Cloud Parameterization in Global Climate Models: Evaluation with Atmospheric Radiation Measurement Data

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The goal of our work is to develop and evaluate a prognostic aerosol model and to parameterize the effects of aerosols, acting as cloud condensation nuclei (CCN), on initial cloud droplet concentrations. This, together with global climate models which are able to treat the microphysics of clouds, should allow us to improve the climate models' representation of clouds to enable them to more accurately predict future climate change. Development of models capable of treating the microphysics of clouds after the initial size distribution is determined is proceeding under separate investigators within the ARM program.

The prognostic aerosol model is being developed using the GRANTOUR model as a framework (Penner et al. 1991). In our model simulations, sulfur compounds in the atmosphere are supplied by 1) anthropogenic sulfur (SO_2) from fossil fuel combustion, industry, and biomass burning, and 2) natural sources such as biogenic ocean sulfur (DMS), terrestrial soils (DMS, H_2S), vegetation (H_2S) and volcanoes (H_2S, SO_2). These sources are adapted from the inventories by Spiro et al. (1992) and Benkovitz (1982). For surface-based sources, trace species are input into GRANTOUR with a vertical profile that is assumed to be constant in mixing ratio in the lowest 100 mb. We note that the source of DMS is highly uncertain. Estimates range from 10 to 45 Tg S/yr. This wide range in emission estimates together with postulated reactions of SO_2 with sea salt particles and the reaction of DMS with Cl, NO_3, or other oxidants, as well as the uncertain yield of SO_2 from the oxidation of DMS, gives rise to a major uncertainty in the budget of sulfur over the oceans. For this reason, our chemical model was simplified. Thus, we include only the gas phase reactions of DMS and SO_2 with OH, using a background concentration of OH that is specified according to the latitudinally and seasonally varying calculated concentrations from the LLNL 2-D model (see Penner et al. 1991) and a total source strength for DMS of 23.7 Tg S/yr. The reactions of DMS with NO_3 and Cl would mainly affect concentrations of $SO_4^=$ over ocean areas and in the winter hemisphere where DMS is the main source of sulfate.

The aqueous reaction to convert SO_2 to $SO_4^=$ in clouds is assumed to have an average e-folding lifetime of 30 hours at 40° N at the surface in summer. This e-folding lifetime is scaled to be proportional to the square of the locally specified concentration of OH at other locations. This formulation is a simplified attempt to account for the observed seasonal variations in sulfate concentrations and wet deposition in North America by assuming that the rate at which $SO_4^=$ forms in clouds depends on how frequently the cloud is intercepted by an air parcel (average cloud-free periods vary from 10 to 80 hours [Lelieveld and Crutzen, 1990]) and on the local concentration of H_2O_2 (which was assumed to be proportional to the square of the specified concentration of OH). This formulation does not account for variations in the rate at which sulfate is produced due to in-cloud oxidation by O_3 or trace metals which may be important at high concentrations of SO_2 after H_2O_2 is consumed. On the other hand, it is not yet clear whether chemistry determines the average rate of in-cloud conversion or whether the frequency of interception of air parcels and clouds determines the overall rate (Chin et al. [Accepted]; Langner and Rodhe 1991). Our parameterization produces surface sulfate concentrations that are quite similar to those in other sulfur models (e.g., Langner and Rhode 1991; Chin et al. [Accepted]). Our total burden and global budget of SO_2 and $SO_4^=$ is similar to that in the model of Chin et al. (Accepted) and to the "slow" oxidation case of Langner and Rhode (1991). Our predicted $SO_4^=$ concentrations in air and in precipitation are in reasonable agreement with observational data, except that our seasonal cycle in Europe is stronger than observations indicate (Penner et al. 1994).

SO_2 and $SO_4^=$ may also be scavenged by precipitation. The scavenging coefficients are set to 0.8 cm hr^{-1} and 1.5 cm hr^{-1} for SO_2 in stratiform and convective precipitation, respectively, and to 5.0 cm hr^{-1} and 1.5 cm hr^{-1} for sulfate in these two precipitation types. The values for SO_2 are approximately 30% of the values previously used for the much more soluble gas HNO_3 (which were based on observations of nitrate scavenging in various types of precipitation

[see Penner et al. 1991]). The values chosen for $SO_4^=$ are a factor of 2.4 higher than values chosen for carbonaceous aerosols. This choice reflects the fact that measured washout ratios for $SO_4^=$ are typically much higher than those for black carbon (Penner et al. 1993) and that, as a result of its high solubility, most of the mass of $SO_4^=$ is expected to be incorporated in cloudy air as a result of nucleation scavenging. In addition, when air parcels are in the lowest 100 mb, deposition velocities of 0.1 and 0.8 cm s^{-1} are applied to $SO_4^=$ and SO_2, respectively. These values do not account for variations in deposition that result from surface and vegetation types, but represent reasonable averages for important anthropogenic source regions.

Besides the aforementioned sulfur species, air parcels also carry anthropogenic black and organic carbon particles as well as natural organic matter particles. Anthropogenic black and organic carbon particles are mainly from sources such as biomass burning and fossil fuel combustion, whereas natural organic particles come from photochemical conversion of gaseous emissions from vegetation and from direct emission of particles from plants. Our model procedures and emission inventories for black and organic carbon particles follow those developed by Liousse et al. (In press). Black and organic carbon particles are assumed to have the same dry deposition velocity as specified for $SO_4^=$, while their scavenging coefficients are assumed to be 2.1 cm hr^{-1} in stratiform and 0.6 cm hr^{-1} in convective precipitation based on measured washout ratios (Penner et al. 1993).

The key issue in assessing the radiative effects of aerosol on cloud optical properties is the relationship between cloud drop number concentration and anthropogenic sulfate mass concentration. However, this relationship depends on the individual aerosol size distribution and, therefore, on the processes that form particulate sulfate. It also depends on the preexisting aerosol. In our model, we parameterize the fraction of sulfate formed by aqueous processes, allowing that portion to deposit on pre-existing aerosols of radius >0.1 μm. The portion from homogeneous gas phase production of sulfate may deposit over the entire size distribution of pre-existing aerosol.

Because of the nonlinear relationship between CCN and cloud drop number concentrations, the presence of substantial concentrations of pre-existing particles that are able to act as CCN must lower the estimates of indirect forcing by anthropogenic $SO_4^=$. To illustrate this possibility, we have calculated the response of drop concentrations of a hypothetical stratus cloud (1000-m thick, 0.2 g m^{-3} liquid water content) to changing anthropogenic $SO_4^=$ concentrations in the presence of natural $SO_4^=$ and organic aerosols. In these

calculations, we have used mass concentrations of 300 ng m^{-3} for $SO_4^=$ (assumed to be in the form of ammonium sulfate) and 500 and 1000 ng m^{-3} for OC.

The calculations were performed by assuming that the natural organic and sulfate aerosol components are distributed over the typical size distribution measured in marine situations adopted by Chuang et al. (Accepted). The number concentrations of the aerosols derived in this manner are 94, 336, and 557 cm^{-3} for 300 ng m^{-3} $SO_4^=$, 500 ng m^{-3} OC, and 1000 ng m^{-3} OC, respectively. We assumed that 75% of anthropogenic $SO_4^=$ is produced by aqueous, in-cloud oxidation (sulfate formed through this pathway should not form additional, new CCN) and that the remainder, in form of sulfuric acid, is deposited on the pre-existing natural aerosols.

In Figure 1 we show the calculated dependencies of cloud drop concentrations. These results were obtained for vertical (updraft) velocities of 10 and 50 cm sec^{-1} by following the procedure and assumptions similar to those in our global model (Chuang et al. [Accepted]). The drop concentrations due to 300 ng m^{-3} of natural $SO_4^=$ are expected to be about 40 cm^{-3} and 58 cm^{-3}, respectively, for the two updraft velocities. When 500 ng m^{-3} of natural OC is added to natural $SO_4^=$ the corresponding drop concentrations are 103 and 165 cm^{-3}. Changes in drop concentrations due to inclusion of anthropogenic sulfate strongly depend on the assumed preexisting aerosol concentrations.

We hope to use this strong relationship to validate our parameterization with ARM data. For example, if ARM data can be sorted by cloud thickness, liquid water content, and aerosol size distribution, we should be able to observe a
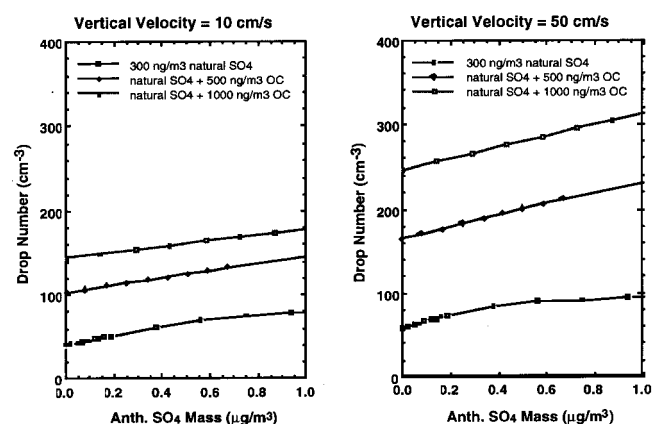

Figure 1. The relationship between increases in sulfate mass and droplet concentration in cloud for three difference assumed background aerosol concentrations and substances.

relationship between the satellite-sensed reflected radiation and the aerosol number concentration. The anticipated relationships are shown in Figure 2. This figure also shows the range of uncertainty in the predicted radiation introduced by a range of updraft velocities from 10 to 50 cm/s.
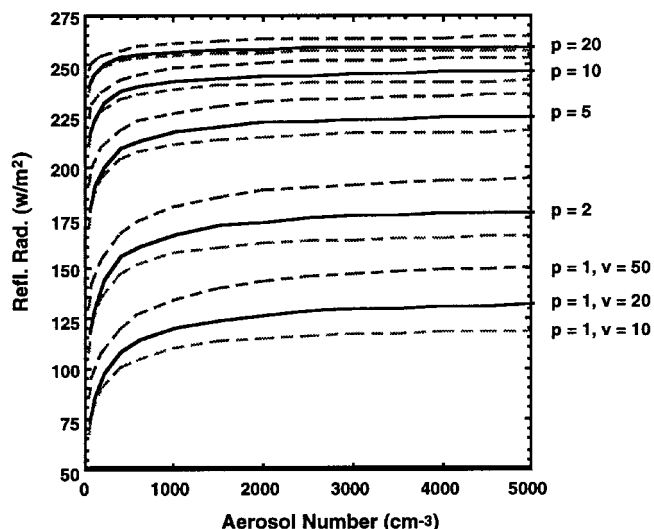

Figure 2. The relationship between reflected solar radiation from clouds and aerosol number concentration at cloud base. The parameter p is given by

$$P = h \left[\frac{9 \pi w_L^2}{2\rho^2} \right]^{L_3}$$

where w_L is the liquid water content of the cloud, h is the depth, and ρ is the density of water. The reflected radiation for updraft velocities ranging from 10 to 50 cm/s is also shown.

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