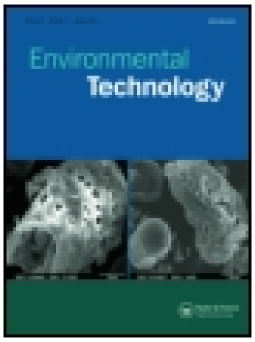

Environmental Technology

ISSN: 0143-2060 (Print) (Online) Journal homepage: https://www.tandfonline.com/loi/tent19

Sulfate aerosol formation rate in an oil fired power plant plume

V.P. Aneja , J.H. Overton Jr. , J.A. Anderson & J.L. Durham

To cite this article: V.P. Aneja , J.H. Overton Jr. , J.A. Anderson & J.L. Durham (1987) Sulfate aerosol formation rate in an oil fired power plant plume, Environmental Technology, 8:1-12, 167-180, DOI: 10.1080/09593338709384476

To link to this article: https://doi.org/10.1080/09593338709384476

Environmental Technology Letters, Vol. 8, pp. 167-180
© Science & Technology Letters, 1987

SULFATE AEROSOL FORMATION RATE IN AN OIL FIRED POWER PLANT PLUME

V.P. Aneja* and J.H. Overton, Jr.
Environmental Sciences, Northrop Services, Inc., Research Triangle Park, NC 27709, U.S.A.

J.A. Anderson
Meteorology Research, Inc., Altadena, CA 91001, U.S.A.

J.L. Durham
Atmospheric Sciences Research Laboratory, U.S. Environmental Protection Agency,
Research Triangle Park, NC 27711, U.S.A.

(Received 9 October 1986; accepted 24 February 1987)

ABSTRACT

In February 1977, a study was conducted to obtain the atmospheric rate of sulfate formation in an oil-fired power plant plume. The power plant studied is located 70 km northwest of Tampa, Florida, and during the study period it was burning fuel oil that contained about 2 percent sulfur and significant concentrations (~ 250 ppm) of vanadium. Aerial sampling was performed to obtain the sulfur dioxide and sulfate concentrations in the plume. The wind flow field was also measured. Analysis of the data indicates no conversion (0.00014 hr^{-1} ± 0.00053) of SO_2 to sulfate in that region of the plume where data was collected (plume age > 20 minutes). However plume fall-out of particulate sulfate before sampling began is not ruled out.

* Present Address: General Electric Company
Corporate Research and Development
Schenectady, NY 12301.

INTRODUCTION

The emission of sulfur into the atmosphere, in connection with various industrial processes and energy production for industry as well as for household consumption, is a major environmental problem. One difficulty is that sulfur dioxide is oxidized to sulfuric acid in the atmosphere. This strong mineral acid may seriously impair the environment and human health (1,2,3) and welfare (4,5,6).

A field study was undertaken to better characterize stationary pollutant sources and to promote a better understanding of the physico-chemical processes occuring in oil-fired power plant plumes; since power plant plumes have proven to be a convenient method, by many investigators (7,8,9) for field studies of particulate sulfate formation. In this study the wind field and the aerosol's physical and chemical properties were measured. The sampling techniques were refined to sample a cross-section of the plume, and measurements were made in approximately the same air mass as it moved downwind. The source chosen was a power plant that uses a mixture of Venezuelan and Middle East fuel oil containing ~250 ppm of vanadium (10), which has been proposed as a catalyst for the oxidation.

Arin et al. (11) first sampled an oil-fired plume for SO_2 conversion. The samples were collected in a "Saran" bag and transferred (over a period of ~15 min) through the West-Gaeke impingers. No appreciable conversion was observed.

The rate of oxidation of sulfur dioxide to sulfate in oil-fired power plant plumes was studied by Newman et al. (12) by sampling with a single-engine aircraft. A mechanism was postulated whereby the sulfur dioxide is in equilibrium with water on particulates and is subsequently catalytically oxidized to sulfate. Aerosols containing vanadium, originating from the oil, were proposed to serve as a catalyst. A pseudo-second order mechanism depending on sulfur dioxide and particulate concentrations is reported. They also concluded that the reaction is independent of relative humidity. However, Freiberg (13) indicates that such is not the case and that humidity indeed plays a significant role in the rate of atmospheric formation of sulfate in the work done by Newman et al. (12).

Air samples collected isokinetically from a DC3 aircraft have led Flyger et al. (14) to the conclusion of a 0.5 hr half life for SO_2 from the Stigsnaes oil-fired power plant on the coast of New Zealand. Experiments carried out by Forrest et al. (15) at the same power plant and during the same time period as the present study indicates, that within the plume where the data was collected, no measurable conversion rate of SO_2 to sulfate.

The discrepancy on the measurable conversion rate in these plume studies casts doubt on the universality of the results on conversion in oil-fired plumes, which are summarised in Table 1. Hence additional research is needed to further elucidate plume chemistry. Also, the role of vanadium as a catalyst in the plume is unclear. To date, only one laboratory study of the vanadium catalyzed oxidation of dissolved SO_2 has been reported. Bracewell and Gall (16) investigated the catalytic oxidation rate of dissolved SO_2 by eight metal ions and found the reactivity decreased in the following order: Fe(II), Fe(III), Mn(II), Co(II) Ni(II), Cu(II), Zn(II), Cr(III), and V(V). The extent of reaction was determined by measuring the conductivity of the solution as a function of time during the reaction. The Assumption was made that the formation rate of H_2SO_4 was equivalent to the elementary chemical oxidation rate of dissolved SO_2 to H_2SO_4. The mass transfer of O_2 from the bubbles to solution was completely ignored. It is likely for fast-reacting systems (such as Fe and Mn) that the conversion rate was O_2 mass transport limited. Although their results should not be accepted as precise, the differences in reactivity between metal ions is a useful indicator. The catalytic activity of V(V) was less than 0.5% of that for Fe and Mn ions.

Table 1

STUDIES OF SO_2 CONVERSION IN OIL-FIRED POWER PLANT PLUMES

Performing Organization	Date of Final Report	Approach	SO_2 Measurement	SO_2 Conversion
Arin *et al.* GSA	1969	SO_2/SF_6	Sign-X West-Gaeke	No Conversion Obesrved
Newman *et al.* BNL	1975	SO_2/SF_6 SO_2/SO_4^{2-} S^{32}/S^{34}	Collection	Pseudo Second order
Forrest *et al.* BNL	1979	SO_2/SO_4^{2-}	Collection	< 0.25 %/hour
Flyger *et al.*	1977	SO_2/SF_6	Collection	75 %/hour
Present Study EPA		SO_4^{2-}/SO_2	Theta	~0 %/hour

EXPERIMENTAL

The experiments were conducted at a power plant on the coast of Florida, located about 70 km northwest of Tampa (Figure 1). The exhaust gases of the power plant exit through a 153 m tall tapered stack having an exit diameter of 5.2 m. The maximum capacity of the plant is 525 MW. This plant uses no pollution control devices. One CEA forced-circulation, drum-type boiler, designed to deliver about 3.5 million pounds of super-heated steam per hour, generates steam for the plant. The fuel generally utilized contains about 2% S, significant quantities of vanadium (~250 ppm), and other trace elements (17).

The main objective of this study was to determine the rate of formation of sulfate in a plume containing vanadium. To accomplish this objective, sampling was performed under both day and night time regimes and under clean and dirty air conditions. The plume pollutant concentration profiles were measured with a fixed-wing single-engine Cessna 206, equipped with instruments for measuring airborne pollutants, meteorological parameters, and location. the specific measurements and instruments used are presented in Table 2. More extensive descriptions have been presented elsewhere (18).

The plume sampling methods used both horizontal traverses at various plume altitudes and vertical spirals through the plume. A combination of several traverses and spirals allows a detailed mapping of the plume's pollutant concentrations at a fixed distance downwind of

TABLE 2

AIRCRAFT INSTRUMENTS

POLLUTANT PARAMETER	INSTRUMENT	TIME RESPONSE (90% for usual range)
Light scattering coefficient	MRI Integrating Nephelometer	1 sec
Aerosol number condensation	Environment One Condensation Nuclei Monitor (Rich 100)	5 sec
Ozone	REM 612 Ozone Monitor	5 sec
NO/NO_x	Monitor Labs 8440 NO/NO_x	5 sec
SO_2	Theta Sensor LS-400 SO_2 monitor	5 sec
SO_4^{2-} Collection	MRI TWO MASS Aerosol Sampler	
SO_4^{2-} Analysis	Ion Chromatography	
Meterology and position	MRI Airborne Instrument Package:	
	Temperature	5 sec
	Humidity	30 sec
	Turbulence	3 s(to 60%)
	Altitude	1 sec
	Indicated Airspeed	1 sec
Dewpoint	Cambridge Systems 137 Dewpoint Hygrometer	0.5 sec/°C
Data acquisition system	Metrodata M/8 V OR Analog Converter	1 sec
Data acquisition system	Metrodata 620 Data Logger (20 channels)	48 channels/sec
Data acquisition system	Linear Instruments 485 Strip Chart Recorder (2 channels)	

the source. Repeated plume mappings at several downwind distances then gives a characterization of the three-dimensional variation of pollutants in the plume. A typical flight pattern is shown in Figure 2.

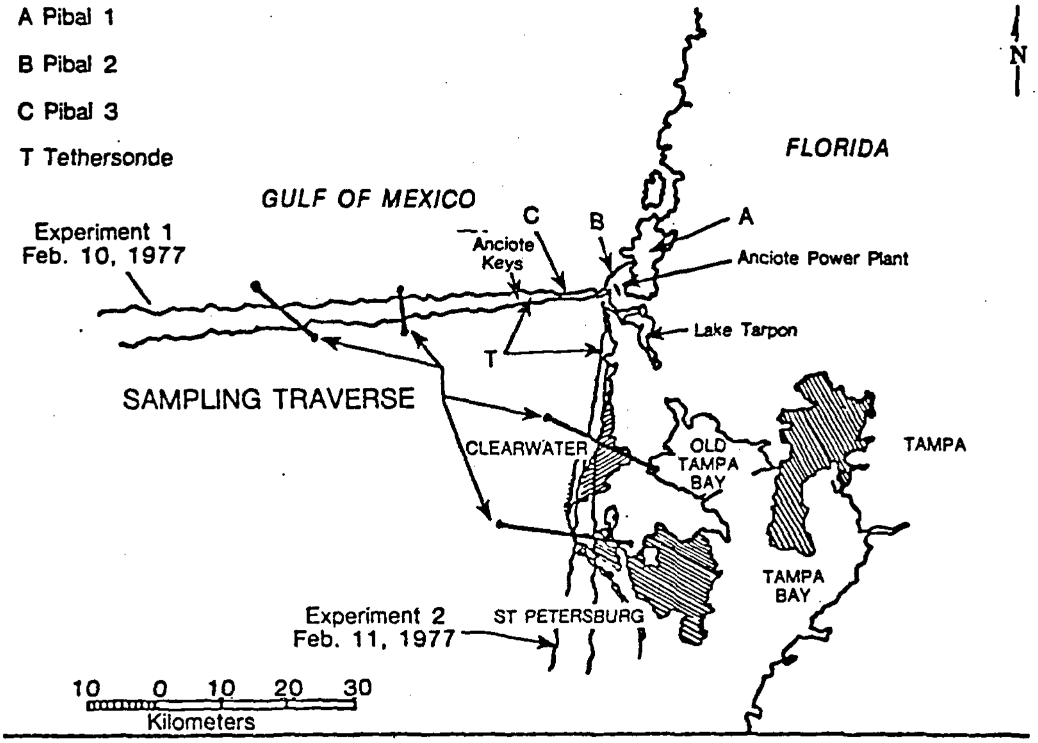

Figure 1. Sampling traverse paths.

A typical experiment lasted about four hours and consisted of making a background spiral from 920 m down to 30 m to obtain pollutant concentrations upwind of the plant. Next, the plume was sampled at two to four locations downwind. The sampling was done in approximately the same air mass as it moved downwind. Calibration of the gaseous monitors (SO_2, NO, NO_x, O_3) were performed immediately following an experiment, using calibration gases and a portable calibration system.

Wind speed and directions were obtained by three pilot balloon measurement crews located in the vicinity of the power plant. These measurements were made every half hour, starting two hours before the experiment, and lasting one hour after the experiment. A research marine vessel made similar measurements with a tethersonde in the event that the plume should blow over water. During the run at least one temperature sounding was taken and the relative humidity recorded. An analysis of the meteorological conditions and a description of the experiment days have been presented elsewhere (19).

RESULTS

A total of nine experiments were conducted; each experiment consisted of traverses and spirals as previously described. Of the nine

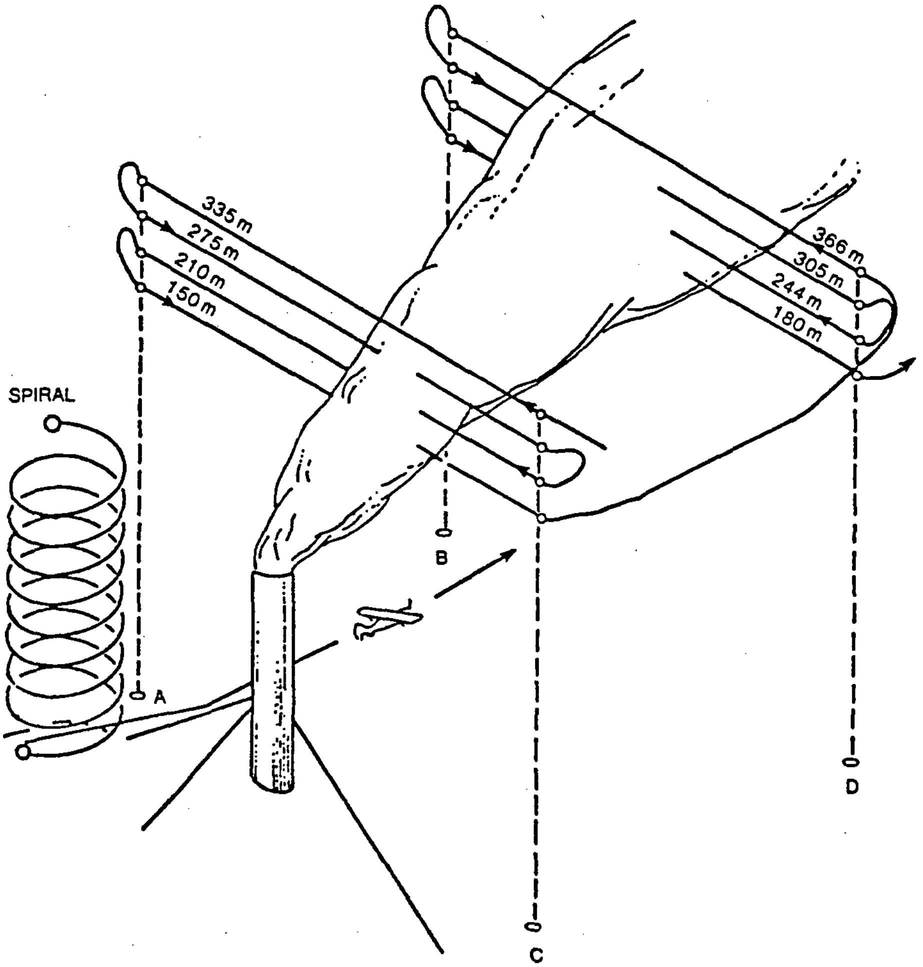

Figure 2. Traverse flight pattern for plume sampling. Points A, B, C, and D are ground reference points laid out before flight.

experiments, 16 observations (each corresponding to a fixed distance from the source) from seven experiments were of sufficient quality to be of use in an analysis. The two experiments that were omitted resulted from poor wind trajectories condition, and/or damage to the collected samples. Table 3 presents the data used in the analysis.

Table 3

DATA USED IN ANALYSIS

Experiment Identity	Day	Time Span of Experiment	u (Average Wind Speed m/s)	Average Wind Direction (degrees)	Observation Number	x* (Distance From Source-km)	$\overline{[SO_2]}$* $\mu g/m^3$)	$\overline{[SO_4^{2-}]}$* $\mu g/m^3$	$\overline{x[SO_2]/u}$* $(\mu g\text{-hour})/m^3$
202	8	1323-1655	6.0	70	1	40.4	432	12.7	830
					2	69.3	197	15.0	631
					3	76.8	112	11.5	398
204	10 and 11	2330-0243	13.0	80	4	34.2	411	1.4	304
					5	50.2	202	1.8	217
205	11	1614-1928	6.0	4	6	17.0	168	4.1	132
					7	22.8	272	4.4	290
					8	42.7	184	5.4	359
206	12	0713-1435	9.0	161	9	5.8	1940	13.0	308
					10	42.1	162	.7	210
					11	130.0	86	1.3	7228
208	15	1427-1709	2.5	320	12	0.64	976	12.6	60
					13	9.0	244	4.3	250
210	17	1414-1755	5.8	343	14	0.24	807	13.0	11
					15	7.2	798	4.9	275
211	18	0718-1047	7.0	217	16	26.7	136	4.66	144

*Defined in the Appendix.

Based on a first-order chemical reaction model (See Appendix) there is no evidence of the conversion of SO_2 to sulfate for plume ages greater than about 20 minutes. (Most of the data collected was from the part of the plume with an age greater than five minutes). The results of the analysis suggests that background sulfate plus that sulfate produced in the stack or in the first five minutes of plume travel explains the data.

These conclusions are based on the results of regression techniques applied to the following equation (which is derived in the Appendix, Eq 8):

$$\overline{[SO_4^{2-}]} = [B]_i + a\overline{[SO_2]} + c\{\overline{x \bullet [SO_2]/u}\}$$

where a = it is related to the initial ratio, A, of SO_2 to total sulfur, $\{A=1/(1+a)\}$
c = is related to the first order rate constant, $k=c \bullet A$
i = the experiment identity index (first colum, Table 3)
$[B]_i$ = background sulfate
The overbar indicates the average over the path that the sulfate was

collected. x is the distance from the source, along the wind diection, to the point of collection and u is the average wind speed, k and A are assumed constant and independent of experiment; $[B]_i$ is assumed dependent on the experiment (essentially on when the experiment was performed). A more detailed explanation of the variables and the equation can be found in the appendix. The values of the independent and dependent variables are given in Table 3.

Two regressions were performed. the first was applied to the equation as given with the following results:

$$\text{Multiple } R^2 = 0.96$$
$$a = 0.0071 \pm 0.0019^*$$
$$c = 0.00014 \pm 0.00053^*$$

i	$[B]_i$	σ_B^*
202	11.20	1.67
204	-0.62	2.00
205	3.10	1.61
206	-0.52	2.83
208	4.12	2.24
210	3.24	2.44
211	3.67	2.68

* Standard error of the estimate.

Because $A = 1/(1+a) \approx 1$, and the parameter c is not statistically significant, then the rate constant, $k=c \cdot A$, is not statistically significant. Since its units are hr^{-1}, its value is not physically or chemically significant either.

A second regression was performed with the parameter $c=0$ (i.e., the dependent variable $\overline{x \cdot [SO_2]/u}$ omitted), experiments 204 and 206 combined with $[B]_{204,206} = 0$, and experiments 210 and 211 combined with $[B]_{210,211} = 0$. The results for this model:

$$\overline{[SO_4^{2-}]} = [B]_i + a\,\overline{[SO_2]}$$

$$\text{Multiple } R^2 = 0.96$$
$$a = 0.0068 \pm 0.00105^*$$

i	$[B]_i$	σ_B^*
202	11.4	1.3
204 206	0	-
205	3.2	1.3
208	4.3	1.6
210 211	3.6	1.4

* Standard error of the estimate ($p<0.02$ for all parameters).

All these parameters are considered statistically significant.

The first regression establishes that the first order rate constant is essentially zero (Figure 3); while the second indicates that the data can be explained in terms of sulfate background and the sulfate produced in the stack which, based on the parameter a, is about 0.5%.

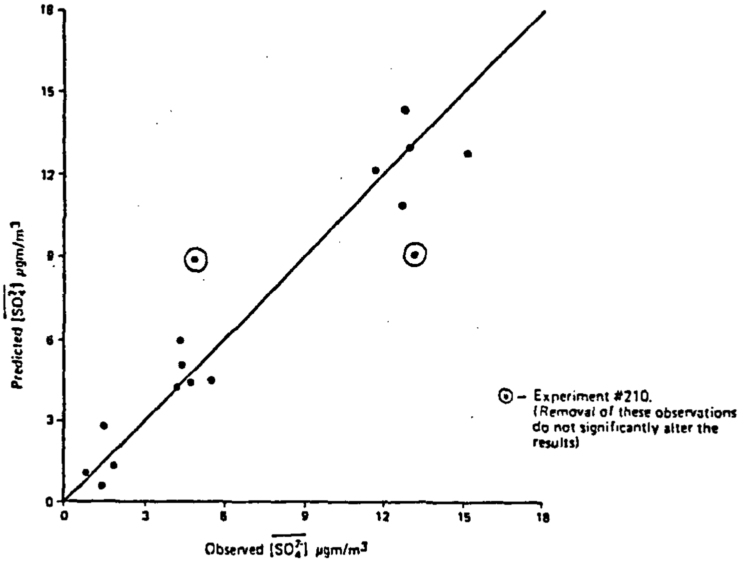

Figure 3. Comparison of estimated and experimental sulfate values for SO_2 to SO_4^{2-} reaction rate ≅ 0.

DISCUSSION

Oil-fired power plant plumes have been studied in some detail. However it is too early to draw any firm universal conclusions of sulfate formation in these plumes. Previous studies have reported relatively high conversion rates in an oil-fired power plant (12). The present study was undertaken to see if indeed vanadium would enhance the conversion rate of SO_2 to SO_4^{2-}; since vanadium in fuel oil had been thought to be an important contributor in primary sulfate formation (20).

Our analysis of the data for sixteen experiments indicates that the oxidation rate of SO_2 for plume age greater than five to fourty minutes is negligible (0.00014 hr^{-1} ±0.00053. The plume age for 4 out of the 16 experiments was less than 40 minutes). However three other important factors in formation of sulfate in power plant plume, in the present study, need addressing. Firstly plume drop out prior to sampling, which was not considered in the present study of oxidation rate of SO_2, could possibly give rise to measurements indicating reduced atmospheric oxidation than actually occurs. This may be a significant process that should be considered in future analysis. Moreover sulfate fallout anywhere in the plume may be important; since we can expect lower SO_4^{2-} calculated values above the SO_2 plume axis and higher values below the SO_2 plume axis than would occur with no fall-out. If measurements were made in the upper portion of the plume a smaller value of k, the reaction rate constant, would be estimated than one determined from data taken in the lower portion of the SO_2 plume. It is therefore necessary to examine the extent of plume sulfate dropout. Some of the earlier studies (15) tend to suggest that indeed particulate sulfate was dropping out of the plume.

Secondly, while our analysis took background SO_2 and SO_4^{2-} concentrations into account, and unless substantial plume fall-out has occurred, the result of the analysis suggests that most of the conversion has taken place in the stack or shortly after emission (within first five minutes of plume travel) from the stack; and no further oxidation took place for plume travel as long as ~40 minutes.

Finally, it is well documented from studies on large scale that a great deal of the emitted SO_2 gets converted to sulfate eventually. Indeed Wilson <u>et al.</u> (21) observed enhanced sulfate formation rate in a plume after an initial delayed reaction. However unfavorable meteorological conditions prevented us from tracking the plume further than ~40 minutes of travel time to investigate the possibility of a sharp increase. We therefore speculate that probably our tracking time was still within the initial delayed reaction time for sulfate formation in the plume. This is consistent with predictions by Schwartz and Newman (8) and Freiberg (22).

The conversion of sulfur dioxide to sulfate in the aerosol phase in the ambient atmosphere is important, amongst other things, for health, economic, and aesthetic reasons. Complex physico-chemical processes complicate the mechanisms for conversion. There still continue to be large discrepancies between the results from different investigations dealing with SO_2 conversions even in power plant plumes which have proven to be convenient system for field studies.

APPENDIX

Physico-Chemical Model

Strong inversion conditions with average wind speeds of 5-10 m/s gave very cohesive plume configurations during this period. The vertical thickness of the plume at the sampling points was often about 50 meters. Often the plume exhibited looping characteristics. These conditions made it impossible to know the location of a horizontal traverse with respect to the plume's axis. These problems were overcome by the development of the following mathematical model which permitted the analysis of the averaged sulfate aerosol data.

Assumptions:

1. The only important sulfur compounds in the plume are SO_2 and SO_4^{2-}.
2. The ratio of SO_2 to total sulfur emitted by the power plant is constant during the sampling run.
3. The reaction orders of dissolved SO_2 oxidation in transition metal ion solutions usually range from 0-2 with respect to the S(IV) concentration. Due to the lack of knowledge of the vanadium catalyzed reaction, we have no insight to the dissolved SO_2 reaction order that should be expected in the plume. For the initial analysis, we assume that the rate is first order with respect to SO_2 concentration, and

that the rate is equal to a constant times the SO_2 concentration.

4. Sedimentation of aerosols in the plume is not important.
5. The boundary conditions for the two sulfur species are of the same form.

For unsteady state transport with chemical reactions the instantaneous equation of continuity for SO_2, SO_4^{2-}, and total sulfur, S, are:

$$(\partial/\partial t + \underline{v} \cdot \nabla)\,[i] = D\,\nabla^2\,[i] + k_i\,[i] \qquad (1)$$

where t = time
∇ = gradient operator
∇^2 = Laplacian
$\underline{v}$ = instantaneous wind velocity $(\nabla \cdot \underline{v} = 0)$
i = sulfur species: SO_2, SO_4^{2-}, or total sulfur, S
$[\ ]$ = instantaneous molar concentration
D = molecular diffusion coefficient $(D_{SO_2} = D_{SO_4^{2-}} = D_S)$
k_i = 1st order rate constant of the ith species: $k_{SO_4^{2-}} = -k_{SO_2} = k;\ k_S = 0$
$[S] = [SO_2] + [SO_4^{2-}]$ = instantaneous total sulfur

A Cartesian coordinate system is chosen as follows: Divide the wind velocity into two components, $\underline{u}$ and $\underline{v}_1$, such that $\underline{u}$ is a constant vector and $\underline{v} = \underline{u} + \underline{v}_1$. The x-axis is taken to be in the $\underline{u}$ direction and the z-axis is the vertical axis. The origin, x=y=z=0, is located at the virtual point source of the plume.

A function g is defined such that

$$[S] = e^{kx/u}\, g(\underline{r},t) \qquad (2)$$

where $\underline{r}$ = spatial coordinate vector
$u = |\underline{u}|$.

The following is obtained if Eq (2) is inserted into Eq (1) with i = S:

$$\frac{\partial g}{\partial t} + u(1 + v_{1x}/u - 2Dk/u^2)\frac{\partial g}{\partial x} + \underline{v}_1 \cdot \nabla_1 g = D\,\nabla^2 g + k[Dk/u^2 - v_{1x}/u - 1]g \qquad (3)$$

where v_{1x} = x-component of $\underline{v}_1$

$$\nabla_1 = \left(\frac{\partial}{\partial y}, \frac{\partial}{\partial z}\right)$$

The assumption is made that $\underline{u}$ can be chosen (e.g. average wind velocity vector) or condition exist such that $|v_{1x}/u| \ll 1$ and $|Dk/u^2 \ll 1$. If these assumptions can be met then g satisfies (approximately the same equation that SO_2 satisfies. Requiring that

$$[SO_2] \propto g \qquad (4)$$

and using Eqs (2) and (4), then

$$[SO_2] = Ae^{-kx/u}\,[S] \qquad (5)$$

A = $[SO_2]/[S]$ evaluated at the virtual source (x=y=z=0). Since $[S] \equiv [SO_2] + [SO_4^{2-}]$ we can relate $[SO_4^{2-}]$ to $[SO_2]$:

$$[SO_4^{2-}] = A^{-1}(e^{kx/u} - A)\,[SO_2] + [B] \qquad (6)$$

[B] has been added to the equation to account for a constant background of sulfate.

Assuming the exponent in Eq (6) is small the term on the right hand side is expanded in a power series and only the first degree term retained:

$$[SO_4^{2-}] \approx [B] + A^{-1}(1 - A + kx/u)\,[SO_2]$$

or

$$[SO_4^{2-}] \approx [B] + a[SO_2] + c(\tfrac{x}{u}\,[SO_2]) \qquad (7)$$

where a = (1 - A)/A, and c = k/A

SO_2 data was taken pointwise along traverses or spirals; however, the sulfate data was not. It was accumulated on filters along the same path. Thus, to use Eq (7) to estimate a,B, and c (or A,B, and k) the variables $[SO_4^{2-}]$, $[SO_2]$ and $x \bullet [SO_2]$ were averaged over the same path as that in which SO_4^{2-} was collected. Therefore for each set of traverses or observations we have an equation of the form,

$$\overline{[SO_4^{2-}]}_{ij} = [B]_i + a\overline{[SO_2]}_{ij} + c\{\overline{(x \bullet [SO_2])}/u\}_{ij} \qquad (8)$$

The bar indicates the average, i the experiment, and j indicates a set of traverses or particular observations. Thus, the background is assumed to change from experiment to experiment; whereas the parameters a and c are assumed to characterize all of the experiments and are, therefore, taken to be independent of the observations and experiments.

REFERENCES

1. Amdur, M.O., T.R. Lewis, M.P. Fitzland and K.I.Campbell. Publication No. AP-111, U.S. Environmental Protection Agency, Research Triangle Park, NC (1972).

2. Newill, V.A. Pollutions price - the cost in human health. Science and Public Affairs, pp47-49 (1973).

3. Rall, D.P. A review of health effects of sulfur oxides. Office of the Management and Budget, Washington, D.C., U.S.A. (1973).

4. Bolin, B. Air pollution across national boundaries, the impact on the environment of sulfur in air and precipitation. Report of the Swedish Preparatory Committee for the U.N. Conference on Human Environment, P.A. Norsted & Soner, Stockholm (1971).

5. Hill, A.C., JAPCA <u>21</u>, pp. 341-346 (1971).

6. Likens, G.E., Chemical and Engineering News, 54(48), pp 29-44 (1976).

7. Husar, R.B., D.E. Patterson, J.D. Husar, N.V. Gillani, and W.E. Wilson, Jr., Atmospheric Environment 12, pp 549-568 (1978).

8. Schwartz, S.E. and L. Newman., Envir. Sci. Technol. 12, pp 67-73 (1978).

9. Lusis, M.A. and C.R. Phillips, Atmospheric Environment, 11, pp 239-241 (1977).

10. Nader, J.S., J.B. Homolya, K.T. Knapp and J. Wagman, Presentation before the Division of Environmental Chemistry, American Chemical Society Meeting, New Orleans, Louisiana, March 20-26 (1977).

11. Arin, M.L., C.E. Billings, R. Dennis, J. Driscoll, D. Lull, F.A. Records, P. Warneck and J.E. Wilder, Study of reactions of sulfur in stack plumes. GCA-TR-69-12-G, Contract No. PH-86-67-125. Second Annual Report (1969).

12. Newman, L., J. Forrest and B. Manowitz, Atmospheric Environment, 9, pp 959-968 (1975).

13. Freiberg, J., Atmos. Environ., 10, pp 672-673 (1976).

14. Flyger, H., E. Lewin, E. Lund Thomsen, J. Fenger, E. Lycke and S.E. Gryning. Physical and chemical processes of sulfur dioxide in the plume from an oil-fired power station. Risoe Rep. pp 328-384 (1977).

15. Forrest, J., R. Garber, and L. Newman, Atmospheric Environment, 13, pp 1287-1297 (1979).

16. Bracewell, J.M. and D. Gall, Proc. of the Symp. on the Physico-chemical Transformation of sulfur compounds in the atmosphere and the formation of acid smogs. (Organization for Economic Co-operation and Development, Directorate for Scientific Affairs.) pp 17-26 (1967).

17. Bennett, R.L. and K.T. knapp, Proceedings of the 4th National Conference on Energy and Environment. October 4-7, Cincinnati, Ohio (1976).

18. White, W.M., J.A. Anderson, W.R. Knuth, D.L. Blumenthal, J.C. Hsiung, and R.B. Husar, Environmental Protection Agency, ORD, Report No. EPA-600/3-76-110 (1976).

19. Hull, L., W. Dennevick and R. Woodford, Environmental Protection Agency, ORD, Report No. EPA-600/7-78-117, p 71 (1978).

20. Dietz, R.N., R.F. Wieser and L. Newman. Presented at a Workshop on Measurement Technology and characterization of Primary Sulfur Oxides Emission from Combustion Sources, Southern Pines, North Carolina, Brookhaven National Laboratory Report No. 24366 (1978).

21. Wilson, W.E., R.J. Charlson, R.B. Husar, K.T. Whitby, and D. Blumenthal. EPA report No. 600/7-77-021, Research Triangle Park, NC (1977).

22. Freiberg, J., Nature, Lond. 274, pp 42-44 (1978).