HTML ABSTRACT

Modelling and Observations of Aerosol Properties in the Clean and Polluted Marine Boundary Layer and Free Troposphere

Elisabetta Vignati, Frank Raes, Rita Van Dingenen,
and Jean-Philippe Putaud

*European Commission, Joint Research Centre, Environment Institute, TP 280, Via E. Fermi,
21020 Ispra (VA), Italy*

Abstract. The characteristic time of many of the microphysical aerosol processes is days up to several weeks, hence longer than the residence time of the aerosol within a typical atmospheric compartment such as the marine boundary layer, the free troposphere etc. To understand aerosol properties, one cannot confine the discussion to such compartments. This paper presents simulations using a box model that describes aerosol microphysics within the context of atmospheric dynamics that connects those compartments. The model results for a Clean Marine and a Polluted Continental air mass are compared to observations.

INTRODUCTION

In the atmosphere, particles are produced from anthropogenic activities, such as fuel combustion and also from natural sources, such as dust, sea spray, and due to volcano activities. They are also formed in the atmosphere by gas-to-particle conversion processes. Particles evolve in size and composition through condensation of vapour species or by evaporation, coagulating with other particles, by chemical reaction, or by activation in the presence of supersaturated water. Aerosols are critical players in the hydrological cycle and climate system. It is therefore needed to understand their cycling in the atmosphere, and to be able to predict their characteristics. This presentation is focused on modelling the microphysics of particle formation and evolution through the Hadley/Walker cells, and comparison with observations.

CP534, *Nucleation and Atmospheric Aerosols 2000: 15th Int.'l Conf.*, edited by B. N. Hale and M. Kulmala

Modeling of Global Sulfate Aerosol Number Concentrations

Michael Herzog[1], Joyce E. Penner[1], John J. Walton[1],
Sonia M. Kreidenweis[2], Debra Y. Harrington[2]

[1] *University of Michigan, Ann Arbor, MI*
[2] *Colorado State University, Fort Collins, CO*

Abstract. A two-moment, two-mode model of sulfate aerosol dynamics has been added to the University of Michigan three-dimensional chemical transport, transformation and deposition model, GRANTOUR. The two-moment model predicts both aerosol number and mass concentrations, and was chosen based on its computational efficiency and the small number of prognostic variables, which reduce storage requirements in the large-scale model. Simulations were performed to investigate the processes that control predicted number concentrations, and comparisons with observations were used to suggest additional features that should be added to the coupled gas-phase chemistry / aerosol model to bring the predictions more in line with observations.

INTRODUCTION

There has been increased recognition of importance of aerosols to climate, and a corresponding interest in including the indirect and direct effects of particles in models for climate and climate change prediction. Sulfate compounds have been of particular interest for a number of reasons: they generally constitute a major mass fraction of particulate matter; sulfur has both natural and anthropogenic sources; and sulfate particle formation is important in determining aerosol number concentrations, which are needed to compute indirect effects.

Aerosol number concentrations should respond to variations in sources and sinks of particles and account for the observed significant temporal and spatial variability in aerosol distributions (d'Almeida et al., 1991). Thus, in further developing an understanding of how the atmospheric aerosol will respond to changes in source strengths, a method is needed for predicting not only particulate mass production and loss rates, but also particulate number production and loss.

MODEL DESCRIPTION

GRANTOUR Transport and Chemistry

GRANTOUR is a global transport, transformation and deposition model which has been applied to three-dimensional tropospheric chemistry studies (Walton et al., 1988; Penner et al., 1994; Chuang et al., 1997). It is formulated as a Lagrangian parcel model, typically run with 50,000 air parcels which represent constant air mass parcels in the atmosphere. Each parcel carries trace constituents represented by mass mixing ratios. Advection of the parcels is carried out on an Eulerian grid. Parcel species concentrations are mapped to the Eulerian grid in order to calculate changes due to diffusional mixing

CP534, *Nucleation and Atmospheric Aerosols 2000: 15th Int.'l Conf.*, edited by B. N. Hale and M. Kulmala

(Walton et al., 1988); these changes are then applied to the parcels. The model runs off-line by using wind and precipitation data from a GCM.

The species that have been included in the global sulfur cycle model are sulfur dioxiode (SO_2), dimethylsulfide (DMS), and sulfuric acid (H_2SO_4), or sulfate. The gaseous emissions are treated in three families divided by source type: anthropogenic, oceanic and vegetative, and volcanic. All sources are imposed at the center of grid boxes and, except for the volcanoes, are assumed to be well mixed in the lowest 100 mbar. The volcanic sources are assumed to be well mixed below their height of injection, which was determined as in Spiro et al. (1992).

To account for fast conversion of SO_2 to $SO4^{2-}$ in combustion plumes, based on observations of power plant plumes (Meagher et al., 1978), 3% of the mass of anthropogenic and biomass burning SO_2 emissions was removed from the gaseous source file and input as primary emissions of particles. In accordance with the methodology used by Binkowski and Shankar (1995) in their regional model, 20% and 80% of the mass of the direct particle source was input to modes 1 and 2, respectively, in number concentrations determined by assuming lognormal modes with geometric number mean diameters of 0.01 and 0.07 μm and standard deviations of 1.6 and 2.0 μm, respectively.

The scavenging coefficients were set to reproduce measured washout ratios (Penner et al., 1994); we applied the scavenging coefficients to the sulfuric acid vapor and to mode 2 particles, which are more likely to serve as CCN.

Sulfate Aerosol Dynamics

The model of sulfate aerosol dynamics is essentially that used by Kreidenweis and Seinfeld (1988) and Kreidenweis et al. (1991), and is described fully in Harrington and Kreidenweis (1998). Two modes that represent Aitken and accumulation mode particles in the fine particle fraction of the aerosol are modeled; a representation of coarse mode particulate matter is not yet included. The model is driven by a chemical source rate of $H_2SO_4(g)$, supplied from the chemistry model, and includes parameterized equations for binary nucleation of sulfuric acid particles from the vapor phase, condensational growth of each mode, and inter- and intra-modal coagulation (Youngblood and Kreidenweis, 1994; Harrington and Kreidenweis, 1998; Kreidenweis and Harrington, 1998). The aerosol modes are characterized by the time-varying mass mean particle size in each mode, determined from mass and number concentration of each mode, rather than by fixed mean diameters or bin boundaries for the two modes, as has been done in other studies (e.g. Raes and Van Dingenen, 1992; Russel et al., 1994).

The addition of particulate mass via the aqueous transformation of SO_2 is assumed to increase only the mass of mode 2 particles over the chemistry-aerosol coupling timestep. The rationale behind this treatment is that the larger mode 2 particles are the likely cloud condensation nuclei (CCN), and thus would receive this additional sulfate mass during a cloud cycle.

RESULTS

Results from the final, converged, annual cycle are reported below.

Aerosol Number Concentrations

Although the modeled particulate sulfate mass exhibits an annual cycle because of seasonal differences in transport and in the magnitude of oxidation and removal pathways, the simulated surface number concentrations over most of the globe exhibit very little month-to-month variability.

This is because they are dominated by the direct emissions from the anthropogenic SO_2 sources, which have no seasonal dependence in the source files used here. Annual averages of the predicted total number concentrations (N_{tot}) at the surface and at 355mbar are shown in Figures 1a and 1b, respectively.

The results shown in Figure 1a indicate that the model reproduces expected gradients in N_{tot} at the surface, with highest values over industrialized source regions and decreasing concentrations with increasing distance from the sources (Pruppacher and Klett, 1997). Peaks in modeled annual average

N_{tot} occur over the eastern U.S., eastern Europe, and China, and exceed 10,000 cm^{-3}, consistent with observations of rural and urban aerosols. The maximum concentration for particles aged an hour, the coupling timestep used in our model, is about 10^6 cm^{-3} (Hinds, 1982); considering this, and taking into account the large grid volumes and thus large spatial averaging used in the global model, we do not expect to reproduce localized very high N_{tot} such as those observed near city centers and in plumes.

Modeled estimates of 1,000-5,000 cm^{-3} over remote land regions (e.g., Alaska, northeastern Asia, N. Africa, and Australia) are similar to the range reported in the literature. In polar regions, the model reproduces expected lower Antartic particle number concentrations (100-200 cm^{-3}) and higher Artic N_{tot} (500-several thousand cm^{-3}). Observed and simulated particle concentrations generally decrease with altitude. In the upper troposphere, modeled N_{tot} have a stronger seasonal dependence; annual averages range from 200-1,200 cm^{-3}, again in good agreement with the literature estimates.

A number of observations and model predictions at the surface and at other levels have been compiled, data are displayed in Figure 2. The best agreement between simulations and observations was in marine regions and in the free and upper troposphere. The number concentrations in the accumulation mode over the central U.S. and Canada were simulated better at upper levels than they were in the boundary layer, where they were underpredicted. The largest differences between observations and simulations occur in polar regions, particularly in the annual cycle. Peak number concentrations are observed in late summer in Antarctica, but while modeled number concentrations were similar to observations for the winter and fall, they did not have a pronounced annual cycle and did not reproduce the observed maxima. In the Artic, the model again failed to produce an annual cycle, but in that case overpredicted number concentrations significantly.

DISCUSSIONS AND CONCLUSIONS

Results from sensitivity tests demonstrate the importance of proper treatment of the surface sources of particulate matter, and of the competition between new particle formation and condensational growth for condensable vapor. First, other sources of particulate mass and number concentration that interact with the sulfur emissions should be added to the model, including sea salt, dust, and carbonaceous particles, and the hygroscopic growth of mixed particles estimated. Second, processes that occur on sub-grid scales must be properly parameterized. The sulfur emissions occur on spatial scales much smaller than the grid sizes used in the global model, and injecting the gaseous emissions immediately into the surface grid boxes dilutes surface species concentrations artificially. It is known that some conversion of sulfur gases to particles occurs in plume, probably due to both homogeneous nucleation and to conversion on particles emitted with the gases; by accounting for this with the assumed 3% rate, a more realistic simulation of surface number concentrations is achieved. However, the most appropriate mass conversion percentages, their variation with location and time, and the input size distribution of these particles, should be developed using data and models with higher resolution.

Our studies indicated that number concentrations in the upper troposphere were not strongly affected by the treatment of surface sources, and that nucleation played a large role in determining number concentrations at higher altitudes. The nonlinearity of the assumed nucleation rate expression, with respect to relative humidity, temperature and $H_2SO_4(g)$ concentration, was particularly influential, and this sensitivity emphasizes the need for a better understanding of the factors controlling new particle formation in the atmosphere.

The results presented here are not intended to represent a fully realized formulation of the complex atmospheric aerosol system, or even the sulfate aerosol fraction. They are, however, a first step towards adding prognostic aerosol equations to a large-scale model.

ACKNOWLEDGEMENTS

This material is based upon work supported by the NASA AEAP Program.

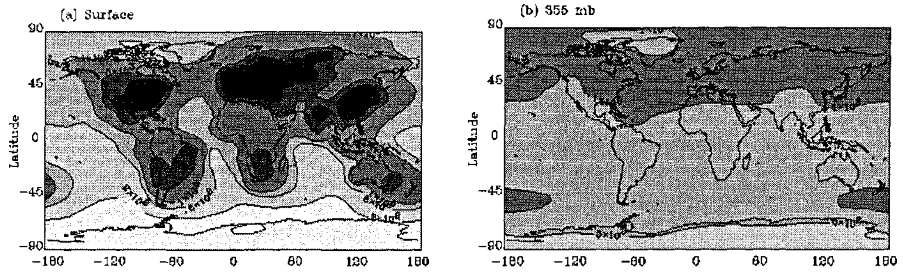

FIGURE 1. Annual average total particle number concentrations [cm^{-3}], base case simulation: a) surface, b) 355 mbar.

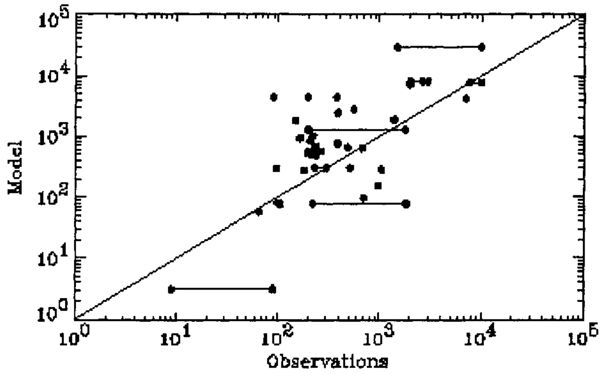

FIGURE 2. Comparison of model predictions of particle number concentrations [cm^{-3}] with observations.

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