THE UNIVERSITY OF MICHIGAN
DEPARTMENT OF ATMOSPHERIC, OCEANIC, AND SPACE SCIENCE

Space Physics Research Laboratory
2245 Hayward Street
Ann Arbor, Michigan 48109-2143

Contract/Grant No.:	NAG5-3771
Project Name:	" Global Studies of the Sulfur Cycle Including the Influence of DMS and Fossil Fuel Sulfur on Climate and Climate Change"
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Report Preparation Date:	12/22/98
Report Type:	Final Report
Period Covered:	2/1/97-1/31/98
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Indirect climate forcing by anthropogenic aerosols: A mechanistic treatment

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Abstract. The indirect effect of anthropogenic aerosols, wherein aerosol particles are thought to increase cloud droplet concentrations and cloud lifetime, is the most uncertain component of climate forcing over the past 100 years. Here, for the first time, we use a mechanistic treatment of droplet nucleation and a prognostic treatment of the number of cloud droplets to study the indirect aerosol effect from changes in carbonaceous and sulfate aerosols. Cloud droplet nucleation is parameterized as a function of total aerosol number concentration, updraft velocity and a shape parameter, which takes into account the mechanism of sulfate aerosol formation, while cloud droplet number depends on the nucleation as well as on droplet sinks. Whereas previous treatments have predicted annual average indirect effects between -1 and -2 W m^{-2}, we obtain an indirect aerosol effect between -0.14 Wm^{-2} and -0.42 Wm^{-2} in the global mean.

Anthropogenic aerosols such as sulfate or carbonaceous aerosols have substantially increased the global mean burden of aerosols from pre-industrial to present-day. While the change in solar radiation at the top of the atmosphere by absorption and scattering of anthropogenic aerosols (direct aerosol effect) remains uncertain [*Houghton et al.*(1996); *Penner et al.*(1998)], the change associated with the indirect effect, where anthropogenic aerosols act as cloud condensation nuclei (CCN) and thereby determine the initial droplet number concentration, albedo, precipitation formation and lifetime of warm clouds is far more uncertain [*Penner et al.*(1994); *Pan et al.*(1998)]).

Several studies of the indirect effect of anthropogenic aerosols have concentrated on the effects of anthropogenic sulfate aerosols, examining the difference in initial droplet concentration [*Chuang et al.*(1997); *Jones and Slingo*(1996)], while, to our knowledge, only one study has examined the indirect effect of anthropogenic carbonaceous aerosols [*Penner et al.*(1996)]. These studies indicated that anthropogenic sulfate might lead

to an indirect forcing that ranged from -0.4 Wm^{-2} to -1.5 Wm^{-2}, with smaller forcing (-0.4 to -0.6 Wm^{-2}) associated with those simulations in which the mechanism for sulfate formation is taken into account. In the latter sulfate mass when formed within drops, simply adds mass to pre-existing aerosols, but does not increase aerosol number concentration. Larger indirect effects (up to -1.5 Wm^{-2}) is associated with either treating the sulfate aerosol as an external mixture (adding aerosol number concentration proportional to sulfate mass) or in an empirical manner that relates sulfate mass directly to droplet concentration. In contrast to sulfate, carbonaceous aerosols add directly to CCN concentrations since they are mainly emitted as primary aerosol particles. Thus, even when a mechanistic formulation of cloud droplet nucleation was used, [*Penner et al.*(1996)] predicted that the initial droplet concentration formed from adding these aerosols to the pre-industrial atmosphere might lead to a forcing ranging from -2.4 to -4.4 Wm^{-2}. However, no sinks for cloud droplets were considered in these experiments, so that the forcing is an upper bound.

[*Lohmann and Feichter*(1997)] and [*Rotstayn*(1998)] included contributions to the indirect effect from anthropogenic sulfate aerosols from an increase in cloud lifetime due to slower precipitation formation as well as the change in cloud albedo associated with droplet concentration change. These studies used a diagnostic relationship between sulfate mass and droplet concentration and resulted in a total forcing between -1.4 and -4.8 W m^{-2} in the global mean. The magnitude of the indirect effect in these studies depends crucially on the autoconversion rate and cloud cover parameterization, but, both these sulfate aerosol studies [*Lohmann and Feichter*(1997); *Rotstayn*(1998)] and the one study of carbonaceous aerosols [*Penner et al.*(1996)] indicate that the indirect effect could be significantly larger than the uncertainty limits set in [*Houghton et al.*(1996)].

Here, we examine the indirect effect of both anthropogenic carbonaceous aerosols and sulfate aerosols employing the mechanistic parameterization of cloud droplet nucleation developed by [*Chuang et al.*(1997)]. Thus, droplet nucleation is a function of total aerosol number concentration, updraft velocity and a shape parameter, which takes into account the change in aerosol composition and size distribution that results from the formation of sulfate on pre-existing CCN within drops as well as the formation of sulfate by homogeneous gas phase processes. Total aerosol number is obtained assuming a uniform aerosol size distribution over land and ocean. In contrast to previous studies, here we in-

troduce a prognostic equation for the number of cloud droplets [*Lohmann et al.*(1998)] in the ECHAM4 GCM [*Roeckner et al.*(1996)] (referred to as PROG) to be able to consider sources and sinks for cloud droplets. With this treatment we are able, for the first time, to account for the effect of a mechanistic treatment of droplet nucleation as well as the effect of different aerosol species on the precipitation formation and lifetime of clouds. This differs from the model used to calculate the indirect effect in [*Lohmann and Feichter*(1997)] (referred to as DIAG) because that model parameterized the lifetime and precipitation formation in terms of liquid water content and cloud droplet number concentration empirically obtained from the mass of sulfate aerosols. In PROG the total number of aerosols is obtained as the sum of marine sulfate aerosols produced from dimethyl sulfide, natural and anthropogenic hydrophylic carbonaceous aerosols, dust and sea salt aerosols. Because of high continental aerosol concentrations, we assume that anthropogenic sulfate aerosols only add mass to the pre-existing aerosols and do not form new particles. Thus, we assume that an internally mixed aerosol particle forms when sulfuric acid with anthropogenic origin condenses onto any pre-existing aerosol particle (in agreement with measurements from [*Hudson and Da*(1996)]). Dust and sea salt are prescribed as three-dimensional monthly mean data while prognostic equations are solved to calculate the mass of sulfate and carbonaceous aerosols. The individual aerosol species are described in detail in [*Lohmann et al.*(1998)]. Their sources and global burdens are summarized in Table 1.

A validation of this new scheme with present-day emissions in terms of cloud physical and optical properties is discussed in [*Lohmann et al.*(1998)].

The results presented below are based on five-year integrations at T30 resolution after a three month spin-up. In the experiments with pre-industrial emissions, fossil fuel use is set to zero and biomass burning emissions are assumed to be 10% of the present-day emissions (cf. table 1).

Figure 1 shows the annual and global mean vertical profiles of the cloud droplet number concentration (CDNC), vertical velocity, the number of aerosols and liquid and ice water content from PROG and DIAG for present-day and pre-industrial aerosol concentrations, respectively. The difference in the number of aerosols decreases with altitude in PROG, from 1500 cm^{-3} near the surface to less than 100 cm^{-3} at 500 hPa. As the vertical velocity has its maximum at 950 hPa the largest number of cloud droplets are activated in the bound-

ary layer. The change in CDNC from pre-industrial to present-day is similar to the mean profile of CDNC driven by the vertical velocity profile and the availability of aerosols. Thus, it is largest below 900 hPa and decreases rapidly aloft. In DIAG the change in CDNC is significant everywhere, because the mass of sulfate aerosols differs in all levels between pre-industrial and present-day. The resulting change in liquid water content in DIAG has its maximum between 750 hPa and 900 hPa, increasing up to 20% from pre-industrial to present-day. In PROG the increase in liquid water content is substantially smaller, because the maximum increase in CDNC occurs at 950 hPa, where the liquid water content is rather small.

The resulting annual global mean indirect aerosol effect, defined as the change in shortwave cloud forcing at the top of the atmosphere between pre-industrial and present-day, amounts to only -0.15 W m^{-2} in PROG, while it is -1.4 W m^{-2} in DIAG (cf. table 2). The much smaller indirect effect in PROG than in DIAG results from difference in the vertical distribution of CDNC in PROG and DIAG (cf. fig. 1). Because the change in CDNC in PROG is much lower than in DIAG above 850 hPa, the increase in liquid water path in PROG between pre-industrial and present-day is only 1.5 g m^{-2} as compared to 10.7 g m^{-2} in DIAG.

To understand the difference between PROG and DIAG better, we conducted two sensitivity experiments. In one pair of experiments (pair referring to one experiment with pre-industrial emissions and one with present-day emissions) we allowed anthropogenic sulfate to form new aerosol particles, i. e. assumed an external mixture. This is certainly an overestimation of the number of aerosols, but is closer to the design of DIAG (experiment PROG-ext). In the other pair of experiments we empirically related the mass of sulfate aerosols (mSO_4^{2-}) to the number of aerosols (N_a) (experiment PROG-emp) as obtained from data from different field experiments (Leaitch/Banic/Li, personal communication, 1998):

$$N_a = 340 \cdot (mSO_4^{2-})^{0.58} \qquad (1)$$

where N_a is in cm^{-3} and mSO_4 in $\mu g\ m^{-3}$. This experimental design is as close to DIAG as possible, because both in PROG-emp and DIAG sulfate mass is taken as a surrogate of total aerosol mass assuming that the fraction of sulfate which condenses on aerosols remains constant. However in PROG-emp aerosol number is obtained from sulfate mass, whereas it is CDNC in DIAG.

In figure 2 the percentage increase from pre-industrial levels for the number of aerosols, CDNC, liquid water content and cloud cover is shown for all experiments.

The change in the number of aerosols varies considerably between the different pairs of experiments, increasing by more than 200% near the surface in PROG-ext to 150% in PROG and PROG-emp. The resulting change in CDNC in all pairs of experiments with PROG is mostly confined to below 600 hPa. More CDNC than in PROG are activated if either anthropogenic sulfate is allowed to produce new aerosol particles as in PROG-ext or if sulfate mass is taken as a surrogate for total aerosol mass as in PROG-emp, so that in these experiments the increase in CDNC in the boundary layer is larger than in DIAG.

The change in liquid water content, however, is significantly smaller in all experiments with PROG than in DIAG, even in the boundary layer. A possible explanation is that in DIAG the larger number of cloud droplets in mid level clouds slows down precipitation formation and, thus, the accretion of rain drops with cloud droplets of low level clouds is reduced. In PROG, however, the precipitation formation in mid level clouds hardly changes, so that the low level clouds are more depleted by accretion than in DIAG. Cloud cover increases between 0.2 and 0.5% in all experiments. As a result of small changes in liquid water content in all pairs of experiments with PROG (cf. table 2) the indirect aerosol effect is significantly smaller than in DIAG.

The larger indirect effect in PROG-emp than in PROG-ext results from a slightly larger increase in total cloud cover in PROG-emp. Moreover, the increase in CDNC in PROG-emp occurs at lower values of pre-industrial CDNC, so that the clouds affected by changes in CDNC are more susceptible [*Platnick and Twomey*(1994),].

In conclusion, the indirect aerosol effect as calculated from different aerosol species in conjunction with a prognostic treatment of the number of cloud droplets with the ECHAM GCM is much smaller than previously estimated, because the change in the number of cloud droplets is much smaller and mainly confined to boundary layer clouds. The mechanistic treatment of droplet formation (in particular, the dependence of droplet concentration on updraft velocity) together with the prognostic treatment of cloud droplets is responsible for the very small indirect aerosol effect of between -0.14 and -0.42 W m^{-2} for the different experiments with PROG.

Clearly the indirect effect deserves further study. The updraft velocity in the GCM as well as the total aerosol number and size distribution is crucial in determining our results. Also ECHAM, as most GCMs underestimate the extend and liquid water amount of stratocumulus, which are thought to be affected most by changes in anthropogenic aerosol concentrations. However, our

results indicate that the magnitude of the indirect effect may be significantly smaller than previously thought.

Acknowledgments. UL is grateful for support from NSERC, JF from the EC SINDICATE program and JEP is grateful for support from the DOE ARM program and the NASA ACMAP program.

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(Received _____)

Table 1. Global emissions of sulfur gases and other aerosol types

Species	Source	Source strength [Tg yr^{-1}]		Burden [Tg]	
		present-day	pre-industrial	present-day	pre-industrial
DMS	marine biosphere	17.3	17.3		
DMS	terrestrial biosphere	0.9	0.9		
SO_2	Non-eruptive volcanoes	8.0	8.0		
SO_2	Fossil fuel use	66.8	0.		
SO_2	Biomass burning	2.5	0.25		
Total sulfate		95.5	27	1.0	0.38
BC	Fossil fuel use	6.1	0.		
BC	Biomass burning	5.6	0.56		
Total BC		11.7	0.56	0.26	0.01
OC	natural	16.2	16.2		
OC	Fossil fuel use	29.6	0.		
OC	Biomass burning	59.3	5.93		
Total OC		105.1	22.1	1.87	0.11
Sea salt (0-1 μm)	bursting of whitecap bubbles	76	76	0.73	0.73
Dust (0-1 μm)	deserts	250	250	5.23	5.23

Table 2. Annual global mean change in liquid water path, cloud cover and indirect aerosol effect for PROG, DIAG and the sensitivity experiments

Experiments	PROG	PROG-ext	PROG-emp	DIAG
Change in liquid water path [g m^{-2}]	1.5	2.2	2.5	10.7
Change in cloud cover [%]	+0.19	+0.17	+0.24	+0.5
Indirect aerosol effect [W m^{-2}]	-0.15	-0.14	-0.42	-1.4

Figure 1. Annual global mean vertical profiles for PROG-present-day (solid), PROG-pre-industrial (dashed), DIAG-present-day (dotted) and DIAG-pre-industrial (dot-dashed) of the number of cloud droplets [cm^{-3}] (a), vertical velocity [m s^{-1}] (b), the total number of aerosols [cm^{-3}] (c), sum of liquid and ice water content [mg kg^{-1}] (d) and cloud cover [%] (e).

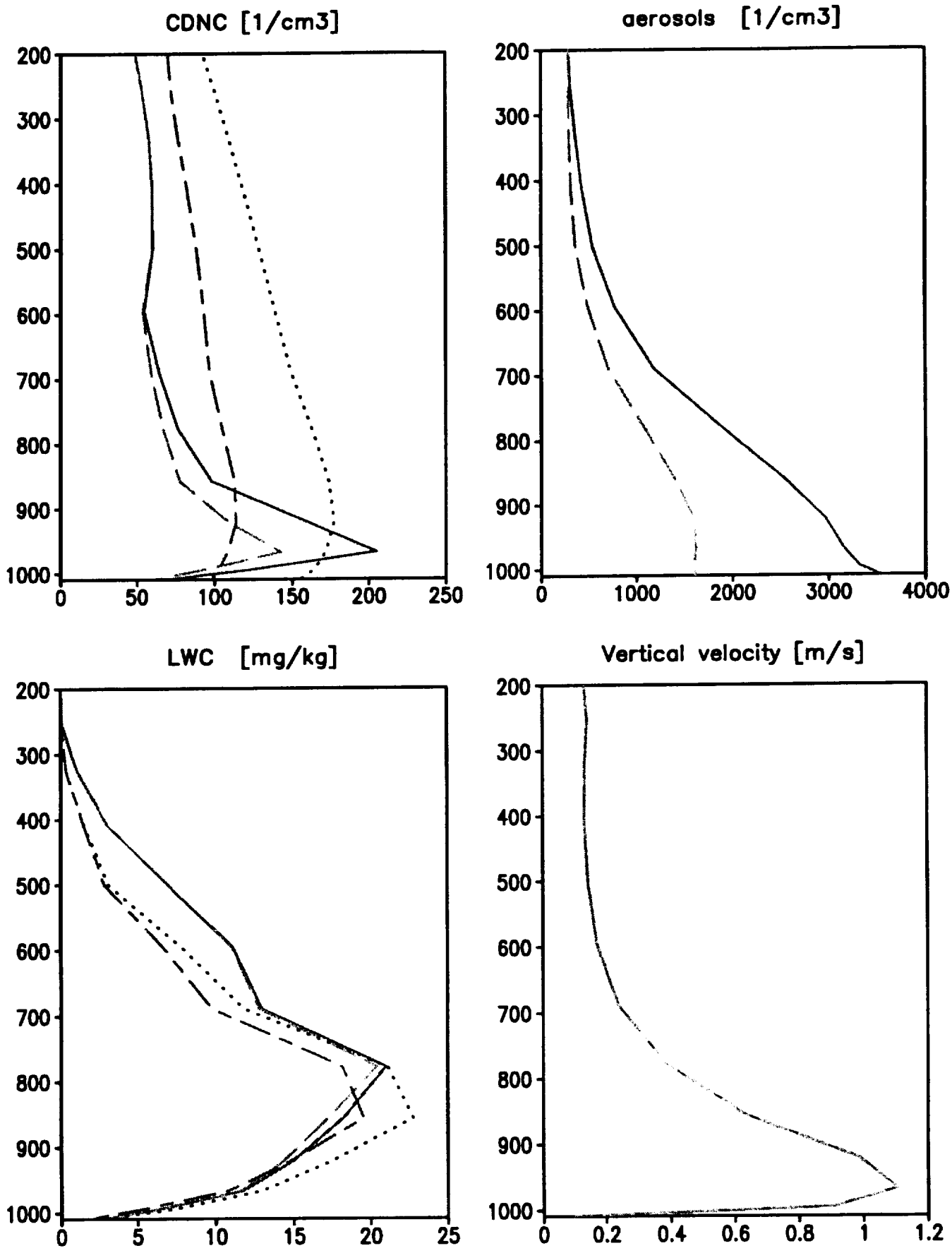
Figure 1. Annual global mean vertical profiles for PROG-present-day (solid), PROG-pre-industrial (dashed), DIAG-present-day (dotted) and DIAG-pre-industrial (dot-dashed) of the number of cloud droplets [cm^{-3}] (a), vertical velocity [m s^{-1}] (b), the total number of aerosols [cm^{-3}] (c), sum of liquid and ice water content [mg kg^{-1}] (d) and cloud cover [%] (e).

Figure 2. Annual global mean difference of vertical profiles between pre-industrial and present-day as percentage of the present-day values for the sensitivity experiments: PROG (solid), PROG-emp (dashed), PROG-ext (dashed-dotted), DIAG (dotted).

Figure 2. Annual global mean difference of vertical profiles between pre-industrial and present-day as percentage of the present-day values for the sensitivity experiments: PROG (solid), PROG-emp (dashed), PROG-ext (dashed-dotted), DIAG (dotted).

Annual global mean profiles of:

PROG–PD (solid), PROG–PI (dashed), DIAG–PD (dotted), DIAG–PI (dash–dot)



Annual global mean differences between present-day and pre-industrial:
PROG (solid), PROG-emp (dashed), PROG-ext (dash-dot), DIAG (dotted)