

**EVALUATION OF SULFATE AEROSOL OPTICAL DEPTHS OVER THE NORTH ATLANTIC AND COMPARISON WITH SATELLITE OBSERVATIONS**

**C. M. Berkovitz and S. J. Ghan  
Pacific Northwest Laboratory  
P.O. Box 999  
Richland, WA 99352**

and

**C. M. Berkovitz, R. Wagener, S. Nemesure, and S. E. Schwartz  
Environmental Chemistry Division  
Brookhaven National Laboratory  
Upton, NY 11973-5000**

November 1993

**Submitted for inclusion in  
Proceedings of the American Meteorological Society  
74th Annual Meeting,  
Nashville, TN  
Jan. 23-28, 1994**

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

**This research was performed under the auspices of the U.S. Department of Energy under Contract No. DE-AC02-76CH00016.**

**MASTER**

**DISCLAIMER**

# EVALUATION OF SULFATE AEROSOL OPTICAL DEPTHS OVER THE NORTH ATLANTIC AND COMPARISON WITH SATELLITE OBSERVATIONS

CARL M. BERKOWITZ\*, STEVEN J. GHAN  
Pacific Northwest Laboratory<sup>†</sup>  
Richland, Washington

CARMEN M. BENKOVITZ, RICHARD WAGENER, SETH NEMESURE  
AND STEPHEN E. SCHWARTZ  
Brookhaven National Laboratory<sup>‡</sup>  
Upton, New York

## 1 INTRODUCTION

It has been postulated that scattering of sunlight by aerosols can significantly reduce the amount of solar energy absorbed by the climate system. Ball and Robinson (1982) have estimated that the reduction in the annual average insolation could be as much as 7% over the eastern United States. Others (Bolin and Charlson, 1976; Charlson *et al.*, 1992; IPCC, 1990) have used climatological values of aerosol mass loading, cloud cover and albedo to suggest that radiative cooling associated with the scattering of solar radiation by anthropogenic aerosols is comparable to the radiative warming associated with absorption of terrestrial radiation by radiatively active gases (the "greenhouse" effect).

Aerosol measurement programs alone cannot provide all the information needed to evaluate the radiative forcing due to anthropogenic aerosols. Surface-based aerosol monitoring programs do not provide global coverage from which to evaluate these postulated effects. Although satellites also provide global aerosol measurements (Durkee *et al.*, 1991), estimates of scattering by aerosols are restricted to the marine atmosphere and many questions related to source attribution and anthropogenic vs. natural contributions cannot be addressed. Thus, comprehensive global-scale aerosol models, properly validated against surface-based and satellite measurements, are a fundamental tool for evaluating the impacts of aerosols on the planetary radiation balance.

Models with various degrees of detail have been used to evaluate the radiative forcing. Charlson *et al.* (1992) used a back-of-the-envelope approach.

\* Corresponding author address: MS K6-08, Pacific Northwest Laboratory, P. O. Box 999, Richland, Wa., 99352

<sup>†</sup>Pacific Northwest Laboratory is operated for the U.S. Department of Energy by Battelle Memorial Institute under Contract DE-AC06-76RLO 1830

<sup>‡</sup>Brookhaven National Laboratory is operated for the U. S. Department of Energy by Associated Universities, Inc. under Contract No. DE-AC02-76CH00016.

Charlson *et al.* (1991) used a three-dimensional meteorological/chemical model to evaluate the global distribution of aerosol forcing. Using a statistical description for removal by precipitation, monthly mean winds and fixed deposition velocities, they estimated peak aerosol optical depths over North America of  $\tau_a \approx 0.12$  at visible wavelengths. Geographical distributions of aerosols agreed well with earlier measurements of turbidity. They estimated that the uncertainty of their calculations of global radiative forcing by aerosols was a factor of two. Kiehl and Briegleb (1993) found, by accounting for the variability of optical depth with wavelength, an aerosol radiative forcing that is a factor of two smaller than that of Charlson *et al.* ( $0.3$  vs.  $0.6 \text{ W m}^{-2}$ ).

Confidence bounds for models describing the spatial, temporal and physicochemical variation of aerosols must be evaluated before the models can be used for either scientific or policy related purposes. Here we present preliminary results of such an evaluation.

## 2 APPROACH

Analyzed meteorological fields from the European Centre for Medium-Range Weather Forecasts (ECMWF) are used to drive a modified version of the PNL Global Chemistry Model (Luecken *et al.*, 1991) applied to the atmospheric sulfur cycle. The resulting sulfate fields are used to calculate aerosol optical depths, which in turn are compared to estimates of aerosol optical depth based on satellite observations from the NOAA-9 Advanced Very High Resolution Radiometer (AVHRR) (Wagener *et al.*, this meeting). Here we describe the sulfur cycle model, the calculation of optical depth from the simulated sulfate, and the calculation of the "observed" optical depth from satellite observations.

## Chemistry/Transport Model

Details of the Global Chemistry Model (GChM) are presented elsewhere (Luecken *et al.*, 1991; Benkovitz *et al.*, this meeting). Only an overview of the model is presented here.

GChM is a three-dimensional Eulerian kinematic code with an explicit description of cloud and precipitation scavenging. Previous applications have used meteorological fields from a general circulation model. A refinement to this approach was necessary to allow a comparison of satellite observations of optical depth at specific locations and times. The meteorological fields were derived from the 6-hour forecast model of the ECMWF. In the present application, we report on results using meteorological analysis prior to and coincident with the times of AVHRR radiometric observations.

The distribution of OH is based on results from a photochemical model using meteorological conditions obtained from the Goddard Institute for Space Studies General Circulation Model (Spivakovsky *et al.*, 1990). The new GChM calculations use 24-h average OH fields that allow us to include the influence of length of daylight and solar zenith angle on oxidant concentrations. Values of the rate constant of SO<sub>2</sub> oxidation by OH were modified to be a function of temperature and pressure based on NASA (1991).

A refinement was also made to the treatment of dimethylsulfide (DMS) oxidation by allowing the formation of either methanesulfonic acid (MSA) or SO<sub>2</sub>, based on the DMS chemistry described by Yin *et al.* (1990a; 1990b). The fractional yield of SO<sub>2</sub> and MSA from DMS oxidation is based on expressions given in Hynes *et al.* (1986). SO<sub>2</sub> can subsequently oxidize to SO<sub>4</sub> aerosol or dry deposit.

The deposition velocity for SO<sub>2</sub> and sulfate aerosol is calculated using a time- and space-dependent formulation of Walcek *et al.* (1986) and Wesely (1989). Input data for these calculations are meteorological fields from the ECMWF, global albedo data from Matthews (1985) and land use classifications derived from Wilson and Henderson-Sellers (1985).

U.S. and Canadian emissions data were based on the National Acid Precipitation Assessment Program's 1985 emissions inventory (NAPAP; Saeger *et al.*, 1989). European sources were obtained from the inventory of the Co-Operative Programme for Monitoring and Evaluation of the Long Range Transmission of Air Pollutants in Europe (EMEP; Iversen, 1990) and natural emissions were based on Bates *et al.* (1992).

## Simulated Optical Depth

GChM simulations provide the spatial and temporal variations in the concentration of sulfate aerosol, C<sub>SO<sub>4</sub></sub> (g m<sup>-3</sup>), for October 1986, over a domain encompassing 140°W to 62°E, and from 12°N to 81°N. The vertical range extends to p ≥ 100 hPa. We use a simple algorithm based on Charlson *et al.* (1992) to calculate the aerosol optical depth, τ<sub>a</sub>, as a function of relative humidity, RH, and height, z,

$$\tau_a = \int f(RH) \sigma_{dry} C_{SO_4} dz. \quad (1)$$

where the dry aerosol specific extinction cross section, σ<sub>dry</sub>, is calculated from the Mie theory for pure ammonium sulfate aerosols. The refractive index of ammonium sulfate is 1.53. We assume a particle radius of 0.15 μm and calculate the specific extinction at a wavelength of 0.65 μm (which corresponds to the mean wavelength of the satellite channel used to measure observed optical depth). This calculation yields a value of σ<sub>dry</sub> = 5.3 m<sup>2</sup> g<sup>-1</sup> dry sulfate. The correction term f(RH) is necessary to account for the growth of soluble aerosols through the absorption of water vapor. We have calculated the correction term based on field measurements for relative humidity ranging from 40% to 90% (Charlson *et al.*, 1984). The correction term ranges from unity for RH below 50% to 3 for RH above 90%. Relative humidity is taken from the ECMWF analysis.

## Observed Optical Depth

The observed clear-sky aerosol optical depth is derived from NOAA-9 AVHRR radiance measurements obtained from a polar sun-synchronous orbit that crosses the equator at approximately 14:30 LST. Using the single-scattering approximation, the aerosol optical depth is expressed in terms of the aerosol single-scattering albedo, the aerosol-scattering phase function, and the solar radiation scattered toward the satellite by aerosols. The radiation scattered by aerosols is determined as the difference between the radiance observed by the satellite and the radiation scattered by the surface and by air molecules, with the latter parameterized using the classical theories of Fresnel and Rayleigh. The method applies only to clear sky (low optical depth) scenes over the oceans (known surface reflectance and spatial uniformity). Data are subject to a number of checks to assure suitability, including tests for the presence of high (optically thin) clouds and sunglint, both of which would corrupt the calculation of the aerosol optical depth. Details are provided by Wagener *et al.* (this meeting).

### 3 RESULTS

Figure 1 shows the observed and simulated aerosol optical depth for October 23, 1986. Points that satisfy none of the criteria for measuring optical depth are unshaded in both plots. Thus, all land is unshaded, and much of the oceans is unshaded because of obscuration by cloud cover. Several large patches remain where optical depth can be measured for this particular date.

The largest observed optical depths are in the western North Atlantic, offshore from the industrial sulfur sources in the eastern United States. Optical depths approach unity in a few pixels. Observed optical depths are substantially lower across most of the North Atlantic Ocean, with values of typically 0.1. However, much larger values (exceeding 0.3) are observed near the west coast of Africa, presumably due to westward transport of dust from the Sahara (Rao *et al.*, 1989).

The simulated optical depth exhibits a similar pattern to the observations, with the largest values again offshore from the east coast of North America. Values approach 0.7 in a few pixels. The simulated optical depth decreases to about 0.05 in the central and eastern North Atlantic, with lower values in the subtropics than in midlatitudes. Because the model treats only sulfur species, it does not simulate the high optical loading observed off the west coast of Africa. The simulated optical depth appears to be generally lower than that observed.

The spatial structure of the bias can be assessed by examining the ratio of the simulated optical depth to that observed. The bottom panel of Figure 1 shows the  $\log_{10}$  of the ratio. A log scale is used because it provides a uniform resolution of positive and negative fractional errors.

The simulated optical depth is somewhat greater than the observed loading over several patches in the western North Atlantic. It is somewhat lower in nearby pixels, so that the errors in local magnitude might simply be due to small errors in position. The bias in the large patches near the Azores and adjacent to western Europe is rather uniform, with the simulated optical loading about 50-70% of that observed. The bias is stronger in the subtropics, both immediately west and far to the west of Africa, where the simulated optical depth is roughly 20% of that observed.

To provide more comprehensive spatial coverage in our evaluation, we have collected values at the first points during the period Oct 23-30 where aerosol optical depth at that point could be measured. Increasing coverage over the model domain

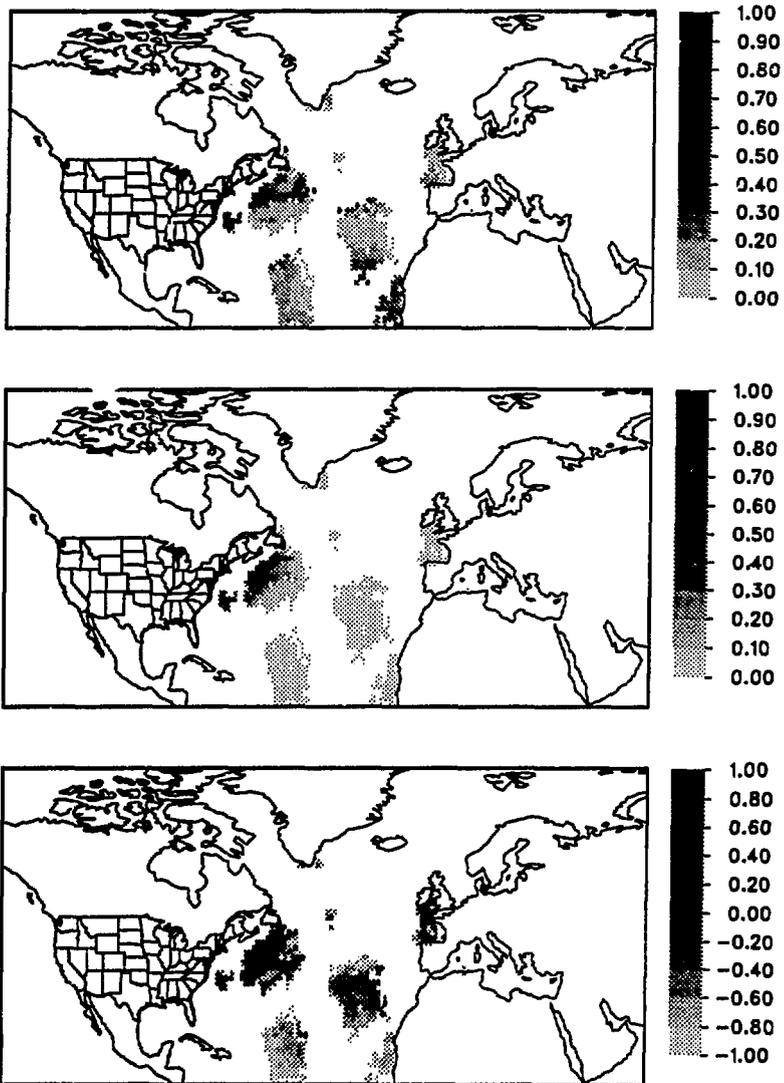
was provided as additional points from each day of satellite observations were used to develop Figure 2. This method of filling in a relatively sparse data set was selected over schemes using time averaging because the latter would mask the time-resolving capability of the model. By presenting the data this way, a comparison between satellite and modeled aerosol optical depth can be done over most of the North Atlantic south of 50°N during this period, with exceptions in the Gulf of Mexico and scattered patches across the central Atlantic, for which there are no satellite observations of aerosol optical depth.

The patterns of optical depth evident in Figure 1 are repeated for the same points in Figure 2. The spatial extent of Saharan dust is more evident in the composite, as is the distribution of aerosol in the western North Atlantic. The simulated aerosol extends farther offshore from the east coast of North America than the observed aerosol, except for a patch to the southeast of Newfoundland that is not that extensive in the simulated values. The observed optical depth exhibits more spatial structure than the simulated values over the central and subtropical Atlantic; the simulated optical depths are systematically too low.

The  $\log_{10}$  of the optical depth ratio (simulated/observed) is also illustrated for the composite analysis of Figure 2. The ratio is spotty, with positive biases adjacent to negative biases in the western Atlantic and in the eastern Atlantic in midlatitudes. We suspect that this structure is due to small offsets in the predicted positions of aerosol mass relative to observed values. Simulated optical depths are generally lower than observed, particularly in the subtropical Atlantic, where typical satellite values are on the order of 0.1 to 0.25, and the modelled values are relatively uniform with values of 0.08.

The ratio of simulated/observed optical depth decreases eastward across most of the central and eastern North Atlantic, reflecting the lower simulated optical loading over this part of the model domain. A possible source of bias is excessive precipitation scavenging in the simulation. The model treats the precipitation rates archived by the ECMWF forecast model every six hours as constant for each six-hour period; a more realistic treatment of the sporadic nature of precipitation would remove less aerosol and hence yield higher simulated aerosol loading. Other factors contributing to the bias could be neglected aerosol types in the simulation (dust, sea salt, carbonaceous aerosol, volcanic aerosol) and neglect of aqueous chemistry in non-precipitating clouds (Dennis *et al.* 1992).

The ratio of simulated/observed optical depth is



**Figure 1: Observed (top) and simulated (middle) optical depth and the  $\log_{10}$  of the ratio simulated/observed (bottom) for October 23, 1986.**

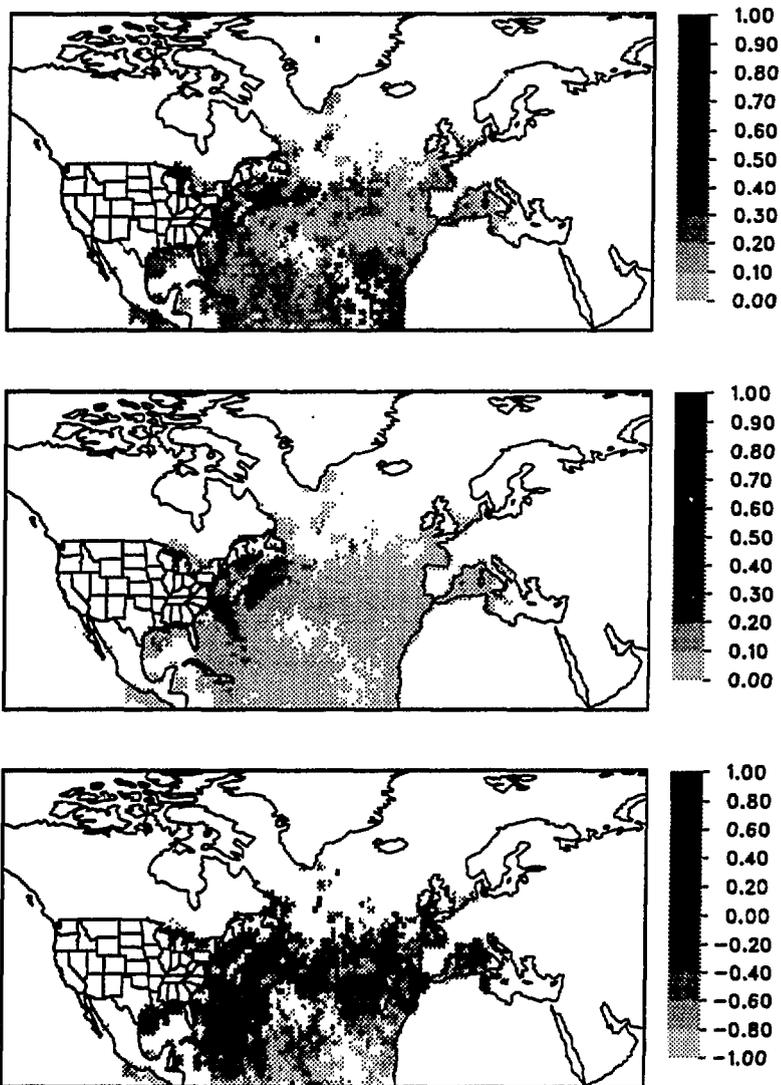


Figure 2: Composite observed (top) and simulated (middle) optical depth and the log<sub>10</sub> of the ratio simulated/observed (bottom) for the period October 23-30, 1986.

lowest in the subtropics, particularly just off the coast of West Africa, where it drops to 0.15. Evidently desert dust, which is not represented in the simulations, is the dominant contributor to optical loading in the subtropical North Atlantic.

#### 4 SUMMARY

Although the results we are presenting must be regarded as preliminary, they demonstrate a general correspondence between simulations and observations in regions contaminated by anthropogenic sulfur aerosol. The biases evident in other regions can be explained in terms of either aerosols not considered in the simulation, or potentially correctable deficiencies in the sulfur model. Thus, we conclude that the approach we have taken is a viable method of evaluating aerosol models. However, we will not consider the evaluation complete until we have addressed the problems identified in this paper.

#### ACKNOWLEDGMENTS

Authors C. M. Berkowitz and S. J. Ghan gratefully acknowledge the financial support provided by the Laboratory Directed Research and Development program at Pacific Northwest Laboratory.

#### REFERENCES

- BALL, T. S. AND G. D. ROBINSON (1982). The Origin of Haze in the Central United States and Its Effect on Solar Irradiation, *J. Appl. Meteorol.* 21, 171-188.
- BATES, T. S., B. K. LAMB, A. GUENTHER, J. DIGNON AND R. E. STOIBER (1992). Sulfur Emissions from Natural Sources, *J. Atmos. Chem.* 14, 315-337.
- BENKOVITZ, C. M., C. M. BERKOWITZ, R. C. EASTER AND S. E. SCHWARTZ (1994). Fine Resolution Atmospheric Sulfate Model Driven By Operational Meteorological Data: Comparison with Observations, *Conference on Atmospheric Chemistry, 74<sup>th</sup> AMS Annual Meeting, 29-28 January 1994, Nashville, Tennessee*. American Meteorological Society.
- BOLIN, B. AND R. J. CHARLSON (1976). On the Role of the Tropospheric Sulfur Cycle in the Shortwave Radiative Climate of the Earth, *Ambio* 5, 47-54.
- CHARLSON, R. J., D. S. COVERT, AND T. V. LARSON (1984). Observation of the Effect of Humidity on Light Scattering by Aerosols. In: *Hygroscopic Aerosols* (eds. L. H. Ruhnke and A. Deepak), A. Deepak Publishing, Hapton, VA. 35-44.
- CHARLSON, R. J., J. LANGNER, H. RODHE, C. B. LEOVY, AND S. G. WARREN (1991). Perturbation of the Northern Hemisphere Radiative Balance by Backscattering from Anthropogenic Sulfate Aerosols, *Tellus 43A-B*, 152-163.
- CHARLSON, R. J., S. E. SCHWARTZ, J. M. HALES, R. D. CESS, J. A. COAKLEY, J. E. HANSEN AND D. J. HOFMANN (1992). Climate Forcing by Anthropogenic Aerosols, *Science* 255, 423-430.
- DENNIS, R. L., J. N. MCHENRY, W. R. BARCHET, F. S. BORKOWSKI AND D. W. BYUN (1992). Correcting RADM's Sulfate Underprediction: Discovery and Correction of Model Errors and Testing the Corrections through Comparisons against Field Data, *Atmos. Environ.* 27A, 975-997.
- DURKEE, P. A., F. PFEIL, E. FROST AND R. SHEMA (1991). Global Analysis of Aerosol Particle Characteristics, *Atmos. Environ.* 25A, 2457-2471.
- HYNES, A. J., P. H. WINE AND D. H. SEMMES (1986). Kinetics and Mechanisms of OH Reactions with Organic Sulfides, *J. Phys. Chem.* 90, 4148-4146.
- INTERGOVERNMENTAL PANEL ON CLIMATE CHANGE (IPCC) 1990. *The IPCC Scientific Assessment*, J. T. Houghton, G. T. J. Jenkins, J. J. Ephraums, Eds. (Cambridge Univ. Press., Cambridge).
- IVERSEN, T., N. E. HALVORSEN, J. SALT BONES AND H. SANDNES (1990). *Calculated Budgets for Airborne Sulphur and Nitrogen in Europe*, EMEP/MSC-W Report 2/90.
- KIEHL, J. T., AND B. P. BRIEGLER 1993. The Relative Roles of Sulfate Aerosols and Greenhouse Gases in Climate Forcing, *Science* 260, 311-314.
- LUECKEN, D. J., C. M. BERKOWITZ AND R. C. EASTER (1991). Use of a Three-Dimensional Cloud-Chemistry Model to Study the Transatlantic Transport of Soluble Sulfur Species, *J. Geophys. Res.* 96, 22477-22490.
- MATHEWS, E. (1985). *Atlas of Archived Vegetation, Land-Use and Seasonal Albedo Data Sets*, Goddard Space Flight Center Institute for Space Studies. Technical Memorandum 86199. New York, N. Y.
- NASA (1991). *Chemical Kinetics and Photochemical Data for Use in Stratospheric Modeling*, JPL Publication 85-37. Jet Propulsion Laboratory. JPL 90-1. Pasadena, CA.

- RAO, C. R. N. , L. L. STOWE, AND E. P. MCCLAIN (1989). Remote Sensing of Aerosols over the Oceans Using AVHRR Data: Theory, Practice, and Applications, *Int. J. Remote Sensing* 10, 743-749.
- SAEGER, M., J. LANGSTAFF, R. WALTERS, L. MODICA, D. ZIMMERMAN, D. FRATT, D. DULLEBA, R. RYAN, J. DEMMY, W. TAX, D. SPRAGUE, D. MUDGETT AND A. S. WERNER (1989). *The 1985 NAPAP Emissions Inventory (Version 2): Development of the Annual Data and Modelers' Tapes*. U. S. Environmental Protection Agency. EPA-600/7-89-012a, Washington, D. C.
- SPIVAKOVSKY, R. Y. C. M., J. A. LOGAN, S. C. WOFSEY, M. B. MCELROY AND M. J. PRATHER (1990). Tropospheric OH in a Three-Dimensional Chemical Tracer Model: an Assessment Based on Observations of  $\text{CH}_3\text{CCl}_3$ , *J. Geophys. Res.* 95, 18441-18471.
- WAGENER, R., S. NEMESURE, C. M. BENKOVITZ, S. E. SCHWARTZ, C. M. BERKOWITZ AND S. J. GHAN (1994). Cloud-free Aerosol Optical Depth Determination over Oceans from Satellite Radiometry, *Conference on Atmospheric Chemistry, 74<sup>th</sup> AMS Annual Meeting, 29-28 January 1994, Nashville, Tennessee*. American Meteorological Society.
- WALCEK, C. J., R. A. BROST, J. S. CHANG AND M. L. WESELY (1986).  $\text{SO}_2$ , Sulfate and  $\text{HNO}_3$  Deposition Velocities Computed Using Regional Landuse and Meteorological Data, *Atmos. Environ.* 20, 949-964.
- WESELY, M. (1989). Parameterization of Surface Resistances to Gaseous Dry Deposition in Regional-Scale Numerical Models, *Atmos. Environ.* 23, 1293-1304.
- WILSON, M. F. AND A. HENDERSON-SELLERS (1985). A Global Archive of Land Cover and Soils Data for Use in General Circulation Models, *J. Climatol.* 5, 119-143.
- YIN, F., D. GROSJEAN, R. C. FLAGAN AND J. H. SEINFELD (1990). Photooxidation of Dimethyl Sulfide and Dimethyl Disulfide II: Mechanism Evaluation, *J. Atmos. Chem.* 11, 365-399.
- YIN, F., D. GROSJEAN AND J. H. SEINFELD (1990). Photooxidation of Dimethyl Sulfide and Dimethyl Disulfide I: Mechanism Development, *J. Atmos. Chem.* 11, 309-364.