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The effects of atmospheric sulfur on the radiative properties of convective clouds: a limited area modeling study

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Abstract. Convective clouds in tropical areas can be sensitive to the atmospheric sulfate loading, particularly during enhanced sulfate episodes. This assertion is supported by simulations with a high resolution limited area non-hydrostatic model (LAN) employing a detailed sulfate-cloud microphysics scheme, applied to estimate the effects of sulfate on convective clouds in a case study from the Tropical Ocean Global Atmosphere Coupled Ocean Atmosphere Response Experiment (TOGA COARE). Results show that a change in sulfate loading for scenarios using the minimum to the maximum observed values produces a change in the average net flux of shortwave radiation above clouds. This time-average change was estimated between -1.1 and -0.3 Wm^{-2} over the integration domain.

Introduction

In addition to the direct atmospheric radiative forcing [*Charlson et al.*, 1992], sulfate aerosols interact with clouds, primarily by providing cloud condensation nuclei (CCN). An increase of sulfate in the atmosphere produces an increase in CCN [*Leaitch et al.*, 1992; *Hegg*, 1994]. CCN in turn, determine the cloud droplet concentration and the size of droplets causing an indirect radiative forcing (IRF) [*Twomey*, 1974; *Slingo*, 1989]. The global average IRF was estimated between -0.5 and -1.5 Wm^{-2} by *Jones and Slingo* [1996]. Most of the previous studies of IRF focused on the dominant role of the stratocumulus clouds in the global climate.

In this study we use the high resolution GFDL LAN model to estimate the effect of sulfate upon the radiative fluxes in tropical convective systems. We attempt these experiments using detailed dynamics of convection driven by observations during TOGA COARE and with sulfate chemistry driven by observations during Pacific Exploratory Mission in the western Pacific Ocean phase B (PEM-West B).

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Method

The LAN model developed at GFDL is a high resolution dynamic model designed for the studies of convective clouds and its essential physics is described by *Held et al.* [1993]. A sulfate-cloud interaction module was added based on *Hegg et al.* [1984] that includes nucleation scavenging of dry aerosol by cloud droplets, rain and snow, aqueous oxidation of SO_2 by H_2O_2 and O_3, and removal by precipitation. In absence of clouds, we included the gas phase oxidation of SO_2 by OH and the condensation of H_2SO_4 on pre-existent sulfate.

A two-dimensional version of the GFDL LAN model is used in this study, with a spatial resolution of 2 km in the horizontal and 500 m in the vertical. The horizontal extent of the domain is 512 km and the vertical extension up to 21 km. The model was integrated to simulate 24 hours, with a time step of 2 s. The model was initialized with vertical soundings from the TOGA COARE site (2^0S, 156^0E) from December 20, 1992, which provide the base state [*Lin and Johnson*, 1996]. The effects of large-scale convergence on temperature and humidity are taken from analysis of the TOGA COARE region. Measurements of SO_2 and $SO_4^{(2-)}$ were taken from the flight observations over the tropical West Pacific, during February-March 1994 [*Hoell et al.*, 1997].

The boundary conditions for the model include the large-scale forcing of potential temperature, water vapor mixing ratio, and horizontal wind from the TOGA COARE vertical soundings, and SST for the lower boundary. Periodic lateral boundary conditions were applied for all dynamic variables, and a sponge-type condition holds at the top of the model to attenuate the gravity waves induced by convection. The model is initialized by applying random small perturbations in the potential temperature in the boundary layer (BL) to trigger convection [*Haywood et al.*, 1997]. The relationship between the predicted sulfate and CCN is based on the empirical results of *Hegg* [1994]. The effective radius of the cloud droplets, r_e, required by the radiation code, is calculated using the relation $r_e = K(d)\bar{r}$, where $K(d) = (1+3d^2)^{2/3}/(1+d^2)$, $d = \sigma/\bar{r}$, with σ the standard deviation of r, and $\bar{r}$ is the average radius (approximated with the mean volume radius), calculated using the condensate mixing ratio and the CCN concentration. Experimental validation of this relationship includes cases of cumulus clouds with entrainment by *Blyth and Latham* [1991], who found $K \approx 1$, trade-wind cumulus by *Pontikis and Hicks* [1992], who found the most frequent values of K to be ~1.05, stratocumulus clouds by *Martin et al.* [1994], who found K=1.07, and precipitating clouds using reported cloud and rain data and a *Marshall and Palmer* [1948] size distribution, for which we found $K \approx 1$ for a large set of conditions. For ice clouds we use the same relationship with the assumption of effective spheres. In summary, the relationship bwtween r_e and $\bar{r}$ is empirically robust for many types of water clouds, even though experimental data for ice clouds is lacking at this time.

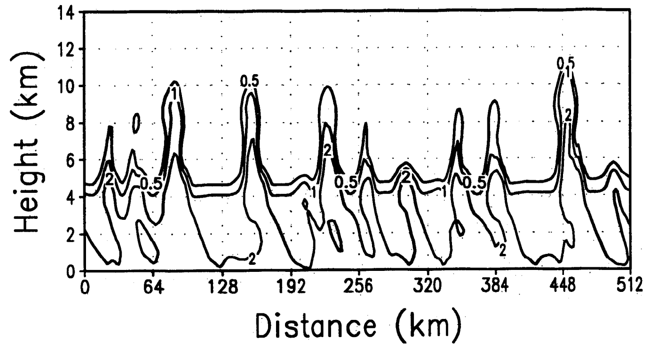

Figure 1. a) Spatial distribution of sulfate (in units of μg kg^{-1}) in the convective cloud field at time t=12 hrs UT, December 20 1992.

Results and discussion

To estimate the effect of sulfate on the LAN model radiative fluxes we compare two runs that correspond to the minimum and maximum observed values of SO_2 and $SO_4^{(2-)}$ during PEM West B in the tropical area. Vertical profiles are generated by an exponential fit of observations with the parameters given in Table 1. The minimum sulfate case will be referred to as Run 1 and is more typical of a clean, remote atmospheric region. The maximum sulfate case, refered to as Run 2 is more typical for polluted scenarios. Another run, corresponding to 10 times the sulfate in Run 2 is refered to as Run 3. This calculation was performed to illustrate the sensitivity of the results to a dramatic increase of sulfate, which is typical in very polluted conditions [*Hegg et al.*, 1984, 1994].

The simulated convective-cloud field for all runs is based on December 20, 1992, of the TOGA COARE experiment. An analysis of the case showed that the relative humidity indicates subsaturated conditions in the first 6 hrs universal time (UT) of the day. Starting at 6 hrs UT, in response to imposed large-scale convergence, conditions for deep convection and precipitation are maintained. A high concentration, (up to ~1.2 g kg^{-1}) of cloud water (CW) is reached between 6 and 9 UT hrs, and values generally decline below 1 g kg^{-1} for the rest of the day. An instantaneous spatial distribution of the total sulfate at time t=12 hrs UT is shown in Figure 1, for the conditions of Run 2. At this time the convective system was fully developed, and the pattern of sulfate concentration resembles that of the clouds. This distribution is mainly the result of transport of sulfate from the BL and incorporation of aerosol in cloud droplets.

Table 1. Parameters of the Initial Vertical Profiles of SO_2 and $SO_4^{(2-)}$ (units: μg kg^{-1})

Run	$SO_2(0)$	$SO_4^{(2-)}(0)$	Comment
1	0.033	0.033	minimum
2	5.061	3.320	maximum

Vertical profiles are given as $SO_2(z)=SO_2(0)\exp(-z/h_{SO_2})$, and $SO_4^{(2-)}(z)=SO_4^{(2-)}(0)\exp(-z/h_{SO_4^{(2-)}})$, with h_{SO_2}=3 km, $h_{SO_4^{(2-)}}$=2 km, and altitude z in kilometers.

The effects of sulfate are illustrated by the differences between Run 1 and Run 2, corresponding to a transition from low to high sulfur loading. Such differences are shown as time series of horizontal averages in Figures 2a, 2b, and 2c, for changes in the effective radius of cloud droplet, r_e, cloud droplet concentration, N_d, and sulfate in cloud water, $q_{SO_4^{(2-)}}$. Figure 2a shows that the increase of sulfate from low sulfate to high sulfate leads to a decrease of r_e of 1-5 μm, with an enhanced difference in the upper troposphere in the second part of the day. (Typical values of r_e are between 3 and 15 μm in the low sulfate case). We estimated that the use of $r_e=K(d)\bar{r}$ in our model introduces an error of about 1 μm for r_e due to uncertainties in K. This error is generally smaller than changes in r_e caused by aerosol variation used in our model calculations. Moreover, it is the changes in r_e between two runs with different sulfate loading which are important in this paper. Our approach gives physically meaningful differences in r_e even if systematic errors in cloud droplet radius are present in both runs. Figure 2b shows the increase of N_d with changes from 0 to about 60 droplets cm^{-3}, which is a result of the availability of more CCN (N_d takes values up to about 50 cm^{-3} in the low sulfate case). Such CCN concentrations are directly caused by the change in sulfate loading, as is illustrated by the $q_{SO_4^{(2-)}}$ in Figure 2c. We note a change up to about 600 ng kg^{-1} which correlates well with both changes in N_d, and r_e ($q_{SO_4^{(-2)}}$ is about 30 ng kg^{-1} in the low sulfate case). According to *Twomey* [1974], for a constant cloud water content, the increase of N_d leads to smaller radii, which has an impact on the radiative fluxes distribution, and particularly on the shortwave radiative flux (SW) above the cloudy domain.

Fluxes of shortwave (SW) and longwave (LW) radiation are calculated above the convective clouds (z=16 km) and at the surface (z=0 km). Each flux has an

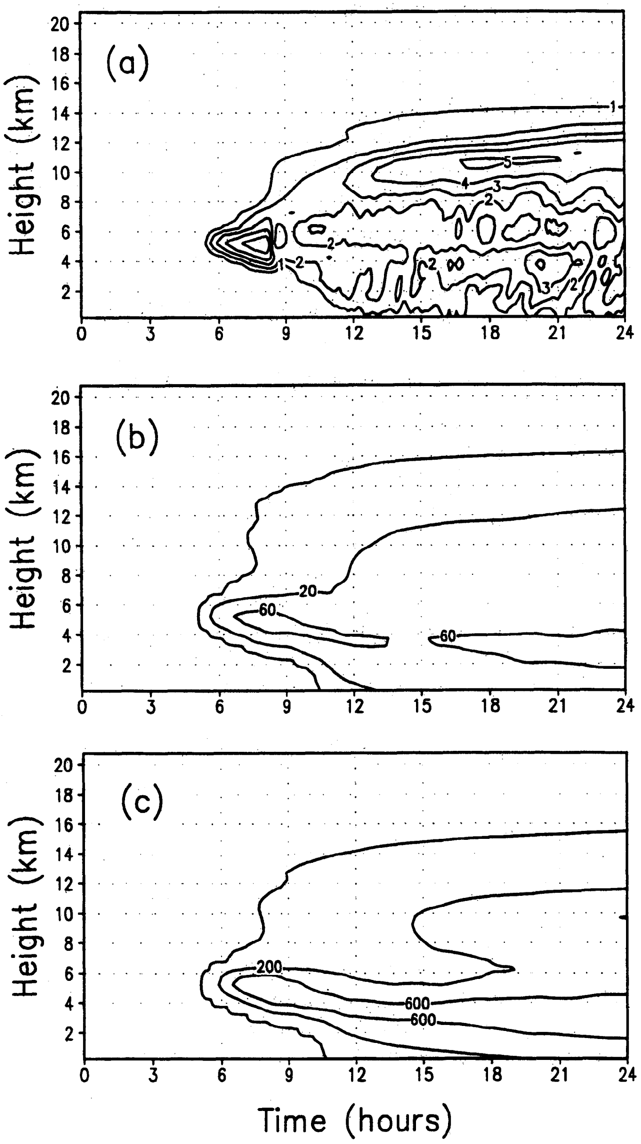

Figure 2. (a) Decrease of the cloud droplet effective radius, r_e (μm), (b) Increase of the cloud droplet number concentration, N_d (droplets cm^{-3}), and (c) Increase of the sulfate concentration in cloud water, $q_{SO_4^{(2-)}}$ (ng kg^{-1}), when sulfate loading changed from the minimum to the maximum observed during the PEM West B. All values are given as a horizontal average over the entire integration domain.

upward (up) and a downward (dn) component. We define $SW_{net}=SW_{dn}\text{-}SW_{up}$, the net SW radiation flux, and $LW_{net}=LW_{dn}\text{-}LW_{up}$, the net LW radiation flux. To obtain a more robust characterization for the entire convective domain, we produce horizontal averages of these fluxes. Moreover, the results are presented in the form of time averages, over several time averaging intervals: 12 hrs (between 12 and 24 hrs UT, an interval dominated by convective activity), 6 hours (between 18 and 24 hrs UT), and 3 hours (between 21 and 24 hrs UT). These last two intervals are chosen to reflect the more prominent effect of sulfate on r_e and N_d, as illustrated in Figure 2.

Changes in radiative fluxes caused by sulfate can be estimated by calculating differences between cases with high sulfate (such as Run 2 and Run 3), and the case with low sulfate (Run 1). The results are given in Table 2. We found that by increasing the sulfate from Run 1 to Run 2, the ΔSW_{net} becomes negative above clouds (illustrated here for z=16 km), estimated between -1.1 Wm^{-2} and -0.3 W m^{-2}, for time averaging of 3 and 12 hours respectively. Systematic negative ΔSW_{net} above the clouds shows that an increase in CCN concentration can lead to an increase in N_d with smaller sizes and thus, can affect the average cloud albedo. Calculations with different averaging intervals show an increase in variability for small-scale averages. An assumed 10 times maximum observed sulfate (Run 3), produces a larger ΔSW_{net}, up to -2.2 W m^{-2} (for a time averaging of 3 hours). Sensitivity of ΔSW_{net} to initial conditions and to variations in CCN-sulfate mass relation as reported by *Hegg* [1994] resulted in negative averages of ΔSW_{net} above clouds, suggesting that the simulated effect of aerosol on cloud albedo is robust.

No changes of ΔLW_{net} were found above the clouds. This is caused by a lack of change in temperature profile above the clouds and by the fact that, to a good approximation, the thick deep convective clouds behave as a black body. Changes in ΔLW_{net} in the regions below the cloud tops were caused by a slight change in the temperature profile as a result of presence of sulfate. For the difference (Run 2-Run 1), ΔLW_{net} has positive and negative signs at the surface depending on the averaging time interval and does not show a systematic

Table 2. Calculated Radiative Flux Changes Induced by Sulfate Increase in Convective Clouds (in W m^{-2})

	(Run2-Run1) [b]		(Run3-Run1) [c]	
Δt [a](hours)	z=0 km	z=16 km	z=0 km	z=16 km
		ΔSW_{net}		
3	-0.6	-1.1	-1.5	-2.2
6	-0.3	-0.5	-0.8	-1.1
12	-0.2	-0.3	-0.4	-0.5
		ΔLW_{net}		
3	-1.1	0.0	1.5	0.0
6	-0.4	0.0	1.3	0.0
12	0.5	0.0	1.0	0.0

ΔSW_{net} and ΔLW_{net} are given as horizontal averages above the clouds (z=16 km), and at the surface (z=0 km).

[a] Δt is the interval of time average.

[b] (Run2-Run1) describes the difference between maximum and minimum observed sulfate.

[c] (Run3-Run1) describes the difference between enhanced sulfate case (10 times the observed maximum) and and minimum observed sulfate.

trend for realistic aerosol concentrations. However, for (Run 3-Run 1), which describes a very significant sulfate aerosol perturbation (not observed in PEM West B data), we found $\Delta LW_{\rm net}$ to be consistently positive. This effect is caused by an increase of temperature in the lower part of the troposphere, an increase of the CW concentration (up to 10 mg kg^{-1}), and an increase of the thickness of anvils. As a result, the LW radiation is emitted toward the Earth, from a region with slightly higher temperature than in the case of low sulfur. While these results suggest that the radiative effect of the sulfate increase is to trap more LW radiation between the clouds and Earth's surface, this process becomes relevant only in intense pollution cases.

Conclusions

The main findings of the impact of sulfate on convective clouds in a TOGA COARE case simulated with the GFDL LAN model are:

1) When changing from a minimum to a maximum-observed sulfate scenario in the tropical West Pacific area, the model indicates a significant decrease of the effective radius of cloud droplets (differences between 1 and 5 μm), an increase in cloud droplet concentration (differences up to about 60 droplets cm^{-3}), and an increase of the sulfate in cloud water (differences up to about 600 ng kg^{-1}). The sulfate distribution in the cloud field resembles the shapes of the cumulus clouds, with enhanced concentrations in updrafts and considerably decreased values in downdrafts.

2) The sulfate increase in the mesoscale convective system produces an average increase of the reflection of SW radiation above the clouds. Time-horizontal averages of SW changes induced by sulfate were estimated between -0.3 (for a time average of 12 hours) and -1.1 W m^{-2} (for a time average of 3 hours). The net LW radiation flux back toward the Earth's surface is increased in conditions of enhanced sulfate loading, an effect caused by the changes in temperature and cloud fields.

While current analysis is restricted to a case study, the sensitivity analysis indicated that the increase of cloud albedo to SW radiation caused by enhanched sulfate aerosol episodes is likely over tropical convective regions. Further work needs to address a better parameterization of ice clouds, and the importance of aerosol variations in convective systems over larger areas.

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