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ON THE FORMULATION OF ATMOSPHERIC  
DEPOSITION PROCESSES IN NUMERICAL MODELS

by

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1. INTRODUCTION

Recent emphasis on evaluating regional-scale and international transport of pollutants emitted from industrial sources has given rise to an increased focus on numerical models. In the last two years, North American interest has been focused on the deposition of acid compounds to sensitive environments, especially in the northeast. In response to the need for accurate deposition models, new deposition sub-routines have been added to many regional scale transport models. These sub-routines vary widely in complexity and in the manner in which they simulate natural processes. In most instances, the models consider the transport and deposition of some selected pollutant. Most commonly the pollutant selected is sulfur, either as gaseous sulfur dioxide or as particulate sulfate. In-air transformation from sulfur dioxide to sulfate is parameterized in a simple fashion. Likewise, the deposition by dry processes of both gaseous and particulate sulfur is simulated. Although the formulation used to describe the deposition of sulfur dioxide is fairly well tied down, there is great uncertainty regarding the best way to formulate the deposition of sulfate particles.

In the case of wet deposition, the acidity emphasis has caused most attention to be given to the pH of rainfall, snowfall, and other precipitation events. In practice, the acidity of precipitation is governed by the ionic balance between many chemical species, such as nitrates, various trace metals, ammonium, and the sulfate material that most of the models are designed to address. In this respect, it is clear that existing sulfate transformation, transport, and deposition models go only part way towards simulating the

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matters of main relevance to the acid deposition concern. It is the formulation of deposition in the models that will be the subject here. The matter is raised following an examination of the deposition subroutines included in models used in the acid deposition negotiations between parties representing the United States and Canada.

## 2. FORMULATIONS IN EXISTING MODELS

Table 1 lists the formulations used to evaluate wet deposition in a selection of regional scale transport models that are potentially suitable for evaluating the transport of acid deposition precursors across the United States/Canada boundary. In every case, the formulation selected is well-founded in the literature, and is based upon an attractive and physically defensible description of the processes involved. However, as will become obvious there seems some confusion regarding the general applicability of these processes.

Consider a hypothetical situation in which clean rain falls through a polluted layer of the atmosphere below cloud base. If the rain is sufficiently widespread, then the effect will be to deplete the atmospheric concentrations of all materials as the rainfall continues. It is easy to see that the situation is somewhat analogous to that of conventional radioactive decay. If rainfall rate is constant, then the rate of removal of material from the air will be proportional to its airborne concentration. The end result of a continuing period of rain is likely to be that the air is completely cleansed of soluble contaminants; thus as a first order approximation the concentration of material in rain will be inversely proportional to the amount of rain itself.

On the other hand, let us consider the case of a strongly convective system, which "inhales" air from a well-mixed polluted layer beneath it. We might view this as an active dynamic system, in which the cloud processes large volumes of air and cleanses pollutants from them, transferring these pollutants to the rain. The conceptual limit of this process is a situation in which a raining cloud system removes material from new air continually as rain continues. In this case, the concentration of pollutant in rain will be independent of the amount of rainfall.

These situations present limits that are unlikely to be attained in nature, however they serve to bracket the natural case in an informative manner. The first example is meant to represent "washout", in which raindrops cleanse air as they fall through it. The second example is an exaggeration of "rainout", in which air is drawn into clouds and pollutants are concentrated into cloud droplets, which then coalesce to form raindrops, and subsequently fall. In both extremes, the concentration of material in rain is going to be governed by the concentration of the same or some precursor material in air. To make a first order allowance for this factor, it is normal to refer to "scavenging ratios" defined as a concentration in rain divided by the related concentration in air. Clearly, there are some subtleties involved in employing scavenging ratios of this kind. In the case of pure washout, scavenging ratios might be relate the concentration in rain to that in air at the time that the rain is falling through it. Scavenging ratios evaluated in this way will appear to be constant, however in the purest situation the concentration of material in rain will fall off exponentially as the concentration in air is depleted. Pure in-cloud scavenging is far less likely to cause a detectable change of concentration with time of ground level air concentrations. It should not then matter

whether scavenging ratios are referred to air concentrations before, during, or even after a rain event.

It must be emphasized that all these comments refer to hypothetical situations, and of course, nature will differ from them. In practice, scavenging will almost never be entirely rainout or entirely washout, but will be some combination of both. Thus a plot of rain concentrations (or appropriate scavenging ratios) against rainfall amount is unlikely to show the pure inverse relationship required by washout, or the lack of a dependence expected if rainout is the only active mechanism. Instead, some behavior between these extremes will be found. Indeed, a variety of power law behaviors have been documented in the literature. Scott (1978) proposes a negative .6 power for such a relationship, Hicks and Shannon (1979) suggest a negative half power. Both results refer to data obtained primarily in the summer months, in conditions dominated by unstable precipitation systems. The agreement is reassuring, but it seems likely that these results are peculiar to the precipitation regimes and to the times in which the data were obtained. One interpretation of these results is that rainout and washout mechanisms are roughly equally important. Clearly, this is not likely to be the case in situations where strong convective activity (such as characterizes the American midwest or much of the northeast) is quite infrequent. In Great Britain, for example, washout mechanisms seem far more likely to predominate, and consequently a much stronger dependence on rainfall amount might be expected. However, in locations, in which convective activity contributes significantly to the net average rainfall, the American results might not be too unrealistic. In winter, when snowfall is a main contributing factor, we might expect washout to become a dominant factor. As yet there is little data to test this conjecture.

In the light of these comments it is informative to review the parameterizations listed in Table 1. It is seen that some models use relationships and formulations that are appropriate for pure washout. Others assume pure rainout.

### 3. IN-CLOUD OXIDATION

Chemical reactions of air pollutants in clouds make rainout a substantially different process than washout, even though many of the processes involved in scavenging particles will be the same. In particular, it is important to consider the manner in which gaseous material such as sulfur dioxide can be taken up, subjected to chemical transformation, and then delivered to the surface as a different chemical species. Every regional scale deposition model approximates chemical transformation during the transport process by some relatively simple expression such as a rate constant for the transformation of sulfur dioxide to sulfate. These constants result from field and laboratory experiments, conducted in circumstances in which clouds or cloud physical processes are not represented. For evidence concerning in-cloud oxidation of sulfur dioxide, we must turn to sources other than the mainstream of atmospheric chemistry literature.

Scavenging of small particles is especially efficient in convective storm systems, such as those which typify summer rainfall in the midwestern USA. Many studies have shown that scavenging ratios of about 400 should be expected in the case of pollutants that originate near the surface, and values of about 800 are characteristic of material derived from the free troposphere aloft (such as fallout radionuclides). The case of NaCl is particularly interesting,

since this wettable and hygroscopic compound is usually present as particles that are substantially larger than the so-called accumulation size range normally associated with sulfate aerosol. Scavenging of sodium chloride particles is therefore quite efficient; scavenging ratios of about 600 seem typical. If studies of some other quantity indicate greater values, then it would be necessary to postulate some mechanism more effective than that which scavenges large, hygroscopic sea-salt particles.

The highly scattered results on particle scavenging in the United Kingdom reported by Cawse (1974) yield geometric means that are not greatly dissimilar from observations in the Midwest, ranging from about 200 ( $\pm$  about 40%) for lead to 400 ( $\pm$  70%) for selenium. However, the data of Cawse indicate ratios of 1460 ( $\pm$  70%) for sulfate and 1630 ( $\pm$  40%) for nitrate. The suggestion that both quantities are scavenged about five times more efficiently than other particles is hard to rationalize unless some kind of in-cloud  $\text{SO}_2$  conversion is postulated. The following points are especially relevant.

1. The finding of a seemingly enhanced sulfate scavenging efficiency is not unique to the Cawse data set.
2. Scavenging of sea salt is not usually found to be enhanced, thus indicating that the mechanism involved in sulfate scavenging enhancement is additional to all those chemical and physical processes that contribute to the removal of the larger and hence more active NaCl particles.
3. Selenium is not removed with the high scavenging ratio of sulfate, but rather as one of the particulate trace metals. It should be noted that selenium and sulfur compounds are chemically similar although the former are more active.  $\text{SeO}_2$  is not a common atmospheric constituent.

The alternative hypothesis that precipitation contains a large sulfur contribution from scavenged  $\text{SO}_2$  which is transformed by in-cloud mechanisms to  $\text{SO}_4^{=}$  remains to be tested. In order to demonstrate the likelihood of this mechanism, Figure 1 presents annual cycles of total sulfur scavenging ratios,



derived from atmospheric and precipitation chemistry data reported for Poland by Hryniewicz (1979) and for the Canadian midwest by Nriagu and Coker (1978). The data show excellent agreement and indicate summer scavenging ratios in the range 400 - 500, precisely as expected. Thus, summer rainfall appears to remove total sulfur from the atmosphere as if all the sulfur were associated with those accumulation size particles that carry trace metals, and almost as rapidly and efficiently as large sea-salt particles. (Note that the values graphed are evaluated as total sulfur based on  $\text{SO}_2$  concentrations only, neglecting airborne particulate sulfur which in this case appears to have been insignificant.)

In the colder months, scavenging ratios referred to  $\text{SO}_2$  drop to about 100 - 200, probably as a direct consequence of a diminished efficiency of in-cloud capture and/or conversion of  $\text{SO}_2$ . Hales and Dana (1979) report maximum concentrations of unoxidized  $\text{SO}_2$  dissolved in rain in the winter months, in support of the contention that reduced conversion causes the lower winter scavenging ratios of Figure 1. It does not appear that the winter sulfur scavenging ratio is overwhelmingly weighted towards particulate scavenging, however, since the Polish data do not differ greatly from the Canadian even though the former are derived from atmospheric sulfur concentrations that include particles whereas the latter are not.

The nitrate data reported by Cawse are not significantly different from his high sulfate values. It is tempting to attribute this to in-cloud processes similar to those that appear to dominate in the case of sulfur, as has been postulated by Marsh (1978), but until nitrate information similar to the sulfate data of Figure 1 becomes available the possibility of a fortuitous result cannot be rejected.

#### 4. DRY DEPOSITION OF GASES

There is rather remarkable similarity between the magnitudes of the dry deposition velocities for sulfur dioxide incorporated in the models of Table 1. It appears that the days of the constant 1.0 cm/sec deposition velocity are long gone; although this value is quite likely to be appropriate in day-time conditions, at night much lower values prevail. In almost all circumstances the factor that controls the uptake of sulfur dioxide is the status of the underlying vegetation. Fully transpiring, summertime conditions will lead to a rather high deposition velocity. At night, stomates are closed and atmospheric stability further limits the flux of sulfur dioxide, so that deposition velocities of the order of 0.1 cm/s are likely to be appropriate. Clearly, we should accommodate not only a time variation of the sulfur dioxide deposition velocity, but also a spatial dependence following the changes in vegetation from location to location. This is likely to be of some importance when attention is focused on specific sensitive areas. Vegetational differences in uptake characteristics for species like  $\text{SO}_2$  must then be taken into account. A large number of appropriate studies have been conducted; it remains to incorporate the results of these studies in the deposition models.

Similar comments could be made regarding the deposition of other gaseous materials, such as nitrogen oxides. As yet there is little experimental evidence to point us in the right direction.

## 5. PARTICLE DRY DEPOSITION

Even a cursory examination of Table 1 will show the wide range in deposition velocity for sulfate that is currently used in the models. It is of interest to investigate the origins of these numbers. The common value of 0.1 cm/s originated partially as a result of adjustments to air quality models developed for application in Europe. There is a considerable body of evidence, both experimental and theoretical, which supports the conjecture that the deposition velocity of small, sub-micron particles might indeed be exceedingly low. The model tuning result indicating a value of the order of 0.1 cm/s is often interpreted as a verification of expectations based on other grounds. However it appears inappropriate to employ as a basic parameter in one model the results of "tuning" another. At the other extreme, some models employ rather high values for the deposition velocity for sulfate. Experimental evidence supporting these higher values is rather sparse, but nevertheless the value 0.1 cm/s could easily be a considerable underestimate.

Table 2 lists a series of experiments on particle deposition that have recently been conducted. Some of these experiments refer to particles in specific size intervals. Others refer to specific chemicals such as sulfur. It has been hypothesized that sulfate deposition velocities should be expected to be considerably higher than the very low values appropriate for some particles in the so-called "accumulation size range", since there will be a large contribution by much larger particles. This conjecture remains to be verified, but it does provide a potential mechanism by which well-aged aerosol may be deposited more rapidly than accumulation size particles of more local origin.

At this time, there seems little alternative than to employ some least-offensive value for sulfate particle deposition velocity, and to retain the option to modify these values as better experimental data become available.

We should also be concerned with the deposition of alkaline pollutants, with special attention on the flux of calciferous, and therefore potentially neutralizing soil particles. While atmospheric sulfate and nitrate particles are normally in the size range from about 0.1 to 1  $\mu\text{m}$ , soil particles are typically between 5 and 50  $\mu\text{m}$  diameter. They deposit largely through gravitational settling; the deposition experiments identified in Table 2 are not indicative of this particular size range, nor are the formulations of the models described in Table 1.

Because of their greater gravitational settling speed, particles of 10 microns or larger are unlikely to be transported for the long distances addressed by the transport models of Table 1. Rather, they are of sufficiently local origin that sources will probably lie within usual grid spacings. Thus it appears that the deposition of such potentially neutralizing properties as soil particles presents a problem that lies outside the range of applicability of the usual long range transport models.

## 6. CONCLUSIONS

The models listed in Table 1 are essentially air quality simulations, to which improved deposition sub-routines have been added in order to obtain a first cut answer to the long-range-transport/acid-deposition question.

However, the deposition of acidity in any location will be influenced by quantities that are more local in origin than the existing models can accommodate. The critical factors in this regard are likely to be the deposition of neutralizing materials, such as the soil particles discussed earlier. Moreover, there is need to recognize that acidic species in the air and in precipitation can be generated within clouds. As yet there is little hard evidence that would allow us to evaluate this factor, let alone determine its impact upon net rates of acid deposition.

Such problems are well appreciated by the modeling community. In response to the deficiencies, specialized deposition models are presently being developed. Eventually these will be coupled with air quality models of the kind listed in Table 1 so that we may derive a defensible, and physically reasonable, representation of the processes that lead to the deposition of acidic compounds.

TABLE 1

Comparison of deposition formulations used in selected long-term transport models. Unless otherwise mentioned, models address sulfur compounds only.

MODEL	DRY DEPOSITION	WET DEPOSITION
ENAMAP (Bhumralkar et al., 1980)	$V_d$ varies with stability, during daytime, similar for both $SO_2$ and $SO_4$ ; about 0.1 cm/s at night.	Calculated as washout, dependent on rainfall rate, different for S as $SO_2$ or $SO_4$ .
ASTRAP (Shannon, 1981)	$V_d$ varies with time of day, similar for $SO_2$ and $SO_4$ , very small at night.	Calculated as rainout, same scavenging ratios for S as $SO_2$ and $SO_4$ , rainfall rate dependent.
TRANS (Weisman, 1980) (Includes $NO_x$ )	$V_d$ not fixed, "standard" values 0.75 cm/s for $SO_2$ , 0.25 cm/s for $SO_4$ , 0.5 cm/s for $NO_2$ .	Calculated as washout, $SO_2$ scavenging pH and rainfall-rate dependent.
AES-LRT (Olson et al., 1978; Voldner et al., 1981)	$V_d = 0.5$ cm/s for $SO_2$ $V_d = 0.1$ cm/s for $SO_4$ .	Scavenging ratio for $SO_2 = 3 \times 10^4$ ; $SO_4 =$ $0.5 \times 10^5$ .

TABLE 2

Examples of recent experimental studies of  
pollutant deposition to natural surfaces.

- Shepherd, J. G., 1974: Sulfur dioxide profile measurements over grass and water.
- Whelpdale, D. M., and R. W. Shaw, 1974: Sulfur dioxide deposition evaluations by the profile method, over grass, snow, and water.
- Dovland, H., and A. Eliassen, 1976: Measurements of the rate of accumulation of particles on a snow surface.
- Garland, J. A., 1977: Profile measurement of sulfur dioxide over a variety of natural surfaces.
- Wedding, J. B., et al., 1977: Particle deposition to foliage determined by leaf washing.
- Wesely, M. L., et al., 1977: Measurements of fluxes of small particles over grassland, by eddy correlation.
- Barrie, L. A., and J. L. Walmsley, 1978: Sulfur dioxide transfer to snow determined from measurements of sulfur-accumulation.
- Eaton, J. S., and G. E. Likens, 1978: Sulfur budget results obtained at a forested watershed.
- Fowler, D., 1978: Profile determinations of sulfur dioxide fluxes to crops.
- Wesely, M. L., et al., 1978: Fluxes of ozone to maize, measured by eddy correlation.
- Galbally, I. E., et al., 1979: Measurements of sulfur turbulent fluxes to forest and pasture, using eddy correlation.
- Lenschow, D. H., et al., 1980: Aircraft measurements of ozone fluxes by eddy correlation.
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## APPENDIX

The results of a field experiment performed during 1978 provide further support for the efficient removal of  $\text{SO}_2$  in clouds. The study was conducted at the meteorological site at Argonne National Laboratory, some 40 km southwest of Chicago, as part of the Multistate Atmospheric Power Production Pollution Study. Adjacent HASL<sup>1</sup> and PNL<sup>2</sup> wet-only automatic samplers were used to collect rain for sulfate analysis, on an event basis. The aspects of the study that are of relevance here are: (i) the agreement between the two sampling systems; (ii) the relationship between concentrations and rainfall amount; (iii) statistical relationships between sulfur concentrations in rain and in air; and (iv) scavenging ratios for total sulfur. The HASL collecting vessels were cylindrical plastic buckets of about 25 cm diameter. The PNL collector utilized a stainless steel collecting vessel, with an opening about 15 cm in diameter. Both devices used sensing arrays which detected raindrops and removed covers from the collection vessels whenever appropriate.

Precipitation samples were collected as soon as possible after every rainfall event. Concentrations of sulfur (as sulfate) in the rainfall were evaluated independently by wet-chemical methods (Tisue and Kacoyannakis,

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<sup>1</sup>For "Health and Safety Laboratory", now the Environmental Measurements Laboratory of the U.S. Department of Energy, New York; for details of design see Volchok and Graveson (1975).

<sup>2</sup>For Battelle, "Pacific Northwest Laboratories"; for details of design see Hales and Dana (1976).



1979). The analysis procedure could not differentiate between sulfate and sulfite; Figure 7 of Hales and Dana (1979) indicates that as much as 5% of the sulfur detected could have been deposited as sulfite during the summer months of this particular experiment. Repeated testing of the chemical procedures indicated that each separate determination of sulfur concentration was probably accurate to  $\pm 10\%$ .

### Results

Table A1 lists the data obtained. Considerable variation is seen between the PNL and HASL sulfate concentrations, however the averages and standard deviations derived from a common set of observations are quite similar. The geometric mean ratio, PNL/HASL, is 0.97 ( $\pm 8\%$ ). If the two kinds of sampler were equally susceptible to error, the standard deviation associated with each measurement would then be about 30%. This is considerably more than the error (about  $\pm 10\%$ ) that can be associated with the chemical analyses. Most of the error must therefore be associated with the automatic sampling procedures.

Occasions of single, uninterrupted rainfall have been extracted from Table A1 and are listed in Table A2. Also listed are the best estimates of  $\text{SO}_4^{=}$  concentrations in the collected precipitation, obtained as averages of concentrations yielded by the HASL and PNL collectors whenever possible. Rainfall amounts listed were obtained independently by means of a standard recording rain-gauge. Average air concentrations of sulfur dioxide throughout the collecting periods are also tabulated; these data were measured at a nearby U.S. EPA monitoring station, located on the outskirts of Joliet, Illinois, about 8 km from Argonne. These concentrations are likely to be

a source of considerable error, partially because of the distance from the Argonne sampling site, and partially because of uncertainty about the period over which concentrations should be averaged. On the assumption that rainout will dominate washout, air concentrations have been averaged through the period of each rainfall. A different averaging procedure would yield results different in detail, but not by a great amount.

The errors involved in deriving the values of Table A2 are sufficient that a detailed statistical analysis is probably not warranted. However, a regression analysis indicates power law relationships of  $\text{SO}_4^{=}$  concentration in precipitation with rainfall and with airborne  $\text{SO}_2$  concentration characterized by exponents of  $0.39 \pm 0.08$  and  $0.84 \pm 0.17$  respectively. The former value is close to the value of  $-0.5$  suggested by earlier studies mainly of radioactive fallout components (see Hicks and Shannon, 1979). The latter value is not significantly different from the proportionality between sulfur concentrations in air and those in rainfall that forms the basis for most parameterizations of precipitation scavenging. However, it is important to emphasize that the airborne sulfur concentrations that are used here are sulfur dioxide data that are largely insensitive to particulate sulfate. When considered in terms of total sulfur, the geometric mean scavenging ratio determined from the data of Table 2 is 400 ( $\pm 28\%$ ). In reality, a small atmospheric particulate contribution was likely, although it cannot be quantified, and thus a value smaller than 400 appears appropriate for the scavenging ratio relative to total sulfur in air. (In this regard, visibility data indicate that aerosol concentrations were not high during the data collection periods). Thus, the total sulfur scavenging ratio is essentially the same as that for small accumulation

size aerosol particles, and hence, in-cloud processes involving the uptake of sulfur dioxide must be highly efficient.

### Conclusion

Scavenging ratios calculated on the basis of total sulfur in rain and gaseous sulfur concentration in air indicate a mean value of  $400 \pm 28\%$ . This value is necessarily an overestimate (although perhaps not by a large amount) of the value appropriate for the removal of total atmospheric sulfur, since it is clear that particulate sulfur will indeed be efficiently scavenged. This is in close agreement with results derived elsewhere for small particles. Hence it seems likely that raining clouds scavenge  $\text{SO}_2$  almost as if it were present in accumulation-size-range particulate range.

The dependence of the quantity of sulfur deposited on the amount of precipitation follows much the same behavior as has been reported elsewhere with a power law exponent, in this case, of about 0.6 (evaluated as  $1.0 - 0.39 \cong 0.6$ ). Previous studies at Argonne (see Hicks and Shannon, 1979) indicated a range of values of the exponent averaging about 0.5 but perhaps slightly higher in the case of sulfur. It is not thought appropriate to vary the previous recommendation of a half-power law on the basis of the two set of sulfur data now available even though a value of 0.6 appears increasingly attractive.

Finally, good agreement was found between average sulfate concentrations determined by the PNL and HASL samplers used here. Individual determinations indicate a  $\pm 30\%$  standard deviation, considerably more than can be accounted for by errors in the chemical analysis.

Acknowledgments

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Table A1

Concentrations of sulfate in rainfall collected at Argonne by use of adjacent PNL and HASL samplers. Precipitation amounts were measured independently.

Date	Precipitation (mm)	Sulfate Concentration ( $\mu$ Molar)	
		PNL	HASL
March 20	2.5	87	-
25-26	16.3	58	-
April 2	13.7	3*	49
4	1.8	147	129
5-6	27.9	54.5	44
10	15.0	49	33
17-18	24.9	46.5	29
18-19	3.3	-	80
20-21	4.1	109	106.5
23	4.1	87	106
24	6.3	104	45
May 4	12.4	30	59
7-8	14.0	31.5	57
9	0.8	-	29
12-14	54.9	35	23
23	2.5	108	168
June 7	4.6	128	96
7	4.1	53	53
16	8.1	52	74
17-18	31.2	40	46
20	21.8	62	65
25-26	11.4	84	70
30	18.8	44	41
July 9	3.0	58	115
12-13	2.8	160	112
19-20	52.6	46	-
20-21	44.2	96	-
21-23	9.4	88	-
26	14.2	60	56
29-2	3.0	85	146

\* Apparently erroneous and omitted from analyses.

Table A1 (continued)

August	2	3.0	44	54
	9	5.8	42	52
	11	16.5	44	46
	15-16	11.9	20	23
	24	6.4	44	-
Sept.	12-13	5.8	66	-
	14-18	31.5	36	-
	20-21	11.2	43	-

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Table A2

Data selected from Table A1 for occasions of single, uninterrupted rainfall periods, with supporting precipitation and SO<sub>2</sub> information, obtained independently. Whenever possible, sulfate concentrations in rain are obtained as the average between the two collectors.

Date and Time (hrs.) (1978)	Sulfur Concentration in Rain ( $\mu$ Molar)	Rainfall (mm)	SO <sub>2</sub> Concentration in Air (ppb)
March 20, 18-22	87	2.5	3.8
April 4, 06-08	138	1.8	11.0
5-6, 18-14	49	27.9	1.4
10, 00-21	41	15.0	1.5
17-18, 23-06	38	24.9	0.8
18, 16-23	80	3.3	4.4
20-21, 15-02	108	4.1	7.4
23, 02-09	97	4.1	9.9
24, 13-15	75	6.3	11.0
May 4, 13-22	45	12.4	11.2
9, 09-10	29	0.8	7.5
23, 02-08	138	2.5	-
June 7, 06-07	112	4.6	20.0
16, 06-10	63	8.1	7.4
20, 13-16	63	21.8	1.2
30, 05-10	43	18.8	5.5
July 9, 02-05	87	3.0	0.6
26, 11-15	58	14.2	33.0
August 2, 15-16	49	3.0	-
9, 01-04	47	5.8	-
11, 00-05	45	16.5	7.0
15-16, 22-02	21	11.9	0.4
24, 05-07	44	6.4	12.3

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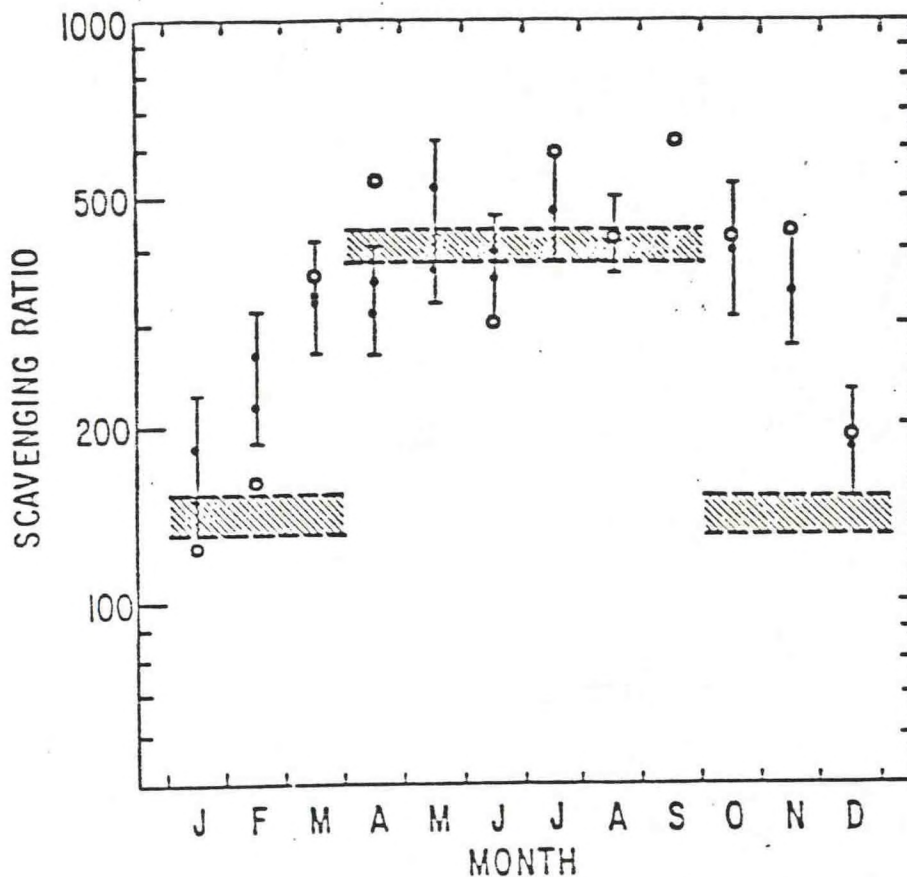


FIG. 1.--Solid circles and error bars: geometric mean scavenging ratios derived directly from data on sulfur concentrations in rain and sulfur dioxide concentrations in air at Canadian stations in the vicinity of the Great Lakes (Nriagu and Coker, 1978). Open circles: scavenging ratios derived from average concentrations of total sulfur in precipitation and in air, as reported for Poland by Hryniewicz (1979) for the year 1976. Shaded bands: long-term average sulfur scavenging ratios evaluated for Poland, over warm and cold six-month periods (see Hryniewicz, 1979).

