The Role of Entrainment and Mixing in Altering the Relationship Between Aerosol Concentration and Cloud Drop Number Concentration

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Introduction

Two basic methods have been used to relate the change in droplet number concentration to aerosol number concentration. The first method uses empirically derived relationships based on either observed aerosol number concentrations and droplet concentrations or observed aerosol sulfate or droplet mass concentrations and droplet number concentrations. This method was followed by Jones et al. (1994) and Boucher and Lohmann (1995). Jones et al. (1994) linked aerosol mass and droplet number concentration, while Boucher and Lohmann (1995) linked sulfate mass to droplet concentrations. The second method for relating aerosol number concentrations and droplet number concentrations uses a mechanistic model that simulates the relationship between aerosol particles and droplets determined at cloud base based on a model of the microphysics of cloud formation. In this method the change in droplet number concentration is associated with a given change in the aerosol concentration, size distribution, aerosol composition, and updraft velocity. While the empirical method is based on observations and may therefore avoid possible inaccuracies associated with the parameterization of microphysics processes in large-scale models, current models must still assume a relationship between aerosol mass and aerosol number to apply the Jones et al. parameterization. Also, the parameterization of Boucher and Lohmann may be inaccurate because the use of sulfate from cloud water measurements to predict N_d is uncertain because of variations in the cloud scavenging efficiencies for aerosol sulfate and because the production of sulfate in cloud water would obscure the relationship between sulfate mass and N_d. The second mechanistic method, has been developed in our work, but still requires some assumptions to determine the aerosol size distribution and chemical composition. Furthermore, the method does not account for mixing processes within the cloud, which may alter the relationship between aerosol number concentration and droplet concentration.

Table 1 summarizes the global modeling studies of indirect climate forcing associated with the indirect effect of anthropogenic aerosols. The estimates for indirect forcing associated with anthropogenic sulfate aerosols lie in the range -0.2 Wm^{-2} to -2.1 Wm^{-2} while the range associated with carbonaceous aerosols lies between -0.9 Wm^{-2} and -1.44 Wm^{-2}, but could be as large as -3.0 Wm^{-2} if we increase the

Table 1a. Comparison of indirect forcing by different models without liquid water path (LWP) feedback.

Model	Pre-Industrial Aerosol (Tg S)	Industrial Aerosol (g/m^2)	Nucleation Parameterization	Cloud Cover and Autoconversion Treatment	Forcing (W/m^2)
Lohmann and Feichter 1997	Sulfate 0.3 Tg S, interactive	Sulfate (0.38), interactive	Boucher and Lohmann (1995)	Beheng (1994) cloud cover = Sundquist et al. 1989	-1.4 (sulfate)
Lohmann et al. 1999	Sea salt 0.79 Tg, **dust 5.23 Tg organic matter,** 0.12 Tg interactive sulfate	Interactive sulfate 1.04 Tg (tot), organic matter 1.69 Tg (tot), black carbon 0.24 Tg	Chuang and Penner 1995	Beheng (1994) cloud cover = Sundquist et al. 1989	-1.1 to -1.9 (sulfate+carbo-naceous)
Jones et al. 1998	Interactive sea salt, sulfate 0.094 Tg	Interactive sulfate (= 0.36 - 0.094)	Jones et al. 1994	Cloud cover (?)	-1.05 (sulfate)
Rotstayn 1999	Monthly average sulfate, 0.25 Tg S	Monthly average sulfate, 0.30 Tg S	Boucher and Lohmann (1995)	Manton and Cotton (1977)	-2.1 (sulfate)

Table 1b. Comparison of indirect forcing by different models that includes LWP feedback.

Model	Pre-Industrial Aerosol (Tg S)	Industrial Aerosol (g/m2)	Nucleation Parameterization	Cloud Cover and Autoconversion Treatment	Forcing (W/m2)
Lohmann and Feichter 1997	Sulfate 0.3 Tg S, interactive	Sulfate (0.38), interactive	Boucher and Lohmann (1995)	Beheng (1994) cloud cover = Sundquist et al. 1989	-1.4 (sulfate)
Lohmann et al. 1999	Sea salt 0.79 Tg **dust 5.23 Tg, organic matter,** 0.12 Tg interactive sulfate	Interactive sulfate 1.04 Tg (tot), organic matter 1.69 Tg (tot), black carbon 0.24 Tg	Chuang and Penner 1995	Beheng (1994) cloud cover = Sundquist et al. 1989	-1.1 to -1.9 (sulfate+carbo-naceous)
Jones et al. 1998	Interactive sea salt, sulfate 0.094 Tg	Interactive sulfate (= 0.36 - 0.094)	Jones et al. 1994	Cloud cover (?)	-1.05 (sulfate)
Rotstayn 1999	Monthly average sulfate, 0.25 Tg S	Monthly average sulfate, 0.30 Tg S	Boucher and Lohmann (1995)	Manton and Cotton (1977)	-2.1 (sulfate)

estimate of Chuang et al. (1997) and Penner et al. (1999) to account for the fact that this model did not include liquid water feedback. Some of these variations are associated with the particular parameterization relating aerosol concentration to droplet number while others may be associated with the concentration of pre-existing aerosol assumed in the calculation. In addition, there are differences associated with whether the model study included feedback to cloud liquid water content (LWC), cloud depth, and cloud lifetime in its quantification of the magnitude of the indirect forcing.

Figure 1 shows the difference in predicted droplet number concentrations as a function of sulfate mass concentration for these different parameterizations. Based on the data they had available, Jones et al. (1994) developed a single parameterization, while Boucher and Lohmann (1995) recommended a different parameterization for marine and continental areas. Chuang et al. (1997) also developed

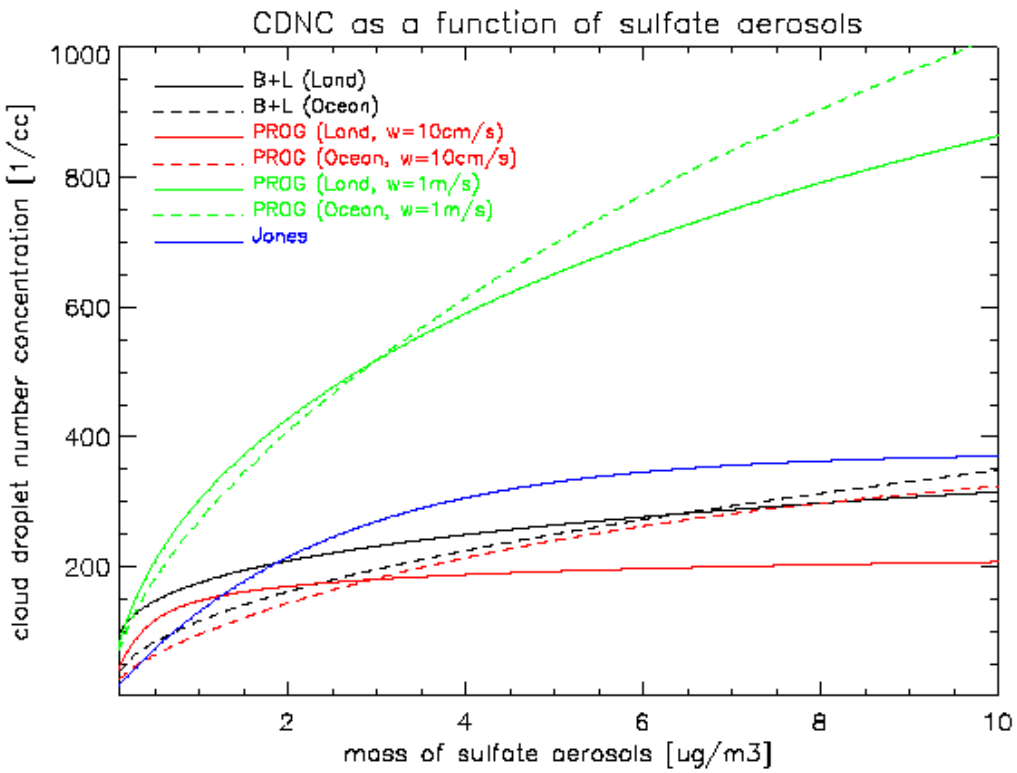

Figure 1. Droplet concentration as a function of anthropogenic sulfate concentration for three different treatments; the empirical treatment of Jones et al. (1994), the empirical treatment of Boucher and Lohman (1995) (denoted B+L), and the mechanistic treatment of Chuang and Penner (1995) (denoted as PROG).

separate relationships over marine and continental areas based on the variation in background aerosol size and concentrations assumed in the two regions. Figure 1 shows the relationship between droplet number concentration and sulfate mass for an updraft velocity of 10 cm/s and 1 m/s according to the parameterization of Chuang and Penner (1995) and the relationship based on the parameterizations of Jones et al. (1994) and Boucher and Lohmann (1995). While these parameterizations are roughly similar for low updraft velocities, there are significantly more droplets formed in the Chuang and Penner formulation than in the empirical relationships if the updraft velocity is as high as 1 m/s.

One possible reason for these differences relates to the occurrence and effects of mixing or entrainment within clouds. Mixing processes deplete the LWC in the cloud, and, in general, lower droplet number concentrations. This is shown in Figure 2, which compares the LWC, number concentration and effective radius from two clouds (Brenguier et al. 1998). The measurements associated with the top panel of figures were made in a nearly adiabatic cloud as shown by comparison of the measured LWC with the line, which depicts the value for an adiabatic cloud. Those from the lower set of panels were made from a set of clouds, which had experienced more entrainment and mixing (Brenguier et al. 1998). Near the top of the cloud shown in the lower panel, droplet number concentrations become significantly smaller on average than at cloud base and droplet effective radius (or diameter) is not increased as rapidly as for the adiabatic cloud.

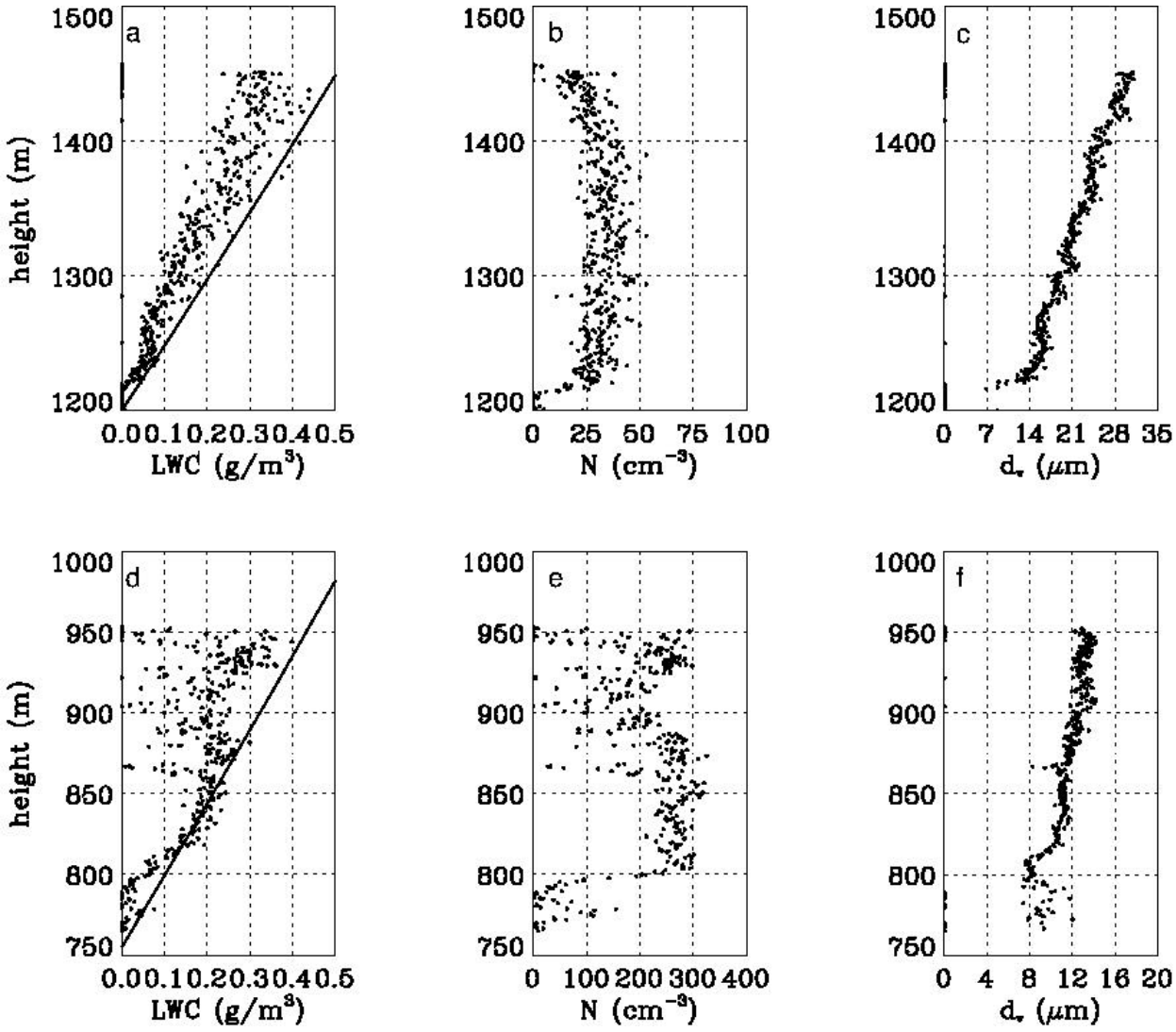

Figure 2. The vertical distribution of cloud liquid water content (LWC, g/m^3), droplet number concentration (N_d, cm^{-3}), and volume-weighted average diameter (d_v, μm) for two different boundary layer clouds: a clean marine case (upper panel) and a polluted case (lower panel). The top panel of figures are from a nearly adiabatic cloud while the second panel of figures are from a cloud that was not as nearly adiabatic. In the latter cloud, mixing with dry air from above cloud lowers the LWC and number concentration of drops toward the top of the cloud. (Figure 4 from Brengueir et al. 1998.)

We have begun exploring how entrainment will affect the predicted droplet concentrations in our microphysics model. In particular, we developed a version of the model, which employs the Baker et al. (1980) assumptions for heterogeneous mixing. Thus, we assumed that air parcels from above cloud base were entrained within the cloud every 10 seconds and that these parcels fully mixed with other cloudy air. Figure 3 shows the result. Droplet concentrations for a given aerosol concentration are significantly smaller in the simulation which allows for entrainment and mixing.

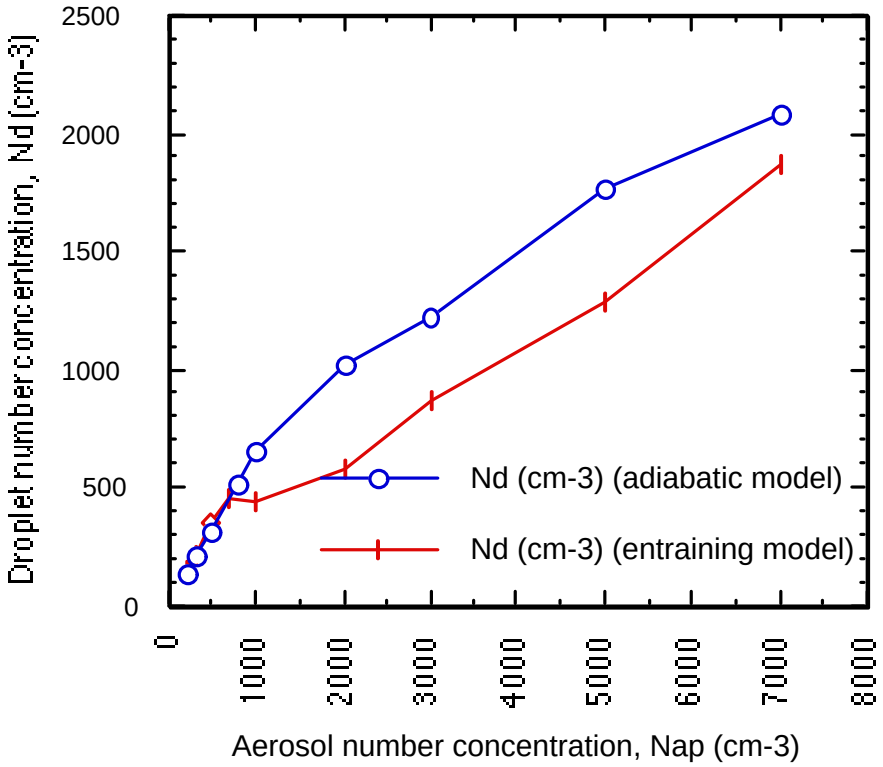

Figure 3. Comparison of initial droplet number in entraining and adiabatic model.

How should this result be incorporated within global models? The answer depends on the manner in which clouds are treated in the global model. Clouds with significant entrainment and mixing are those which are smaller that typical grid sizes. Thus, the incorporation of a parameterization for droplet number in these clouds must be consistent with the manner in which the size of the cloud is determined in the cloud parameterization of the global model. In the future, we hope to explore these issues and to explore whether Atmospheric Radiation Measurement (ARM) data can be used to validate a parameterization for the effects of changing aerosol concentrations on sub-grid scale clouds.

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