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INFLUENCE OF SULFATE AEROSOL PARTICLES ON RADIATIVE PROPERTIES OF CLOUDS

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Abstract. Tropospheric aerosols, from natural and anthropogenic sources, may have a significant influence on the radiative balance of the Earth by their direct and indirect effects.

The aim of this paper is to study the interactions between aerosol particles (SO_4^{2-}) and the cloud radiative properties. Consequently, using the production parameters and burdens of SO_4^{2-} predicted by eleven chemical transport models, we have calculated the microphysical parameters: liquid water content, cloud droplet number concentration, effective radius, cloud albedo, and the susceptibility (the sensitivity of cloud albedo to changes in cloud droplet number concentration(CDNC)). The relationships proposed by Boucher and Lohmann (1995) and by Menon and Saxena (1998) were used to compare the values of CDNC for maritime and continental clouds. The obtained values of the cloud parameters with these relationships are in agreement with the results from satellitare data for small amounts of sulfate mass concentration. Consequently, new approximations or corrections are necessary for the relationships between aerosol sulfate mass and cloud droplet number concentrations, for higher values of sulfate amount.

Keywords: sulfate aerosol, cloud droplet number concentration, optical thickness, liquid water content, cloud albedo, radiative forcing

1. INTRODUCTION

Aerosols have a direct radiative forcing because they scatter and absorb solar and infrared radiation in the atmosphere. Aerosols also alter the formation and precipitation efficiency of liquid-water, ice and mixed-phase clouds, thereby causing an indirect radiative forcing associated with the changes in cloud properties.

The quantification of aerosol radiative forcing is more complex than the quantification of greenhouse gases radiative forcing because aerosol mass and particle number concentrations are highly variable in space and time. The indirect effects (Twomey, 1977, Albrecht, 1989, and Pincus and Baker, 1994) are far more complex and more difficult to assess than the direct effect, because they depend on a chain of phenomena that connect chemical and physical properties of aerosol to concentration of cloud condensation nuclei (CCN), then CCN concentrations to cloud droplet number concentrations (CDNC) and these, in turn, the cloud albedo.

Therefore, it is a great challenge in adequately characterising the nature and occurrence of atmospheric aerosols and in including their effects in models to reduce uncertainties in climate prediction. Large uncertainties appears because aerosols: originate from a variety of sources, are distributed across a wide spectrum of particle sizes and have atmospheric lifetimes that are much shorter than those of most greenhouse gases, their concentrations and composition having great spatial and temporal variability. Satellite-based measurements of aerosols are a necessary but not sufficient component of an approach needed to acquire an adequate information base upon which progress in understanding the role of aerosols in climate can be built (e.g. Han et al., 1998). Aerosols also affect biogeochemical cycles and human health. Main types of natural aerosols are sea salt, soil dust, aerosols produced by volcanic eruptions and forest fires, organic aerosols, and biogenic sulfates. Anthropogenic aerosols include sulfates, nitrates, black carbon, product of biomass burning, and soil dust (produced by agricultural activities).

Several studies suggest that among the anthropogenic aerosols, sulfates may exert a substantial radiative forcing of climate, comparable in magnitude, but opposite in sign, to the forcing from anthropogenic greenhouse gases. They may also play an important secondary role in climate as a source of cloud condensation nuclei. Thus, the man-made and natural aerosols would have major implications on global climate.

Tropospheric sulfate aerosol is formed from SO_2 oxidation in the gaseous phase and in aqueous phase in cloud drops. The aqueous phase reactions have been estimated to contribute over two-thirds of the sulfate yield in general circulation model simulations of the global sulphur cycle (Pham et al., 1995; Feichter et al., 1996). The conversion was seen to depend upon reactant and oxidant concentrations, cloud pH, temperature and cloud liquid water content, all of which varying with season and region over small scales (Langner and Rodhe, 1991; Feichter et al., 1996). The initial aerosol spectrum and composition are affected both the size-dependent sulfate production rates (Hegg et al., 1992; Kreidenweis et al., 1997) and the resulting droplet spectrum characteristics. Cloud microphysics has been also shown to have a significant effect on the chemistry of SO_2 to sulfate conversion in clouds (Hegg and Larson, 1990; Gurciullo and Pandis, 1997; Kreidenweis et al., 1997; Zhang et al., 1999). Explicit microphysical treatment resulted in higher predicted sulfate formation, from size-dependence of pH and reaction rates not yet accounted for in bulk chemistry models.

Published estimates of the indirect forcing by anthropogenic aerosols are the most uncertain (0 to -1.5 W m^{-2}) relative to other known forcings (IPCC, 2001), with “very low” confidence.

In this paper, we have studied the contributions of different amounts of sulfate aerosol to cloud microphysical properties using the parameterization proposed by Boucher and Lohmann and Menon and Saxena. For this purpose, we are analysing outputs of eleven chemical transport models (CTMs) and we have derived the microphysical parameters for stratocumulus maritime and continental clouds. Datasets and methods used are shown in Section 2. The results related to relationship between CDNC, effective radius, optical thickness, as well as links between cloud albedo and CDNC are presented in Section 3. Summary and conclusions are in the last section.

2. DATASETS AND METHOD

The interactions between atmospheric radiation and dynamics, cloud microphysics, aerosols, and thermodynamics are not fully understood. In many cases, predicted climate changes (connected to the radiative effects of aerosols) are dependent on the cloud parameterization used in a particular general circulation model. The link between sulfate aerosols and clouds is an important one because, although there are several types of aerosol particles that can serve as a cloud condensation nuclei, some of the most effective are the sulfate particles formed from the oxidation of SO_2.

Our current research concerns the potential effects of sulfates aerosols on cloud optical properties, the goal being to understand the relative contributions of different amounts of sulfate aerosols to properties of clouds. We have estimated the radiative properties of stratocumulus clouds and we have investigated the relationships among these properties as well as the processes controlling them. We have also investigated the changes in clouds reflectivity for the two spectral intervals of the solar part of electromagnetic spectrum (Fouquart and Bonnel, 1980): the visible (0.2 ÷ 0.68 μm), and near infrared (0.68 ÷ 4.0 μm). In this discussion, we shall ignore the perturbation of the atmospheric sulphur cycle induced by manmade fluxes of SO_2 (mostly from burning of oil and coal) and shall consider the anthropogenic and natural sources of sulphur for the Northern Hemisphere.

The cloud microphysical parameters that we have investigated are: liquid water content (L), cloud droplet number concentration (CDNC), effective radius (R_{eff}), cloud albedo (A), and the susceptibility (S), which means the sensitivity of cloud albedo to changes in cloud droplet number concentration.

The cloud albedo, A, was computed taking into account the two-stream approximation of a nonabsorbing, horizontally homogeneous cloud (Lacis and Hansen, 1974):

$$A = \frac{\sqrt{3}(1-g)\tau}{2+\sqrt{3}(1-g)\tau} \tag{1}$$

where τ is the optical depth of the cloud, and g denotes the asymmetry factor.

The effective radius R_{eff} is defined by the ratio between the third moment of the size spectrum and its second moment (Hansen and Travis, 1974):

$$R_{eff} = \frac{\int_0^\infty N(r) r^3 dr}{\int_0^\infty N(r) r^2 dr} \tag{2}$$

where r is the radius of the cloud droplet and N(r) is the number concentration of cloud droplets.

Han et al. (1998) reported that for most continental clouds and all optically thick clouds ($\tau > 15$) over most the world, cloud albedo increases with decrease of R_{eff}, and for optically thin clouds ($\tau < 15$) (Lohmann et al, 2000) over oceans and tropical rain forest areas, cloud albedo decreases with decrease of R_{eff}, in which case τ varies with square power of R_{eff}:

$$\tau = 2 \cdot \pi \cdot (R_{eff})^2 \cdot N \cdot \Delta z \qquad (3)$$

where N is the cloud droplet number concentration and it is assumed to have a constant value over the vertical extent of cloud. The underlying reason is the correlation between effective radius and liquid water path (the product of liquid water content and the cloud height). The results reported by Han et al. (1998) show that for clouds over most regions of the world and for all seasons, the LWP decrease as far as the effective radius decrease. These results suggest that, as LWP is not always constant, this implies cloud dynamic feedbacks may affect the magnitude of the indirect effect of aerosols. This approximation is a reasonable one for thick boundary-layer clouds over mid-latitude oceans and we shall suppose it valid for lower clouds over the continental areas, too.

For the asymmetry factor, we have considered the values of 0.865 for visible and 0.910 for near infrared domain of spectrum. These are currently used values in the general circulation model (GCM) of the Laboratoire de Meteorologie Dynamique (LMD).

The liquid water content (g/m^3), cloud droplet number concentration (cm^{-3}) and the effective radius (μm) are related by the following equation:

$$L = \frac{4}{3}\pi R_{eff}^3 \rho N \qquad (4)$$

where ρ is the water density.

The aerosol mass, derived from chemical transport models (CTMs), was related to cloud droplet concentration by the relationships proposed by Boucher and Lohmann (equations 5a, abbreviated as BL) and Menon and Saxena (equations5b, abbreviated as MS), as following:

A1: $CDNC_{cont}^{st} = 10^{2.24+0.257\log(mSO_4)}$ - stratiform continental clouds

A2: $CDNC_{cont}^{cu} = 10^{2.54+0.186\log(mSO_4)}$ - cumulus continental clouds

(5a)

A3: $CDNC_{ocean}^{st} = 10^{2.06+0.48\log(mSO_4)}$ - stratiform maritime clouds

A: $CDNC = 10^{2.21+0.41\log(mSO_4)}$ - all types of clouds, to consider the convective processes, and respectively:

B1: $CDNC_{cont}^{st} = 10^{2.15+0.52\log(mSO_4)}$

B2: $CDNC_{cont}^{st} = 10^{2.33+0.47\log(mSO_4)}$

(5b)

B3: $CDNC_{cont}^{st} = 10^{2.29+0.49\log(mSO_4)}$

B: $CDNC = 10^{2.21+0.54\log(mSO_4)}$,

where mSO$_4$ are expressed in μg m^{-3}, and CDNC in cm^{-3}.

The production parameters and burdens of sulphur dioxide and aerosol sulphate as predicted by different chemical transport models, which have included the main oxidation pathways through the sulfate aerosols result from sulphur dioxide by gaseous and aqueous phase oxidations, as well as the sinks by dry and wet depositions are summarized in Table 1.

Table 1
Production parameters and burdens of sulphur dioxide and aerosol sulphate as predicted by eleven different models (from IPCC, 2001).
Model/Reference: A MOGUNTIA/Langner and Rodhe, 1991; B: IMAGES/Pham et al., 1996; C: ECHAM3/Feichter et al., 1996; D: Harvard-GISS / Koch et al., 1999; E: CCM1-GRANTOUR/Chuang et al., 1997; F:ECHAM4/Roelofs et al., 1998; G: CCM3/Barth et al., 2000 and Rasch et al., 2000a; H: CCC/Lohmann et al., 1999a.; I: Iversen et al., 2000; J: Lelieveld et al., 1997; K: GOCART/Chin et al., 2000.

Model	Sulphur source TgS/yr	Precursor deposition %	Gas phase oxidation %	Aqueous oxidation %	SO_2 burden TgS	Sulphate dry deposition %	Sulphate wet deposition %	SO_4^{2-} burden TgS
A	94.5	47	8	45	0.30	16	84	0.77
B	122.8	49	5	46	0.20	27	73	0.80
C	100.7	49	17	34	0.43	13	87	0.63
D	80.4	44	16	39	0.56	20	80	0.73
E	106.0	54	6	40	0.36	11	89	0.55
F	90.0	18	18	64	0.61	22	78	0.96
G	82.5	33	12	56	0.40	7	93	0.57
H	95.7	45	13	42	0.54	18	82	1.03
I	125.6	47	9	44	0.63	16	84	0.74
J	90.0	24	15	59	0.60	25	75	1.10
K	92.5	56	15	27	0.43	13	87	0.63

A short analysis of data presented in Table 1 lead us to the fact that uncertainties in the input parameters in the CTMs lead to differences in the atmospheric burdens of sulfate aerosols. This is, in part, because the CTM models differ from the point of view of horizontal resolutions. Moreover, some of them, such as IMAGES and Harvard-GISS, include a more detailed chemical scheme, with more chemical species, to describe more properly the gas phase chemistry. Other, such as ECHAM 3, ECHAM 4, and GCM 3 offer a detailed representation of processes in aqueous phase. Furthermore, other models have a better representation of the involved physical processes. All these considerations suggest that the amount of burden sulphate aerosol is very dependent of the used chemical transport model. Having briefly discussed the results from the CTMs, we now focus on the estimates of microphysical and optical parameters of clouds.

3. RESULTS AND DISCUSSION

We have examined the effect of holding constant of one of among variables (CDNC, R_{eff}, and L) while the other two changes, for a cloud with a geometrical thickness of 500 m. This type of analysis was begun by Twomey, in 1977. Keeping R_{eff} fixed means that the increase in cloud albedo is due to increase of CDNC. On the other hand, we have considered the effect of holding L constant while CDNC increase. This lead to a decrease in droplet radius in agreement with the results reported by Charlson et al. (1987) for sulfate aerosol resulting from dimethylsulphide (DMS) from oceanic phytoplankton. Such decrease causes an increase in total surface area of cloud droplets and thus an increase of cloud albedo. The values we have considered for R_{eff} and L (Table 2) have been resulted from the analysis of outcomes from literature since 1980s (eg. Martin et al., 1994, Slingo and Schrecker, 1982). The column 4 contains the mediated values of these parameters, which have been used for calculus with formulae A and B (equations 5a and 5b).

Table 2

Values of effective radius and liquid water content for every studied type of cloud

	Continental Stratiform Clouds	Maritime Stratiform Clouds	Continental Cumuliform Clouds	Mediated values
R_{eff} (μm)	7,43	10,5	3,46	9,13
L (g/m^3)	0,10	0,15	0,30	0,18

1. Relationship between CDNC, cloud albedo and sulfate mass concentration

To clarify the role of burdens of SO_4 on clouds reflectivity, we have examined the correlated changes of CDNC and mSO_4 for all cloud types indicated in Table 2. Figures 1÷ 4 show the results for maritime stratiform clouds. It can see that the ratio CDNC (MS) to CDNC (BL) is equal to 1 for $0.5 < mSO_4 < 0.75$ TgS, and is greater (1.75) in the case $0.75 < mSO_4 < 1.1$ TgS. This suggests that results are the same for BL and for MS parameterisations for small amounts of sulfate and lead to very high differences for bigger amount of sulfate.

Concerning the cloud albedo, the relationships MS and BL lead to different values in the sense that the differences are higher for the small concentrations of sulfate aerosols (Figure 2).

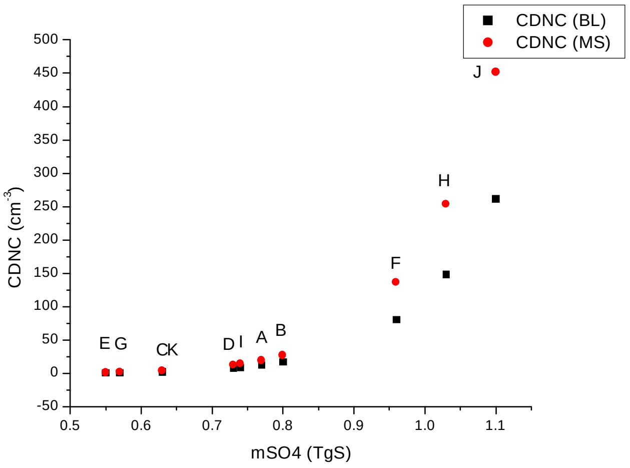

Fig. 1.- CDNC dependence of sulfate mass concentration of SO_4, obtained from models (Table 1) using the Boucher & Lohmann relation (square dots), and Menon & Saxena (circle dots) for maritime stratiform clouds

These results confirm the experimental observations, namely the stratiform maritime clouds albedo increase with the increase of cloud droplet concentration and depend, in fact, of sulfate aerosol mass concentration. The CDNC calculations as a function of mass concentration that we have derived agree with those reported by Haywood and Boucher (2000).

For high values of CDNC, the cloud albedo is not sensitive to variations of cloud droplet number concentration.

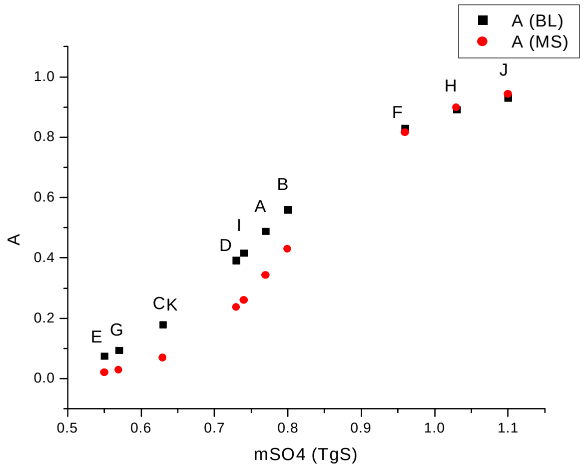

Fig 2. - Estimates of cloud albedo, corresponding to computed CDNCs from Figure 1

This conclusion has been strengthen by the insignificant variation of liquid water content (Figure 3) and of the optical thickness (Figure 4) as a function of sulfate mass concentrations for both relationships sets for small amount of sulfate aerosol, where the liquid water content (L) and optical thickness (τ) are holding constant

The figures 3 and 4 show a very good accordance between the values sulfate mass concentrations computed with the BL and MS parameterizations in the range $0.5 < mSO_4 < 0.75$ TgS, and a good one in the case $0.75 < mSO_4 < 1.1$ TgS.

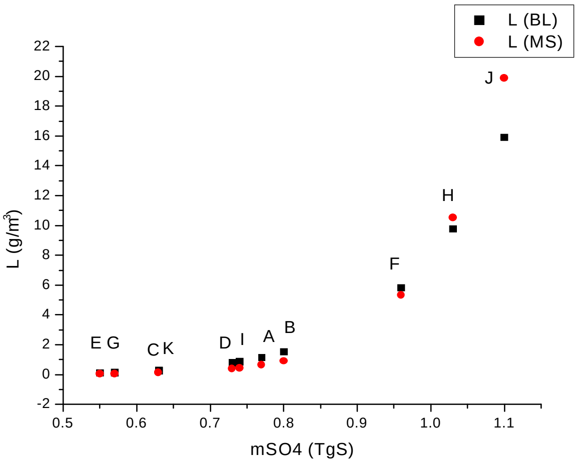

Fig. 3- Estimates of liquid water content, corresponding to computed CDNCs from Figure 1

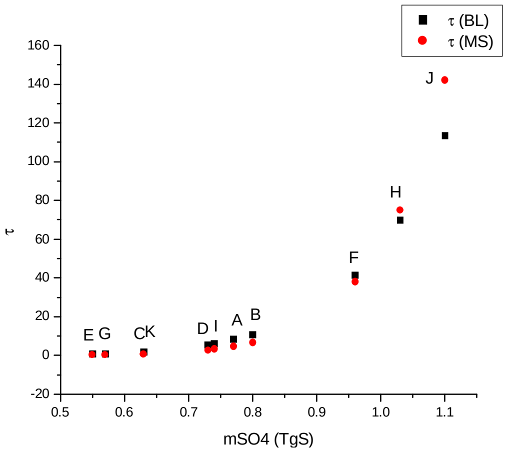

Fig. 4.- Estimates of optical thickness, corresponding to computed CDNCs from Figure 1

Although, the ECHAM 4 model, the CCC model and the model proposed by Lelieveld et al. (1997) predict large values of sulfate aerosol mass concentrations. Correspondingly, we have derived the high values for L and τ with both parameterizations, BL, and MS. This fact lead us to the following idea: the assumption that all particles of sulfate aerosol act as cloud condensation nuclei (CCN), and subsequently all CCN are activated to CDNC, cannot remain valid for $mSO_4 > 0.9$ TgS.

For continental cumuliform clouds (Figures 5 and 6) the results are related to cloud microphysical properties. The relationships proposed by Boucher and Lohmann, and Menon and Saxena reflect in differentiate way the dynamic processes.

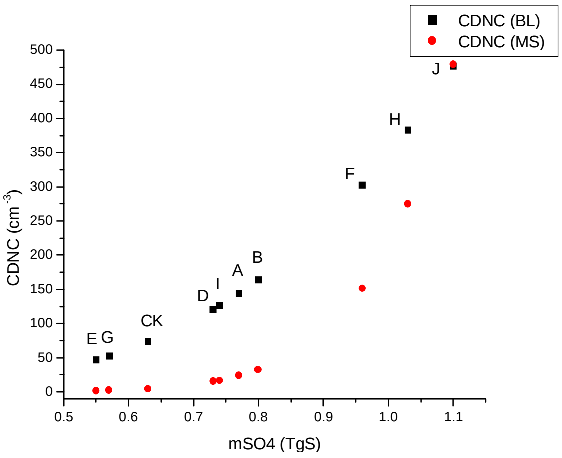

Fig. 5.- Cloud droplet number concentration as a function of sulfate mass concentration of SO_4, using the Boucher & Lohmann relation (square dots), and Menon & Saxena (circle dots)-cumuliform continental clouds

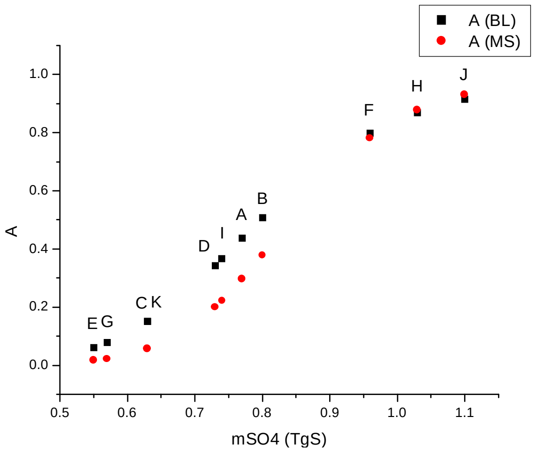

Fig 6- Estimates of cloud albedo, corresponding to computed CDNCs from Figure 5

Therefore, we have obtained very different values for cloud droplet number concentrations and cloud albedo, too. Thus, the ratio CDNC(MS) to CDNC(BL) reaches a maximum value for 0.8 TgS, and is equal to 1 for 1.1 TgS. Concerning the cloud albedo, the ratio A(MS) to A(BL) is relatively constant for $mSO_4 > 0.95$ TgS.

The distribution of CDNC is primarily a reflection of the distribution of sulfate aerosol mass both for continental and for maritime clouds. Moreover, the used parameterizations predict more cloud droplets for continental clouds than those for maritime clouds at low sulfate concentration (Figures 1 and 5). This feature seems to be reasonable because the processes governing the aerosol formation are different, and on the other hand, there are significant differences concerning the cloud dynamics.

Because the magnitude of indirect effect of aerosols may be dependent of susceptibility of clouds to changes in cloud droplet number concentrations, we have computed the cloud susceptibility for two spectral intervals: 0.2 ÷ 0.68 μm (visible), and 0.68 ÷ 4.0 μm (near-infrared).

The absolute cloud susceptibility is defined as the change that takes place in cloud albedo for a given change in cloud droplet number concentration:

$$S = \frac{dA}{dN} = \frac{A(1-A)}{3 \cdot N}$$

(6)

where N denotes the CDNC.

Figures 7 and 8 show the results for the dependence of S (cm^3) of variation of cloud albedo, for BL parameterization. The MS parameterization leads to much closed values (not shown here) of presented estimations.

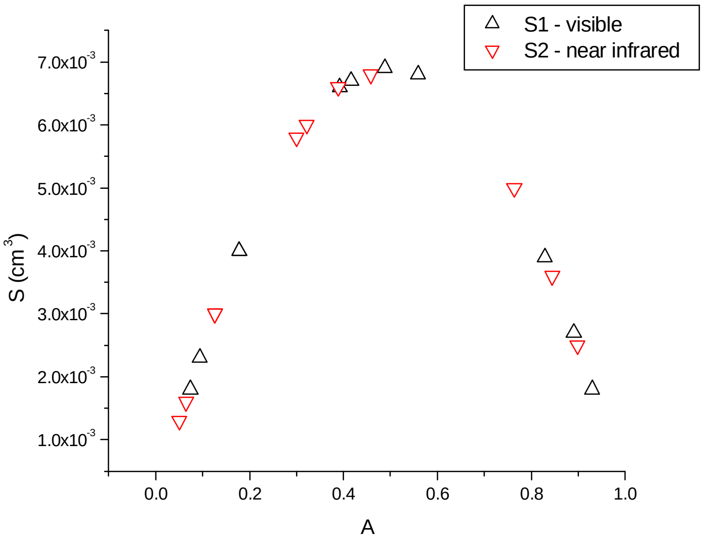

Fig. 7- Calculated susceptibility as a function of cloud top albedo for visible and near-infrared for stratiform maritime clouds for BL parameterization

The magnitude of susceptibility can be very dependently on the assumed relationship between sulfate aerosol mass and CDNC and on the cloud type. There is a difference of one order size between the susceptibility for stratiform as compared with the cumuliform clouds. One can also observe the high susceptibility in the region 0.35 < A < 0.6 for stratiform clouds, and in area of 0.3 < A < 0.7 for cumulus clouds.

This suggests that it may be of great importance to take into account the high albedos of clouds in evaluating the indirect aerosol forcing effect.

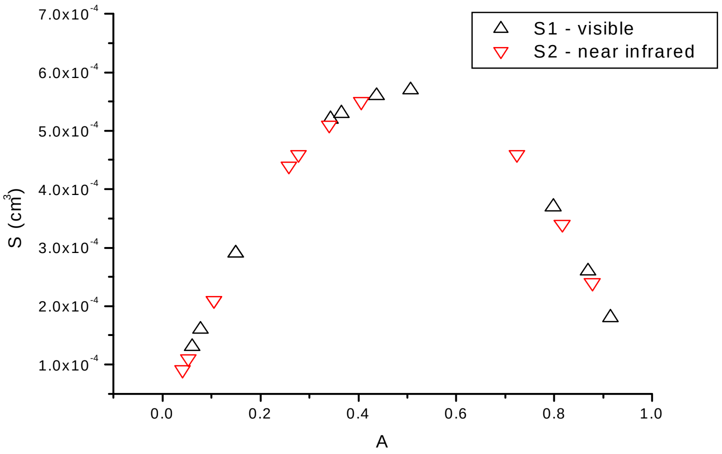

Fig. 8- Calculated susceptibility as a function of cloud top albedo for visible and near-infrared for cumuliform continental clouds for BL parameterization

Our estimates for albedo values as function of CDNC for both spectral intervals (not shown in this paper, see Figures 1, 2 and 5, 6) indicate higher values for the visible range than for near infrared. Moreover, the estimates for cloud albedo are quite high for mSO_4 > 0.9 TgS. This suggests us that the two-stream approximation of a non-absorbing cloud (Lacis and Hansen, 1974) may be a crude one for the 0.68 ÷ 4.0 μm spectral interval, because in this range the absorbtivity of water vapours is high.

However, at high sulfate concentrations, which determine high droplet concentrations, and for higher liquid water content, is possible to appear a competing effect between the multiple scattering (could increase the reflectivity) and the absorption properties of aerosol and of liquid water (could decrease the cloud reflectivity).

b. Relationships between effective radius, optical thickness and CDNC

We think that the both the microphysics and the optical properties of clouds are determinant for the estimates of the magnitude of global mean forcing that may be attributable to anthropogenic aerosol. Therefore, we have analysed the connexions between optical thickness, effective radius and cloud droplet number concentrations, for stratiform maritime clouds and for cumuliform continental clouds (Figure 9 and, respectively 10).

The results reported by Lohmann et al (2000) from analysed satellite data show that for maritime clouds with τ > 15 exist a correlation between cloud albedo and the effective radius, and for $\tau < 15$ there is an anticorrelation between these parameters. They attribute this change in sign of the correlation to precipitating versus non-precipitating clouds. They also reported that if $\tau = 10$ is used as a separator, the precipitation formation occur less often in clouds with $\tau < 10$ than in clouds with $\tau > 10$.

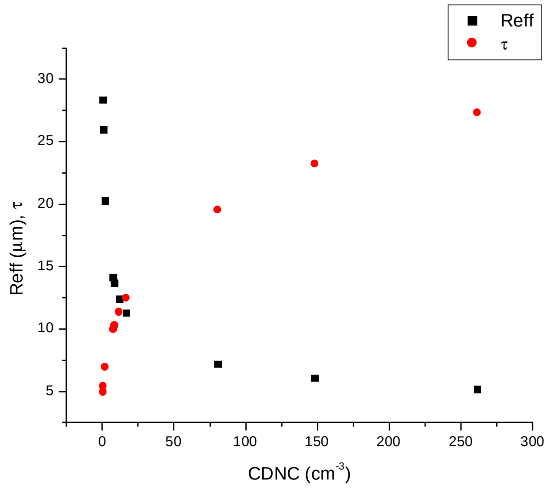

Fig. 9. Calculated effective radius and optical thickness as a function of CDNC for stratiform maritime clouds

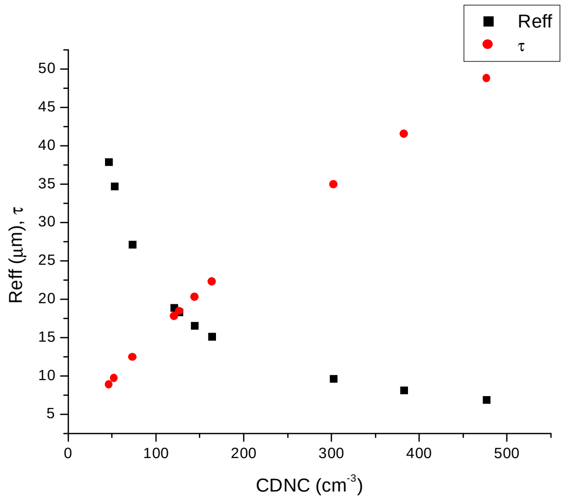

Fig. 10. Calculated effective radius and optical thickness as a function of CDNC for cumuliform continental clouds

Our results show that $\tau = 15$ is obtained for an approximate value of CDNC of 35 cm^{-3}, which correspond to an effective radius of 11 μm in stratus maritime clouds, and for 100 cm^{-3} and 25 μm in cumulus continental clouds. In addition, for the same concentration of cloud droplet, the effective radius is bigger for continental than for maritime clouds. The difference is more pronounced when we choose τ =10 as a separator value, from 15 μm for maritime to 35 μm for continental clouds. One can see that, for both types of clouds, for $\tau > 15$, R_{eff} decrease with the increase of CDNC, and for $\tau < 15$, R_{eff} increase with the decrease of CDNC, increase which is more pronounced for maritime stratiform clouds. These results are in good agreement with that reported by Han et al. (1998), that found that a decrease of R_{eff} does not necessarily lead to a more reflective cloud, and those of Lohmann et al. (2000) .

Nakajima et al. (2000) examined the variation of cloud albedo, droplet concentration, and effective radius with column aerosol concentration, too. They found that the cloud droplet column concentration increased while R_{eff} decreased over a range of values of column aerosol number concentration. They did not find a significant increase in liquid-water path as aerosol number concentration increased. This observation may indicate that the indirect effect is not so important on a global scale; however, further work is needed to confirm this. However, the lower clouds seem to be very sensitive to anthropogenic influences by variation in CDNC, whereby they is possible to have a stronger influence to the planetary albedo. Again, further work is necessary.

4. SUMMARY AND CONCLUSIONS

An assessment of the effects of sulfate aerosol to the reflectivity of clouds has been performed. This study was concentrated on the role of sulfate aerosols but other aerosol components may be important. The inclusion of other aerosol types (nitrates, soot, and dust) remains as a subsequent study, as well as a comparative study on the differences induced by the different emissions of sulphur dioxide for the Northern and Southern Hemisphere.

Our study shows that new approximations or corrections are necessary for the relationships between aerosol sulfate mass and cloud droplet number concentrations, at least for higher values of sulfate amounts. The parameterizations proposed by Boucher and Lohmann (1995), and Menon and Saxena (1998) cannot be considered as universally valid. Furthermore, these relationships do not take into account the links between aerosol mass and cloud condensation nuclei. It is possible that this constitute one of the major uncertainties in estimating the cloud properties changes via changing aerosol concentrations. Clouds of liquid water droplets form only in the presence of CCN and a supplementary uncertainty through all the CCN have activated may be induced by such a supposition.

The results also show the two-stream approximation of a non-absorbing cloud (Lacis and Hansen, 1974) is a crude one for the 0.68 ÷ 4.0 μm spectral interval, because in this range the absorbtivity of water vapours is high.

It is essential that we need more observational data for the critical evaluation of the results of CTMs, in order to strengthen confidence in the ability of CTMs to model constituent fields. The quantification of climate forcing by aerosols, together with climate model calculations of response, provides a unique method of testing climate models. The GCMs allow a consistent interaction between radiative forcings and atmospheric circulation.

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REFERENCES

1. Barth, M.C., P.J. Rasch, J.T. Kiehl, C.M. Benkowitz, and S.E. Schwartz, (2000): Sulphur chemistry in the NCAR CCM: description, evaluation, features and sensitivity to aqueous chemistry, *J. Geophys. Res.*, **105**, 1387-1416
2. Boucher, O., Lohmann, U. (1995): The sulfate-CCN-cloud albedo effect: a sensitivity study with two general circulation models, *Tellus*, 47B, 281-300;
3. Charlson, R. J., Lovelock, J. E., Andreae, M. O., Warren, S. G. (1987): Oceanic phytoplankton, atmospheric sulphur, cloud albedo and climate, *Nature*, 326, 655-661;
4. Chin, M., R.B. Rood, S.-J. Lin, J.-F. Muller, and A. M. Thompson (2000): Atmospheric sulphur cycle simulated in the global model GOCART: Model description and global properties, *J. Geophys. Res.*, **105**, 24671-24687;
5. Chuang, C.C., J.E. Penner, K.E. Taylor, A.S. Grossman, and J.J. Walton, (1997): An assessment of the radiative effects of anthropogenic sulphate, *J. Geophys. Res.*, **102**, 3761-3778;
6. Feichter, J., Kjellstrom, E., Rodhe, H., Dentener, F., Lelieveld, J. and Roelofs, G. (1996): Simulation of the tropospheric sulfur cycle in a global climate model, *Atmos. Environ* 30, 1693-1707;
7. Fouquart, Y., Bonnel, B., (1980): Computations of solar heating of the Earth's atmosphere: a new parameterization, *Beitr. Phys. Atmos.*, **53**;
8. Gurciullo, C. S., and Pandis, S. N. (1997): Effect of composition variations in cloud droplet populations on aqueous-phase chemistry, *J. Geophys. Res.*, 102, 9375-9385;
9. Han, Q., Rossow, W. B., Chou, J. and Welch, R. M (1998): Global survey of the relationship of cloud albedo and liquid water path with droplet size using ISCCP, *J. Climate*, 11, 1516-1528;
10. Hansen, J.E., Travis, L.D., (1974): Light scattering in planetary atmospheres, *Space Sci. Rev.*, **16**, 527–610;
11. Haywood, J., and Boucher, O. (2000): Estimates of the direct and indirect radiative forcing due to tropospheric aerosols: a review, *Rev. Geophys.*, 38,4, 513-543;
12. Hegg, D. A., and Larson, T. V. (1990): The effects of microphysical parameterization on model predictions of sulphate production in clouds, *Tellus* 42B, 272-284;
13. Hegg, D. A., Yuen, P.-F., and Larson T. V. (1992): Modelling the effects of heterogeneous cloud chemistry on the marine particle size distribution, *J. Geophys. Res.*, 97, 12927-12933;
14. Intergovernmental Panel on Climate Change (IPCC), (2001): Climate Change, The Scientific Basis, (2001): Aerosols, their direct and indirect

effects, Intergovernmental Panel on Climate Change, Cambridge University Press, New York;

15. Iversen, T., A. Kirkevåg, J. E. Kristjansson, and Ø. Seland, 2000: Climate effects of sulphate and black carbon estimated in a global climate model. In *Air Pollution Modeling and its Application XIV*, S.-E. Gryning and F.A. Schiermeier, Eds. Kluwer/Plenum Publishers, New York, 335-342;
16. Koch, D., D. Jacob, I. Tegen, D. Rind and M. Chin, 1999: Tropospheric sulphur simulation and sulphate direct radiative forcing in the Goddard Institute for Space Studies general circulation model. *J. Geophys. Res.*, **104**, 23,799-23,822;
17. Kreidenweis, S. M., Zhang, Y., and Taylor, G. R. (1997): The effects of clouds on aerosol and chemical species and distributions 2. Chemistry model, description and sensitivity analysis, *J. Geophys. Res.*, 102, 23867-23882;
18. Lacis, A.A., Hansen, J.E., (1974): A parameterization for the absorption of solar radiation in the Earth’s atmosphere, *J. Atmos. Sci.*, **31**;
19. Langner, J. and Rodhe, H. (1991): A global three dimensional model of the tropospheric sulfur cycle, *Atmos. Chem.* 13, 225-263;
20. Lelieveld, J., G.J. Roelofs, L. Ganzeveld, J. Feichter, and H. Rodhe, 1997: Terrestrial sources and distribution of atmospheric sulphur, *Phil. Trans. R. Soc. Lond. B.*, **352**, 149-158;
21. Lohmann, U., K. Von Salzen, and N. McFarlane, H.G. Leighton, and J. Feichter, (1999): The tropospheric sulphur cycle in the Canadian general circulation model. *J. Geophys. Res.*, **26**,833-26,858;
22. Lohmann, U., Tselioudis, G., Tyler, C., (2000): Why is the cloud albedo-particle size relationship different in optically thick and optically thin clouds?, *Geophys. Res. Lett.*, **27**, 8;
23. Martin, G. M., Johnson, D.W., Spice, A., (1994): The measurement and parameterization of effective radius of droplets in warm stratocumuls clouds, *J. Atmos. Sci.*, **51**, 13, 1823-1842;
24. Menon, S., Saxena, V.K. (1998): Role of sulfate in regional cloud-climate interaction, *Atmos. Res*. 47, 299-315;
25. Nakajima, T., Tsukamoto, M., Tsushima, A.,Sumi, A. (2000): modellimg of the radiative process in an atmospheric general circulation model, *Appl.Opt.*, 39, 4869-4878;
26. Pham, M., Muller, J.-F., Brasseur, G. P., Granier, C. and Megie, G. (1995): A three dimensional study of the tropospheric sulphur cycle, *J. Geophys. Res.*, 100, 26061-26092;
27. Pham, M., J.-F. Müller, G. P. Brasseur, C. Granier, and G. Mägie, 1996: A 3D model study of the global sulphur cycle: Contributions of anthropogenic and biogenic sources, *Atmos. Env.*, **30**, 1815-1822;
28. Rasch, P.J, M.C. Barth, J.T. Kiehl, S.E. Schwartz and C.M. Benkowitz, 2000a: A description of the global sulphur cycle and its controlling processes in the NCAR CCM3, *J. Geophys. Res.*, **105**, 1367-1386;
29. Roelofs, G.-J., J. Lelieveld and L. Ganzeveld, 1998: Simulation of global sulphate distribution and the influence on effective cloud drop radii with a coupled photochemistry-sulphur cycle model, *Tellus*, 50B, 224-242;

30. Slingo, A., Schrecker, H. M., (1982): On the shortwave radiative properties of stratiform water clouds, *Quart. J. Roy. Meteor. Soc.*, **108**, 407–426;
31. Twomey, S., The influence of pollution on the shortwave albedo of clouds, (1977): *J. Atmos. Sci*., **34**, 1149–1152;
32. Zhang, Y., Kreidenweis, S. M. and Feingold, G. (1999): Stratocumulus processing of gases and cloud condensation nuclei 2. Chemistry sensitivity analysis, *J. Geophys. Res*., 104, 16061-16080.