CONF-790571--

BNL-26369

MODELING SULFUR OXIDE CONCENTRATIONS: AN ASSESSMENT OF ENERGY UTILIZATION SCENARIOS

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INTRODUCTION

The long-range transport of sulfur dioxide (SO_2) and sulfate (SO_4) in the atmosphere[1-7] has received increasing attention in conjunction with possible health impacts[8-13], visibility degradation[1,14], and acidification of precipitation[1,3,15,16]. In particular, time and distance scales for the residence of sulfates in the atmosphere suggest that airborne sulfate levels, occurring in the eastern U. S. and elsewhere, may be the result of emissions from locations hundreds or thousands of kilometers upwind.

Sulfate levels in large portions of the eastern U. S. (and elsewhere) have the potential of being strongly affected by emission control regulations as well as by siting and fuel use decisions which will take effect within the next several decades. If ambient standards for sulfate or a surrogate such as fine particulates are promulgated, as some expect will occur in the not too distant future, the relationship of relevant distance scales of atmospheric transport of sulfate to the dimensions of states and air quality control regions must be taken into account. It is the purpose of this paper to document a range of air quality projections which are a consequence of the Department of Energy fuel utilization and emission control scenarios for the year 1990.

Levels of sulfur dioxide and sulfate in the contiguous U. S. have been calculated with the BNL AIRSOX long-range transport model[17] using emission inventories consisting of either major fuel-burning sources or major coal-burning sources, as described in more detail below and elsewhere.

The extended spatial scale over which impacts of sulfur oxide emissions are significant is illustrated via an emitter-receptor matrix. This matrix presents the contribution of exposure to SO_4 in each of 10 receptor EPA Federal regions due to SO_2 emissions from each Federal region.

MODEL METHODOLOGY[17]

For each SO_2 source in the emission inventory, along-wind trajectories are calculated at intervals of every six hours for one month. The trajectories are calculated using July 1974 observed upper air winds, averaged in the vertical through the mixing layer. Horizontal diffusion along the trajectory is parameterized as Gaussian about the center of mass. Vertical diffusion is calculated using a finite difference representation for eddy diffusivity or "K" theory. A bottom boundary deposition velocity condition for the vertical diffusion is used to account

(1) Superior numbers refer to similarly-numbered references at the end of this paper.

DISCLAIMER

for dry deposition. A constant linear conversion rate of SO_2 to SO_4 is assumed. In addition, representative amounts of SO_2 are assumed to be converted to primary SO_4 in stack. A linear rainout model is used to remove SO_2 and SO_4 from the vertical column whenever a plume segment encounters rainfall. Model parameters have been chosen on the basis of literature values, sensitivity runs, and a comparison of calculated SO_2 and SO_4 concentrations with observations from the NASN and EPRI networks. Table 1 provides the values of model parameters used in the present calculations.

Table 1. Model Parameters

	Parameter	Value
1.	Stack or release height	
	utility	200 meters
	industrial	100 meters
2.	Amount of SO_4 in effluent	2% of emitted SO_2 by mole
3.	Meteorology and precipitation data	July 1974
4.	Mixing layer height	1000 meters
5.	Atmospheric stability	neutral
6.	Vertical levels	12
7.	Conversion rate of SO_2 to SO_4	0.57%/hour
8.	Dry deposition velocity	
	for SO_2	3.4 cm/sec
	for SO_4	0.23 cm/sec
9.	Wet removal rate	
	for SO_2	0.216 P
	for SO_4	0.07 P
		P = rainfall (mm/hour)
10.	Grid resolution, approximate	32 x 32 km

EMISSION INVENTORY

Sulfur oxide air quality projections have been made for 1985 and 1990[18] as part of the two DOE assessment activities, the National Coal Utilization Assessment (NCUA)[19] and the Regional Issue Identification and Assessment (RIIA)[20].

The NCUA had as its goal the quantification of impacts resulting from the coal component of the National Energy Plan (NEP). The NEP specifies energy use, conservation, and supply goals in the U. S. Important components of NEP are increased conservation measures and an increased reliance on coal. A modified NEP has become the National Energy Act (NEA) of 1978. A comparison of NEP and NEA indicates that the environmental impacts are similar. Some of the difficulties of achieving the NEP level of coal use are documented in the NCUA report.

Three scenarios were defined, corresponding to three levels of utility emission controls. The majority of the NCUA analysis was performed upon the mid-range scenario, scenario III, in which the SO_2 emissions were consistent with the projections of the SEAS model. The scenario with higher SO_2 emissions, I, and a scenario with lower SO_2 emissions, II, were also defined as in Table 2. It is evident from the data in Table 2 that relative to 1975 there is projected to be

a large decrease in the amount of SO_2 emitted per BTU for even the worst case scenario, scenario I.

The RIIA program addressed the environmental impacts from a "business as usual" scenario of energy development. The RIIA scenario is based upon the continuation of policies prior to the implementation of the NEA and assumes median estimates of energy supply and demand and constant world oil prices. Major differences between the RIIA and NCUA emissions inventories are the inclusion of oil and gas sources in RIIA and the use of different, more stringent, emission control assumptions for RIIA. In Table 2 the RIIA scenario, as well as the coal-only portions of the RIIA scenario, are compared with the NCUA scenarios.

Table 2. NCUA and RIIA Sulfur Dioxide Emission Inventories for the Contiguous U. S.

Scenario	Utility		Industrial		Total	
	SO_2[1]	BTU[2]	SO_2[1]	BTU[2]	SO_2[1]	BTU[2]
1975						
NCUA	18,623	8,485	3,810	1,980	22,433	10,465
RIIA	20,143	14,639	5,789	10,884	25,931	25,524
1990						
NCUA I	23,094	17,300	7,897	8,370	30,991	25,670
NCUA II	13.938	17,300	7,897	8,370	21,835	25,670
NCUA III	15,900	17,300	7,897	8,370	23,797	25,670
RIIA (coal)	12,424	18,669	2,697	5,664	15,121	24,333
RIIA (total)	14,011	21,498	6,198	20,114	20,209	41,612

[1] SO_2 emissions in 10^3 tons/year
[2] Thermal equivalent in 10^{12} BTU/year

The comparison of the RIIA scenario with the variety of NCUA scenarios shows that uncertainties in projected emission levels are substantially more dependent on the uncertainties surrounding emission controls than on other aspects of implementation (or lack thereof) of NEP (or NEA). In order to put the range of emission projections in prospective, it is noted that even in the worst case, NCUA Scenario I, flue gas desulfurization (FGD) systems are installed in all utility plants built after 1985 and in all plants inventoried by the EPA FGD system status report as of 1978. In the lowest emission case, RIIA, utility plants that were operating prior to 1976 are assumed to meet SIP's. Plants built between 1976 and 1982 are assumed to meet SIP's or the New Source Performance Standards, whichever is more stringent. Plants built after 1982 are assumed to have FGD systems which achieve 85% daily average SO_2 removal. Although the magnitude of uncertainty in the emission controls for the industrial sector is comparable to or larger than in the electric utility sector, only one level of emission controls for the industrial sector has been projected for each of the assessments.

The SO_2 emission inventory considered for RIIA is a fairly complete list of point source emissions due to the combustion of fossil fuel in the U.S. Major categories of emission sources not included in the RIIA or NCUA inventories include process plants such as smelters and oil refineries and area sources, typified by residential heating. Some areas of the U.S. may be impacted by foreign emission sources not included in the RIIA or NCUA inventories.

RESULTS AND DISCUSSION

The BNL AIRSOX model has been used to calculate SO_2 and SO_4 concentrations on a nominal 32 x 32 km (1/10 National Meteorological Center) grid for each of the approximately 1600 point sources in the NCUA and RIIA emission inventories. State and regional population-weighted SO_2 and SO_4 concentrations have been calculated from the individual gridded concentration fields for either an entire emission inventory or specific subsets of an emission inventory. All concentrations have been calculated as monthly averages using meteorological data from July 1974. While there are indeed seasonal trends in emission patterns, meteorological conditions, and observed SO_2 and SO_4 concentrations, the use of July 1974 data is not considered to significantly bias the comparison of the different scenarios presented here.

The 1975 RIIA calculation is based upon the average annual emissions from 1264 fuel-burning utility and industrial point sources. The calculated population-weighted SO_2 and SO_4 concentrations are presented in Table 3. Observed, population-weighted SO_2 and SO_4 concentrations for the month of July 1974 are presented for comparison in Table 4. In the western regions, in which SO_2 emissions are dominated by process sources not included in the emission inventory, the calculated SO_x values are seen to be significantly lower than the observed values. The omission from the RIIA inventory of area sources in densely populated regions probably accounts for much of the difference between the calculated and observed SO_x values averaged over the entire U.S.

Table 3. NCUA and RIIA SO_2 Emissions and SO_x Concentrations

Region	1975 RIIA			1990 RIIA			1990 NCUA I		
	SO_2[1]	$\langle SO_2 \rangle$[2]	$\langle SO_4 \rangle$	SO_2	$\langle SO_2 \rangle$	$\langle SO_4 \rangle$	SO_2	$\langle SO_2 \rangle$	$\langle SO_4 \rangle$
1	322	5.8	6.1	438	4.8	3.9	562	8.8	7.3
2	946	20.1	9.6	629	11.3	5.4	1889	28.6	11.4
3	4015	24.9	14.9	2269	13.0	8.0	4071	25.0	15.6
4	7001	11.5	8.4	6273	10.1	6.6	8499	13.1	9.6
5	10516	28.2	13.8	5094	14.6	7.5	10279	26.6	14.5
6	458	1.0	.6	2221	4.2	1.5	2531	5.5	1.9
7	1900	11.1	5.3	1708	8.1	4.3	2146	10.9	6.5
8	347	1.0	.6	905	2.4	1.4	405	1.9	1.1
9	300	2.3	.6	319	1.9	.5	443	1.9	.5
10	129	.7	.3	354	1.9	.7	166	.8	.4
U.S.	25931	14.5	8.0	20209	9.1	5.0	30991	16.2	9.0

[1] Emissions in 10^3 tons/year.

[2] $\langle\ \rangle$ = population weighted concentration (microgram/cubic meter).

Some critical parameters in the AIRSOX model are chosen from nationwide sulfur budget-type considerations and therefore represent national rather than regional or local effects. In addition, some of the meteorological data which are input to AIRSOX are of too coarse a resolution to resolve potentially important local effects. The implication of these and other such considerations is that the accuracy of long-range transport calculations may be degraded when projections are made for particular regional or local areas.

For 1990, the range of air quality predictions is spanned by calculations using the RIIA and the NCUA Scenario I emission inventories. A comparison of the 1990 population-weighted SO_x concentrations and the corresponding 1975 values is shown in Table 3. Also shown is a comparison of the regional sulfur oxide emissions.

Table 4. July 1974 Observations[1]

Region	$\langle SO_2 \rangle$[2]	$\langle SO_4 \rangle$
1	16.7	11.5
2	28.4	13.2
3	25.1	16.6
4	5.8	12.2
5	28.8	15.6
6	6.4	7.1
7	15.0	8.3
8	2.8	3.3
9	6.2	8.1
10	17.6	4.0
U.S.	17.1	11.8

[1] NASN and EPRI measurements.

[2] $\langle\ \rangle$ = population weighted concentration (microgram/cubic meter).

In RIIA, most of the 1990 regional SO_2 emissions and population-weighted SO_x concentrations decrease relative to 1975, with a notable exception being Region 6 (Southwest) which is now heavily reliant upon gas and oil and which is projected to undergo substantial conversions to coal. Imposition of increased levels of SO_2 controls is particularly effective in the Ohio River Valley and upper Appalachian areas (Regions 3 and 5). The overall improvement in SO_x levels projected for RIIA is particularly impressive in view of the increased fuel use (see Table 1) and the increased reliance on coal in 1990 relative to 1975. A factor of 2.1 decrease in SO_2 emissions per BTU fuel consumed is projected for 1990, relative to 1975.

The SO_x concentrations projected for the 1990 NCUA worst case, Scenario I, are considerably higher than for 1975 or 1990 RIIA, not withstanding the omission of oil and gas fired plants in the NCUA emission inventory. As in RIIA, a major increase in emissions appears in Region 6 (Southwest). In the NCUA study, emissions either increased or remained the same, relative to 1975, in the heavily industrialized Regions 2 to 5. The SO_x air quality impacts occurring in the NCUA, II and III Scenarios, are somewhat higher but qualitatively similar to those projected for RIIA.

The comparison here between a RIIA and an NCUA scenario is not intended to provide an assessment of the merits of the NEP or NEA but instead provides evidence for the deterioration in SO_2 and SO_4 levels which would result if strict emission controls, such as are assumed for RIIA, are not implemented. In order to effect the emission decreases which are incorporated in RIIA 1990, and which are responsible for the SO_x air quality improvement in much of the East,

FGD systems or their equivalent must be fitted to most new and many existing plants. Increased regional SO_x air quality impacts in parts of the West seem unavoidable given projected demographic shifts and given the projected extent of conversion from oil and gas to coal.

It is apparent from Table 3 that regional SO_4 concentrations (and to a lesser extent, regional SO_2 concentrations) are not linearly related to regional emissions. The spatial scales appropriate for the description of the formation of sulfate from emitted sulfur dioxide and its subsequent advection prior to either wet or dry deposition are comparable to the dimensions of EPA Federal Regions. The impacts of inter-regional transport of sulfate, as calculated for the 1975 RIIA emission inventory, are shown in the emitter-receptor matrix in Table 5. Due to the connection with linear health damage functions, exposure in units of (millions of people) x (micrograms/meter cubed) has been used here as a measure of SO_4 impacts. Reading down the columns of the emitter-receptor matrix, we obtain the calculated SO_4 exposure in a given receptor region, due to SO_2 emissions in each emitter region. Reading across the matrix, we obtain the regional distribution of impacts (exposure) from a given emitter region. Values less than .5 are omitted from Table 5.

TABLE 5. EMITTER-RECEPTOR MATRIX FOR SO_4 EXPOSURE (RIIA 1975)

EMITTER REGION	RECEPTOR REGION									
	1	2	3	4	5	6	7	8	9	10
1	7	1								
2	6	64	11	2	1					
3	14	60	151	19	29					
4	8	25	57	204	77	4	5			
5	37	87	131	60	465	1	24			
6				2	3	6	2	1		
7	2	7	6	10	45	2	29			
8					1			2		
9								1	15	
10										2
SUM	74	244	358	296	622	13	61	4	15	2

SEE TEXT FOR EXPLANATION OF TABLE.
EXPOSURE = (MILLION PERSONS) X (MICROGRAM/CUBIC METER)

Regions 3-5 have high SO_2 emissions (see Table 3) and are located upwind of major population centers. These regions "export" exposure to each other and are calculated (1975 RIIA) to account for the majority of the SO_4 exposure in New England and the Mid-Atlantic Regions (1 and 2).

The RIIA 1990 calculations show that relative to 1975 the population-weighted SO_4 concentrations for the U. S. decrease by a greater factor than emissions. The major portion of the calculated improvement in SO_x air quality in RIIA 1990 is due to the large emission decreases in Regions 3-5. Separate calculations indicate that a reduction of emissions in Regions 3-5 is particularly significant since

the SO_4 impacts, per unit emission, from many source locations in these regions are among the highest in the U. S., due to their upwind location relative to major population centers. Conversely, the deterioration of SO_x air quality predicted in conjunction with the NCUA I Scenario is a result of the lack of improvement, relative to 1975, of emissions in Regions 3-5, coupled with the increases in other regions. In 1990, the NCUA Scenario III (the mid-range scenario upon which most of the analysis of NEP was performed) yields an exposure to SO_4 per unit emission that is 17% higher than in RIIA. The comparable figure for NCUA I is also 17% higher than in RIIA. This figure represents the extent to which the NCUA emission sites (as compared with the RIIA emission sites) are located unfavorably with respect to long-range transport of sulfur oxides.

ACKNOWLEDGEMENTS

The submitted manuscript has been authored under contract EY-76-C-02-0016 with the U. S. Department of Energy. Accordingly, the U. S. Government retains a non-exclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so, for U. S. Government purposes.

We acknowledge the support and direction provided by our sponsors at the U. S. Department of Energy, especially Arnold Goldberg of the Technology Assessment Division, Program Manager for NCUA; Arthur Katz of the Regional Assessments Division, Program Manager for RIIA; and Richard Ball of the Environmental Impacts Division, under whose programs portions of the transfer matrix and other methodologies were developed.

We are grateful for the contributions and advice provided by our colleagues at Brookhaven National Laboratory: Ta-Yung Li, Stephen Schwartz, Sam Morris, Leonard Hamilton, Peter Meier, and Paul Michael.

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