sulfate was significantly correlated with elemental carbon deposition, and no systematic spatial or temporal variations in the ratio of excess sulfate to elemental carbon were observed. Comparision of these results with sulfur and elemental carbon emissions in Western Europe suggests that elemental carbon is removed at a slower rate than sulfur for the first day or so after emission.

1. Introduction

Particulate elemental carbon (EC) is an important constituent of the atmospheric aerosol because of its high specific absorption of solar radiation, large surface area available for adsorption of other trace atmospheric species, and the catalytic activity of its surface. The effects of these properties of EC include warming of the atmosphere, reduction of solar radiation reaching the ground, reduction of the albedo of clouds and snow, long-range transport of compounds adsorbed on the EC surface, and enhancement of the rate of oxidation of dissolved sulfur dioxide. Furthermore, the chemical stability of EC suggests that it might be useful as an inert tracer for distinguishing between dilution and removal processes, which affect all particles, and chemical transformations, which affect only the chemically reactive species.

Measurements of the removal rate of EC at an urban site in Seattle and at 12 rural sites in Sweden were reported previously (Ogren et al., 1984). Concentrations of EC in precipitation were comparable in both urban and rural regions, as were the EC wet deposition fluxes. Wet removal was found to be responsible for most of the flux of EC to the glass collection vessels. The results from that study were interpreted to suggest that the same mechanisms were acting to remove EC and soluble particles in Sweden, due to the EC particles becoming coated with hygroscopic substances (e.g. sulfates).

Additional information on the extent of coating of EC particles by hygroscopic substances, and thereby the dominant removal mechanism for EC, is provided by comparing the relative removal rates of EC and sulfates. If the removal rates of EC and sulfates show different variations with space and time, then it is most likely that different mechanisms are acting to remove the two substances. Conversely, a positive correlation of the two removal rates provides support for, but does not prove, the hypothesis that the same mechanisms act to remove EC and sulfates after sufficient time has passed for the EC particles to acquire a hygroscopic coating. Changes in the ratio of sulfur-to-EC from sources in Western Europe to rainwater falling on Sweden provide additional information on changes in the relative rates of removal of the two substances.

2. Experimental approach

In the previous study (Ogren et al., 1984), EC deposition fluxes and concentrations in precipitation were reported for 12 rural sites in Sweden (Fig. 1). The EC samplers were located adjacent to samplers that form a part of the routine air and precipitation chemistry sampling network in

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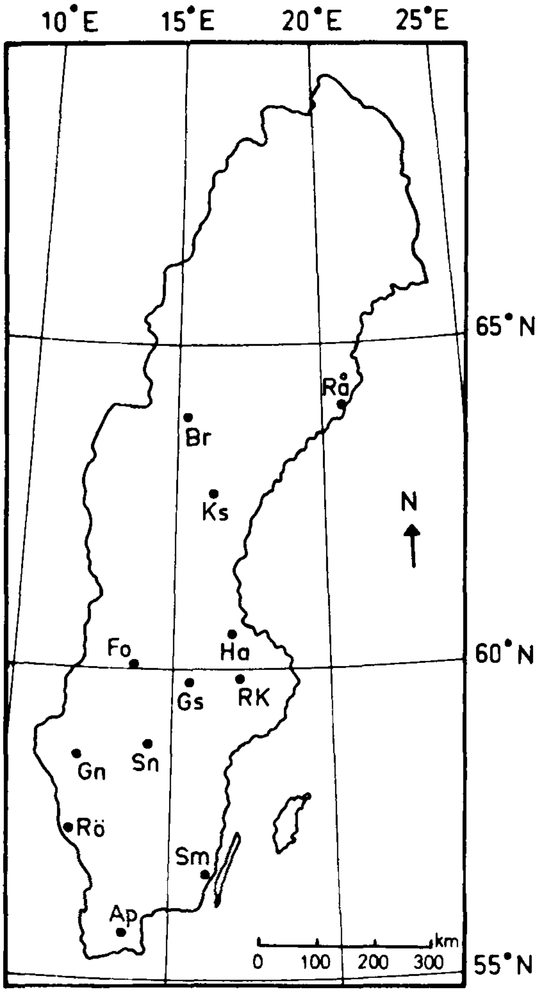
[1] Contribution No. 500.



Site Name	ID
Arup	Ap
Smedby	Sm
Rörvik	Rö
Granan	Gn
Sjöången	Sn
Grimsö	Gs
Ryda Kungsgård	RK
Forshult	Fo
Hedesunda	Ha
Kassjö	Ks
Bredkälen	Br
Rickleå	Rå

Fig. 1. Location of sampling sites in Sweden.

Sweden. This provided the advantages of the network's routine measurements of the chemical composition of precipitation and air, a historical data base, site documentation and logistical support, and, in particular, freedom from local sources. Samples of the monthly average total deposition of EC were obtained by exposing glass jars to the air and precipitation; similar collectors, exposed to the air but protected from precipitation, were used to determine the contribution of dry removal to the total deposition samples. The collectors used in the routine Swedish sampling network for determining the ionic composition of precipitation were exposed only during precipitation events.

A detailed description of the method for determining EC concentration in rainwater was presented earlier (Ogren et al., 1983, 1984). At all sites except Rörvik, samples were analyzed by colorimetric methods (SO_4^{2-}, Cl^-, NO_3^-, NH_4^+), flame emission spectroscopy (Na^+, K^+), and flame atomic absorption spectroscopy (Mg^{2+}, Ca^{2+}) (Granat et al., 1977; Trägårdh and Granat, 1982). Glass electrodes were used to measure pH at all sites and NH_4^+ in the samples from Rörvik; other cations were not determined in the Rörvik samples. Anions (SO_4^{2-}, NO_3^-, Cl^-) in the Rörvik samples were determined using ion chromatography. Two of the samples were rejected because the measured anion and cation equivalent sums were out of balance; this quality check could not be performed on the Rörvik samples because some important cations were not determined in them.

3. Results

Collection of samples on a monthly basis makes it difficult to specify the relative contributions of rain and snow to the measured wet deposition fluxes. Precipitation during the month of April was primarily in the form of snow, while rain was the predominant form of precipitation after the first week of May. The least amount of precipitation fell in April, while June was the wettest of the 5 months studied.

Wet deposition of excess (non sea-salt) sulfate showed a similar pattern to that of EC deposition (Table 1), with a weak gradient from south to north. This behavior has also been noted for excess sulfate deposition in Sweden by Granat (1978). Sea-salt contributed less than 3% of the measured sulfate in all but six samples, with a maximum contribution of 10%. The correction for sea-salt sulfate was based on the chloride concentration in the sample; the difference in excess sulfate values produced by using sodium for the correction was negligible.

A comparison of the wet deposition rates of excess sulfate and EC is shown in Fig. 2. The linear correlation coefficient (r^2) for the entire set of 53 points is 0.29, calculated to be non-zero at the 0.005 level of significance; in other words, the probability that the two deposition fluxes are independent is less than 0.5%. The slope of the least-squares regression line is 4.4, with an intercept of 27. If the labelled outliers are excluded, for the reason that least-squares regression line parameters are strongly influenced by outliers, the regression line passes close to the origin, and shows that the wet deposition flux for excess sulfate (as sulfur) is greater by about a factor of eight than the elemental carbon deposition rate.

We consider the samples from Kassjö (Ks) for May, June, and July to be contaminated, because the wet deposition fluxes of ammonium, magnesium, potassium, and chloride are all higher than typical values for the other sites. In addition, the pH values for these three samples are greater than 6, indicative of contamination by alkaline compounds. One likely source of such compounds is an unpaved road that passes close to the sampler, although agricultural chemicals used on the farm on which the sampler is located might also be a factor.

The remaining unexplained outliers, all from Arup and Rörvik, are excluded from the regression analysis only because the least-squares procedure is very sensitive to them, drawing attention away

Table 1. *Elemental carbon wet deposition flux (mg m^{-2} mo^{-1}). Values in parentheses are corresponding excess sulfate (as sulfur) wet deposition fluxes (mg m^{-2} mo^{-1}). Entries are listed in order of increasing latitude*

	Month (1981)				
Site	4	5	6	7	8
Ap	5 (29)	24 (68)	13 (163)	5 (105)	5 (32)
Sm	4 (17)	4 (43)	10 (127)	8 (43)	NA (24)
Rö	2 (28)	21 (104)	8 (108)	2 (97)	1 (17)
Gn	4 (20)	14 (98)	14 (81)	7 (38)	2 (14)
Sn	3 (11)	11 (98)	9 (80)	7 (72)	3 (18)
Gs	4 (NA)	2 (44)	11 (93)	NA (111)	4 (10)
RK	3 (23)	4 (17)	8 (84)	5 (41)	2 (34)
Fo	1 (NA)	9 (NA)	11 (NA)	9 (46)	1 (6)
Ha	3 (17)	3 (71)	10 (50)	5 (64)	2 (20)
Ks	6 (11)	3 (106)	6 (147)	2 (85)	1 (9)
Br	3 (10)	3 (29)	8 (45)	4 (82)	1 (8)
Rå	4 (29)	2 (NA)	8 (48)	10 (69)	2 (9)

NA: not available.

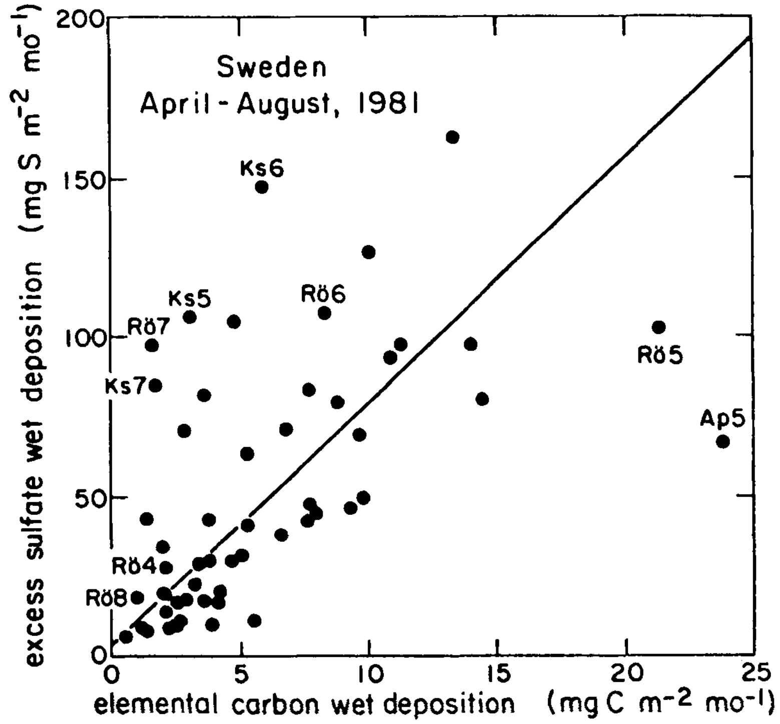


Fig. 2. Wet deposition fluxes of excess sulfate and elemental carbon. Labelled points are excluded from the least-squares line (shown) for reasons discussed in the text. Uncertainties for 95% confidence and regression-line parameters are: $y = (4 \pm 50) + (7.6 \pm 2.0)x$; $r^2 = 0.57$; $n = 44$.

from an overall pattern which needs to be understood. Repeating the line-fitting analysis for all the samples with an algorithm, based on medians, that is less sensitive to outliers (Velleman and Hoaglin, 1981) yields almost the same result as the least-squares procedure with the labelled outliers in Fig. 2 excluded.

No obvious reasons have been found for the high wet deposition rates found at Arup and Rörvik in May, although the proximity of these sites to each other and to the major source regions in Europe suggests the possibility of an isolated instance of transport of air rich in EC, followed by relatively localized deposition. Dry deposition at Rörvik also appeared to be controlled by a different set of factors than those affecting the other sites as three of the six bulk deposition samples which had more than 15% of the EC mass contributed by dry processes were from this site. Further research at Arup and Rörvik is needed to understand the different EC deposition fluxes found there.

Further insight into the relationship between excess sulfate (as sulfur) and EC wet deposition is provided by analysis of their ratio (S^*/C) as a function of site and month. This analysis, conceptually similar to analysis of variance, is based on medians instead of means in order to minimize the influence of outliers. A two-way multiplicative model is used to represent the measured ratio S^*/C for each sample as the product of a common term, a month-dependent effect, a site-dependent effect, and a residual (Tukey, 1977). The residual is the ratio of the measured value and the value predicted for that site and month by the product of the common term, month effect, and site effect. The size of the residuals is a measure of how well the model represents the measured values.

The two-way multiplicative model begins with a table of the logarithms of S^*/C, where each row contains the samples from one site and each column contains the samples from 1 month (similar to Table 1). The median value for each row is subtracted from the entries in that row, and recorded in a separate column as the first estimate of the row effect. Likewise, the median value of each column is subtracted from the entries in that column, and recorded in a separate row as the first estimate of the column effect; the median value of the row effects column becomes the first estimate of the common term, and is subtracted from each of the entries in the row effects column. This procedure is repeated in each direction, with subsequent row (column) medians being added to the row (column) effect and subtracted from the entries in that row (column). Eventually, the median of each row and column gets close to zero, signifying that all the systematic trends in the body of the table have been transferred into the row and column effects, and all systematic trends in the row and column effects have been transferred into the common term. The final step in the analysis is to take the antilogarithm of each value in order to convert the results of the two-way additive model into a two-way multiplicative model.

The results from performing the above procedure twice in each direction are presented in Table 2; repeating the procedure twice more in each direction did not change the results substantially. As an example for interpreting Table 2, the ratio measured at Arup in April (6.2) is the product of the common term (7.44), the month effect for April (0.73), the site effect for Arup (1.09), and the residual (1.04). Entries in the table between 0.6 and 2 are not considered to be significantly different from 1, given the ±50% accuracy of the EC measurements. Slightly different results are obtained if the analysis begins with the columns rather than rows, but the differences are not great enough to alter the conclusions drawn from the analysis.

The value of the common term is 7.4, very close to the slope of the regression line in Fig. 3 and the median of the entire sample set of 6.8. No significant monthly variation is revealed by the analysis. Only two sites are seen to have site effects significantly different from 1: Rörvik and Kassjö. This is in accord with the discussion above, which identified these two sites as being markedly different from the others. The low residuals at Kassjö for April and August are due primarily to a high site effect, although the S*/EC ratio for April is lower than most. The very high EC wet deposition flux at Arup in May is reflected in the residual term, which is the lowest residual for any entry other than those from Rörvik and Kassjö. Two other samples show significantly low residual terms, while three have significantly high values.

Table 2. *Two-way multiplicative model of the ratio of excess sulfate (as sulfur) to elemental carbon in precipitation, with significant residuals in boldface type. Sites are in order of increasing latitude*

Site	Residuals by month					Site effect
	4/81	5/81	6/81	7/81	8/81	
Ap	1.04	**0.26**	1.51	**2.17**	0.79	1.09
Sm	0.81	1.24	1.86	0.67	—	0.92
Rö	1.13	**0.24**	0.84	**3.15**	1.00	**2.08**
Gn	1.16	0.93	1.00	0.82	1.08	0.76
Sn	0.83	0.97	1.35	1.27	0.89	0.90
Gs	—	**2.58**	1.00	—	**0.30**	1.15
RK	1.04	**0.37**	1.15	0.65	1.80	1.27
Fo	—	—	—	0.62	1.57	0.86
Ha	0.94	1.93	**0.53**	1.00	1.00	1.28
Ks	**0.11**	1.03	1.00	1.61	**0.26**	**3.35**
Br	0.96	1.12	0.98	**3.12**	1.00	0.77
Rå	1.74	—	1.03	0.95	0.65	0.81
						Common term
Month effect	0.73	1.33	1.00	1.24	1.00	**7.44**

S*/EC = (common term) (month effect) (site effect) (residual).

This indicates that there may be other independent variables affecting the measured ratio of excess sulfate to elemental carbon in precipitation.

4. Discussion

Sufficient information on source emission factors is available to prepare a preliminary estimate of EC emissions in Europe. This will provide a reference for comparison with the magnitude of EC deposition in Sweden, and will allow an estimate of the relative rates of removal of EC and sulfur by means of comparison with sulfur emissions in Europe. The source emission factors used to calculate EC emissions are shown in Table 3. The value shown for solid fossil fuels is an arbitrary assumption, chosen so that coal (and other solid fossil fuel) combustion could be included in the emission inventory for comparison with other sources, but is consistent with the range of EC emission factors of 0.0005–0.004 g kg^{-1} calculated by Wolff et al. (1981) for coal combustion in Denver. Many of the other values in Table 3 are based on limited measurements, and are probably not better than order of magnitude indicators. In particular, the emission factor for residual fuel oil is considered to be very uncertain, while much higher emission factors for natural gas have been reported for some uses (Hansen et al., 1978). The emission factor for diesel fuel is probably one of the best characterized values in Table 3.

The resulting EC emissions for 1978 are presented in Table 4, based on fuel consumption values reported by the Organization for Economic Cooperation and Development (OECD, 1982a). 1978 was chosen as a reference year because of the availability of sulfur emission estimates for that year (EMEP, 1981). Fuel combustion in 1981 was about 10% lower than in 1978 (OECD, 1982b), so that the 1978 emission estimates should be appropriate for comparison with the 1981 EC deposition measurements. Distillate fuel oil consumption is divided into two categories to account for the large difference in emission factors between transportation (i.e., diesel vehicles) and other uses of distillate fuel oil. The solid fuel category includes hard coal, coke, and lignite. Emissions due to jet fuel are based on the assumption that only 10% of the fuel is burned within the lowest few kilometers of the atmosphere, and hence available for deposition within Europe.

Estimated emissions from diesel engines are seen to dominate, contributing over 80% of the total. Considering that the emission factor for diesel fuel is one of the better characterized values, it seems likely that the total emission estimates are probably lower bounds for the true emissions. Examined in another light, the emission factor for solid fossil fuels would have to be 0.3 g kg^{-1} in order for these fuels to equal the emissions from diesel vehicles; corresponding emission factors (g kg^{-1}) required to equal emissions from diesel vehicles are 0.07 for natural gas, 1.0 for gasoline, 60 for jet fuel, 0.7 for other uses of distillate oil, and 0.5 for residual fuel oil.

The annual wet deposition of EC in Sweden can be estimated as the product of the average wet deposition rate (6 mg m^{-2} mo^{-1}) and the area of Sweden (4.5×10^{11} m^2), or roughly 3×10^7 kg a^{-1}. This is about 20% of the total emissions in the European OECD countries (listed in Table 4),

Table 3. *Source emission factors for elemental carbon*

Fuel	Use	g EC/kg fuel	Reference
Natural gas	All	0.0003	Hansen et al. (1978) Muhlbaier and Williams (1982)
Gasoline	Automobile	0.02	Muhlbaier and Williams (1982)
Distillate	Diesel	2	Muhlbaier and Williams (1982) Springer (1978)
	Other	0.1	Cass et al. (1982) Hersh et al. (1976)
Residual	All	0.02	Cass et al. (1982)
Jet	All	1	Stockham et al. (1979) Cass et al. (1982)
Solid fossil	All	0.001	Assumed

Table 4. *Estimated elemental carbon emissions in western Europe (1978)*

	EC emissions (10^3 kg a^{-1})								
Country	Solid fossil fuels	Natural gas	Gasoline	Jet fuel	Distillate		Residual oil	Total ($\times 10^{-3}$)	Sulfur/EC
					Diesel	Other			
Austria	6	11	46	10	1,800	190	100	2.2	98
Belgium	11	25	62	46	3,400	780	160	4.5	90
Denmark	6	1	35	80	2,000	550	110	2.8	81
Finland	6	4	27	19	2.100	370	87	2.6	100
France	42	60	350	230	18,000	3,200	650	23.0	78
Germany, FR	180	120	470	270	21,000	5,300	520	28.0	65
Greece	22	—	27	110	2,100	210	95	2.6	140
Iceland	—	—	2	8	360	13	3	0.4	15
Ireland	1	—	20	30	720	110	53	0.9	97
Italy	8	62	220	160	16,000	1,700	850	19.0	120
Luxembourg	2	3	5	6	190	47	10	0.3	80
Netherlands	3	80	79	85	5,700	500	110	6.6	36
Norway	1	2	27	25	2,600	240	31	2.9	26
Portugal	1	—	15	54	2,300	29	57	2.5	34
Spain	19	9	110	220	11,000	430	430	13.0	79
Sweden	2	1	74	55	3,300	740	200	4.4	63
Switzerland	—	2	52	76	1,000	710	33	1.9	31
Turkey	13	1	46	17	4,600	250	120	5.0	100
UK	110	100	370	450	16,000	1,400	630	19.0	140
OECD Europe	430	470	2,000	2,000	110.000	17.000	4,300	140.0	87

which seems to be a rather high percentage. By comparison, the annual deposition of sulfur in Sweden is only 2% of the sulfur emitted in OECD Europe (EMEP, 1981). Two factors are likely contributors to this apparently high percentage of total EC emissions deposited in Sweden: underestimates of the EC source strength (due to low emission factors and/or other sources), and greater removal of sulfur within the major source regions than is the case for EC (i.e. relatively less sulfur than EC reaches Sweden).

Sources of EC other than those shown in Table 4 are likely to exist in Western Europe, although insufficient data are available to estimate their magnitude. One such source, which might be important because of its proximity to Sweden, is the poorly-controlled combustion of waste natural gas from the North Sea oil fields. Combustion of biomass (e.g., wood and peat) is another likely source, some of which may be due to natural causes (e.g., forest fires started by lightning); no other natural sources are expected for EC. All of these sources of EC are likely to have a lower ratio of sulfur to EC than the average value shown in Table 4, suggesting that the average ratio in Table 4 is probably an overestimate.

The ratio of sulfur to EC in the sources is estimated to vary from 15 to 140 for the OECD countries of Western Europe, with an emissions-weighted mean value of 87 (Table 4). The variability of this ratio indicates that air parcel trajectories should have some effect on the ratio of excess sulfur to EC measured in precipitation in Sweden. For example, two of the largest contributors to the sulfur deposited in Sweden, West Germany and the United Kingdom, are estimated to have sulfur-to-EC ratios in the sources of 65 and 140, respectively. Rodhe et al. (1972) concluded in an earlier analysis of soot (measured by the British smoke shade method, OECD, 1964) and sulfate data from southern Sweden that different air masses appear to have different characteristic soot-to-sulfate ratios. A subsequent comparison of EC and soot measured by the British smoke shade method (Edwards et al., 1983) suggests that the findings of Rodhe et al. (1972) can also be applied to the ratio of EC-to-sulfate. However, the absence of data on sea-salt sulfate prevents quantitative comparison of the results of Rodhe et al. (1972) with the results of the present study.

The ratio of sulfur-to-EC in western European sources is roughly one order of magnitude greater than that measured in precipitation in Sweden. Although an underestimate of EC emissions probably contributes somewhat to this difference, atmospheric physical and chemical processes are also expected to have an effect. Much of the sulfur near the sources is present as sulfur dioxide, and is removed from the atmosphere by wet and dry processes with a turnover time of about 40 h, roughly twice as fast as the rate for sulfates (Rodhe, 1978). If EC in the sources is hygrophobic and is in the accumulation mode (0.1–1 μm diameter), the removal rate for EC can be much lower than that for sulfates (Ogren and Charlson, 1983). Hence, proportionally more sulfur than EC is removed near the sources, serving to decrease the ratio of sulfur to EC in the air. Eventually, the airborne SO_2 is converted to sulfates, which coat the surface of the EC, after which the two species are expected to be removed at the same rate. However, it can take over 1 day for accumulation-mode EC to acquire this coating in air masses representative of aged urban plumes (Whitby, 1978; Ogren and Charlson, 1983). Considering that 1 day is a realistic transport time from western Europe to Sweden, it is likely that a substantial fraction of the EC reaching Sweden is not yet coated with sufficient hygroscopic material (ca. 10% or more by volume, Junge and McLaren, 1971) to grow significantly at high relative humidities. This is consistent with the results of Covert and Heintzenberg (1984), who found that EC in seven samples collected in Stockholm was relatively non-hygroscopic.

It is interesting to note that the ratio of the mass of excess sulfate (as sulfur) to EC in precipitation in Sweden was below four for only three of the samples, and was never lower than two (Fig. 3). This might suggest that there is some limit to the extent of preferential removal of sulfur. The picture that emerges is one where relatively little removal of EC occurs during the first day or so in the atmosphere, whereas a significant amount of the sulfur is lost. As the air parcel ages, dry deposition of sulfur becomes less important, and the relative rate of removal of EC increases as the EC particles acquire hygroscopic coatings. Farther downwind, the removal rates of sulfur and EC are expected to approach one another as the processes of coagulation and coating by gas-to-particle conversion continue.

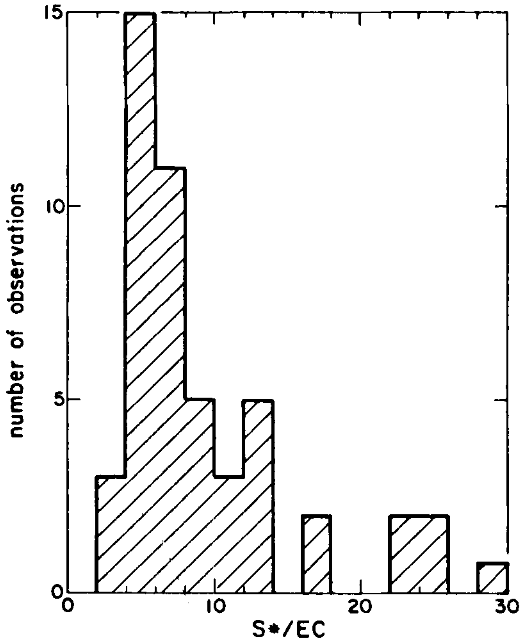


Fig. 3. Frequency of occurrence of excess sulfate (as sulfur) to EC ratio in precipitation. Outliers not shown are from Kassjö (34, 49) and Rörvik (61).

The ratio of sulfate to EC in aerosol particles is controlled by the source ratio of sulfur to EC, the conversion rate of sulfur dioxide to sulfates, and the relative rates of removal of EC, sulfur dioxide, and sulfates. Low values of the ratio of aerosol sulfate and EC concentrations (black episodes) were attributed by Rahn et al. (1982) to relatively unaged emissions (ca. 1 day) with high ratios of gaseous to particulate sulfur, while higher ratios of sulfate to EC (white episodes) were attributed to emissions that had aged for several days, with much of the SO_2 having been converted to sulfate. Considering the findings of the present study, a complete explanation of the causes of black and white episodes must include the effects of spatial and temporal differences in source strengths and removal mechanisms of sulfur and EC. Specifically, dry deposition of sulfur dioxide and a slower removal rate for EC than for sulfates during the first day or so in the atmosphere may counteract to some extent the effect on the sulfate-to-EC ratio of the transformation of SO_2 to sulfates. These opposing processes may thereby increase the time required for black episodes to become white episodes.

5. Conclusion

Excess sulfate (as sulfur) and elemental carbon were significantly correlated in precipitation collected in Sweden, with a typical mass ratio of about 8. No systematic variations of this ratio in space or time were observed. However, several individual samples differed significantly from this ratio, perhaps due to air mass origin and history or to local contaminatin. The lack of systematic variations suggests that, to within the ±50% accuracy of the EC measurements, the rates of removal (% h^{-1}) of EC and sulfate were equal over the study region.

The order-of-magnitude decrease in the sulfur to EC ratio estimated to be from western European sources to Swedish precipitation is probably due in part to a higher relative rate of removal of sulfur in the source regions, and in part due to an underestimate of the EC source strength. EC, on the other hand, may be incorporated more effectively into precipitation in Sweden than was the case for fresh emissions sampled earlier in Seattle. This suggests that some fraction of the EC particles becomes coated with hygroscopic substances (e.g., sulfates) between the source region and Sweden, contributing to the significant correlation between EC and excess sulfate observed in precipitation in Sweden.

The correlation between excess sulfate and EC in precipitation supports the potential for the use of EC as chemically-inert tracer for submicrometer aerosols, although much more needs to be done to understand the relative rates of removal of EC and sulfates. Among the research needs are improved analytical accuracy for EC in rain, simultaneous aerosol and deposition measurements of sulfate and EC (preferably with a greater time resolution than that of the present work), deposition measurements of sulfur and EC in the source regions, measurements of the vertical distribution of EC, and a better characterization of EC emission factors.

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