

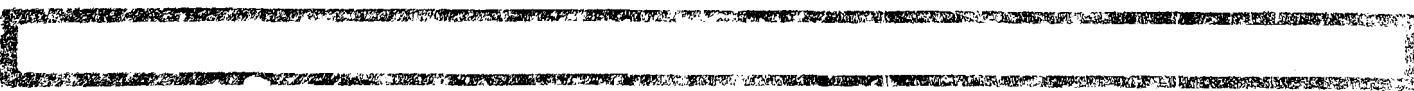
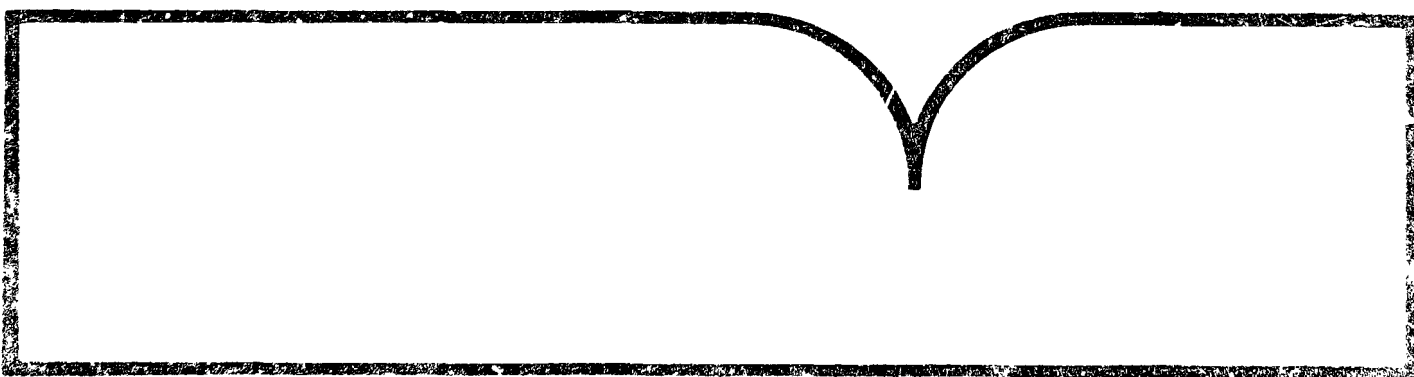
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Radian Corp., Morrisville, NC

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PROCEEDINGS:  
FIRST ANNUAL ACID DEPOSITION  
EMISSIONS INVENTORY SYMPOSIUM

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16 ABSTRACT The proceedings document a 2-day symposium on the progress in implementing the National Acid Precipitation Assessment Program. (NAPAP) Task Group B's emission inventory programs. The meeting was intended primarily for government, academic, and private sector individuals involved in either developing or using atmospheric emission inventories for acid deposition and air quality research. Topics included the development of emission factors for a wide range of pollutant emissions and the use of detailed emission inventories for atmospheric transport, transformation, and deposition modeling. The meeting: 1) provided detailed presentation and information transfer of the NAPAP emission inventory program; 2) illustrated and strengthened the relationship between the emission inventory data base and its users; and 3) compared and contrasted the NAPAP emission inventory with other on-going emission inventory development programs.			
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## ABSTRACT

A two-day symposium discussing the progress in the implementation of the National Acid Precipitation Assessment Program Task Group B emission inventory programs was sponsored by the Environmental Protection Agency's Air and Energy Engineering Research Laboratory, Research Triangle Park, North Carolina, in cooperation with the EPA Office of Air Quality Planning and Standards and the U.S. Department of Energy. The meeting was intended primarily for government, academic, and private sector individuals involved in either the development or use of atmospheric emission inventories for acid deposition and air quality research. Topics included the development of emission factors for a wide range of pollutant emissions and the use of detailed emission inventories for atmospheric transport, transformation, and deposition modeling.

The meeting accomplished the following objectives: 1) to provide detailed presentation and information transfer of the NAPAP emission inventory program; 2) to illustrate and strengthen the relationship between the emission inventory data base and its user; and 3) to compare and contrast the NAPAP emission inventory with other on-going emission inventory development programs.

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## SESSION 1: MAPAP EMISSION INVENTORY ACTIVITIES

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CURRENT NAPAP EMISSION INVENTORY ACTIVITIES

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Presented at:

First Annual Acid Deposition Emission Inventory Symposium  
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## CURRENT NAPAP EMISSION INVENTORY ACTIVITIES

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### ABSTRACT

The most exacting requirements, in terms of disaggregation and specification of emission inventory data within NAPAP, are needed to support the development and testing of the Eulerian acid deposition model. The Initial NAPAP Task Group B emission inventory research program began with a modification and augmentation of the 1980 NEDS inventory to provide outputs suitable for Lagrangian modeling activities and to support preliminary development of the Eulerian model. Current activities are directed at assessing available data for formulation of non-criteria pollutant emission factors, development of the preliminary 1980 NAPAP emission inventory for Eulerian model use, and development of estimation methodologies to assess uncertainties of available emission factors and the 1980 NAPAP emission inventory.

## CURRENT NAPAP EMISSION INVENTORY ACTIVITIES

### INTRODUCTION

In 1980, Congress established the National Acid Precipitation Assessment Program (NAPAP) to coordinate and expand research relevant to the problems posed by acid deposition in the United States. The program is organized and managed through the Interagency Task Force on Acid Precipitation (ITFAP) and 10 subordinate task groups coordinating specific technical areas of research. One of the groups is Task Group B which is responsible for man-made sources. A major objective of Task Group B is the development and maintenance of detailed 1950 and 1984 emission inventories to support acid deposition research and analysis. The most significant uses of the inventories are for policy analysis and to support both Eulerian and Lagrangian long-range transport/deposition models.

### DISCUSSION

The most exacting requirements, in terms of disaggregation and speciation of emission inventory data within NAPAP, are needed to support the development and testing of the Eulerian acid deposition model. Therefore, the needs of other inventory users, with the exception of historical analyses, can be satisfied with an inventory that can meet the requirements of the planned modeling activities. Very detailed emissions information is needed for development, testing, and application of atmospheric models. Detailed emissions data can require a substantial lead time for development. Therefore, the timing of emission inventory needs of atmospheric model development is a key driving force in the emission research strategy. Task Group B planning activities address emission inventory needs of the NAPAP research program, allocation of responsibility for planned activities to meet these needs, and

procedures for quality assurance, maintenance, and distribution of emission inventory information. Group B has been conducting a research program to address the following assessment of priorities in needs:

1. The primary focus of laboratory activities is directed to fulfill the emission data base requirements for the development of an Eulerian acid deposition model. Within the EPA's Office of Research and Development, the Environmental Sciences Research Laboratory has been assigned the lead responsibility to develop an Eulerian model for acid rain which will be based on a framework similar to the Northeast Regional Oxidant Study (NEROS) Eulerian oxidant model. The National Center for Atmospheric Research (NCAR) has been assigned the task of developing the model framework which includes a number of chemical transformation modules. The variety of these modules under development dictates the specific chemical species required in the emission inventory to drive the model input. Development of the Eulerian acid deposition model began in FY-83 with preliminary testing to begin in FY-85.
2. Lagrangian model development and testing parallels work on the Eulerian acid deposition model. Inventory requirements to support Lagrangian models are focused on sulfur dioxide ( $\text{SO}_2$ ), primary sulfate, and the oxides of nitrogen ( $\text{NO}_x$ ). Models being supported by inventory development include ASTRAP (developed by Argonne National Laboratory) and ENAMAP (utilized by the EPA's Environmental Sciences Research Laboratory).
3. Historical emission profiles are needed to assess long-term effects of acidic deposition on materials, and aquatic and terrestrial receptors.

The emission inventory development program within Task Group B began in FY-82 at a resource level of \$85,000. Funded activities included:

1. Development of an historical emission inventory data base for  $\text{SO}_2$  and  $\text{NO}_x$ .
2. Preliminary development of primary sulfate emission factors.
3. Assessment planning.
4. Preparation of the Initial NAPAP acid deposition emission inventory implementation plan.

The Initial NAPAP emission inventory implementation plan outlined a strategy focused on the development of a NAPAP emission inventory for the 1980 base year to be initiated in FY-83. The 1980 NEDS (National Emissions Data System) Inventory was modified and augmented to provide

outputs suitable for Lagrangian modeling activities and to support preliminary development of the Eulerian model.

During FY-83 and FY-84, the following program activities were performed:

1. Assessment of available data for the formulation of non-criteria pollutant emission factors. Detailed assessments were prepared for primary sulfate, ammonia, hydrochloric acid, and hydrofluoric acid.
2. Source measurement tests to complete emission factor development for primary sulfate, ammonia, hydrochloric acid, and hydrofluoric acid. Tests were conducted to fill data gaps identified in the assessments.
3. Development of the 1980 NAPAP emission inventory. The 1980 NAPAP inventory was developed in a phased approach. The initial development was an interim inventory produced to support Lagrangian modeling studies. The data base included an annual inventory of  $\text{SO}_2$ ,  $\text{NO}_x$ , and total volatile organic compounds (VOCs) for the contiguous U.S. The NEDS utility point source inventory was supplanted by the E. Pechar Associates utility emissions inventory prepared for EPA/DOE. Area source emissions were aggregated at the county-centroid level. The second phase in the development and refinement of the 1980 NAPAP emission inventory has produced a preliminary inventory for use in Eulerian model development and testing. The inventory includes point and area sources for the contiguous U.S. gridded to  $1/6$  latitude by  $1/4$  longitude areas for  $\text{SO}_2$ ,  $\text{SO}_4$ ,  $\text{NH}_3$ ,  $\text{NO}_x$ ,  $\text{NO}_2$ , and 10 VOC photochemical reactivity classifications. A temporal allocation profile is provided for a typical summer day.
4. Development of estimation methodologies to assess uncertainties of available emission factors and the 1980 NAPAP emission inventory. As the development, applications, and refinement of the NAPAP emission inventories progress, quality assurance and validation of the data bases are important activities which serve to both improve and define the value of each inventory element.

Figure 1 illustrates the interrelationship of the various tasks performed to produce the preliminary 1980 NAPAP emission inventory for Eulerian model development and testing. A major element of the emission inventory structure is the data handling systems supporting the required temporal, spatial, and species resolution for the NAPAP Eulerian acid deposition model. Since the temporal and spatial resolution requirements of the acid deposition model are similar to those of the Northeast Corridor Regional Modeling Project (NECRMP), the data handling system

used in NECRMP, termed the Regional Model Data Handling System (RMDHS), was adapted for use with the preliminary 1980 inventory. RMDHS was modified to fulfill the following interim emission inventory requirements:

- spatial resolution to 48 states (20 km x 20 km grids);
- NECRMP VOC classes, using national species factors;
- NECRMP NO<sub>x</sub> allocation factors for NO and NO<sub>2</sub>;
- SO<sub>2</sub>;
- primary sulfate;
- ammonia;
- temporal resolution to a single typical summer weekday; and
- four separate time zones resolved to Greenwich Mean Time.

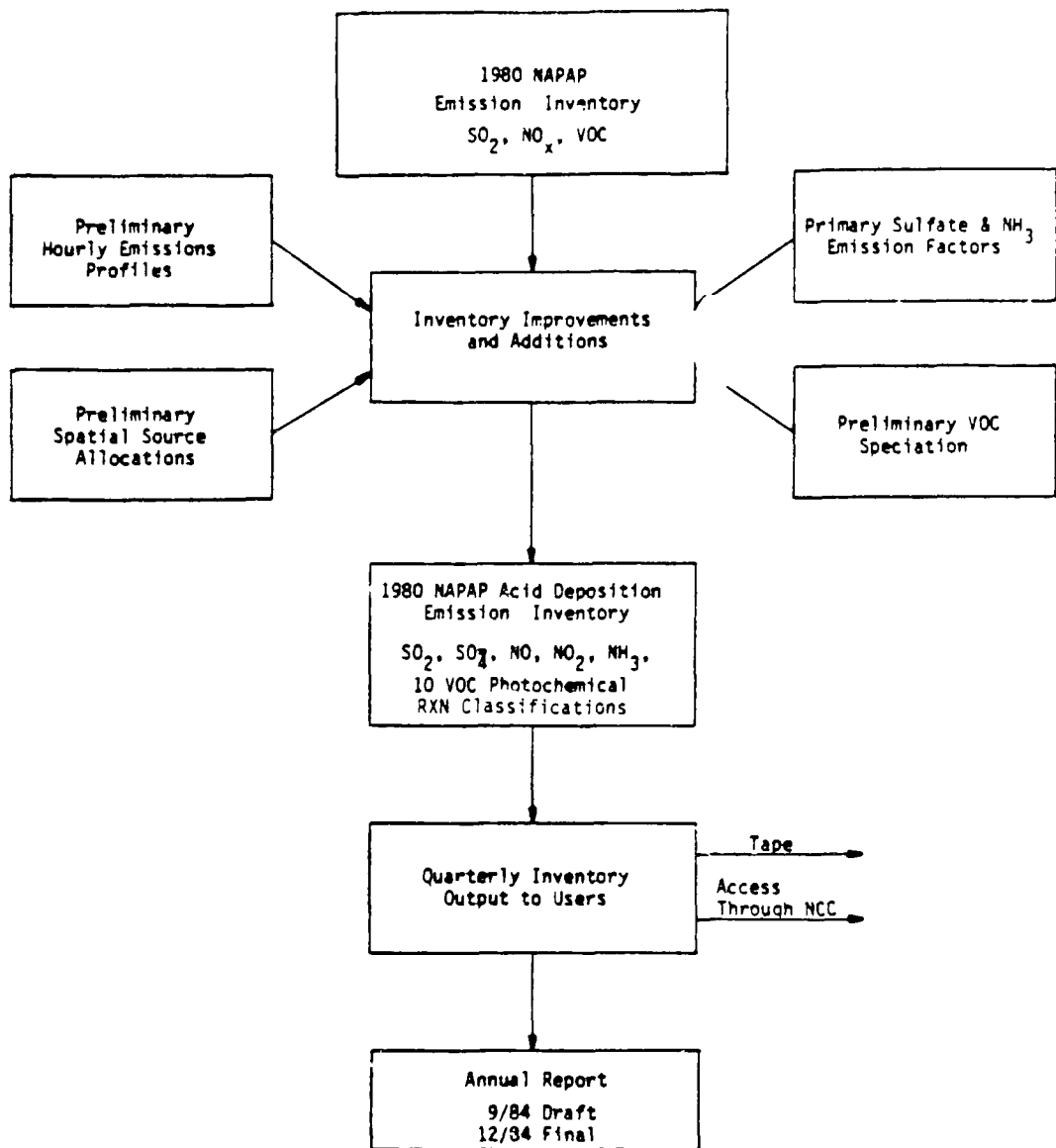


Figure 1. Development of the Preliminary 1980 NAPAP Emission Inventory for Eulerian Model Use.

OVERVIEW OF 1980 NAPAP  
EMISSION INVENTORY

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First Annual Acid Deposition Emission Inventory Symposium  
Raleigh, North Carolina  
December 3-4, 1984



OVERVIEW OF 1980 NAPAP  
EMISSION INVENTORY

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ABSTRACT

This paper addresses the compilation of a 1980 emission inventory for use in the National Acid Precipitation Assessment Program (NAPAP). The current inventory (Version 4.0) contains point source data for 50,200 plants with over 201,000 emission points and area source data for the 3,069 counties in the 48 contiguous states and District of Columbia. Emissions of  $\text{SO}_2$ ,  $\text{NO}_x$ , VOC, sulfates, ammonia, CO, and particulates are included in the inventory, but this paper focuses on  $\text{SO}_2$ ,  $\text{NO}_x$ , and VOC which are of primary interest for acid deposition research. NAPAP Version 4.0 emissions of  $\text{SO}_2$ ,  $\text{NO}_x$ , and VOC are 27.1, 23.7, and 23.3 million tons per year, respectively. Emissions in the NAPAP data base are in reasonable agreement with Work Group 3B and Office of Air Quality Planning and Standards (OAQPS) emission trends estimates. NAPAP fuel use data show reasonable agreement with fuel values in DOE's State Energy Data Report. Version 4.0 of NAPAP represents the best detailed inventory of emissions on a national scale that has been developed to date. Nevertheless, additional improvements are planned, focusing on major point sources. The bulk of future NAPAP resources will be used to meet the needs of Eulerian modeling activities.

## OVERVIEW OF 1980 NAPAP EMISSION INVENTORY

### INTRODUCTION

In 1980, Congress established NAPAP to coordinate and expand research relevant to the problems posed by acid deposition in and around the United States. The program is organized and managed through the Interagency Task Force on Acid Precipitation (ITFAP) and 10 subordinate task groups coordinating specific technical areas of research. One of the groups is Task Group B which is responsible for man-made sources. A major objective of Group B is the development and maintenance of detailed emission inventories to support acid deposition research and analysis.

The objectives of the effort summarized herein were to develop the initial 1980 base year emission inventory, evaluate its quality and comprehensiveness, and identify actions needed for further refinement of the data base in the future. The project required development of a central, quality-assured data base of emissions of pollutants of interest for acid deposition research and modeling. The area covered includes the 48 contiguous states of the U.S. and the District of Columbia.

### DISCUSSION

Version 4.0 of the NAPAP data base was developed starting with 1980 "snapshot" information from the U.S. Environmental Protection Agency's (EPA's) National Emissions Data System (NEDS). The initial data base was improved by incorporating the latest available emission factors, substitution of data from the Northeast Corridor Regional Modeling Project and other NEDS data more representative of 1980, updating electric utility data with the U.S. Department of Energy (DOE) data compiled by E.H. Pechan and Associates, cross-checking data with information from the U.S./Canada Work Group 3B report, and adding county centroid latitude and longitude for sources with missing or incorrect Universal Transverse

Mercator (UTM) coordinates. The NAPAP data are stored in Emission Inventory System (EIS) format on the EPA's IBM computer at Research Triangle Park, North Carolina.

NAPAP data show that SO<sub>2</sub> emissions are dominated by electric utilities, primarily from coal-fired generating stations located in the eastern U.S. For NO<sub>x</sub>, the largest sources are transportation (mostly highway vehicles), electric utilities, and industrial combustion. For VOC, emissions result largely from transportation (again primarily highway vehicles), other industrial processes, and miscellaneous sources which include organic solvent use not accounted for by point sources, retail gasoline service stations, and forest wildfires.

The geographic breakdown of SO<sub>2</sub> emissions in NAPAP indicates that EPA Regions 4 and 5 are the largest contributors. The eastern 31 states account for over 82 percent of nationwide SO<sub>2</sub> emissions. For NO<sub>x</sub>, Regions 4, 5, and 6 are the highest emitters, with the eastern 31 states accounting for about 64 percent of national emissions. Regions 4, 5, and 6 are also responsible for the greatest amount of VOC emissions. The eastern 31 states account for 66 percent of the nation's VOC emissions.

The relative importance of point versus area source emissions varies for each of the three pollutants. Point sources contribute over 90 percent of national SO<sub>2</sub> emissions. For NO<sub>x</sub>, emissions are nearly evenly distributed. Area sources, on the other hand, emit almost 80 percent of total VOC emissions. Ohio, Pennsylvania, and Indiana have the greatest SO<sub>2</sub> emissions. Texas, California, and Ohio are the greatest NO<sub>x</sub> emitters, while Texas and California have the greatest VOC emissions.

Seasonal variations were derived from operating data in the point source inventory and seasonal factors added to the area source file. Seasonal variations are less than expected. For SO<sub>2</sub>, the maximum variation is 3 percentage points, from 24 to 27 percent. The maximum variation for NO<sub>x</sub> and VOC is only 2 percentage points, from 24 to 26 percent. Emissions of SO<sub>2</sub> and NO<sub>x</sub> are greatest in winter and lowest in spring. VOC emissions are the highest in summer and lowest in winter.

Although it is effective plume height that is of greatest interest to modelers, only stack height data are included in NAPAP. Thus, only emissions by stack height range could be summarized. These data show that nearly all VOC emissions (both point and area) are emitted below

120 feet. Over two-thirds of the total emissions of  $\text{NO}_x$  also are released below 120 feet. On the other hand, nearly 40 percent of all  $\text{SO}_2$  is emitted at heights above 480 feet.

County emission densities were calculated and density ranges summarized to obtain more information on the concentration of emissions. For  $\text{SO}_2$ , counties in the highest emission density range result from power plants located in the East, primarily in the Ohio Valley and Great Lakes areas. These counties represent only 4 percent of those in the nation but have 54 percent of the total emissions. For  $\text{NO}_x$ , the counties in the highest range result from either power plants or highway vehicles. These counties represent only 3 percent of those in the nation but contribute 27 percent of the total emissions. For VOC, the counties in the highest range result from solvent use and highway vehicles. These counties represent only 2 percent of all counties in the nation but contribute 29 percent of the total emissions.

A comparison of NAPAP, Trends, and Work Group 3B emissions of  $\text{SO}_2$  and  $\text{NO}_x$  shows reasonable agreement. NAPAP total  $\text{SO}_2$  emissions are greater than Work Group 3B by 3 percent and than Trends by 5.5 percent. NAPAP and Work Group 3B/Trends emissions compare well for all categories except non-utility combustion. NAPAP total  $\text{NO}_x$  emissions are greater than Work Group 3B by 11.8 percent and greater than Trends by 4 percent. The greatest difference between NAPAP and Work Group 3B occurs for the electric utility category. The differences between NAPAP and Trends occur for the electric utility, non-utility combustion, and transportation categories. Some of the variation for utilities is a result of fuel differences; but most is likely to be caused by different emission factors and control efficiencies. The non-utility combustion category variation occurs for the same reasons as the variation in the utility category except that the non-utility category may be more affected by fuel differences. The transportation category variation occurs because more detailed traffic data, available only on a nationwide basis, are used in developing the Trends estimate.

## CONCLUSIONS

Version 4.0 of NAPAP represents a detailed inventory of emissions on a national scale for the 1980 base year. Over 80 percent of the NAPAP emissions truly represent 1980. Over 90 percent are in the range from 1978 to 1981. Many of these large emitting facilities are electric utility plants and nonferrous smelters for which extensive quality assurance efforts have already been performed. About 80 percent of point source  $\text{SO}_2$  and  $\text{NO}_x$  emissions occur at sources with complete stack data and valid UTM coordinates.

Future resources will be used to try to improve NAPAP to meet the needs of Eulerian modeling activities. Additional quality assurance efforts are planned for the 1980 data base focusing on major point sources. Additional pollutants not now in NAPAP, speciation of VOC and  $\text{NO}_x$  emissions, hourly temporal resolution of emissions, and spatial resolution of data into small grid zones covering the entire U.S. will be required. Other activities include incorporation of emission data for Canada into NAPAP, coordination with Task Group A to include natural emission sources into NAPAP, and a statistical evaluation of the uncertainty of NAPAP emission estimates.

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## UTILITY POINT SOURCE EMISSION INVENTORY

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## UTILITY POINT SOURCE EMISSION INVENTORY

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### ABSTRACT

Estimates of 1973-1982 annual SO<sub>2</sub> emissions from electric utility plants are presented in this paper. Results are based on analyses of plant level data collected by the U.S. Department of Energy on consumption and quality of fuels burned. Emissions are estimated from known information about fuel consumption, sulfur content, ash content, and control equipment. Results show that these reductions were due to the use of lower sulfur coals and to the operation of flue gas desulfurization equipment.

## UTILITY POINT SOURCE EMISSION INVENTORY

### INTRODUCTION

In the United States, the electric utility industry is responsible for approximately two-thirds of  $SO_x$  emissions (where  $SO_x$  is expressed as  $SO_2$ ). Therefore, it follows that much of the interest in the acid rain debate has focused on electric utilities. A number of researchers in the field are likely to have an interest in knowing how utility emissions have behaved in the last decade.

This paper provides detailed and definitive information on annual  $SO_2$  emissions from generating plants operated by the electric utility industry over the period 1973 through 1982. To produce these emission estimates, a detailed data base at the individual plant level of detail was used. For this paper, annual  $SO_2$  emission estimates are presented at the State level. More detailed information (e.g., plant level emissions) is available from the authors.

The utility data base used to develop the  $SO_2$  emission estimates presented in this paper is also being used as part of the NAPAP Acid Deposition Emission Inventory.

### DISCUSSION

A variety of sources were employed to develop the data base used in this study. Results are based on the authors' analyses of primary data collected and automated by the U.S. Department of Energy's (DOE) Energy Information Administration (EIA). These analyses included all fossil-fueled units, both steam and nonsteam. Data were merged to create a file containing a single record for each plant with all data elements of interest. The definition of a plant used for this analysis is identical to that used by DOE in the assignment of plant codes. That is, data from units at a single site were aggregated even if these units



were operated by different utilities. In addition, facilities owned by an industrial user were excluded even if they exist at a site that is also producing electricity for public distribution.

Primary data sources for this work include Federal Power Commission (FPC) Forms 4 and 423,<sup>1</sup> and the EPA's Utility Flue Gas Desulfurization (FGD) Survey.<sup>2</sup> FPC Form 4 (which in 1983 was renumbered and is now called Form EIA 759) is used to survey all generating electric utilities. This form reports the consumption and stocks of coal and other fuels at each plant; respondents account for 100 percent of total electric utility generation. FPC Form 423 surveys all fossil-fueled plants with a total generating capacity of 25 MW or more and reports the cost and quality (sulfur content) of fuels delivered to a plant.

EPA's Utility FGD Survey supplies current data on operating and planned domestic utility FGD systems. It summarizes information contributed by the utility industry, system and equipment suppliers, system designers, research organizations, and regulatory agencies. The survey includes data on system design, fuel characteristics, operating history, and actual system performance.

Emissions were calculated using information on fuels used for generating electricity (supplied by FPC Form 4) and fuel sulfur content (supplied by FPC Form 423). For those cases in which fuel quality data were not provided, State average sulfur content values were used. (The quantities of fuels for which sulfur contents were not available was small over the entire study period - between 1 and 1.5 percent of coal use was not accounted for; the comparable figures for oil were slightly higher.)

Sulfur dioxide emission calculations were straightforward. For coal, the quantity of fuel burned was multiplied by twice the sulfur content (1 ton of sulfur burned will produce approximately 2 tons of SO<sub>2</sub>). For coal-fired plants, an SO<sub>2</sub> ash retention value of between 5 and 25 percent was assumed based on coal quality. For bituminous coals (as defined by heating value), a 5 percent ash retention was used. Fifteen percent was assumed for subbituminous coal, and 25 percent for lignite. For oil and gas-fired plants, standard EPA emission factors and calculation methods were employed.

Emission reductions resulting from SO<sub>2</sub> scrubbing systems were also taken into account. For each plant with SO<sub>2</sub> scrubbing, a total equivalent net capacity of "SO<sub>2</sub> free" generation was based on the capacity scrubbed, date of commercial operation, and SO<sub>2</sub> removal rate. The total uncontrolled SO<sub>2</sub> emissions for the plant were then adjusted to account for removal of scrubbed SO<sub>2</sub>.

## CONCLUSIONS

Table 1 presents estimates produced in the present analysis of utility SO<sub>2</sub> emissions during the 1973-1982 period. Table 1 shows that total U.S. SO<sub>2</sub> emissions from electric utilities decreased 16 percent from 1973 to 1982 - from almost 19.3 million tons to 16.2 million tons. At the same time, the total heating value of the fuels used increased by 10 percent. Coal, oil, and gas used increased by 19 percent from 1973 to 1980, but decreased between 1980 and 1982. The reduction in SO<sub>2</sub> emissions while fuel use was generally increasing is due both to the use of lower sulfur coals and to the operation of FGD equipment.

The decrease in emissions occurred during a period with significant changes in the mix of fuels and in electric generation. Use of oil as a fuel for electricity generation has declined dramatically from 560 million barrels in 1973 to 250 million in 1982. The decline from 1978, the year before oil prices doubled, is even more dramatic - 1978 oil usage was nearly 640 million barrels.

Coal use in electricity generation has continued to climb, with an increase of more than 44 percent in the 1973 to 1982 period. Natural gas use decreased during the mid-1970's because of lack of availability, and increased in the 1979 to 1981 period as supplies became more abundant. Its use decreased again in 1982 because prices climbed due to decontrol. These natural gas usage changes had minimal effect on SO<sub>2</sub> emissions, however.

Coal-fired units are the dominant source of SO<sub>2</sub> among utilities. The percentage of total utility SO<sub>2</sub> emissions due to coal burning has remained relatively constant at approximately 90 percent. This has occurred despite the increase in the quantity of coal burned relative to other fuels. Reasons for this behavior include the overall decline of

sulfur content in coal and the increase in the number of coal-fired units equipped with FGD systems. The average sulfur content of coal declined by 36 percent between 1973 and 1982. During the same period, the average heating value of coals delivered to utilities declined by 5 percent. Even though the use of lower heating value western coals increased over this period, the majority of the large overall decrease in average coal sulfur content is not due to extensive use of western coals. Close examination of Table 1 shows that almost all of the utility SO<sub>2</sub> emissions decrease occurred in the eastern States (all States east of the Mississippi River plus one tier of States west of the Mississippi River).

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3. "Compilation of Air Pollutant Emission Factors, Third Edition." AP-42 (including Supplements 1-14) Office of Air Quality Planning and Standards, Environmental Protection Agency. Research Triangle Park, North Carolina. (1983).

Table 1. Utility SO2 Emissions from 1973 to 1982 (1000 Tons per Year)

State	1973	1974	1975	1976	1977	1978	1979	1980	1981	1982
Alabama	846.8	815.4	735.9	704.9	735.3	530.6	521.3	543.1	554.9	407.9
Alaska	12.2	13.1	13.0	12.8	12.4	7.7	12.2	11.7	12.3	13.3
Arizona	19.9	38.2	52.4	68.5	78.3	59.5	81.4	87.5	111.9	109.2
Arkansas	21.9	21.7	16.7	38.3	54.3	58.9	40.6	26.6	43.7	49.8
California	120.9	104.3	123.3	151.7	156.2	107.6	108.0	77.9	50.8	20.8
Colorado	47.3	43.6	59.2	67.9	78.5	76.6	77.4	77.5	71.6	78.6
Connecticut	52.2	58.3	32.3	25.2	23.5	26.0	27.6	32.1	30.2	46.4
Delaware	57.8	69.2	62.0	60.9	59.2	55.6	61.1	52.5	69.8	57.0
District of Columbia	20.9	16.3	7.0	7.2	12.6	10.4	6.7	4.6	2.6	0.8
Florida	748.3	725.4	656.4	673.2	657.8	595.1	658.7	725.9	753.9	648.5
Georgia	445.3	525.5	464.1	499.1	581.2	616.2	666.1	736.7	843.7	787.9
Hawaii	29.6	30.8	26.0	40.1	42.7	38.4	46.1	41.6	23.6	22.9
Idaho	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Illinois	1,618.6	1,598.9	1,423.6	1,428.8	1,367.0	1,292.9	1,167.7	1,125.6	1,001.2	1,037.8
Indiana	1,519.5	1,378.8	1,463.8	1,443.1	1,457.6	1,351.2	1,536.9	1,539.6	1,447.3	1,291.9
Iowa	226.8	170.3	186.3	217.8	230.0	263.7	230.8	231.3	193.0	189.2
Kansas	56.4	62.5	100.6	121.1	138.9	159.3	142.3	150.1	151.1	140.8
Kentucky	1,350.3	1,407.7	1,360.5	1,512.3	1,356.5	1,210.0	1,130.0	1,007.6	1,028.8	935.5
Louisiana	16.2	17.3	11.9	32.9	58.5	63.8	39.6	24.8	22.0	30.5
Maine	35.7	29.2	20.1	13.0	9.9	8.7	10.9	16.3	13.4	12.6
Maryland	204.6	239.0	191.7	218.2	198.0	220.5	205.2	223.2	197.2	203.0
Massachusetts	119.6	118.4	109.6	159.6	160.4	258.9	264.5	275.5	264.9	262.2
Michigan	1,107.8	1,064.8	1,011.6	887.6	905.1	806.9	741.0	565.4	598.7	588.9
Minnesota	277.9	222.0	205.6	230.6	230.5	190.3	163.6	177.4	150.3	128.1
Mississippi	78.5	118.6	132.6	154.1	198.0	208.7	166.1	129.2	99.6	99.4
Missouri	955.0	952.7	1,075.6	1,179.4	1,201.9	1,013.6	1,076.2	1,140.5	1,091.2	1,093.8
Montana	10.8	12.1	13.2	16.8	22.8	21.8	22.9	23.4	21.6	15.8
Nebraska	47.8	22.8	23.7	27.4	31.4	37.9	38.8	49.5	45.0	36.7
Nevada	31.1	32.2	32.7	33.6	35.6	38.6	47.0	39.5	42.4	58.8
New Hampshire	56.2	55.0	59.3	50.5	59.4	52.3	78.9	80.5	68.0	60.1
New Jersey	122.8	165.4	107.5	113.2	128.4	115.3	105.1	110.2	103.6	96.0
New Mexico	83.3	85.8	76.2	87.1	103.7	81.4	76.7	84.6	74.7	92.4
New York	534.3	594.0	538.1	512.8	548.8	520.0	508.1	480.3	524.7	469.8
North Carolina	440.8	449.7	373.9	410.2	427.2	396.4	379.5	435.4	444.8	409.7
North Dakota	49.6	52.7	41.5	58.7	65.2	71.6	82.2	82.5	74.6	89.9
Ohio	2,584.8	2,649.0	2,710.4	2,749.8	2,686.1	2,462.6	2,514.5	2,171.6	2,179.1	2,120.9
Oklahoma	0.4	1.4	0.3	0.3	3.1	13.0	19.5	37.7	56.0	72.2
Oregon	0.0	0.1	0.0	0.0	0.3	0.1	0.9	3.3	6.8	3.4
Pennsylvania	1,624.0	1,442.1	1,435.9	1,432.0	1,381.1	1,322.7	1,415.1	1,466.1	1,296.8	1,261.7
Rhode Island	7.7	7.2	4.1	3.0	3.6	3.4	2.8	5.2	5.0	3.1
South Carolina	163.6	157.0	139.6	162.2	194.4	192.9	191.1	213.1	231.5	197.2
South Dakota	3.8	3.3	21.4	33.9	30.2	32.4	27.9	28.6	22.1	31.0
Tennessee	1,139.4	1,107.2	1,030.3	1,228.3	1,257.6	1,033.1	893.3	933.7	875.9	629.0
Texas	56.2	54.5	84.8	117.3	143.3	179.8	221.7	302.8	332.3	356.5
Utah	10.8	12.1	19.8	12.8	32.2	29.9	30.4	22.1	25.3	24.7
Vermont	1.3	0.7	0.3	0.4	0.4	0.3	0.4	0.5	0.2	0.4
Virginia	276.6	261.5	209.1	224.9	238.0	223.9	203.2	163.7	140.5	123.0
Washington	37.7	33.0	34.8	37.0	53.9	69.8	79.3	69.4	61.7	49.7
West Virginia	972.8	1,071.6	1,029.7	1,010.4	1,001.4	895.5	955.9	944.2	944.4	885.7
Wisconsin	509.0	497.4	467.8	469.7	514.7	471.7	496.3	485.7	397.8	361.8
Wyoming	50.2	52.1	59.0	80.9	97.7	95.7	111.1	120.9	119.9	110.9
United States	18,805.1	18,661.7	18,355.2	18,821.4	19,070.8	17,593.2	17,684.9	17,378.5	16,922.5	15,817.2

DEVELOPMENT OF TEMPORAL, SPATIAL, AND  
VOLATILE ORGANIC COMPOUND ALLOCATION FACTORS  
FOR THE NAPAP EMISSION INVENTORY

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Presented at:

First Annual Acid Deposition Emission Inventory Symposium  
Raleigh, North Carolina  
December 3-4, 1984

DEVELOPMENT OF TEMPORAL, SPATIAL, AND  
VOLATILE ORGANIC COMPOUND ALLOCATION FACTORS  
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ABSTRACT

The Eulerian acid deposition model being developed for use in the National Acid Precipitation Assessment Program (NAPAP) will require more resolved emission data than are available in the NAPAP emission inventory. The NAPAP emission inventory, which separately covers annual emissions from point and countywide area sources, had to be apportioned to reflect hourly emissions with area source emissions assigned to grid squares. Hourly emissions of volatile organic compounds (VOC) then had to be allocated into photochemical reactivity classes, and nitrogen oxide ( $\text{NO}_x$ ) emissions separated into  $\text{NO}$  and  $\text{NO}_2$ . This paper describes the processes, assumptions, and data sources used in developing the NAPAP temporal, spatial, and species allocation factors.

DEVELOPMENT OF TEMPORAL, SPATIAL, AND  
VOLATILE ORGANIC COMPOUND ALLOCATION FACTORS  
FOR THE NAPAP EMISSION INVENTORY

INTRODUCTION

The most extensive use of the NAPAP emissions inventory will be to support the Eulerian acid deposition model currently under development. The NAPAP inventory, compiled using EPA's Emission Inventory System (EIS), contains annual emissions from point and area sources. Area sources are compiled on a county total basis, while point source data are compiled for individual sources. Emission totals in the NAPAP inventory for VOC and  $\text{NO}_x$  actually represent composites of various individual species. To support the Eulerian model, further temporal, spatial, and species resolution is required.

GCA utilized the Regional Model Data Handling System (RMDHS) to resolve the NAPAP inventory for use as a Eulerian model input tape. RMDHS calculated hourly emission totals of  $\text{NO}_x$ ,  $\text{SO}_2$ ,  $\text{SO}_4^{=}$ ,  $\text{NH}_3$ , and VOC, allocated VOC and  $\text{NO}_x$  into photochemical reactivity classes, separated out major point sources, and assigned minor point sources and area sources to grid cells. The major inputs that enabled RMDHS to generate the Eulerian modeler's tape from the NAPAP annual emission inventory were temporal, spatial, and pollutant species allocation factors, whose development is described below.

DISCUSSION

Temporal Allocation Factor Development

RMDHS apportioned the NAPAP annual emission totals into hourly totals for a typical summer weekday by applying the NAPAP temporal allocation factors, a series of fractional multipliers, to the EIS emission file. First, a seasonal fraction is applied to determine

quarterly emissions for the summer season. Next, a daily fraction is applied which apportions the seasonal total to a daily total for a typical weekday:

$$\text{daily fraction} = \frac{1}{(13 \text{ weeks/season}) (\text{number of operating days/week})}$$

Similarly, hourly totals are calculated by multiplying the daily totals by one of 24 hourly fractions representing an entire diurnal pattern. For example, if all of a plant's emissions occur during an 8 a.m. to 5 p.m. workday, the hourly fraction for each of these hours would be:

$$\text{hourly fraction} = \frac{1}{9 \text{ operating hours/day}} = 0.111$$

The hourly fraction for the 15 hours of nonoperation would, of course, be zero.

RMDHS can generate default temporal factors based on operating rates contained in EIS point source records or uniform emission distributions for area sources if no patterns are supplied. Therefore, primary emphasis was placed on developing temporal factors for the 54 NAPAP area source categories. Since the temporal distribution of emissions most often directly reflects the temporal patterns of the activities that cause the emissions, related categories were grouped together.

GCA developed temporal factors based on literature and data sources published by the U.S. Department of Energy, Department of Transportation, Civil Aeronautics Board, National Weather Service, and Bureau of the Census. Also examined were previously compiled regional-scale inventories such as the Northeast Corridor Regional Modeling Project (NECRMP), the Regional Air Pollution Study (RAPS), and the Sulfate Regional Experiment (SURE), and inventories developed by several States in support of their State implementation plans (SIP's).

Since the NAPAP study area spans four time zones, temporal factors were standardized to reflect Greenwich Mean Time (GMT). This was accomplished by creating four separate, time-zone-specific temporal factor files, each with local time adjusted to reflect GMT, and processing the EIS data accordingly. Thus, hourly emissions in the Eulerian model input tape reflect GMT.



### Spatial Allocation Factor Development

Spatial allocation factors were developed to apportion NAPAP area source emissions from counties to individual grid cells. The NAPAP grid system is comprised of 37,440 grid cells (156 rows, 240 columns) approximately 20 x 20 km, extending from 65° to 125° west longitude and from 25° to 51° north latitude.

Each spatial allocation factor assigns a portion of a particular county's area source emissions to a specific grid cell. Generally, since the actual subcounty distribution of area source emissions is unknown, emissions are assumed to be distributed according to the known distribution of some surrogate indicator (e.g., population).

The objective in NAPAP was to develop as many surrogate values as possible for each county to allow maximum flexibility in assigning county level area source emissions to specific grid cells. The surrogate indicators used in NAPAP include housing and population counts, total land area, and 10 land use classifications. Once the distribution of the surrogate indicators was known, county level area source emissions were spatially distributed by matching area source emission categories to the most appropriate surrogate indicators.

Housing and population surrogates were derived from the 1980 Census by assigning housing units and population counts to grid cells based on the latitude and longitude of the centroid of each enumeration district. Land use surrogates were derived using Landsat land use percentages for each grid cell and grid/county relationships in the following algorithm:

$$SPAFACT_{CSi} = \frac{(A_{Ci})(A_{Si})}{\sum_i^n (A_{Ci})(A_{Si})}$$

where:

$SPAFACT_{CSi}$  = the spatial allocation factor for county C, land use type S, and grid i;

$A_{Ci}$  = the portion of county C that falls within grid i;

$A_{Si}$  = the portion of grid i with land use type S; and

$n$  = the total number of grids covering county C.

The final step in defining spatial allocation factors was development of the surrogate factor selection file, which assigns each of the 54 NAPAP area source categories to the most appropriate surrogate indicator.

#### Species Allocation Factor Development

The NAPAP emission inventory includes annual emission rates for  $\text{NO}_x$  and VOC. The Eulerian acid deposition model requires disaggregation of VOC emissions into photochemical reactivity classes and separation of  $\text{NO}_x$  into NO and  $\text{NO}_2$ . There are numerous possible VOC speciation schemes based on different modeling chemistries. To provide the flexibility of developing and testing a number of reactivity schemes in NAPAP, it was decided to provide a general species listing for NAPAP point and area source classes, which in turn could be adapted to fit any particular modeling requirements. This objective was achieved by coding a set of "species profiles," each of which provides a typical list of VOC's for a given process. Each specie is defined by its SAROAD code, molecular weight, and weight percent of total VOC emissions.

A separate SCC Index File was created to link the emission inventory emissions classes (referenced by SCC) to the most appropriate species profiles. Finally, a photochemical class assignment file assigns each VOC specie (referenced by SAROAD code) to the appropriate reactivity class. This approach was taken to provide flexibility in establishing SCC-profile-reactive class relationships.

STATIONARY SOURCE EMISSION FACTOR DEVELOPMENT

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First Annual Acid Deposition Emission Inventory Symposium  
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## STATIONARY SOURCE EMISSION FACTOR DEVELOPMENT

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### ABSTRACT

The Eulerian atmospheric model under development for acid deposition analyses requires emission data bases of certain chemical species which act as direct acidic emissions to the atmosphere, scavengers of primary or secondary acids in the atmosphere, or catalysts in atmospheric transformation processes. Most of the chemical compounds or classes of compounds needed for input into the model chemical transformation/deposition modules are non-criteria pollutants. Limited information is available concerning emission factors for most species. This paper presents a discussion of the NAPAP Task Group B emission factor development project to assess and develop emission factors for ammonia, primary sulfate, alkaline dust, and specific organic compounds.

## STATIONARY SOURCE EMISSION FACTOR DEVELOPMENT

### INTRODUCTION

The National Acid Precipitation Assessment Program (NAPAP) was established by Congress in 1980 to coordinate and expand research on problems posed by acid deposition in and around the United States. The Interagency Task Force on Acid Precipitation manages the program by coordinating the activities of 10 task groups having the specific technical responsibilities. Task Group B (Man-Made Sources) is charged with providing a complete and accurate United States and Canadian inventory of emissions from man-made sources thought to be important in acid-deposition processes. Two distinct types of inventory development programs are required to address the research and assessment needs within NAPAP:

- Detailed, multicomponent point and area source disaggregated inventories for defined annual base years to support the development and testing of atmospheric transport and transformation models to predict acid deposition; and
- Retrospective inventory summaries of acidic and acid-precursor emissions over the past 80 years to support historical analyses of material damage and to aid in developing policy assessments.

### DISCUSSION

The Eulerian atmospheric model under development for acid deposition analyses requires emission data bases of certain chemical species which act as direct acidic emissions to the atmosphere, scavengers of primary or secondary acids in the atmosphere, or catalysts in atmospheric transformation processes. The preliminary 1980 NAPAP emission inventory for Eulerian model use contains point and area source emissions for:

1. sulfur dioxide ( $\text{SO}_2$ ),
2. primary sulfate ( $\text{SO}_4^{=}$ ),

3. nitric oxide ( $\text{NO}$ ),
4. nitrogen dioxide ( $\text{NO}_2$ ),
5. ammonia ( $\text{NH}_3$ ), and
6. volatile organic compounds (VOC) disaggregated according to 10 photochemical reactivity classes.

With the exception of  $\text{SO}_2$ , the remaining species within the inventory are either non-criteria pollutants or criteria pollutants which have been further disaggregated. For example, the oxides of nitrogen ( $\text{NO}_x$ ) and VOC speciation factors were developed under the Northeast Corridor Regional Modeling Program (NECRMP). Each VOC is defined by its SAROAD code, molecular weight, and weight percent of total emissions. These profiles are independent of any reactivity scheme, and may be manipulated by the modeler into any reactivity classification. The  $\text{NO}$  and  $\text{NO}_2$  allocation factors were also contained within NECRMP at the point and area source classification code (SCC) level and were subsequently incorporated into the preliminary 1980 inventory.

An assessment was prepared to review available  $\text{SO}_4$  emissions data. Adequate data exist for coal- and oil-fired utility sources operating without flue gas scrubbers. Emission factors for these sources were calculated and incorporated into the NAPAP inventory. Limited data were available for other fossil-fuel-fired combustion sources, and qualitative emission factors were formulated. Many remaining SCCs lacked any measurement data and only "best estimates" of  $\text{SO}_4$  emission factors could be provided.

A similar assessment study was prepared for  $\text{NH}_3$  emissions. Major sources of  $\text{NH}_3$  include livestock wastes, coal combustion, ammonium nitrate manufacture, anhydrous ammonia fertilizer application, petroleum refineries, urea manufacture, coke manufacture, and ammonium phosphate manufacture. Emission factors were calculated based upon available data.

The final 1980 NAPAP emission inventory will contain 23 chemical species or components:

- |                     |                        |
|---------------------|------------------------|
| 1. Sulfur dioxide   | 13. Aromatics          |
| 2. Primary sulfate  | 14. Aldehydes          |
| 3. Nitric oxide     | 15. Formaldehyde       |
| 4. Nitrogen dioxide | 16. Organic acids      |
| 5. Ammonia          | 17. Formic acid        |
| 6. Carbon monoxide  | 18. Propane            |
| 7. Total VOC        | 19. Butane             |
| 8. Methane          | 20. Benzene            |
| 9. Alkanes          | 21. Acetic acid        |
| 10. Alkenes         | 22. Alkaline dust      |
| 11. Propylene       | 23. Total particulates |
| 12. Ethylene        |                        |

Specific organic compounds or classes of compounds have been requested by the National Center of Atmospheric Research (NCAR), who has the responsibility for development and integration of specific chemical modules within the Eulerian model development program. Therefore, the emission factor development and formulation activities over the next 2 years will focus on the organic compound and alkaline dust emission factor development as the highest priority needs. Assessments of available data will be prepared with recommendation for the conduct of emission factor testing where necessary to develop data for important SCCs lacking information. In addition, an analysis of existing continuous emission monitoring (CEM) data for  $\text{SO}_2$  and  $\text{NO}_x$  will be conducted to verify the representativeness of NAPAP emission factors for major utility and industrial point sources.

SESSION 2: NAPAP EMISSION INVENTORY ACTIVITIES (continued)

Chairman: John Bosch, Chief  
National Air Data Branch  
U.S. Environmental Protection Agency (MD-14)  
Office of Air Quality Planning and Standards  
Research Triangle Park, NC 27711



USERS' GUIDELINES FOR ACCESS OF THE  
1980 NAPAP EMISSIONS INVENTORY

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Presented at:

First Annual Acid Deposition Emission Inventory Symposium  
Raleigh, North Carolina  
December 3-4, 1984

## USERS' GUIDELINES FOR ACCESS OF THE 1980 NAPAP EMISSIONS INVENTORY

### INTRODUCTION

An interim 1980 NAPAP emissions inventory data base is stored in the Emissions Inventory System (EIS) formats for point and area sources on the U.S. Environmental Protection Agency (EPA) IBM computer at Research Triangle Park, NC. The NAPAP data were developed starting with information from the EPA's National Emissions Data System (NEDS). These data have been improved by incorporating the latest available emission factors, substitution of data from the Northeast Corridor Regional Modeling Project and other more representative of 1980 NEDS data, cross-checking the electric utility data with the U.S. Department of Energy (DOE) data compiled by E.H. Pechan and Associates, cross-checking data with information from the U.S./Canada Work Group 3B report, and adding county centroid latitude and longitude for sources with missing or incorrect Universal Transverse Mercator (UTM) coordinates. The current data are preliminary and not suitable for defining 1980 emissions. Any use of the data should note the preliminary nature of the information. Revisions and additions to the data will be made based on information to be received from some States, and other changes will result from ongoing NAPAP data improvement activities.

Currently, NAPAP reports annual emissions of SO<sub>2</sub>, NO<sub>x</sub>, VOC, particulates, CO, primary sulfates, and ammonia. Additional pollutants are to be added in the future per NAPAP FY '85 and FY '86 program plans.

Since NAPAP emission inventory development activities began in 1983, a number of standard versions of the inventory has been made available to users. A summary status report, covering the data in standard version 3.0 (data in NAPAP as of May 1984), has been produced and distributed. In the future, updated standard versions of the NAPAP inventory will be made available to users.

To obtain NAPAP data, requestors may address their requests to:

Mr. Charles O. Mann  
U.S. Environmental Protection Agency (MD-14)  
Research Triangle Park, North Carolina 27711  
(Phone 919/541-5694, FTS 629-5694)

NAPAP data can be provided either as computer files on magnetic tape (standard EIS/PS and EIS/AS master file formats) or in a number of standard hardcopy formats. Normally, magnetic tape files will be provided as 9-track, EBCDIC characters, nonlabeled, 6250 bpi density unless the requestor specifies otherwise. The magnetic tapes contain data for individual point and area source records in standard EIS format. The entire file may be provided or only selected records retrieved by the standard EIS retrieval language. Requestors who want to obtain data on magnetic tape should forward their own tapes on which to write the selected data.

There are a number of standard format hardcopy reports that can also be provided. These reports may be created using the EIS retrieval language as well. A partial list of the report formats that are available is:

- EIS/PS detailed masterfile listing,
- EIS/PS condensed masterfile listing,
- EIS/PS emission summary by geographic area,
- EIS/PS emission summary by SCC,
- EIS/PS rank-ordered listing of emissions by plant/point,
- EIS/AS detailed masterfile listing, and
- EIS quick look report (one line listing of user-specified data items; available for both point and area source data).

(Examples of these report formats will be presented.)

Requests for NAPAP data that can be satisfied by any of these standard reports will be processed by the OAQPS National Air Data Branch as expeditiously as possible. Computer accounts have been established to cover the cost of computer resources, so there will not be any charge to the data requestor. Requests that cannot be satisfied by a standard format report may instead be completed by providing a magnetic tape file

to the requestor who will be responsible for development of his own software to process the data. Alternatively, requests from within the NAPAP user community that require development of special software or job control language to complete will be considered on a case-by-case basis. All such requests must be approved by the NAPAP emission inventory project manager (David Mobley, IERL-RTP) who will decide whether Task Group B contract resources can be committed to completion of such nonstandard data requests.

Since some of the data in the NAPAP point source file originated at State agencies who submitted the data to NEDS, the EPA is obligated to honor claims of confidentiality for these data indicated by the States. Data items that may be claimed as confidential include annual operating rate, maximum design rate, boiler capacity, emission estimation method code, and percent space heat. These data will be blanked out on reports or files that are provided to requestors who are not authorized to receive confidential data. Only Federal employees and contractors performing work for the Federal government are authorized to receive confidential data. These personnel must submit in writing a statement that they will use the data only for government-related work and will not release the data to anyone else.

In the future, NAPAP retrieval and reporting capabilities may be expanded to include more standard report formats and computer graphics capabilities. Availability of these new capabilities is dependent upon the level of resources that will be available in FY '85 and FY '86.

HISTORIC EMISSIONS OF SO<sub>2</sub> AND NO<sub>x</sub> SINCE 1900

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Contract No. 68-02-3511  
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First Annual Acid Deposition Emission Inventory Symposium  
Raleigh, North Carolina  
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## HISTORIC EMISSIONS OF SO<sub>2</sub> AND NO<sub>x</sub> SINCE 1900

by: G. Gschwandtner  
PES, Incorporated

### ABSTRACT

Historic emissions of sulfur dioxide (SO<sub>2</sub>) and nitrogen oxides (NO<sub>x</sub>) were estimated for Task Group B, Man-made Sources, of the National Acid Precipitation Assessment Program for each State of the conterminous United States. Historic emissions were estimated by individual source category on the State level from 1900 to 1980 for every fifth year and for 1978. The source categories included power plants, industrial boilers, industrial processes, commercial and residential heaters, natural gas pipelines, highway vehicles, off-highway diesel engines, and all other anthropogenic sources. These emissions were calculated from salient statistics indicative of fuel consumption or industrial output, estimations of average statewide fuel properties, and estimations of emission factors specific to each source category over time. The emission estimates were then aggregated to show the estimated emission trends by state, region, and all States combined.

This paper summarizes the estimated historic emission trends on a state, regional, and national scale. The trends are presented by source category and by major fuel type. The emission estimates allow temporal and regional trend comparisons between SO<sub>2</sub> and NO<sub>x</sub> emissions and chemical and biological effect trends being derived in other studies. They provide a basis for assessing pollutant damage, for studying trends in stream chemistry and deposition monitoring data, and for evaluating proposed mitigation and control strategies.

## HISTORIC EMISSIONS OF SO<sub>2</sub> AND NO<sub>x</sub> SINCE 1900

### INTRODUCTION

Sulfur dioxide (SO<sub>2</sub>) and nitrogen oxides (NO<sub>x</sub>) are considered primary precursors of acidic precipitation. The anthropogenic emissions of these pollutants are suspected causes of many biological and chemical effects observed in recent years. Understanding the historic emission trends is important to understanding the development of acid-precipitation-related problems and causes of observed environmental effects.

Annual quantities of emissions of SO<sub>2</sub> and NO<sub>x</sub> are presented for each of the conterminous 48 states, including the District of Columbia. Emissions of each pollutant were estimated by source category for every fifth year from 1900 to 1980 and for 1978. Total state emissions were interpolated for the other years based on national consumption levels of fuels. Five-year intervals from 1900 to 1980 were selected to provide an indication of the emission trends sufficient for most effects studies and to develop a methodology that could be applied to all other years. The state level was selected because it provides the most complete and consistent body of information on an historic basis and collectively covers all geographic regions of the country.

### DISCUSSION

Average emission rates for each study year were estimated for individual source categories for each state. The source categories are listed in Figure 1 according to the type of fuel consumed. These categories represent all types of boilers, furnaces, engines, processes, and other man-made emission sources. The basic steps involved in calculating state emissions are:

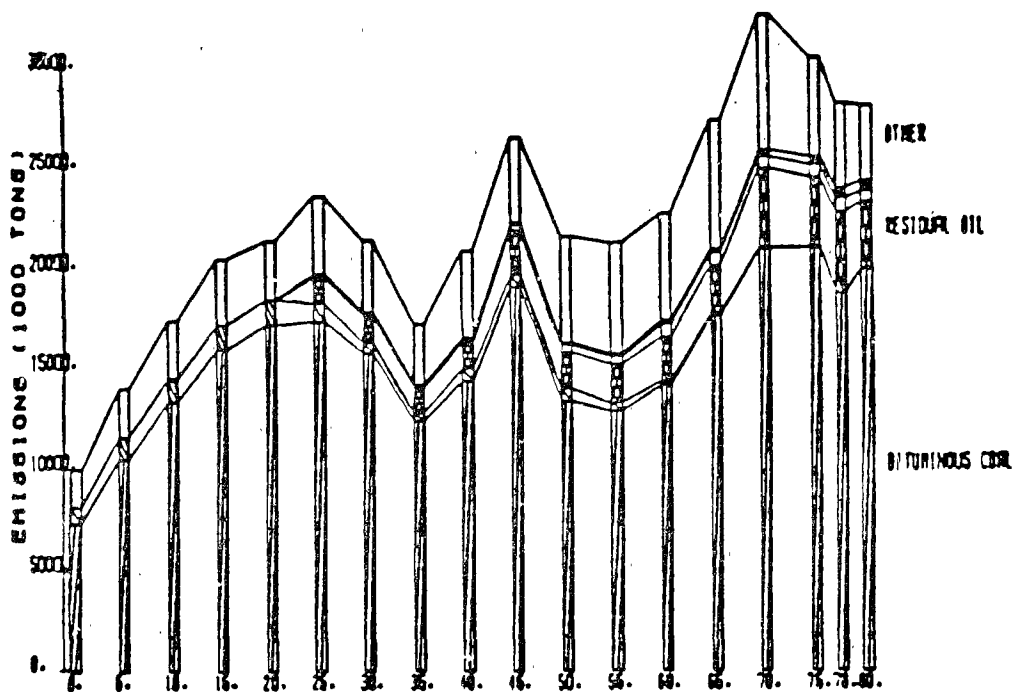


Figure 1. Overall trend in SO<sub>2</sub> emissions from 1900 to 1980 for the United States by year and by fuel type.



1. obtain state level information on fuel use;
2. allocate fuel quantity used by each source category;
3. develop source category emission factors;
4. determine fuel sulfur content by state for each category; and
5. calculate emissions, after emission controls.

The actual procedure varied somewhat depending on the usefulness and availability of information. It can generally be described in more detail for two time periods: 1) 1950 to 1980, and 2) 1900 to 1945.

For each source category, the annual fuel consumption (FC) was multiplied by a representative emission factor (EF) for each pollutant. For SO<sub>2</sub>, the emission factor was scaled by the average statewide sulfur content values (S) of the particular fuel. The calculation procedure can generally be expressed as:

$$\text{SO}_2 \text{ Emission} = \text{FC}_{i,j,k} \times \text{EF}_{\text{SO}_2} \times S_{i,j,k} \quad (1)$$

$$\text{NO}_x \text{ Emission} = \text{FC}_{i,j,k} \times \text{EF}_{\text{NO}_x} \quad (2)$$

where:

i = study year,

j = source category, and

k = state.

While these equations generally describe the approach, variations occurred depending on the nature of the source category and available information. For a complete discussion of the specific methodology for each source category, the EPA Project Officer may be contacted for additional descriptions which may be available. This presentation focuses primarily on the results obtained to-date.

Figures 1 through 4 show the estimated trend in emissions of each pollutant from 1900 to 1980. The estimated trend is shown on the national level (the result of aggregating all state emission estimates), on the regional level, and on the state level for selected states. Total estimated emissions are further divided according to major source and

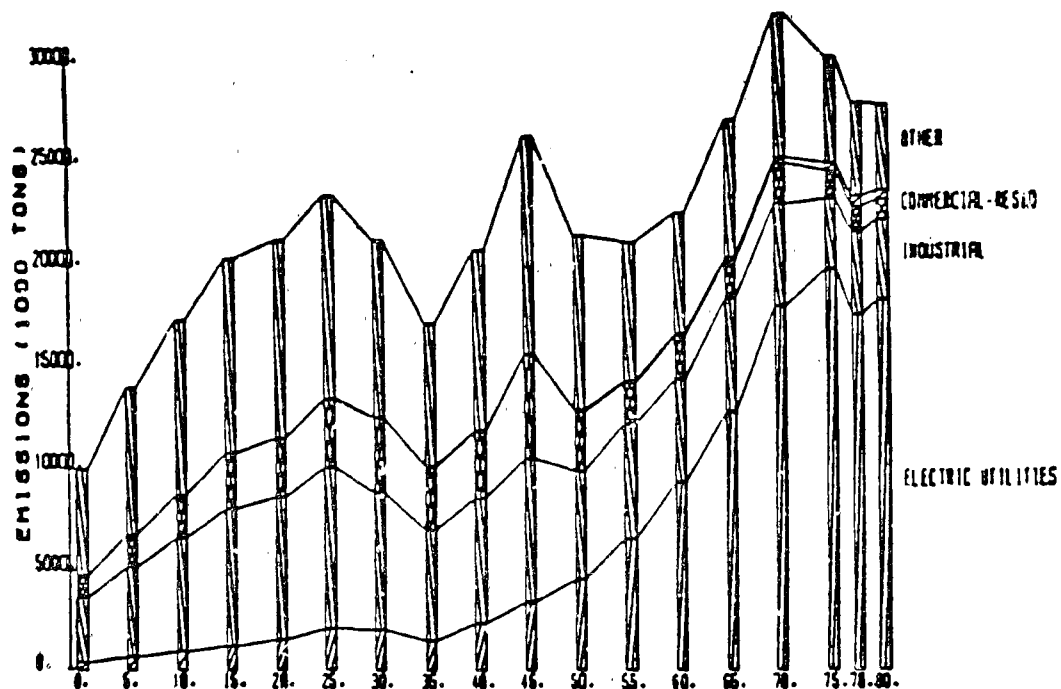


Figure 2. Overall trend in SO<sub>2</sub> emissions from 1900 to 1980 for the United States by year and by source category.

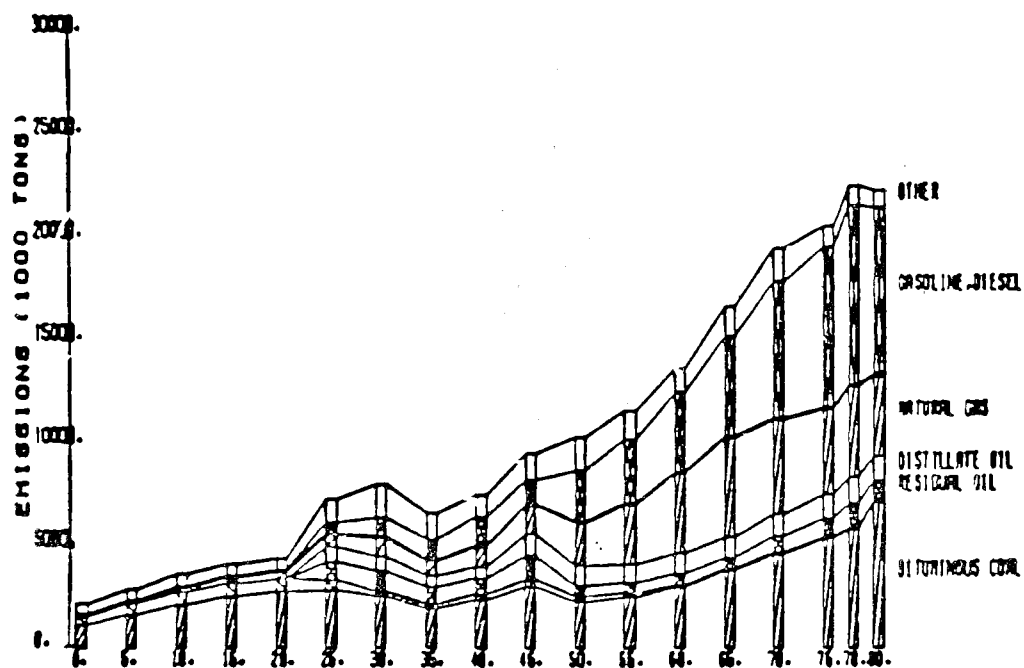


Figure 3. Overall trend in NO<sub>x</sub> emissions from 1900 to 1980 for the United States by year and by fuel type.

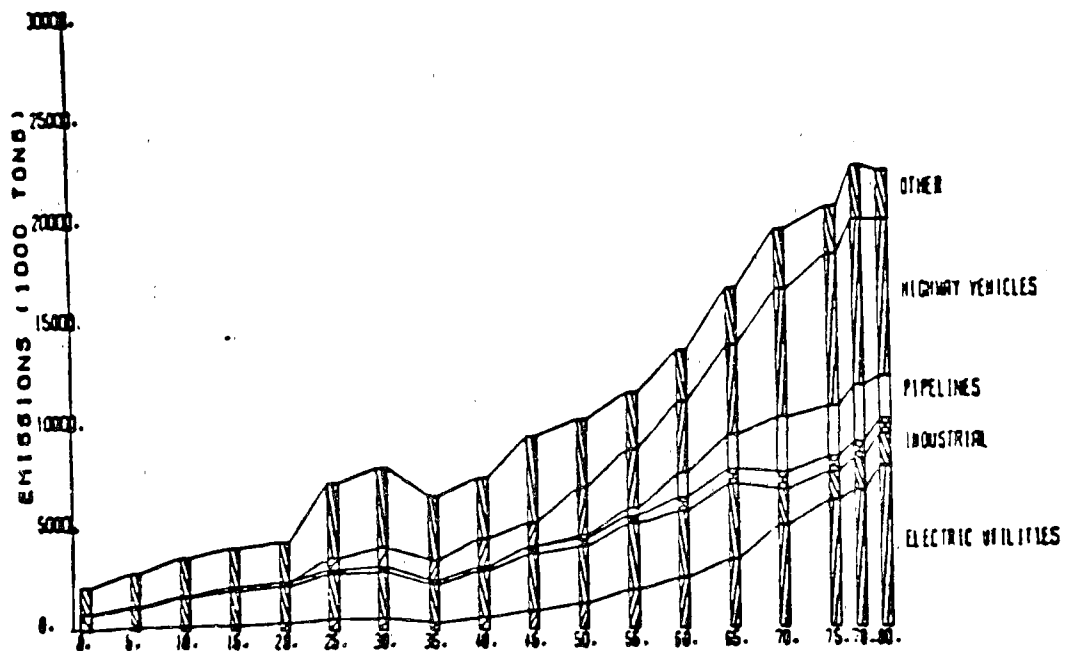


Figure 4. Overall trend in NO<sub>x</sub> emissions from 1900 to 1980 for the United States by year and by source category.

fuel categories to show the role of historic emitter groups, the effect of fuel switching, the effect of technological changes, and (recently) the effect of fuel mixing and SO<sub>2</sub> controls.

## CONCLUSIONS

The historic emission estimates presented are consistent in the estimating methodology used. They provide a basis for studying the relationship between emissions in the past and observed environmental effects including tree-ring growth patterns, material damage, and acidic deposition. The emission quantities shown in this presentation should be considered the best available estimates at this time, but it should be remembered that, as work in this area continues, more refined estimates may be made.

The historic emission estimates show a definite trend in terms of the total nation. State emission trends vary significantly depending on the state's location and geographic size, population, industries, and other factors. NO<sub>x</sub> emissions appear to have been increasing almost steadily in all states, while SO<sub>2</sub> emissions appear to have decreased, most recently since around 1970.

DEVELOPMENT OF A MONTHLY HISTORICAL  
EMISSIONS INVENTORY

Duane Knudson  
Energy and Environmental Systems Division  
Argonne National Laboratory

Presented at:

First Annual Acid Deposition Emission Inventory Symposium  
Raleigh, North Carolina  
December 3-4, 1984

DEVELOPMENT OF A MONTHLY HISTORICAL  
EMISSIONS INVENTORY

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ABSTRACT

As atmospheric transport and deposition modeling capabilities increase and monitoring data are accumulated, it becomes desirable to also refine emissions inventories. One such refinement is the application of monthly fuel use and industrial production data to define the intra-annual variability of emissions. This is the general approach being taken to portion the 1980 NAPAP SO<sub>2</sub> emissions to monthly values. The data bases for disaggregating annual emissions are Energy Information Administration Form 759 (formerly Form 4) for electric utilities; the Quarterly Coal Report for coal use by industrial and commercial/institutional sources; Federal Reserve Board (FRB) monthly industrial production indices for industrial processes; industry-specific fuel use and FRB monthly production indices for oil and gas use by industrial boilers; and heating degree-day accumulation for space heating for all fuels. Monthly SO<sub>2</sub> inventories are being prepared for utility and nonutility sectors for the period 1975 to 1983. This paper presents 1980 monthly SO<sub>2</sub> emissions for New York and West Virginia for the subject sectors.

## DEVELOPMENT OF A MONTHLY HISTORICAL EMISSIONS INVENTORY

### INTRODUCTION

Atmospheric deposition patterns are largely determined by meteorological conditions and precursor emissions. The variability of meteorological conditions controlling acidic deposition has been recognized, with the consequence that several years of meteorological data are being readied for model analysis. To complement these data, monthly emissions inventories of  $\text{SO}_2$  and  $\text{NO}_x$  for the period 1975 through 1983 are being prepared. These inventories are designed to reflect actual intra-annual and yearly emissions variability, and when used with the actual meteorological data in model simulations will eliminate some unnecessary assumptions.

The methodology used to portion the annual values is the focus of this paper. Review of the method will deal first with  $\text{SO}_2$  emissions from electric utilities, next with application of a number of data bases to portioning annual  $\text{SO}_2$  emissions from industrial (process and combustion) institutional/commercial and residential source categories, and finally for computation of monthly  $\text{NO}_x$  emissions (yet to be undertaken). State total monthly  $\text{SO}_2$  emissions will be presented for New York and West Virginia, as examples of the type of results obtained.

### DISCUSSION

The 1980 NAPAP emission inventory (Version 3) was used as a starting point to define State total  $\text{SO}_2$  emissions by 6-digit SCC class. The following paragraphs present methodology and pertinent data bases for computing monthly  $\text{SO}_2$  emissions for the utility sector and specific SCC classes. Data bases used for portioning annual nonutility  $\text{SO}_2$  emissions, discussed in the succeeding paragraphs, are listed in Table 1 for respective SCC's.



**Table 1 Data Sources for Estimation of Nonutility Monthly SO<sub>2</sub>  
Emission Fractions for NAPAP Source Categories**

SCC Code	Description		Data Source or Approach for Estimating Monthly Fractions
<u>POINT SOURCES</u>			
102001	Industrial	Anthracite	Quarterly Coal Report - 1981
102002		Bituminous	Quarterly Coal Report - 1981
102003	External	Lignite	Quarterly Coal Report - 1981
102004		Resid. Oil	Adjusted FRB monthly production indices
102005	Combustion	Dist. Oil	Adjusted FRB monthly production indices
102006		Nat. Gas	Adjusted FRB monthly production indices
102007	Boilers	Process Gas	Adjusted FRB monthly production indices
102008		Coke	Adjusted FRB monthly production indices
103001	Commercial/	Anthracite	Quarterly Coal Report - 1981
103002	Institutional	Bituminous	Quarterly Coal Report - 1981
103003	External	Lignite	Quarterly Coal Report - 1981
103004		Resid. Oil	Unresolved
103005	Combustion	Dist. Oil	Unresolved
103006		Nat. Gas	Unresolved
103007	Boilers	Process Gas	Unresolved
103008		Coke	Unresolved
105001	Ext. Combust. Space Htr - Industrial		Local Climatological Data - Heating Degree Day Accumulation
105002	Ext. Combust. Space Htr - Com/Inst.		Local Climatological Data - Heating Degree Day Accumulation
202001	Int. Combust. Dist. Oil - Industrial		Unresolved
203001	Int. Combust. Dist. Oil - Com/Inst.		Unresolved
301	Industrial chemicals: sulfuric acid, plastics, organic chemicals, explosives, carbon black, printing ink		FRB monthly production indices for SIC 2819: inorganic chemicals
302	Food product processing		FRB monthly production indices for SIC 209: miscellaneous food preparation

Table 1 Continued

SCC Code	Description	Data Source or Approach for Estimating Monthly Fractions
303	Primary Metal: Coke manufacturing, steel production, copper smelters, zinc, and other primary metals	FRB monthly production indices for SIC 331; basic steel, coking, and mill production
304	Secondary smelting: aluminum, copper, lead, etc.	FRB monthly production indices for SIC 333-6, 9; nonferrous metals
305	Mineral Products: glass, fiber-glass, gypsum products, cement, brick, pottery	FRB monthly production indices for SIC 102-5, 108, 109; non-ferrous ores: 326-9; concrete and miscellaneous clay: 3151; brick: 324; cement
390	In-Process fuel use	Same as SCC 305
306	Petroleum refining	FRB monthly production indices for SIC 291-9; petroleum refining
307	Wood and paper products (including Kraft pulping)	FRB monthly production indices for SIC 261-263; pulp and paper
310	Crude oil and natural gas extraction (including gas sweetening)	FRB monthly production indices for SIC 131; crude oil and natural gas extraction

AREA SOURCES

90100101	Residential	Anthracite coal	Same as SCC 105002
102	Combustion	Bituminous coal	Same as SCC 105002
103		Distillate oil	Same as SCC 105002
104		Residual oil	Same as SCC 105002
105		Natural gas	Same as SCC 105002
106		Wood	Same as SCC 105002

Table 1 Continued

SCC Code	Description		Data Source or Approach for Estimating Monthly Fractions
90100207	Commercial/	Anthracite coal	Same as SCC 103001
208	Institutional	Bituminous coal	Same as SCC 103002
209	Combustion	Distillate oil	Same as SCC 103005
210		Residual oil	Same as SCC 103004
211		Natural gas	Same as SCC 103006
90100313	Industrial	Anthracite coal	Same as SCC 102001
314		Bituminous coal	Same as SCC 102002
315		Coke	Same as SCC 102008
316		Distillate oil	Same as SCC 102005
317		Residual oil	Same as SCC 102004
318		Natural gas	Same as SCC 102006
320		Process gas	Same as SCC 102007
90200121	Residential Incineration		Divide annual values by 12.
90200122	Industrial Incineration		Divide annual values by 12.
90200123	Commercial/Institutional Incineration		Divide annual values by 12.
903	Transportation-related		Unresolved

For the utility sector, annual emissions were those developed by E. H. Pechan and Associates for the years 1975 through 1982, with 1983 utility emissions computed by Argonne using Pechan's approach.<sup>1</sup> Portioning annual emissions to monthly values relied on fuel consumption data presented in EIA Form 759. Form 759 provides information on the quantity of fuel consumed. Fuel quality information is available only for fuel delivered (from Form 423). This discrepancy necessitates the assumption that the quality of fuel consumed in a given month is approximated by the annual average quality of that type of fuel. The implication of this assumption on monthly emissions estimates is being investigated.

Annual coal combustion emissions from industrial and commercial/institutional boilers (SCC classes 102001-102003 and 103001-103003) were adjusted using quarterly coal consumption data from 1981.<sup>2</sup> The 1981 data are the first year of available data for coal consumption. Monthly fractions were derived from the quarterly data by dividing by three.

Annual emissions from oil, natural gas, process gas, and coke combustion in industrial boilers (SCC's 102004-102008) were apportioned to monthly values using fractions based on State-specific annual industrial fuel consumption and monthly industrial production data. Since the NAPAP inventory does not differentiate fuel combustion in industrial boilers by industry, State fuel use by 2-digit SIC industrial classification was used to determine weighting factors for use with the monthly Federal Reserve Board (FRB) production indices. Annual industrial fuel consumption, for use in weighting the monthly production indices, was for SIC classes 20, 26, 28, 29, 32, and 33. State-level industrial fuel use statistics are compiled by the Commerce Department and reported in the Annual Survey of Manufactures Fuels and Electric Energy Consumed.<sup>3</sup> The weighting factors adjust the monthly production fractions for the identified SIC classes by the relative amount of fuel consumed and specific industrial class in each State. The monthly FRB production indices are compiled by the Commerce Department based on output of specific industrial classes.<sup>4</sup>

The computation of monthly emissions for space heating in industrial and commercial/institutional sectors follows the NEDS/NAPAP methodology. That methodology holds 25 percent of the annual State SO<sub>2</sub> emissions constant and apportions the other 75 percent on a seasonal basis using heating degree day accumulation. A difference in this methodology was to apportion to monthly values. A representative station for heating degree day accumulation was selected for each region.

State total annual SO<sub>2</sub> emissions for industrial processes (SCC's 301, 302, 303, 304, 305, 306, 307, 310, and 390) were apportioned to monthly values through use of appropriate national average monthly FRB production indices.

Relevant data on which to base a derivation of monthly emissions for commercial/institutional boilers burning oil, gas, and coke (SCC 103004-103008) and internal combustion for industrial and commercial/institutional sectors (SCC's 202001 and 203001) have not been found. Two options being considered for commercial/institutional boilers are to use the coal-derived monthly fractions for the entire category or simply to divide the oil, gas, and coke-related emissions by twelve. Tentative plans for treating internal combustion of distillate oil in the industrial and commercial/institutional sectors are to divide the annual emissions by twelve to get monthly values.

Computation of monthly emissions for area sources of residential, commercial/institutional, and industrial fuel combustion follows the methodology taken for corresponding point source categories. Annual emissions from residential, commercial/institutional, and industrial incineration were disaggregated to monthly values by dividing the annual estimates by twelve.

Two major assumptions must be made to allow the use of 1980-specific monthly fractions for computations of monthly emissions for the other years in the analysis. First, it must be assumed that relative monthly activity in the specific categories is constant throughout the period of interest (1975-1983). The second assumption is that relative industrial activity among the States remains constant for the same period. Given these assumptions, the procedure for computing monthly emissions for the subject SCC classes for the non-1980 years uses the national totals presented in the report "National Air Pollutant Emission Estimates,

1974-1982"<sup>5</sup> to adjust the 1980 SCC-specific emissions for each year. Relative changes to national emissions for each SCC are applied equally to each State to compute year-specific State total SO<sub>2</sub> emissions. The monthly apportioning then proceeds according to the process used for 1980.

The use of the same monthly fractions for all years in the analysis means that such influences on emissions as industry-wide strikes are not reflected in the monthly emissions data. Conversely, any anomalies specific to 1980 are propagated through the entire period. This is not strictly acceptable, but is a reasonable preliminary approximation considering budgetary and schedule constraints. It is planned to reevaluate this assumption later, and, if appropriate data are available, compute year-specific monthly fractions for years other than 1980.

Results of application of the monthly portioning are presented in Table 2 for New York and West Virginia. Monthly emissions estimates for individual source classification codes have been consolidated into source category totals. As previously mentioned, satisfactory information for portioning of annual SO<sub>2</sub> emissions from commercial and institutional fuel combustion has not been discovered. Annual emissions for this source category are thus assumed to be uniformly distributed throughout the year. The other source category presenting some problems is the potpourri of area sources. In some States, emissions from transportation-related activities can contribute substantially to total area source emissions. To date, however, project efforts have focused on stationary sources, necessitating an assumption of uniformly distributed transportation SO<sub>2</sub> emissions.

The monthly variability of utility emissions is the major controlling influence on monthly SO<sub>2</sub> emissions for New York and West Virginia, which is the situation for most Eastern U.S. States. The combined contribution of nonutility sectors in New York has the potential to significantly influence monthly emissions. However, the monthly variability of SO<sub>2</sub> emissions from these categories is small. This suggests, although it does not prove, that simplifying assumptions for nonutility source categories may be acceptable. Work continues on these questions.

**Table 2 Preliminary Estimates of 1980 Monthly SO<sub>2</sub> Emissions  
For New York And West Virginia (Short tons)**

	<u>Jan</u>	<u>Feb</u>	<u>Mar</u>	<u>Apr</u>	<u>May</u>	<u>Jun</u>	<u>Jul</u>	<u>Aug</u>	<u>Sep</u>	<u>Oct</u>	<u>Nov</u>	<u>Dec</u>	<u>Total</u>
<u>New York</u>													
Utility	49050	40800	43180	41550	34550	38430	41550	43660	36360	34520	34500	45360	483520
Industrial Fuel Combustion	16247	16493	16704	16470	16080	16050	15030	15646	16201	16005	15904	15356	192186
Commercial/Institutional Fuel Combustion	5963	5963	5963	5963	5963	5963	5963	5963	5963	5963	5963	5963	71556
Industrial Processes	2757	2736	2774	2678	2517	2422	2315	2409	2471	2599	2674	2644	30996
Area Sources	5939	5939	5668	5126	4584	4381	4313	4381	4584	4855	5397	5939	61106
Total	79956	71931	74289	71787	63694	67246	69171	72059	65579	63942	64438	75262	838364
<u>West Virginia</u>													
Utility	80170	65420	73230	67120	62420	68440	88620	88890	79030	80820	84580	100070	938820
Industrial Fuel Combustion	8457	8478	8514	6904	6798	6758	6907	6938	6986	7040	7091	7071	87942
Commercial/Institutional Fuel Combustion	70	70	70	70	70	70	70	70	70	70	70	70	840
Industrial Processes	4773	4913	5211	5055	4522	4174	3705	3115	4090	4432	4804	4794	53583
Area Sources	1542	1411	1367	1015	817	729	707	707	773	1024	1235	1520	12852
Total	95012	80292	88392	80164	74627	80171	100009	99720	90949	93386	97780	113525	1094042

## CONCLUSIONS

Several data bases were accessed to provide information for portioning State total annual SO<sub>2</sub> emissions to monthly values. The data can generally be characterized as fuel use and activity indicators.

## REFERENCES

1. Pechan, E. H. and J. H. Wilson, Jr. "Estimates of 1973-1982 Annual Sulfur Oxide Emissions from Electric Utilities." JAPCA Vol. 34, No. 10, pp. 1075-1078. (October 1984).
2. U.S. Department of Energy, Energy Information Administration, Office of Coal, Nuclear Electric, and Alternate Fuels. Quarterly Coal Report. DOE/EIA-0/21 (82/1Q), (82/2Q), (82/3Q), (82/4Q).
3. U.S. Department of Commerce, Bureau of the Census. 1980 Annual Survey of Manufactures Fuels and Electric Energy Consumed. M80(AS)-4.2. (Issued October 1982).
4. Board of Governors of the Federal Reserve System, Division of Research and Statistics. "Industrial Production Indexes, 1980." (September 1981).
5. U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards. "National Air Pollutant Emission Estimates 1940-1982." EPA-450/4-83-024. (February 1984).



QUALITY ASSURANCE OF THE  
NAPAP MAN-MADE EMISSIONS DATA BASE

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Presented at:

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Raleigh, North Carolina  
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## QUALITY ASSURANCE OF THE NAPAP MAN-MADE EMISSIONS DATA BASE

### INTRODUCTION

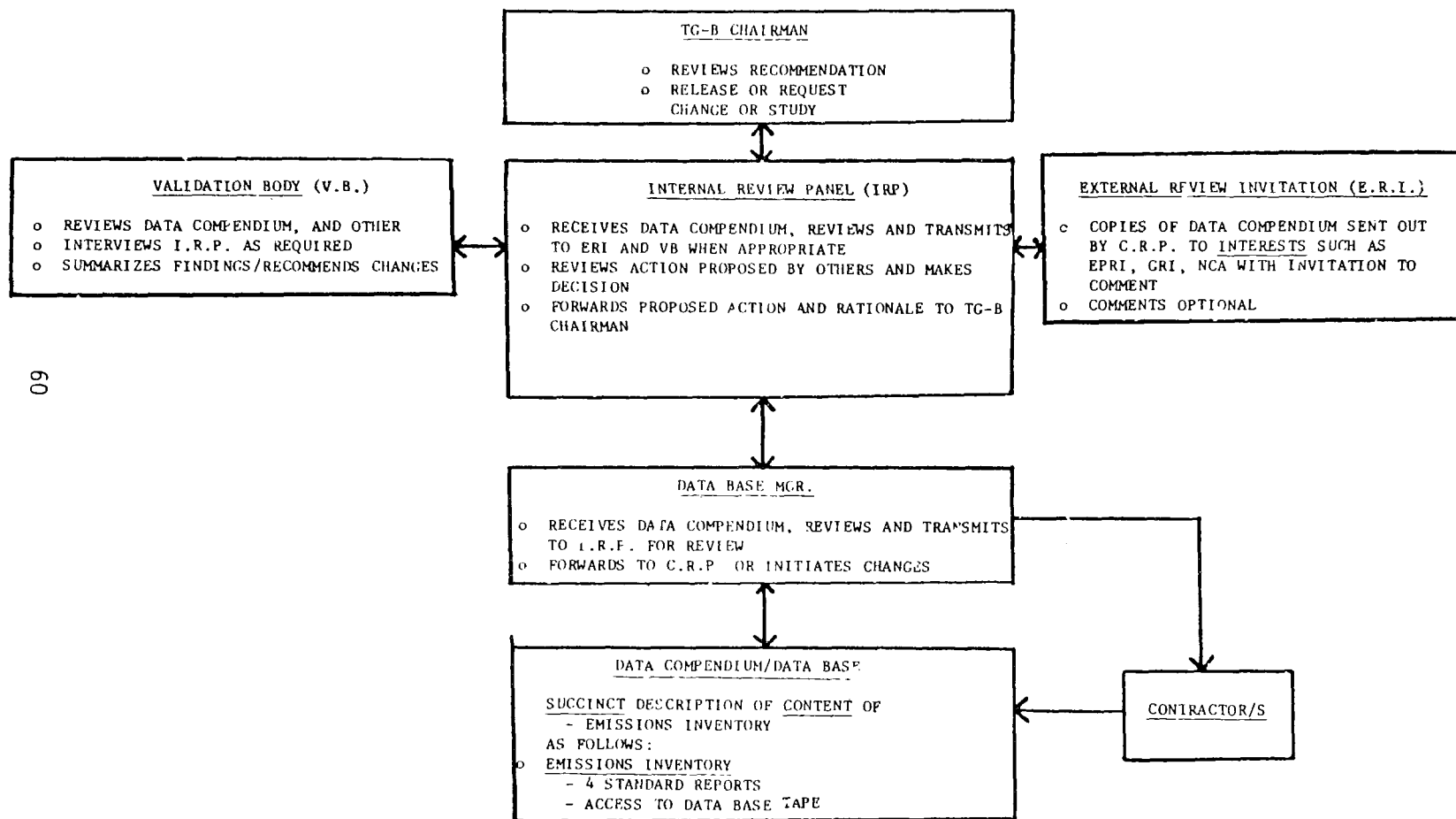
The pursuit of quality in the creation of man-made emissions data bases brings with it the need for unique approaches. While these approaches are applied within the principals of traditional quality assurance practices, it must be recognized up front that we must pursue our objectives without the benefit of significant absolute standards against which we can measure the quality of our product. The generation of our product, as previously discussed, is primarily a matter of building from a framework of test data a highly disaggregated data base through the application of estimates and assumptions. It follows then that quality must be pursued as a constructive direction and not as a quantitative objective. The direction we have chosen provides the framework for iterative improvement through visibility and review. Coupled with this is a program of accuracy assessment. By making our assumptions and results visible, we are trying to encourage constructive feedback which will lead to a more accurate product. By pursuing a program of accuracy assessment, we will be providing the consumers of our data our best assessment of its true accuracy at the time.

### DISCUSSION

#### Review/Reconciliation

Our review program can perhaps be best understood by making reference to Figure 1. This figure is a simplification of the review/reconciliation procedure put in place last December for the review of Task Group B emissions data bases prior to their release for use. A similar procedure was also established for the Task Group B Model Set/Data Base. It is significant to note that these procedures are end product review procedures and are applied in addition to the normal quality control practices followed in the development of the data.

FIGURE 1  
REVIEW/RECONCILIATION FLOW DIAGRAM  
EMISSIONS INVENTORY



Referring now to Figure 1, it can be seen that the major participants in the procedure are the Data Base Manager, the Internal Review Panel, the External Reviewers, a Validation Body (if needed), and the Task Group B Chairman. The major products are the Data Base being reviewed, and a Data Compendium. The Internal Review Panel members are generally persons who have not been involved directly in the creation of the product under review.

Procedural action begins with the Data Base Manager who, when he believes his data base is ready for release, assembles a Data Compendium succinctly describing the content, important rationale and summarized output from the data base and arranges for access to the Data Base computer tapes. This data is then received by the Internal Review Panel where it undergoes reviews and perhaps some modification.

The next step in the procedure takes place when the panel decides the data base is ready for external review. Copies of the Data Compendium, and information on access to the Data Base tape, are sent out to all known interested parties with an invitation to review and comment.

Comments received by the panel and discussions are made with regard to changes or future reviews. The panel might choose to involve a Validation Body in further review. When satisfied with the reviews, the panel forwards its recommendations to the Task Group B Chairman with regard to release of the data base along with its rationale.

Final release is made by the Task Group B Chairman.

#### Accuracy Assessment

Our accuracy assessment program should be recognized as a modest step forward in estimating the accuracy of our end product, but obviously limited in terms of real emissions data against which to measure true accuracy. Our objective is to provide an uncertainty or precision value to coincide with each emission value coming from the emissions data base. This will be achieved by the construction and interconnection of an Uncertainty Data Base such that the call for a report of an emissions value engages the Uncertainty Data Base in which resides predetermined values of uncertainty based on the resolution, source type and emissions species being reported. The construction of the Uncertainty Data Base will be accomplished through the use of "Expert Teams" who will judge

the precision with which we know such things as operating habits, fuel quality, feed qualities and the emitting community as well as the normal assessments of activity level, emission factors and control efficiency.

The interconnection of the Uncertainty Data Base is seen in Figure 2. The kinds of factors which will be judged for each speci/resolution cell will include those listed in Figure 3.

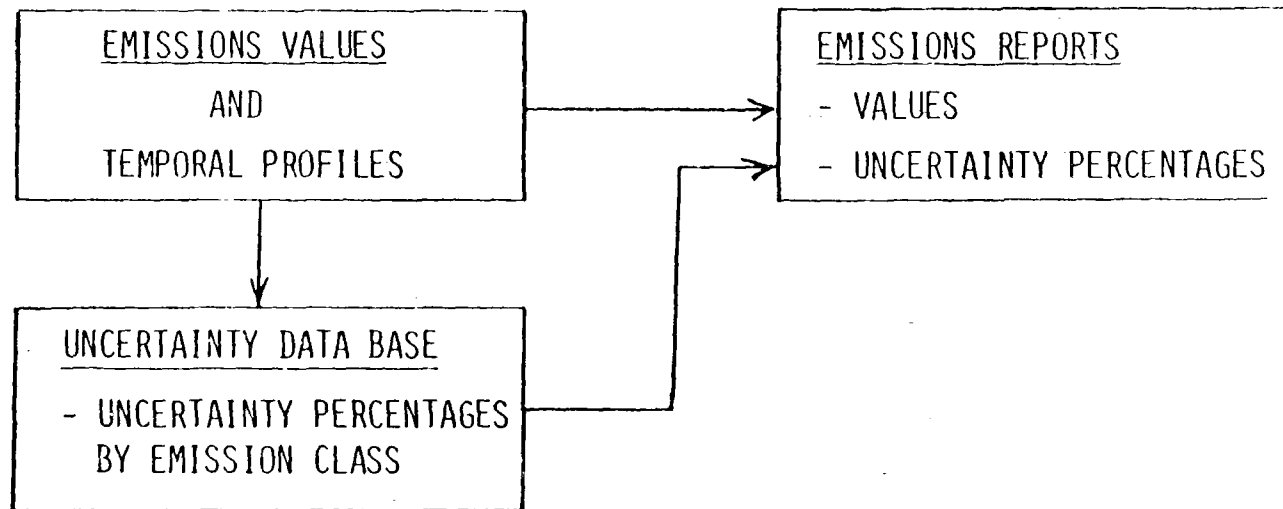
Further information on the uncertainty data base will be provided in the next presentation by Carmen Benkovitz.

### CONCLUSION

In conclusion, it should be recognized that the Quality Assurance Program, like the data base work it serves, is in a relatively early stage in its development. Its objectives at this time are to make modes and general improvements. The approach being taken follows the general guidelines of quality assurance in terms of high reporting levels, avoidance of conflicts of interests and visibility. It relies a great deal on stimulating corrective feedback from external reviewers. It could benefit greatly from increased measurement and feedback whereby estimated emissions are compared with true measurements.

FIGURE 2

UNCERTAINTY ESTIMATES, EMISSION DATA BASE



UNCERTAINTY ESTIMATES, EMISSIONS DATA BASE

FACTORS WHICH AFFECT ACCURACY OF ESTIMATES

DEVIATIONS ON:

- (G) ● EMISSION FACTORS: AVAILABILITY, ACCURACY, DEVIATION, VARIABLES
- (A) ● FUEL/FEED; QUANTITY DEVIATIONS FROM ASSUMED VALUE
  - A ● FUEL QUALITY; S, N CONTENT DEVIATIONS
  - B ● ASSUMED IDENTIFY OF EMITTERS OR SOURCES
  - C ● INLET AIR TEMP. RELATIVE TO ASSUMED VALUE
  - D ● EXCESS AIR RELATIVE TO ASSUMED VALUE
- E ● PRODUCTION ACTIVITY; QUANTITY DEVIATIONS FROM ASSUMED VALUE
- F ● OPERATING PROFILES; RELATIVE TO ASSUMED VALUE
- H ● CLIMATIC VALUES; RELATIVE TO ASSUMED VALUE
- J ● VMT/POPULATION
- K ● POPULATION
- L ● FAR/POPULATION
- M ● OTHER
- (RE) ● CONTROL TECHNOLOGY EFFECTIVENESS

ESTIMATION OF UNCERTAINTY WITHIN  
NAPAP EMISSION INVENTORIES

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Presented at:

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ESTIMATION OF UNCERTAINTY WITHIN  
NAPAP EMISSION INVENTORIES

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ABSTRACT

A major goal of Task Group B is the development and maintenance of detailed inventories of anthropogenic emissions in support of acid deposition research. The acid deposition emission inventory plan, prepared as one of the EPA contributions to Task Group B, identifies the need for emission inventories of current base years to support both assessment activities and the development, evaluation, and use of long range transport and transformation models. The uncertainty estimates of emission data are an integral part of the inventory. The objective of this project is to develop the methodologies needed to evaluate the uncertainties associated with the emission data as presented in the 1980 base year NAPAP emission inventory and to implement a prototype system to calculate these uncertainties.

## ESTIMATION OF UNCERTAINTY WITHIN NAPAP EMISSION INVENTORIES

### INTRODUCTION

The NAPAP emission inventory was based on the National Emissions Data System (NEDS) currently operated by the Office of Air Quality Planning and Standards (OAQPS) of the EPA (National Air Data Branch, 1983). NEDS provided the basic data from which all other levels of aggregation or disaggregation will be calculated. The basic NEDS data are statistical averaged parameters which allow the calculation of yearly emissions of the five criteria pollutants (particulates,  $\text{SO}_2$ ,  $\text{NO}_x$ , VOC, and CO) on an individual source/process basis for point sources and on a county level for area sources. Current plans call for the application of spatial, temporal, and species disaggregation algorithms which will be based on disaggregation factors (or modifications thereof) developed for the Northeast Corridor Regional Modeling Project (NECRMP). Higher levels of aggregation will be calculated as sums of the NEDS data.

Calculation of the uncertainty of the emissions values will be based on the statistical formulas expressing the variance of a function based on the expected values and variances of the parameters used to calculate the function. Application of these techniques will start with the algorithms used to calculate the yearly emission values and will be extended to include currently known algorithms for spatial and temporal aggregation and for spatial, temporal, and species disaggregation as applicable.

### DISCUSSION

The statistical formulas to be used are those expressing the variance of a function based on the expected values and the variances of the parameters used to calculate the function. For  $N$  independent parameters

and a function of the form  $x = \sum_{k=1}^N U_k$ , the variance is given by:

$$V(x) = \sum_{k=1}^N V(U_k) \quad (1)$$

For a function of the form  $x = \prod_{k=1}^N U_k$ , the variance is given by:

$$V(x) = \prod_{i=1}^N \{ [E(U_i)]^2 + V(U_i) \} - \prod_{i=1}^N [E(U_i)]^2 \quad (2)$$

where  $E( )$  represents the expected value and  $V( )$  represents the variance of a population. The sample mean and the sample variance can be used as estimators of the population expected value and variance; Equations (1) and (2) are exact for these estimators as they are for the population moments. These equations are applied as appropriate to annual estimates of point and area source emissions as well as to the temporal, spatial, and speciation disaggregation to determine the overall variance of the estimates.

#### Review of Previous Work

Several previous projects have addressed the problem of assigning, in statistical terms, quantitative values to the errors in emission data. Final reports from the following projects were reviewed.

1. Weighted Sensitivity Analysis of Emissions Data, project conducted by IBM Corporation under contract to the Office of Air Quality Planning and Standards (OAQPS), EPA, July 1983.
2. Source Inventory and Emission Factor Analysis (SIEFA), project conducted by PEDCo - Environmental Specialists, Inc. under contract to OAQPS, September 1974.
3. Emissions Inventory for the SURE Region, project conducted by GCA Corporation under contract to the Electric Power Research Institute (EPRI), April 1981.
4. Emissions, Costs, and Engineering Assessment, Work Group 3B, U.S./Canada Memorandum of Intent (MOI) on Transboundary Air Pollution, June 1982.

5. Preliminary Evaluation of Acidic Deposition Assessment Uncertainties, project conducted by Argonne National Laboratory (ANL) under contract to the U.S. Department of Energy, November 1982 (preliminary report).

All these projects have based their calculations on the statistical formulas for error propagation as derived for "small" values of the errors; i.e., the Taylor series expansion included only the first derivatives of the function. Some of the error values used in subsequent calculations have been as high as 80 to 90 percent of the mean. For this size errors, the exact equations derived in this project represent more accurate solutions.

#### Acquisition of Required Data

Information currently available in the NAPAP emission inventory includes data on yearly emissions of point sources at the individual source level and on yearly emissions of area sources at the county/category level. Parameters used to calculate emission values include emission factors, fuel, process or activity rates, ash or sulfur content of fuels (if appropriate), and efficiency of control equipment for the appropriate point sources or category adjustment factors for area sources.

The development of the variance values for emission estimates or for the parameters needed for the calculation of the emission values is not within the scope of this project. These values are to be developed by subsequent projects.

Spatial, temporal, and speciation calculations are scheduled to be implemented as add-on systems to the basic NAPAP data. Disaggregation factors that were developed for other emission inventory projects are being studied and adapted for use in the NAPAP inventory. Estimation of the variances of all disaggregation factors is being included as part of the output from these projects.

#### Implementation

The final task of this project addresses the design and implementation of the basic framework of computer software needed to calculate uncertainties associated with yearly emission values for both point and area sources. The conceptual design is independent of the software system used to support the NAPAP inventory; the design and implementation of the software modules will allow portability between computer systems and will be as independent as possible of the current NAPAP inventory software system.

Uncertainty values associated with each parameter used in calculating emission values are expected to be applicable for whole "classes" of these parameters rather than just to individual sources. To avoid needless repetition of data (with the associated increase in storage costs and the ever present danger at update times), a file with uncertainty "profiles" will be designed. This file will contain all the variance information needed to calculate the uncertainty value associated with the yearly emissions of a point source or an area source category. Each uncertainty profile will be identified by a unique code; at retrieval time the appropriate uncertainty profiles can be referenced to allow the calculation of uncertainty values associated with the reported emissions.

The current version of the NAPAP emission inventory has been implemented based on the Emission Inventory System (EIS) on the EPA IBM computer at Research Triangle Park, NC. Only currently unused space in EIS data files will be used for needed pointers; all the data needed for uncertainty calculations will be stored outside EIS. Programs needed to create, maintain, and access the uncertainty files, to maintain the uncertainty pointers and flags in the EIS files, and to calculate uncertainty values are being implemented as modular programs using the standard FORTRAN 77 language.

REVIEW OF APPROACHES TO VOC SPECIATION

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Presented at:

First Annual Acid Deposition Emission Inventory Symposium  
Raleigh, North Carolina  
December 3-4, 1984

## REVIEW OF APPROACHES TO VOC SPECIATION

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### ABSTRACT

This paper presents the results of a study that was conducted to evaluate the various approaches that have been used by atmospheric scientists to speciate VOC emissions. A wide diversity exists in speciation approaches amongst studies completed to date due primarily to the requirements of the atmospheric model being used. Recommendations are that full speciation be encouraged in future program efforts to provide the flexibility and accuracy required to accommodate the more sophisticated models and mechanisms, and to provide data for air toxic assessments.

## REVIEW OF APPROACHES TO VOC SPECIATION

### INTRODUCTION

The Reagan Administration has established an Interagency Task Force to investigate technical and policy issues associated with the formation and mitigation of acid precipitation. One of the concerns of this task force is the emission of volatile organic compounds (VOC), which are thought to participate in atmospheric reactions with nitrogen and sulfur compounds resulting in the formation of acid precipitation. VOC's are also known to contribute substantially to photochemical reactions that produce ozone.

Although wide agreement exists on the importance of VOC in these reactions, opinions differ markedly on the approach to speciation for VOC. Researchers have performed modeling and monitoring studies using data bases with different degrees of speciation. The objective of this paper is to identify and review these studies and approaches, and to provide the Department of Energy (DOE) with preliminary recommendations on the type of approach or approaches to be used for future VOC programs.

### DISCUSSION

The purposes of this study are to (1) identify the various approaches to VOC speciation used currently by atmospheric scientists, (2) describe the rationale for each speciation approach, and (3) evaluate what degree of speciation will be required for future photochemical and acid precipitation modeling and monitoring programs.

To fulfill the first two objectives, six studies were examined in this paper including:

- the Northeast Regional Study (NEROS and NECRMP),
- the Houston Area Oxidant Study (HAOS),



- the EPRI/ERT VOC emissions inventory data base,
- Harris County (Houston, TX) VOC emissions inventory,
- Harris County Monitoring Study, and
- a modeling study supporting a Louisiana State implementation plan for ozone.

These studies focused primarily on photochemical modeling to establish the dependence of ozone formation on VOC emissions. Although these studies do not directly address acid precipitation formation, they were selected for review because models and mechanisms specific to acid precipitation formation and VOC are in the early stages of development. Further, many of the VOC speciation issues are common to those encountered for ozone formation.

The better known chemical mechanisms included in atmospheric models were also reviewed. These mechanisms include:

- Dodge;
- Demerjian;
- California Institute of Technology (CIT);
- Carbon Bond III (CB III);
- Atkinson, Lloyd, and Wines; and
- National Center for Atmospheric Research (NCAR).

These mechanisms were reviewed to evaluate the different speciation requirements of each.

Review of the six studies and the six chemical mechanisms revealed that a great diversity exists in the requirements for speciation in the current ozone and acid precipitation models and mechanisms. Table 1 summarizes each study, the speciation scheme, and the rationale for selecting this scheme. With respect to the chemical mechanisms, the speciation requirements vary from two specific VOC species and one VOC class in the simplest mechanism to 12 VOC species and classes in the most sophisticated mechanism. The VOC species and classes used in each chemical mechanism are selected by a modeler to reflect his understanding of the atmospheric chemistry involved.

Table 1. SUMMARY OF VOC SPECIATION APPROACHES AND RATIONALE

Study	VOC Species Classes	Rationale
NEROS	Approximately 150 individual species	Dictated by the number of individual species contained in the speciation profiles from each point and area source
HAOS	Detailed speciation grouped into seven classes	Driven by photochemical model requirements
EPRI/ERT	10 species classes	Driven by input requirements for the Atkinson, Lloyd, Wines chemical mechanism
Harris County Emissions Inventory	133 individual species	Dictated by the number of individual species contained in the speciation profiles for each source
Harris County Ambient Monitoring Program	38 individual species monitored continuously  100 individual species in C <sub>2</sub> -C <sub>10</sub> monitored discretely	Limits of analytical techniques and cost
Louisiana SIP Modeling Study	Detailed speciation grouped into five classes	Driven by input requirements for the Carbon Bond III chemical mechanism

Notes: NEROS - Northeast Regional Oxidant Study  
 HAOS - Houston Area Oxidant Study  
 EPRI/ERT - Electric Power Research Institute/Environmental Research and Technology, Inc. data base

A second factor that has influenced VOC speciation approaches is the development of more specific analytical methods. In the past, some classes of VOC (e.g., paraffins, olefins, aromatics) could be directly measured by colorimetric methods. These are no longer widely used, however, since more accurate techniques that analyze specific VOC species are now available. Such techniques include gas chromatography with flame ionization/photoionization detectors or gas chromatography with mass spectrometry confirmation. As a result, the input requirements for a photochemical or acid precipitation model are satisfied by grouping the individual VOC species into the necessary reactivity classes. Thus, for most modeling studies, full speciation usually precedes rather than follows the development of reactivity classes.

Where the regional VOC species data are unavailable to photochemical modelers, government agencies, or others interested in VOC emissions the source most widely used to estimate these data is the VOC Species Data Manual published by the EPA and developed by KVB, Inc. This manual contains speciation profiles for a large number of VOC-emitting source types. If the mass of total hydrocarbon is available for a source, the relative fractions of individual VOC species (or reactivity classes) are given by the speciation profile. It should be noted, however, that although the manual is the best available source for speciation data, it has several broad data gaps as well as inaccuracies that result from a lack of actual sampling data. In many cases, no sampling data at all are available; in these instances, literature values and engineering judgment are used, leading to inherent uncertainties in the speciation profile.

## CONCLUSIONS

Based on the results of this study, full speciation should be pursued for future VOC monitoring and modeling programs. This preliminary recommendation is based on four factors.

1. Currently available analytical methods suggest individual speciation over the older, less accurate methods for directly measuring VOC classes. Data from the older methods on such classes would likely satisfy input requirements for only the simplest chemical mechanisms.

2. The broad diversity in the requirements for speciation in the current set of ozone and acid precipitation mechanisms strongly suggests that full speciation be used. Identifying exact species gives the flexibility of being able to use the data with a variety of mechanisms.
3. As ozone and acid precipitation models evolve, the need for full speciation will increase to accommodate the more sophisticated models and mechanisms.
4. Although not directly associated with acid precipitation, toxic air pollutant assessments are possible if full speciation is utilized in ambient air monitoring and source sampling programs.

These conclusions are corroborated by the increasing trend toward full speciation in most studies performed by most atmospheric scientists and modelers.

It should also be noted that the number of VOC species that are actually measured in a monitoring program will be constrained by the available budget and the ultimate use of the data. Therefore, the thousands of VOC species that could potentially be measured must be prioritized into a scheme that is consistent with the available budget and the technical requirements of the program.

NAPAP EMISSION INVENTORY DEVELOPMENT FOR FY-85/86

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Presented at:

First Annual Acid Deposition Emission Inventory Symposium

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NAPAP EMISSION INVENTORY DEVELOPMENT FOR FY-85/86

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ABSTRACT

The major inventory development activities of the National Acid Precipitation Assessment Program Task Group B for FY-85/86 are designed to fulfill the emission data base requirements for the development of an Eulerian acid deposition model. A data handling system will be developed to provide the necessary spatial, temporal, and species resolution of the inventory. Planned quality assurance activities will focus on establishing a QA expert team for review of the inventory development. Emission factor developments will include VOC allocation factors and an assessment of the use of continuous emission monitoring data for  $\text{SO}_2$  and  $\text{NO}_x$  as a measure of comparison with the temporal allocation algorithms integrated in the inventory data system.

## NAPAP EMISSION INVENTORY DEVELOPMENT FOR FY-85/86

### INTRODUCTION

In 1980, Congress established the National Acid Precipitation Assessment Program (NAPAP) to coordinate and expand research relevant to the problems posed by acid deposition in and around the United States. The program is organized and managed through the Interagency Task Force on Acid Precipitation (ITFAP) and 10 subordinate task groups coordinating specific technical areas of research. One of these 10 groups is Task Group B, Man-Made Sources, chaired by a representative of the Office of Fossil Energy, Department of Energy. The task group includes members representing the Environmental Protection Agency, the Tennessee Valley Authority, and the National Laboratory Consortium. The major objectives of Task Group B are:

1. Provide an accurate and complete inventory of emissions from man-made sources believed to be important in acid deposition processes. The inventories are to be provided with adequate geographic, temporal, and sectoral resolution.
2. Provide models which predict how acidic and acid-precursor emissions may be altered by factors such as economic growth, fuel supply, emissions regulations, and control techniques. These models will have the capability to permit the calculation of alternative control strategies.

The specific objectives of the Task Group B emission inventory implementation program are:

1. Support the 1985 assessment to be performed by the ITFAP.
2. Provide the necessary inventory refinements and resolution for the ITFAP 1987 and 1989 assessments.
3. Support the emission inventory needs for the Eulerian model development and validation programs.

## DISCUSSION

The primary focus of FY-85/86 Inventory activities is directed to fulfill the emissions data base requirements for the development of an Eulerian acid deposition model. Within the EPA's Office of Research and Development, the Environmental Sciences Research Laboratory has been assigned the lead responsibility to develop an Eulerian model for acid rain which will be based on a framework similar to the Northeast Regional Oxidant Study (NEROS) Eulerian oxidant model. The National Center for Atmospheric Research (NCAR) has been assigned the task of developing the model framework which includes a number of chemical transformation modules. The variety of these modules under development dictates the specific chemical species required in the emission inventory to drive the model input. Development of the Eulerian acid deposition model began in FY-83, with preliminary testing to begin in FY-85.

A major element of the emission inventory structure is the data handling systems supporting the required temporal, spatial, and species resolution for the NAPAP Eulerian acid deposition model. Since the temporal and spatial resolution requirements of the acid deposition model are similar to those of the Northeast Corridor Regional Modeling Project (NECRMP), the data handling system used in NECRMP, termed the Regional Model Data Handling System (RMDHS), was adapted for use with the preliminary 1980 inventory.

In order to meet FY-85 and FY-86 NAPAP emission inventory requirements, substantial modifications to RMDHS would be needed, including:

- VOC species resolution into specific chemical classes,
- Additional inorganic pollutant species,
- Derivation of alkaline dust emissions,
- Integration of Canadian emission inventory, and
- Integration of natural source emission inventory.

Table 1 is a summary of planned tasks for the development of acid deposition emission inventories for atmospheric modeling and historical emission profiles during the FY-85 and FY-86 timeframe. In parentheses after each task is a program code identifier consistent with the FY-86 Interagency Budget Proposal for Task Group B.



Table 1. ACID DEPOSITION EMISSION INVENTORY TASKS

- 
1. NAPAP emission inventory for 1980 and 1984 (or 1985) base years.
    - A. Develop spatial/temporal allocation system (B1-19b).
    - B. Generate inventory report and improve software (B1-19a).
    - C. Develop and apply 1984 SO<sub>2</sub>/NO<sub>x</sub> allocation factors (B1-13a).
    - D. Integrate Canadian and natural source inventories (B1-16).
    - E. Update annual data base (B1-31).
  2. Quality assurance and inventory review/certification.
    - A. Document QA/QC of SO<sub>2</sub>/NO<sub>x</sub> inventory (B1-18a).
    - B. Third party review and certify inventory (B1-18b).
    - C. State review 1984 SO<sub>2</sub>/NO<sub>x</sub> inventory (B1-18c).
    - D. QA-expert team review inventory (B1-20).
  3. Emission factor development and formulation.
    - A. Develop and apply VOC allocation factors (B1-14).
    - B. Develop allocation factors for SO<sub>4</sub>, NH<sub>3</sub>, and alkaline dust emissions (B1-15).
    - C. Conduct emission factor tests (B1-22).
    - D. Develop fuel use/emitter characteristics (B1-21).
    - E. Analyze existing CEM data (B1-13b).
  4. Historical emissions inventories.
    - A. Increase accuracy of SO<sub>2</sub>/NO<sub>x</sub> inventory (B1-17a).
    - B. Develop special historic inventories (B1-17b).
    - C. Develop monthly SO<sub>2</sub>/NO<sub>x</sub> emission data base (B1-23).
-

The inventory development program is also directly supportive of certain activities of Task Group 1, "Assessments and Policy Analysis." The inventory needs for assessment purposes are generally less-detailed and require fewer resources to develop, but are nonetheless extremely important to the overall NAPAP program. Assessment projects use more aggregated emission information in combination with models or other analytic techniques to evaluate the relative importance of various pollutants, regions, types of sources, etc., to the acid deposition problem. Results of these studies have implications for planning additional research needed and development of potential control/mitigation strategies.

During FY-85, the 1980 NAPAP emission inventory will undergo numerous improvements. Major inventory activities will focus on the development of a data handling system (IA, IB, and IC) which will overcome the limitations of RNDHS. Figure 1 illustrates the interrelationships of the various elements of the data handling system to produce the required temporal and spatial disaggregation for the FY-80 NAPAP acid deposition emission inventory of 23 pollutant species. Planned quality assurance activities will focus on establishing a QA-expert team for review of the inventory development.

Emission factor development activities will be directed at VOC allocation factors with field measurement/verification tests to be conducted as appropriate. A program task will also be initiated to examine the availability and quality of existing continuous emission monitoring (CEM) data for  $\text{SO}_2$  and  $\text{NO}_x$  as a measure of comparison with the temporal allocation algorithms integrated in the inventory data system. Historical emission inventory activities will include improvement to the current inventory produced in FY-84 as well as the development of a monthly  $\text{SO}_2/\text{NO}_x$  emission data base.

Figure 2 depicts the interrelationships of FY-86 activities designed to produce the 1984 or 1985 NAPAP emission inventory. The present approach calls for a 1984 base year of record for development of the inventory. However, an assessment is currently being prepared which indicates that a calendar 1985 base year may be more appropriate for the inventory by allowing greater lead time for implementation of a state review plan. The major activity in the inventory development will be

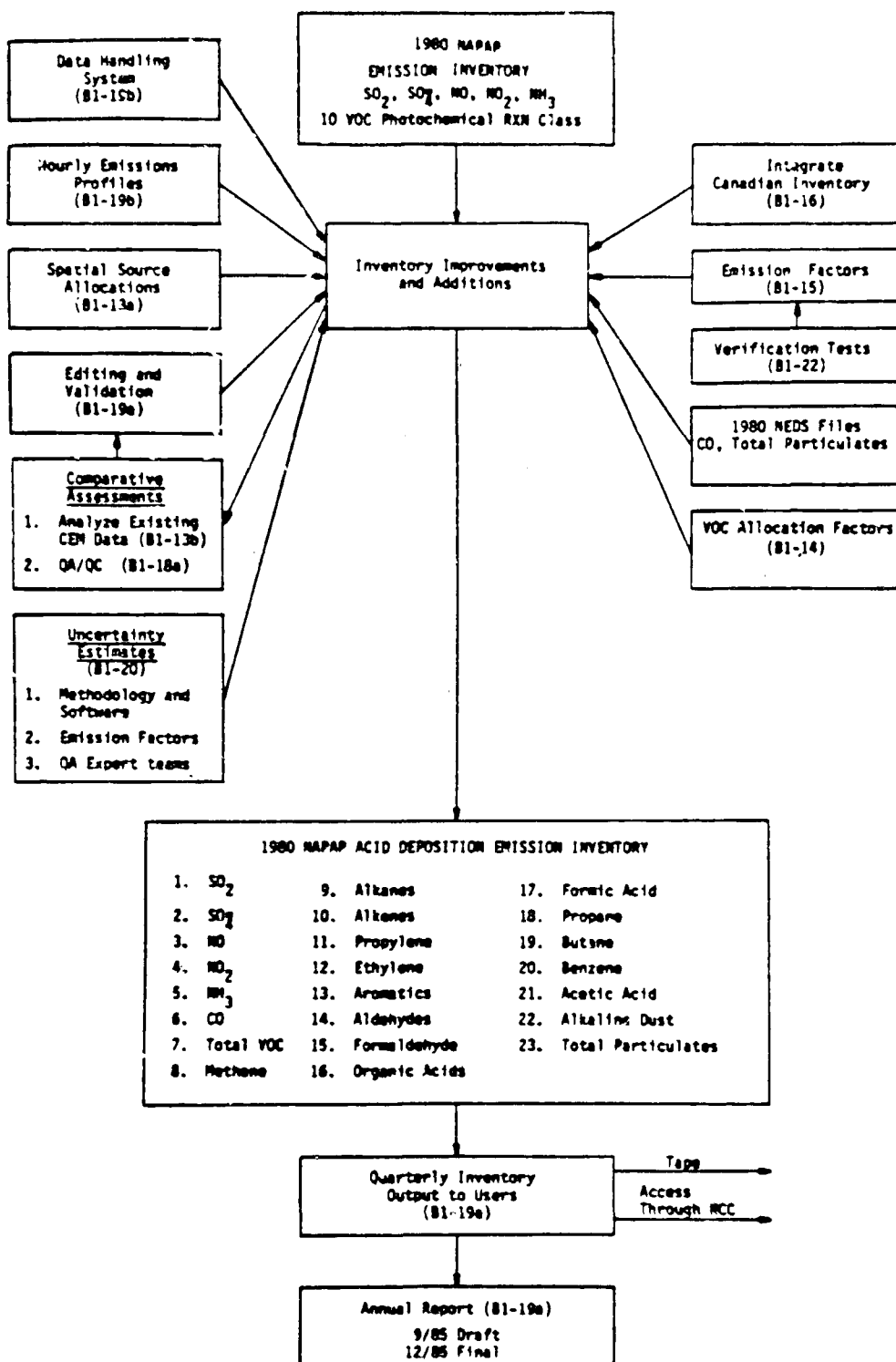


Figure 1. FY-85 activities in developing the final 1980 NAPAP emission inventory.

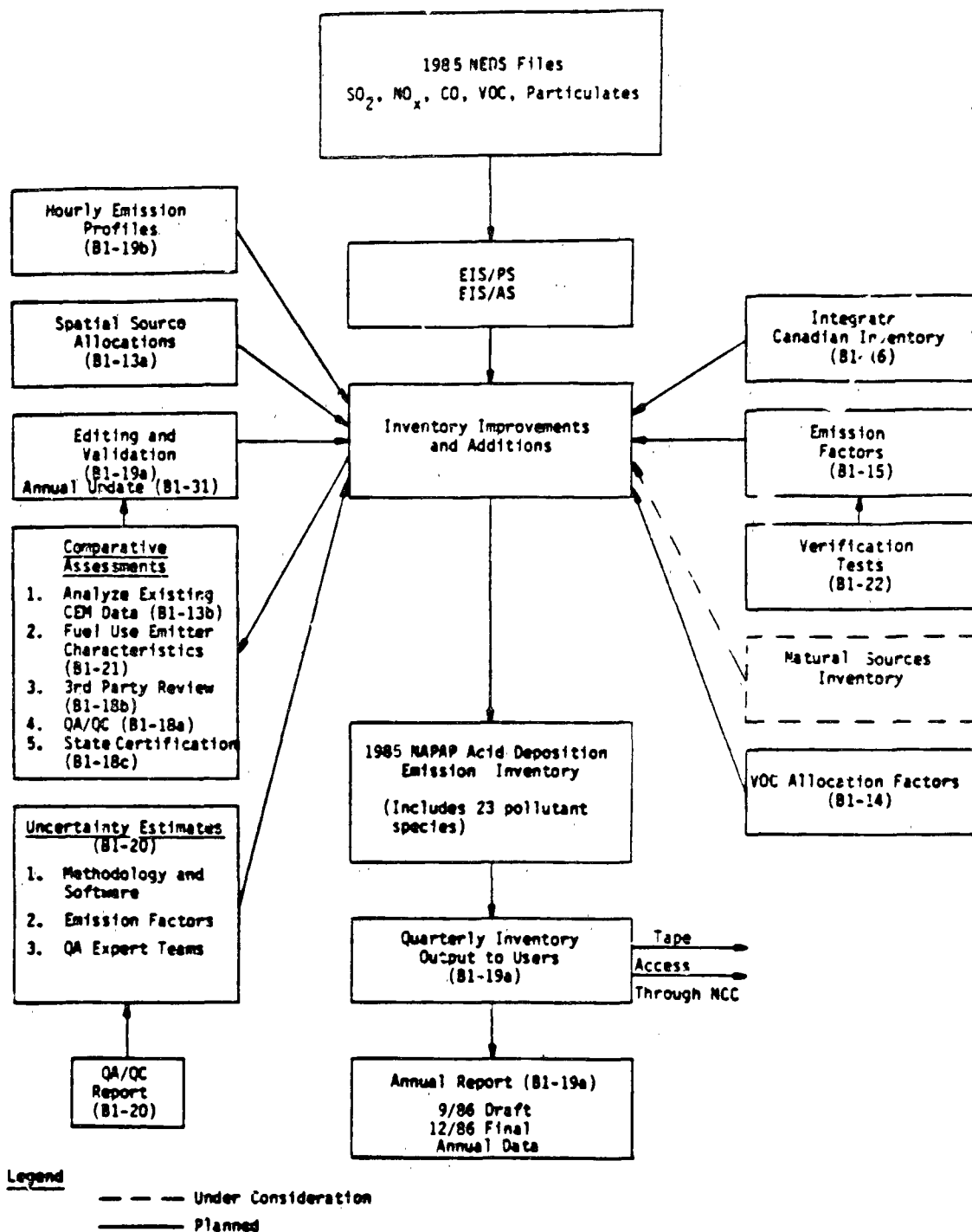


Figure 2. FY-86 activities in developing the 1985 NAPAP emission inventory.

the formulation of detailed allocation factors for  $\text{SO}_2$ ,  $\text{NO}_x$ , and all VOC species required for the Eulerian model. Intensive QA/QC of the Inventory will be provided through both third party review of the data base and specific state review of the  $\text{SO}_2/\text{NO}_x$  Inventory components. In addition, the CEM data evaluation will be intensified in FY-86 pending assessment of the results from the initial FY-85 program applied to the 1980 NAPAP emission Inventory.

SESSION 3: APPLICATION OF EMISSION INVENTORIES FOR SCIENTIFIC PURPOSES

Chairman: Ed Trexler  
U.S. Department of Energy  
Office of Planning and Environment  
Mail Stop FE-13, Room B-120  
Washington, D.C. 20545

SULFUR DEPOSITION MODELING WITH THE NAPAP EMISSION INVENTORY

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Presented at:

First Annual Acid Deposition Emission Inventory Symposium  
Raleigh, North Carolina  
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## SULFUR DEPOSITION MODELING WITH THE NAPAP EMISSION INVENTORY

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### ABSTRACT

Version 2.0 of the 1980 NAPAP SO<sub>2</sub> emission inventory has been selected for use in the International Sulfur Deposition Model Evaluation (ISDME). Appropriate emission data from this inventory have been processed to create model input files to be used directly by Lagrangian/Eulerian ISDME regional-scale sulfur deposition models. This paper identifies the NAPAP emissions inventory elements useful to these models and summarizes the emissions data processing procedures.



## SULFUR DEPOSITION MODELING WITH THE NAPAP EMISSION INVENTORY

### INTRODUCTION

The Environmental Protection Agency and Environment Canada are coordinating the International Sulfur Deposition Model Evaluation (ISDME) as an extension of an earlier model evaluation effort.<sup>1</sup> One goal of the current study is to assess model performance and credibility via statistical evaluation of the 1980 seasonal and annual model results from as many as 15 Lagrangian and two Eulerian regional-scale sulfur deposition models.<sup>2</sup>

The evaluation year of 1980 was selected on the basis of the significant increase in the number of operational precipitation chemistry monitoring sites in eastern North America during that year and the availability of suitable U.S. and Canadian SO<sub>2</sub> emission inventories for that year.<sup>3,4</sup>

Before any air pollution model can be applied, point and area source emissions data from an appropriate inventory must be aggregated or processed, usually via a model "preprocessor," to create a more compatible input data file. In an evaluation study involving a group of models, it is essential that one processing algorithm be used to avoid extraneous differences between model results. Therefore, a uniform processing algorithm was developed and applied by Benkovitz<sup>5</sup> to create four sets of seasonal and annual emission grids, any one of which would be compatible with the ISDME models.

#### NAPAP Emission Data Processing

Essentially, ISDME data processing involved extracting appropriate inventory elements (Table 1), apportioning SO<sub>2</sub> emissions to grid cells, and creating model input data grids with configuration required by the ISDME models. Spatial resolution of these grid configurations ranged from 70 to 127 km.

Table 1. ELEMENTS OF THE NAPAP EMISSION INVENTORY  
APPROPRIATE TO REGIONAL-SCALE SULFUR DEPOSITION MODELING

Element Name	Reason For Need
State Code	Source Identification
County Code	Source Identification
Plant Code	Source Identification
NEDS Point Code	Source Identification
Date of Record	Uncertainty Assessment
SCC Code	Emission Apportionment
SIC Code	Emission Apportionment
Point Source Location	Emission Apportionment
Percentage Annual Throughput	Emission Apportionment
Normal Operating Rate	Emission Apportionment
Annual SO <sub>2</sub> Point Source Emission Rate	Emission Apportionment
Annual SO <sub>2</sub> Area Source Emission Rate	Emission Apportionment
Points With Common Stack	Emission Apportionment

For point sources, as a means of data quality assurance, the inventory source coordinates were screened to determine if indeed the source was located within the designated county. When the coordinates were missing or the inventory source coordinates were located outside a quadrangle whose perimeter was at least  $\frac{1}{2}$  degree latitude/longitude from any point along the county border, the county centroid coordinates were substituted as a correction. For some states, county centroids were used for over half the point sources (e.g., Iowa, Kansas, Michigan, Nebraska, North Carolina, South Carolina, and Wisconsin). These point source location adjustments have been incorporated into subsequent versions of the NAPAP inventory by Engineering Science.

After the source location was established, an annually averaged effective stack height was computed for each point source using the inventory stack parameters and climatological atmospheric stability data. Frequently, the set of stack parameters was incomplete so national average stack parameters for the appropriate SIC were substituted.

Next, the seasonal SO<sub>2</sub> emission rates were determined from percentage annual throughput data and allocated to the proper layer (0-200, 201-500, and above 500 m) of the grid cells containing the source. Seasons were defined thusly: winter December, January, and February, and so forth.

Area source emissions, which were available only on a county basis, were apportioned to grid cells according to the apportionment of 1970 county population based on U.S. Census Bureau data. Emissions from area sources were assumed to be injected in the 0-200 m layer

Upon completion of the point and area source emissions data processing, a quality assurance program was implemented. As a result, for each grid cell, the sum of the seasonal emissions equalled the annual emissions. In addition, the distribution of ISDME annual emissions compared very well with that of the MOI emission inventory <sup>1,4</sup>

## CONCLUSIONS

As a result of the ISDME emission data processing effort, Lagrangian and Eulerian sulfur deposition modelers have at their disposal 1980 seasonal and annual 3-layer SO<sub>2</sub> point source and 1-layer SO<sub>2</sub> area source emissions grids for North America. Four different grid configurations, ranging in resolution from 70 to 127 km, are available.

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EMISSION INVENTORY APPLICATIONS TO  
REGIONAL ACID DEPOSITION MODELING

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EMISSION INVENTORY APPLICATIONS TO  
REGIONAL ACID DEPOSITION MODELING

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ABSTRACT

A comprehensive Regional Acid Deposition Model (RADM) is being developed and a simpler fast-turn-around "engineering" model(s) (EM) is being designed by the National Center for Atmospheric Research as part of the National Acid Precipitation Assessment Program (NAPAP). This paper describes how potential assessment applications and research questions affect those model designs and the subsequent emission inventory requirements. Finally, a recommendation is made for the EPA to consolidate inventory development efforts to produce an agency emission inventory applicable to the wide spectrum of models and applications throughout the EPA.

## EMISSION INVENTORY APPLICATIONS TO REGIONAL ACID DEPOSITION MODELING

### INTRODUCTION

The scope and complexities of the acid rain issue in the United States present modelers with conflicting requirements. The models are expected to be accurate and scientifically credible as well as easy to use for assessment purposes. Task Group C of NAPAP Atmospheric Processes is responsible for coordinating the model development and research efforts. The primary model development activities are being performed at the National Center for Atmospheric Research (NCAR) in Boulder, Colorado. These activities include the development of a comprehensive Regional Acid Deposition Model (RADM) and a simpler fast-turn-around "engineering" model(s). These models have great potential to provide information needed to achieve many of the major goals of NAPAP.<sup>1</sup>

- understand the basic chemical and physical processes related to acid deposition,
- assess the severity of acid deposition effects,
- determine the causes of acid deposition,
- assess the relative importance of different causes of acid deposition,
- assess the effects of man's activities on acid deposition, and
- determine effective control strategies.

### DISCUSSION

The purpose of this discussion is to present an overview of the current modeling approach with special emphasis on emission inventory requirements. The scientific community agrees that a rigorous treatment of processes relevant to acid deposition is essential for credible

results. Thus, the RADM will include a comprehensive description of all the major physical and chemical processes currently known to effect acid deposition. However, incorporation of this level of detail on a regional scale extends the model execution time beyond what is considered reasonable to address the policy and assessment questions; these require calculation of long term averages with sufficient speed to permit extensive experimentation with alternative solution strategies.

The EM, therefore, is being specifically designed to address the policy and assessment applications. This model will incorporate condensed representations of several simple nonlinear physical and chemical processes contained in the RADM and consequently will require a relatively short execution time. The credibility of the EM can, at a minimum, be evaluated by comparison with the more accurate RADM results.

Recent reviews<sup>1,2</sup> have shown that a three dimensional Eulerian framework is the most feasible general approach for both RADM and EM. This decision has significant impact on spatial resolution requirements of emissions. The current RADM domain covers the entire continental United States and southern Canada. Theoretical considerations suggest a  $20 \times 20 \text{ km}^2$  grid resolution, however, due to computer time and space limitations, a compromise of  $80 \times 80 \text{ km}^2$  grid spacing was chosen. A larger grid size may give substantial inaccuracies. Major point source emitters and many urban areas, nevertheless, must be treated individually to determine the impact of specific sources and to correct for the effect of chemical and physical phenomena on a subgrid scale. Within a grid cell, inhomogeneities may exist in emissions, chemical reaction types and rates, deposition, and transport phenomena. Development of an independent module to calculate correction factors for each grid due to these inhomogeneities is being planned. Thus, a  $20 \times 20 \text{ km}^2$  grid resolution for area emissions is required to adequately parameterize the effect of subgrid scale emission patterns. The RADM has up to 15 vertical layers and can incorporate emissions into each of these layers. Most models assume emissions at the surface with instantaneous mixing aloft. Multiple levels of emissions should have a definite accuracy advantage, especially near the source.

The RADM, because of its size and complexity, is typically executed in an episodic mode for a 2 to 3 day period. Annual or long term averages can be approximated by the weighted average of representative episodic model results. The RADM typically requires hourly emissions for these episodic applications. A representative weekday/weekend diurnal pattern for each season is adequate to produce the required temporal variation of emissions.<sup>3</sup>

A comprehensive chemical reaction scheme detailed enough to handle nonlinearities resulting from sulfur, nitrogen oxide and ozone chemistry, and cloud chemistry in both the gaseous and aqueous phases is necessary to achieve many of the major objectives of NAPAP. An emissions inventory is being developed which is capable of addressing the above issues on the required temporal and spatial time scales. It will include:

- major acid precursors ( $\text{SO}_2$ ,  $\text{NO}_x$ );
- sulfate aerosols because of their contribution to the total sulfur budget;
- volatile organic compounds and carbon monoxide because of their importance in the oxidation process;
- reactive hydrocarbons and nitrogen oxides because of their role in oxidant production;
- direct sources of acid species such as organic acids, and hydrochloric and hydrofluoric acid whose vapors are incorporated into clouds and precipitation;
- coarse aerosols ( $>2.5 \mu\text{m}$ ) and particles important to acid formation;
- ammonia for its role as a buffering agent in cloud chemistry;
- other buffering aerosol species such as alkaline dust;
- aerosol components key to catalytic conversion in aqueous reactions (soot, iron, manganese);
- naturally emitted precursors to acid rain such as reduced sulfur species; and
- other naturally emitted species related to acid or oxidant formation.



In contrast, emission requirements for the EM are less stringent. The model domain remains the same as RADM, however, grid resolution may be reduced. The EM will have only three to five vertical levels and each specific version would treat only one family of chemical species. Parameterization of transformation and deposition processes would include simplifications of the nonlinear chemistry based on knowledge gained from RADM executions and observation data.<sup>2</sup> Therefore, emission species requirements and, in fact, all other emission requirements, are easily satisfied by the RADM emissions data set.

## CONCLUSIONS

The goals of the NAPAP are too broad to rely on a singular solution. This brief overview of the atmospheric modeling approach illustrates the conflicting demands of the research and assessment applications and their implications for emissions inventory development. The inventory developers must provide an emissions data base at a level of resolution that meets the most stringent requirements, and yet be flexible enough to transform those base emissions into appropriate spatial, temporal, and species resolutions to satisfy the full spectrum of needs of the scientific and assessment/policy communities. They must also be able to provide estimates of uncertainty associated with the emissions.

Finally, acid rain is only one of the many air pollution problems facing the EPA today. Modeling assessment of other problems such as ozone, sulfate, visibility, particulate, etc. have many of the same basic emission inventory requirements even though differences do exist. I recommend that the EPA integrate emission inventory development activities to provide an agency inventory that will satisfy the various emission requirements of current and anticipated modeling and assessment programs.

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THE USE OF EMISSION INVENTORIES  
FOR EFFECTS STUDIES

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Presented at:

First Annual Acid Deposition Emission Inventory Symposium  
Raleigh, North Carolina  
December 3-4, 1984

## THE USE OF EMISSION INVENTORIES FOR EFFECTS STUDIES

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### ABSTRACT

Understanding effects of atmospheric deposition on terrestrial and aquatic ecosystems requires that we understand how systems are responding presently when compared to past response. Effects should be coupled to deposition wherever possible; however, deposition data are incomplete both spatially and temporally. Attempts to identify, for example, changes overtime in deposition of  $\text{SO}_4^{2-}$  and/or  $\text{H}^+$  ions are confounded by differing methods of collecting rainfall (e.g., bulk vs. wet-only, event vs. weekly or monthly, etc.) and techniques for measuring analysis of precipitation chemistry. The lack of evidence for a strong nonlinearity in the relationship between emissions and deposition in eastern North America suggests that nonlinearity is probably not significant for annual average deposition in eastern North America (National Research Council, 1983). When used with appropriate consideration for the uncertainties involved, this conclusion provides a powerful research tool as the emissions data base is much more complete than either historical or regional deposition data bases. The relationship of  $\text{SO}_2$  emissions on a State-wide or regional basis to changing tree ring chemistry and water quality is discussed as examples of the potential role of emissions inventories in ecosystem assessment.

## THE USE OF EMISSION INVENTORIES FOR EFFECTS STUDIES

### INTRODUCTION

Evaluation of emissions on a State-wide, regional, and national level is a major research activity for determining trends and patterns of acidic deposition. Direct effects of emissions on ecosystems are not as much a concern within current research programs as the transport and transformation of emissions and the subsequent deposition of substances on terrestrial and aquatic ecosystems. However, deposition data bases are incomplete with regard to various components, specifically dry deposition and metals. If a consistent relationship between emissions and deposition can be defined, then researchers can use emissions data as a surrogate for deposition to describe spatial and temporal trends in ecosystem response. The remainder of this paper will discuss specific examples of ecosystem research where emissions inventories are appropriate.

### DISCUSSION

#### Trends in Aquatic Chemistry

Acid deposition effects were first brought to the attention of the scientific community when Adirondack lake waters were reported to have decreased in pH to levels deleterious to fish. As research in aquatic ecosystems has expanded, we are not only trying to measure responses to present deposition levels but also evaluating current responses in terms of historical levels of deposition. Changes in lake or stream chemistry reflect the amount of modification imposed on deposition by the terrestrial ecosystem. Changes in deposition quality may be immediately reflected by changes in aquatic chemistry (direct response), or the effects observed currently may be the result of long term inputs of acidic or acidifying compounds (delayed response). The chemical characteristics of very few systems are known prior to 1975/1980. Thus,

trend analyses of lake and stream chemistry remain a dominant theme in aquatic ecosystem research both from the standpoint of evaluating the historical record and being able to predict responses at changing deposition rates.

As historical deposition data for many regions of concern are not available, the relationship between trends in emissions and changes in surface water chemistry is of particular interest. Two recent reports have evaluated the relationship between these data: 1) Hendrey, et al. (1983) developed the ACID data base to examine regional response, and 2) Smith and Alexander used the USGS benchmark stations coupled with State-wide SO<sub>2</sub> emissions for site-specific trends in stream chemistry. The USGS sites have an advantage over ACID sites because sampling and analytical methods have been consistent over the 10-15 year record, data are quality assured, and the watersheds have experienced little or no changes in land use. Thus, although Smith and Alexander (1983) had data for fewer sites (47 in the U.S.), the data and trend analyses for any given site are probably more reliable.

A comparison of emissions, deposition, and water quality trends for several southern States is presented in Table 1. North Carolina and Virginia stream sulfate concentration follows the emissions pattern; however, inconsistencies in the other States make generalizations difficult. Predicting future response in other aquatic systems may be possible as long as land use history is considered. Because of the many potentially confounding factors, detection of trends from atmospheric pollution is best accomplished through consideration of streams with negligible upstream disturbance.

#### Forest Response

Recent concern over the impact of atmospheric deposition on forested ecosystems has increased as reports of forest decline from Germany and from the Appalachian Mountains of the U.S. increase. Causal mechanisms for forest decline have not been identified although many hypotheses have been proposed. Because trees are long-lived and respond to many environmental changes, recent declines must be put in a historical context in order to evaluate the role of anthropogenic pollutants.

Table 1. Comparison of trends in S emissions, S deposition, and sulfate concentrations in surface waters.

State	SO <sub>2</sub> - S Emissions Density <sup>a</sup> (MT-S/km <sup>2</sup> )							Sulfur Deposition (MT-S/km <sup>2</sup> )						Sulfate Trend in Surface Waters		
	1955	1960	1965	1970	1975	1980	% increase	1953 <sup>b</sup>	1954 <sup>b</sup>	1955 <sup>b</sup>	1963 <sup>c</sup>	1979 <sup>d</sup>	1980 <sup>d</sup>	1981 <sup>d</sup>	Hendrey et al.	Smith and Alexander
NC	0.89	0.84	1.10	1.80	1.77	1.84	107	0.56	0.36	0.71	0.78	0.86 <sup>e</sup>	0.79 <sup>e</sup>	0.68 <sup>e</sup>	NS	+
VA	1.3 <sup>f</sup>	1.47	1.76	1.98	1.48	1.24	0		1.63	2.22	0.88		.88	1.13	++	-
GA	0.48	0.52	0.88	1.14	1.90	2.36	392		0.55	0.88			1.05	1.01	++	NS
KY	2.20	2.60	3.88	5.75	5.80	5.02	128		0.98	1.72					NS	
SC	0.62	0.66	0.72	1.12	1.12	1.66	168	0.53	0.41	1.05			1.04	1.00	++	++
TN	1.98	3.31	3.18	4.24	6.02	4.74	140			1.44			1.55	1.13	+	-

<sup>b</sup> Jordan et al. 1959; bulk deposition measurements<sup>c</sup> Gambell and Fisher 1966; modified bulk deposition measurements, see discussion in text<sup>d</sup> MAOP monitoring sites; wet only measurements<sup>e</sup> data averaged for sites only in eastern North Carolina in order to be comparable to the Gambell and Fisher (1966) monitoring network

+ Positive trend significant at p = 0.05; ++ positive trend significant at p = 0.01

- Negative trend significant at p = 0.05

NS Not significant

Tree response over time can be assessed through an analysis of long-term growth patterns by measuring annual ring widths. Changes in this dendrochronologic record can be statistically analyzed and compared to changes in stand history, climate, and air pollution. However, this comparison assumes an adequate record exists. While long-term climatology data are available for correlation, the same is not true for atmospheric deposition. Several studies are using available emissions data as an indicator of local or regional air quality. Assuming that certain metals can be used as tracers of emissions, trends in the concentration of metals in tree rings can be used to track air quality conditions. Baes and McLaughlin (1984) have shown a good correlation between  $\text{SO}_2$  emissions and Fe content in tree rings (Figure 1). Similar trends for metals mobilized by acidification (e.g., aluminum) may aid in our understanding of forest response to acid deposition on a regional scale where local source emissions are not implicated.



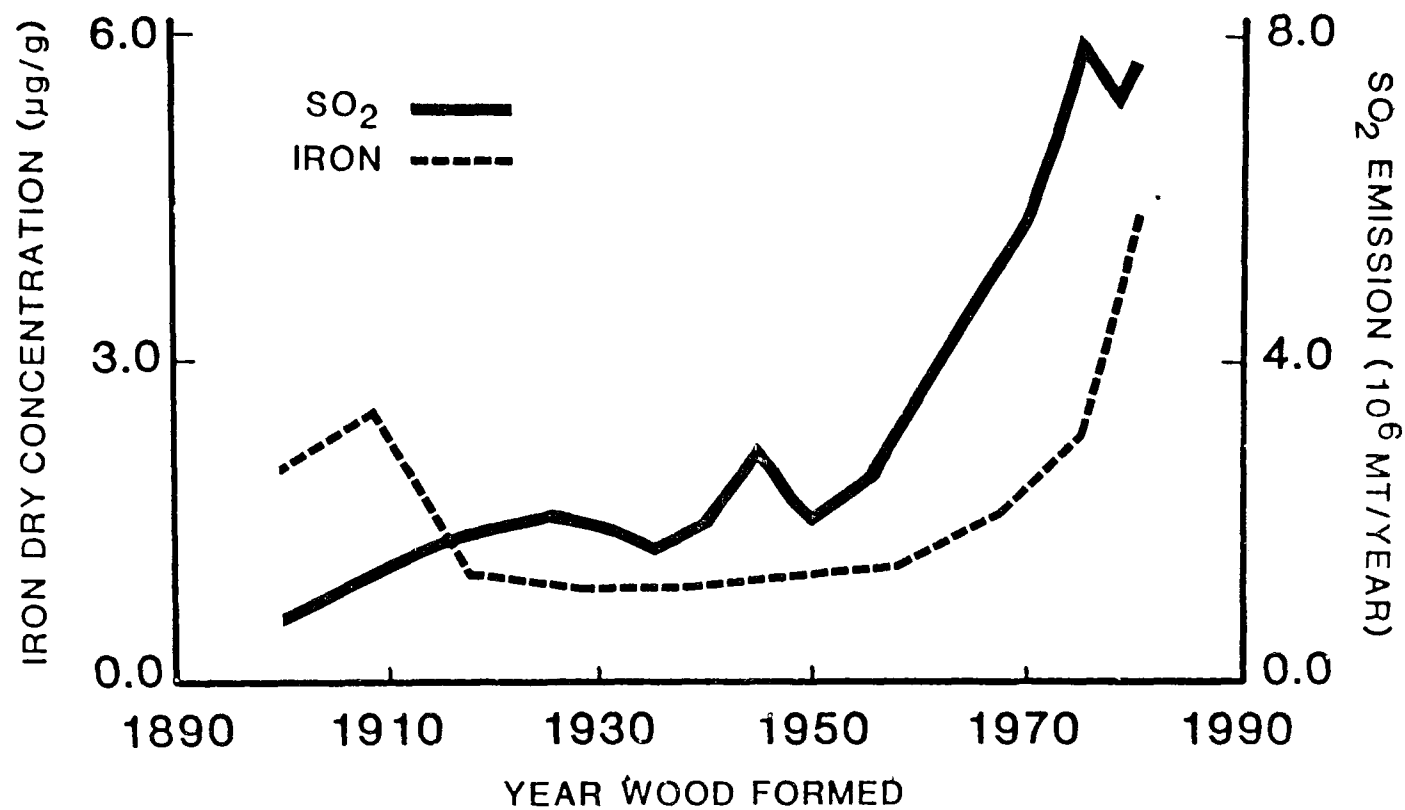


Figure 1. Iron concentration in short-leaf pine tree rings at Cades Cove, Tennessee and estimated SO<sub>2</sub> emissions from within 900 km of Cades Cove (adapted from Baes and McLaughlin 1984)

EMISSION INVENTORY REQUIREMENTS IN SUPPORT OF ACID  
DEPOSITION ASSESSMENTS AND POLICY DEVELOPMENT

by

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SESSION 4: RELATED EMISSION INVENTORY DEVELOPMENT ACTIVITIES

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DEVELOPMENT OF  
THE CANADIAN ACID DEPOSITION EMISSION INVENTORY

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Presented at:

First Annual Acid Deposition Emission Inventory Symposium  
Raleigh, N.C.

December 3-4, 1984

## ABSTRACT

An overview of Canadian activities to develop emission inventories for the acid deposition program is presented. Initial efforts emphasized the development of emission inventories of acid-causing pollutants to assess the importance of emission sources in the long range transport of air pollutants. The information obtained was also used in simple atmospheric models simulating the transport, transformation and deposition, predominantly of  $\text{SO}_2$ . Currently Canada is developing: a) comprehensive inventories of defined temporal and spatial resolution to input to more complex models that predict acid deposition and oxidant concentrations; b) an emission forecasting model incorporating all major emission sources. We are also maintaining a current inventory on the major sources of  $\text{SO}_2$ .

## INTRODUCTION

The principle objective of the Canadian Long Range Transport of Air Pollutants (LRTAP) program is to obtain reductions in emissions contributing to the long range transport of air pollutants in order to reduce environmental loadings to levels which ecosystems can tolerate. In support of this objective, considerable Canadian effort has been put into establishing emission inventories. Such inventories are needed:

- (1) as input to atmospheric models to establish the relationships between emissions and ambient concentrations and deposition rates;
- (2) to assess the relative importance of emission sources for various pollutants; and,
- (3) in order to project the impact of changes in emissions of existing sources, coupled with contributions that new sources will be making.

The discussion to follow will centre largely on the emission inventory activities that support atmospheric modelling.

## DISCUSSION

Environment Canada has carried out emission inventories of the common pollutants every two years in cooperation with the provincial agencies since 1970. This includes a detailed emission inventory for SO<sub>2</sub> used as input to the atmospheric models that were examined under the United States-Canada Memorandum of Intent on Transboundary Air Pollution. The information compiled for point sources includes geographical location, SIC/SCC codes, stack parameter data, process data, emission factors, control equipment efficiencies, and annual emission values. Area sources can be resolved spatially to a 127 km

grid and the file includes information on SIC codes, base quantity data, emission factors and annual emission values. The information is maintained in the National Emissions Inventory System.

These inventories are being maintained and expanded to include other pollutants relevant to both acid deposition and oxidants to support the development and testing of an Eulerian atmospheric model in Canada. (The province of Ontario, Environment Canada and the Federal Republic of Germany are co-sponsoring the development of this model.) The pollutant species inventoried for the base year 1980 include  $\text{SO}_2$ ,  $\text{SO}_4$ ,  $\text{NO}_x$ , speciated VOC's,  $\text{NH}_3$  and particulate matter (alkaline fraction identified separately). Inventories quantifying the contribution from natural sources of these pollutant species have been compiled based on generally sparse literature data and consequently can only be considered as order of magnitude estimates.

Testing and validation of the Eulerian model necessitates that emission inventories be resolved spatially to a 127 km grid and temporally to provide diurnal, weekday and weekend variations in emissions. The model will initially be tested against field observations taken during August 1980. Work is currently underway in cooperation with provincial agencies to develop three-hourly profiles for the major point sources in Eastern Canada of  $\text{SO}_2$ ,  $\text{NO}_x$ , VOC's and TSP for that time period; typical diurnal, weekday, weekend and seasonal emission profiles are being developed for area sources for the base year 1980. Standard procedures for reporting uncertainties in these inventories will also be examined. This work is scheduled to be completed by March 1985. The development of a 1984 baseline inventory to support the analysis of abatement strategies will begin in FY 85/86 and is scheduled for completion in FY 86/87.

In addition to the inventory activities supporting the modelling program, other inventory projects are also being done. Future emissions may increase due to the installation of new sources or an increase in industrial production or they may decrease due to a lagging economy. It is important that

these changes be estimated as they may affect concentrations and depositions. Environment Canada is currently developing an emission forecasting model which accounts for the major emission sources. The work is being done in collaboration with Statistics Canada who, through their Socio-Economic Resource Framework (SERF) model, can provide the economic forecasts to drive the emissions database at Environment Canada. This work is scheduled to be completed in FY 85/86. Canada is committed to decreasing Eastern Canadian emissions to 2300 kt SO<sub>2</sub> by 1994. On-going periodic emission inventories of the major sources of SO<sub>2</sub> are being maintained in order to monitor our progress in achieving this commitment.

### CONCLUSION

The Canadian emission inventory activities in support of the LRTAP program have been summarized. A substantial part of the current and future effort will be devoted to supplying the data needs of the Eulerian model. Once this model is operational, the processes that govern the atmospheric transport, transformation and deposition of pollutants should be better understood. In this way the Eulerian model can be used to improve the simpler LRT models that are used to generate annual and seasonal source-receptor relationships and which are more practical for assessing abatement strategy options.



EMISSION INVENTORY REQUIREMENTS  
FOR DEPOSITION AND REGIONAL AIR QUALITY  
MODEL DEVELOPMENT: A SUMMARY

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Presented at:

First Annual Acid Deposition Emission Inventory Symposium  
Raleigh, North Carolina  
December 3-4, 1984

EMISSION INVENTORY REQUIREMENTS  
FOR DEPOSITION AND REGIONAL AIR QUALITY  
MODEL DEVELOPMENT: A SUMMARY

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ABSTRACT

Although several regional and national emissions inventories have been prepared, none of them fulfill all of the requirements for deposition and air quality modeling. They are limited with regard to contemporaneity of data, pollutants included, spatial and temporal resolution, or data reliability. In order to try to overcome those limitations, a project is underway to develop procedures to produce inventories suitable for modeling air quality and deposition in the contiguous United States and Canada during 1982. The inventory includes total emitted particulate matter, alkaline particulate matter, primary sulfate, sulfur dioxide, nitric oxide, nitrogen dioxide, ammonia, and hydrocarbons classified by photochemical reactivity. The nominal spatial resolution is 100 meters for point sources and one-quarter degree longitude by one-sixth degree latitude for area sources. Average emission rates during each of the eight 3-hour time periods of a day have been estimated for weekdays and weekends in each season. Traceability of the data to their origin has been maintained during the development to enable differences between the inventory and other inventories to be resolved. This traceability also allows the inventory to be updated to other time periods without major new developmental efforts.

EMISSION INVENTORY REQUIREMENTS  
FOR DEPOSITION AND REGIONAL AIR QUALITY  
MODEL DEVELOPMENT: A SUMMARY

INTRODUCTION

Precise, accurate emission inventories are of great importance in effectively evaluating air quality control strategies. Source-oriented models use pollutant emission rates and atmospheric dispersion characteristics to calculate source contributions at receptors. The precision and accuracy of those calculations can be no better than those of the emission information.

Regional and national emission inventories have been prepared for many parts of the United States and Canada. The most comprehensive inventories include the National Emissions Data System (NEEDS), and inventories developed for the Sulfate Regional Experiment (SURE), the Multistate Atmospheric Power Production Pollution Study (MAP3S), the Northeast Corridor Regional Modeling Program (NECRMP), The Memorandum of Intent on Transboundary Air Pollution (MOI), and the National Acid Precipitation Assessment Program (NAPAP). None of these existing inventories completely fulfill all of the needs for regional deposition and air quality modeling. As recognized by their compilers, each is limited with respect to contemporaneity of the data, chemical substances included, spatial resolution, temporal resolution, or data reliability.

To improve upon the situation, the Electric Power Research Institute (EPRI) is sponsoring a project to develop and apply procedures for the compilation of emissions inventories for 1982 for the contiguous United States and southeastern Canada.

## DISCUSSION

The inventory compilation procedures for the project are intended to satisfy several major requirements in the following areas:

Materials: Total emitted particulate matter, primary sulfate, alkaline particulate matter, sulfur dioxide, nitric oxide, nitrogen dioxide, ammonia, and hydrocarbons classified according to photochemical reactivity.

Time Period: The most recently available point source inventories are being obtained from NEDS and State agencies. Because some data in those inventories are older than 1982, State agencies and emitting facilities are being contacted to obtain more recent data for 200 major point sources of nitrogen and sulfur oxide. Approximately two-thirds of the point source data is expected to pertain to 1982. The 1982 NEDS area source inventory is also being used.

Spatial Resolution: Point source locations are nominally resolved to 100 meters. Area source emissions are estimated by county. To provide higher geographic resolution, factors are being developed to allocate county emissions to elements of a one-quarter degree longitude by one-sixth degree latitude grid system. The factors are based upon the fraction of county geographic area that falls in each grid element. Also included are data needed to calculate point source emission injection height (stack height, diameter, flow rate, and temperature). The nominal resolution for stack height is 1 foot.

Temporal Resolution: NEDS and State point source inventories contain annual emission rates. Seasonal, weekly, and daily operating schedules in those inventories are used to calculate average diurnal emissions (eight 3-hour time periods) for weekdays and weekends during each season. The NEDS area source inventory does not include operating schedules. Therefore, operating schedules for each area source category are being developed to calculate emissions with the same resolution as point sources. For example, 1982 heating degree days in each county during each season are used to calculate average seasonal emission rates for residential fuel use.

Missing Values: Data for some point sources in the NEDS and State inventories are not complete. Facilities and State agencies are being contacted to obtain most missing data for 200 plants. When possible, typical values are being inserted for missing location (city or county centroid), exhaust flow rate (based upon operating rate and engineering practice), exhaust temperature (based upon type of process and emission control device), control device removal efficiency (based upon type of control device), and fuel sulfur and ash content (based upon type of fuel) for other point sources.

Erroneous Data: A series of 90 tests is applied to data for each point source emission point to identify suspect values. The tests compare values to normal ranges and check the consistency among different quantities. The values tested include exhaust parameters, locations, control efficiencies, operating rates, and reported emission rates. Most suspect data are being corrected for 200 major plants through contacts with State agencies and plant operators. Test results are recorded in the inventory to evaluate data accuracy.

Data Precision: Point source emission rate precisions are being estimated by applying propagation-of-errors methods to the calculations and measurements used to determine them.

Data Source Documentation: The origin of each value is recorded in the inventory. When a value is changed, the origin of the new value is recorded, and a record of the change is automatically produced. Because this information is maintained, comparisons can be made with other inventories, and differences resulting from different sources of information can be resolved.

Data Retention: All information used to compile the inventory has been retained in it. This allows it to be improved in two ways. Firstly, data older than 1982 can be identified and replaced with 1982 data as they become available. Secondly, procedures used to calculate various quantities in the inventory, such as diurnal emissions from annual emissions, can be changed when better procedures are developed. The revised procedures can then be applied to the data to recalculate those quantities.

## CONCLUSIONS

A 1982 emissions inventory is being developed to satisfy most of the requirements for regional modeling. It is an improvement over currently available regional inventories because of the number and types of materials included, its geographic coverage, its spatial and temporal resolution, concurrency of the data, reduction in the number of missing values, and the corrections that have been made to suspect values. All data in the inventory are traceable to their origins, which enables comparison with other inventories and resolution of differences. All data used in the compilation are retained in the inventory, which allows improvement by substitution of 1982 data for older data. Procedures to calculate various quantities in the inventory can also be revised, as better procedures are developed, and applied to the data to calculate more reliable values.

Acknowledgement: This work is sponsored by Electric Power Research Institute Contract RP1630-23.

DEVELOPMENT OF THE NATURAL SOURCES EMISSIONS INVENTORY

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Presented at:

First Annual Acid Deposition Emission Inventory Symposium  
Raleigh, North Carolina  
December 3-4, 1984

## DEVELOPMENT OF THE NATURAL SOURCES EMISSIONS INVENTORY

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### ABSTRACT

Both natural and man-made sources influence precipitation acidity. The former have been considered generally to be small, but the uncertainties in these natural source strength estimates are very large. The research of the Natural Sources Task Group is addressing these uncertainties by new instrument development, technique intercomparison, and additional measurements. As a result of these efforts, a large part of the existing body of data on biogenic sulfur emissions must be revised upward. New measurements in the summer of 1985 will test the laboratory-determined correction factors. Lightning appears to be a negligible source of nitrate to the northeast, based on an estimate of the global lightning-produced nitrate deposition patterns that have been established by data from remote areas. Airborne dust from unpaved roads appears to be a significant source of neutralizing material and hence needs to be better quantified. Further work is also required before the roles of biogenic nitrogen and ammonia in acid precipitation can be considered to be established.

1



## DEVELOPMENT OF THE NATURAL SOURCES EMISSIONS INVENTORY

### INTRODUCTION

It is well known that both natural and man-made sources release chemical species that can modify acidic deposition. If a controlled reduction in man-made emissions were to occur, natural sources will continue to contribute to acidic deposition. As a result, the benefits that would be anticipated from such a controlled reduction could be erroneously optimistic if natural sources make a significant contribution to the present total acidic budget. Therefore, the research of the Natural Sources Task Group of the National Acid Precipitation Assessment Program is focused on one key policy question:

Are the natural emissions of acid precursors and alkaline materials significant relative to man-made emissions?

The natural emissions that are most relevant to precipitation acidity are those of sulfur, nitrogen, and alkaline materials. These sources have several general characteristics that shape the approach required to define their significance. First, there is great variety: e.g., terrestrial and oceanic biogenic activity, lightning, and airborne soil and water aerosols. Second, the sources can have large spatial extent and temporal variations. Third, while broadly distributed, the fluxes are generally low at a given site, thereby requiring state-of-the-art measurements. Consequently, the research projects of the Natural Sources Task Group have involved survey searches to identify the major sources, rigorous laboratory and field assessments of the reliability of existing data sets, instrument and technique development, and additional field measurements.

## DISCUSSION

Since the source identification, measurement technique and reliability, and the extent of existing data vary considerably for the various species, the approach has been different for each.

### Sulfur

Natural sulfur emissions from terrestrial and tidal areas have been studied rather extensively by several investigators, most notably in the SURE study in the U.S. southeast. The Task Group research focuses on an assessment of the reliability of that data set. Current improved standards, preconcentration techniques, and sampling methods are being compared to those used in the SURE study and are resulting in an upper revision of those earlier flux values. These revisions will be checked by side-by-side measurements in the southeast in the summer of 1985.

The influx of sulfur species from oceanic sources is being examined in two ways. First, direct measurements of the production of sulfur-containing species have been made in nutrient-rich oceanic areas. These provide estimates of the source potential. Secondly, the influx of airborne sulfur species into the southeast from the Gulf is being examined directly. The first study was in the summer of 1984 and the next will be in 1985.

### Nitrogen

The major sources of natural nitrogen compounds appear to be biogenic emissions from soils and production by lightning, both of which are only crudely known, largely due to lack of measurement methods. Thus, the Task Group has focused on the development of sensitive instruments and flux-measurement techniques applicable to biogenic nitrogen emissions. The field trials of two methods - box and gradient - will occur in the summer of 1985. For lightning, the approach has been to develop an estimate that is independent of the current one, which has relied on estimates of production per stroke and stroke frequency. The Task Group has examined the deposition of nitrate in extremely remote oceanic areas, where lightning is likely to be the only source, and has determined a global lightning-produced nitrate deposition pattern. For the U.S., this independent estimate supports the earlier assessment that this source is a minor contribution to east-coast nitrate deposition.

### Alkaline Aerosols

So little is known about the potential of alkaline aerosols to neutralize acidity in precipitation that the present Task Group activity has been to try to better define the problem. For example, an assessment has been made of the potential contribution of dust from unpaved roads to airborne alkalinity. The data suggest that such material, as well as that from tilled fields, is a significant regional source and hence must be better quantified. Lastly, ammonia emissions are being addressed by the Task Group. With the initial goal of identifying major sources, ammonia measurement techniques are being developed and tested.

### CONCLUSIONS

Biogenic sulfur emissions are likely to be larger than originally thought, based on intercomparisons of new techniques with those used in the earlier investigations. Further lab tests and field measurements in 1985 will establish the revised emission values. Ocean-to-land influx of sulfur compounds appears to be small, but a recheck is scheduled in 1985. Lightning contributes negligibly to northeastern nitrate. Biogenic nitrogen emissions remain poorly quantified and an assessment must await additional field measurements. Dust from unpaved roads is a significant source of airborne alkaline materials and deserved better quantification.

SESSION 5: PANEL DISCUSSIONS

Moderator: John Fink, Chief  
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## PANEL DISCUSSIONS

### EMISSION INVENTORY APPLICATIONS

Joan Novak, EPA

### EMISSION FACTOR DEVELOPMENT

Jim Homolya, Radlan Corporation

### HISTORICAL EMISSION INVENTORIES

Gerhard Gschwandtner, Pacific Environmental Services

### QUALITY ASSURANCE AND ESTIMATION OF UNCERTAINTIES

Carmen Benkovitz, Brookhaven National Laboratory

APPENDIX  
ATTENDEES

1ST ANNUAL ACID DEPOSITION EMISSIONS INVENTORY SYMPOSIUM

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