Constraining the first aerosol indirect radiative forcing in the LMDZ GCM using POLDER and MODIS satellite data

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[1] The indirect effects of anthropogenic aerosols are expected to cause a significant radiative forcing of the Earth's climate whose magnitude, however, is still uncertain. Most climate models use parameterizations for the aerosol indirect effects based on so-called "empirical relationships" which link the cloud droplet number concentration to the aerosol concentration. New satellite datasets such as those from the POLDER and MODIS instruments are well suited to evaluate and improve such parameterizations at a global scale. We derive statistical relationships of cloud-top droplet radius and aerosol index (or aerosol optical depth) from satellite retrievals and fit an empirical parameterization in a general circulation model to match the relationships. When applying the fitted parameterizations in the model, the simulated radiative forcing by the first aerosol indirect effect is reduced by 50% as compared to our baseline simulation (down to −0.3 and −0.4 Wm^{-2} when using MODIS and POLDER satellite data, respectively). **Citation:** Quaas, J., and O. Boucher (2005), Constraining the first aerosol indirect radiative forcing in the LMDZ GCM using POLDER and MODIS satellite data, *Geophys. Res. Lett.*, *32*, L17814, doi:10.1029/2005GL023850.

1. Introduction

[2] The aerosol indirect effects are estimated to be the most uncertain forcings of the Earth's climate, with estimates deduced from modeling studies varying from −2 to 0 Wm^{-2} [*Ramaswamy et al.*, 2001] for the first aerosol indirect effect. Recent studies comparing forward and inverse model calculations suggest that the aerosol radiative forcings may be overestimated in current climate models [*Anderson et al.*, 2003]. The first aerosol indirect effect is defined as the reduction in cloud droplet size with (anthropogenically) increasing aerosol concentration at fixed cloud water due to the ability of aerosols to serve as Cloud Condensation Nuclei (CCN) [*Twomey*, 1974]. The anti-correlation between cloud droplet size and aerosol concentration might thus be defined as a measure of the first aerosol indirect effect [e.g., *Feingold et al.*, 2003] and expressed as IE = $\Delta \ln r_e / \Delta \ln (\alpha\tau_a)$, where r_e is the cloud droplet "effective" radius (CDR, in μm) and $\alpha\tau_a$ the aerosol index (AI), which may be interpreted as the product of the Ångström coefficient α and the aerosol optical depth (AOD, τ_a). Alternative definitions use the aerosol optical depth or the column aerosol number concentration instead of the aerosol index. The CDR to AI relationship has been established on large scales using Advanced Very High Resolution Radiometer (AVHRR) data by *Nakajima et al.* [2001], who found IE = −0.17 over ocean, and using POLarization and Directionality of the Earth's Reflectances (POLDER) measurements by *Bréon et al.* [2002] giving IE = −0.085 over oceans and −0.04 over land. For the coarse resolution which is also used in this study, *Quaas et al.* [2004] give a smaller value deduced from POLDER data of −0.04 over oceans and −0.01 over land. Similarly, *Sekiguchi et al.* [2003] show that the absolute value of IE is smaller for a coarser horizontal resolution.

[3] Comparing the CDR to AI relationships from simulations with the ECHAM GCM and from POLDER satellite data, *Lohmann and Lesins* [2002] find that the relationship is too steep in their GCM. They scale the radiative forcing simulated by their model by the factor equal to the ratio of the IE deduced from POLDER observations and from the model simulation. This leads to a reduction in the forcing from −1.28 to −0.98 Wm^{-2} over oceans and from −1.62 to −0.53 Wm^{-2} over land, yielding a global mean value of −0.85 Wm^{-2}. Using a radiative transfer model and aerosol optical depth to CDR relationships derived from the same POLDER observations, *Sekiguchi et al.* [2003] derive radiative forcings due to the aerosol indirect effect (AIE) of −0.14 to −0.28 Wm^{-2} depending on the method used. These values are also much smaller than the ones derived from current GCMs confirming our suspicion that the AIE may be overestimated in current models.

[4] In the present study, we seek a parameterization of the AIE for a GCM which is able to reproduce the CDR to AI (or AOD) relationship found by the satellite observations in order to constrain the aerosol indirect radiative forcing (AI and AOD are applied when using the two different satellite datasets, respectively). Current satellite data are now well suited to evaluate parameterizations in global models. We use for this purpose satellite retrievals from the POLDER and MODerate resolution Imaging Spectroradiometer (MODIS) instruments. We focus here on the "first" AIE, and which has been included in many of the recent IPCC climate simulations. The mechanisms and magnitude of the second aerosol indirect effect (or cloud lifetime effect) are much more uncertain, and parameterizations are not well evaluated in global models. We neglect this effect therefore in the present study, and assume in particular that it does not

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have a large influence on the relationships we investigate. This assumption has to be evaluated when more observational constraints for the cloud lifetime effect exist.

2. Method

2.1. POLDER and MODIS Satellite Data

[5] The POLDER-1 instrument on board the ADEOS-1 platform provided observational data from November 1996 until June 1997. Here we use the POLDER aerosol index (AI), which is retrieved using the total and polarized reflectances at 670 and 865 nm over ocean, and the polarized reflectances only over land [*Deuzé et al.*, 1999]. The cloud droplet effective radius (CDR) is retrieved from the rainbow features of the cloud droplet polarized phase function [*Bréon and Goloub*, 1998; *Bréon and Colzy*, 2000]. The retrieval is representative of the cloud top and is limited to liquid water clouds which are homogeneous at a horizontal scale of 150×150 km^2.

[6] MODIS data are available since March 2000 from the Terra satellite. We use data for the March 2000–February 2001 period. Aerosol optical depth is derived at 550 nm [*Remer et al.*, 2005]. Cloud droplet effective radius is derived in the absorbing band at 2.1 μm assuming plane-parallel homogeneous clouds above a black surface in combination with a non-absorbing band at 0.65, 0.86, and 1.2 μm over land, ocean, and ice surfaces, respectively [*Platnick et al.*, 2003]. We use here the quality-assured products.

[7] As pointed out by *Rosenfeld and Feingold* [2003] who investigated differences in IE stemming from POLDER and AVHRR data, POLDER may underestimate the average cloud top droplet radius due to the sampling of clouds which are homogeneous on a large scale. Detailed comparisons also show a systematic positive bias for MODIS CDR as compared to POLDER CDR which has not been fully elucidated [*Bréon and Doutriaux-Boucher*, 2005], although assumptions on the width of the droplet size distribution and cloud homogeneity are thought to play a role. *Rosenfeld and Feingold* [2003] further argue that POLDER aerosol index retrieval over land may be less reliable, showing more variability, and the POLDER-derived IE over land may thus be too small. There are also differences in the retrievals of the AOD between POLDER and MODIS [*Myhre et al.*, 2004]. However a detailed analysis of the differences in POLDER and MODIS data is beyond the scope of this study. We rather seek to analyze the uncertainties introduced by these differences on the radiative forcing by the AIE. All satellite data used in this study are gridded to the model horizontal resolution, with daily temporal resolution.

2.2. The LMDZ GCM

[8] We use the Laboratoire de Météorologie Dynamique (LMD-Z) GCM [*Li*, 1999] in a resolution of 96×72 grid points horizontally with 19 vertical layers. Here, we use the standard version of the model, which includes a bulk scheme for the precipitation formation in which cloud droplet size does not affect precipitation formation. We are therefore looking exclusively at the first aerosol indirect effect in this study. The model results are from simulations using SST for 1997 and aerosol emissions valid for the 1990s.

[9] We calculate the cloud droplet effective radius at the top of liquid water clouds using the random overlap assumption which is applied in the model parameterizations. A daily coverage of the globe is sampled in the same way as in the observational data by using the satellite over-passing local time, which is 10.30 a.m. for both POLDER and MODIS. To meet the homogeneity criteria applied by *Bréon et al.* [2002], we use the CDR only for liquid water clouds which fill at least a quarter of the grid cell (the results are not sensitive to the exact choice of the threshold). The aerosol index is estimated from the simulated aerosol concentrations as in the POLDER retrieval from the aerosol optical depths at 670 and 865 nm.

[10] We are firstly interested in finding a robust and realistic parameterization to use in long climate change simulations including the first AIE. Many climate simulations recently conducted for the IPCC Fourth Assessment Report used prescribed rather than interactive aerosol concentrations. For this reason, we first use here pre-calculated monthly mean distributions of sulfate aerosol mass concentrations valid for the end of the 20th century [*Boucher and Pham*, 2002]. For future climate change simulations and process studies, it is, however, valuable to simulate the aerosol atmospheric cycles on-line. This is done in the present study using the multi-component aerosol model of *Reddy et al.* [2005], using the emissions as in their study. Optical properties of the aerosols such as AOD and AI are estimated as in the satellite observations. The radiative forcing is diagnosed by applying the radiation parameterization twice, using the present-day aerosol concentrations to calculate cloud optical properties for the simulations, and using pre-computed daily distributions for natural aerosol sources only for the diagnosis. We "nudge" the model to ECMWF reanalysis of the wind fields in order to ensure that the aerosol transport is similar under pre-industrial and present-day conditions.

2.3. Parameterization of the Aerosol Indirect Effect

[11] We apply a link between sulfate aerosol mass concentration (m_{SO4}) and cloud droplet number concentration (N_d) following *Boucher and Lohmann* [1995, hereinafter referred to as BL95]:

$$N_d = \exp(a_0 + a_1 \ln(m_{SO4})) \quad (1)$$

where a_0 and a_1 are empirically derived parameters, which may be considered globally constant (formula "D" of BL95), or different above continents and oceans (formulae "A" to "C" of BL95), where also stratiform and convective clouds may be distinguished. The parameters of formula "D" used in the reference cases are $a_0 = 5.1$ and $a_1 = 0.41$. The cloud droplet effective radius (r_e) is calculated assuming spherical particles:

$$r_e = \epsilon \sqrt{\frac{q_l \rho_{air}}{\frac{4}{3}\pi \rho_w N_d}} \quad (2)$$

where q_l is the cloud liquid water mixing ratio (kg kg^{-1}), ρ_{air} and ρ_w the densities of air and water, respectively (kg m^{-3}), and $\epsilon = 1.1$ is a factor taking into account the ratio between the effective radius and the volume-mean droplet radius [*Pontikis and Hicks*, 1993]. By combining equations (1) and (2), we get

$$r_e \propto \exp\left(-\frac{a_0}{3} - \frac{a_1}{3} \ln(m_{SO_4})\right) \quad (3)$$

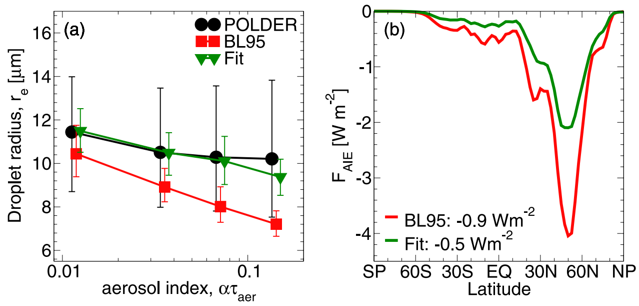

Figure 1. (a) Fit of the CDR to AI relationship (black: POLDER data, red: model using the original BL95 parameters, green: model using the fitted parameters). Error bars show the mean absolute deviation for CDR larger and smaller than the mean value within each bin of aerosol index. For the sake of readability, the three plots are slightly shifted along the x-axis. (b) Resulting zonal annual mean radiative forcing by the aerosol indirect effect (red: model using the original parameters, green: model using the fitted parameters). This model version uses prescribed monthly-mean sulfate distributions.

The aim of the present study is to constrain the values of the parameters a_0 and a_1 in the above formula using satellite observations in order to get a realistic parameterization of the first aerosol indirect effect in the GCM.

3. Results

[12] Figure 1a shows the relationship between cloud top droplet radius and aerosol index from the POLDER-1 measurements, the same relationship from the model using the original parameters in equation (1) as given by BL95, together with the modeled relationship using adjusted parameters (a_0 = 3.9, a_1 = 0.2). Figure 1b shows the zonal annual mean radiative forcing by the first AIE for the simulation with pre-calculated sulfate aerosol mass using the standard parameters and the adjusted parameters, respectively. The global annual mean is reduced from −0.9 to −0.5 Wm^{-2}. This adjusted BL95-type parameterization has been used in the recent transient IPCC simulations with the Institut Pierre Simon Laplace (IPSL) coupled model yielding results which agree well with observations in terms of the global temperature record since 1860 (J.-L. Dufresne et al., Contrast of the climate effects of anthropogenic sulfate aerosols between the 20th and the 21st century, submitted to *Geophysical Research Letters*, 2005, hereinafter referred to as Dufresne et al., submitted manuscript, 2005). For more detailed process studies, it is necessary to calculate the aerosol atmospheric cycles on-line in the model, thus allowing for interactions between aerosol concentrations and other model variables such as precipitation. In addition, it is becoming more clear that aerosol types other than sulfate have indirect effects as well. For this purpose we slightly modify equation (1) by choosing the maximum of the masses of all hydrophilic aerosols in the model (sulfate, the hydrophilic fractions of black and organic carbon, and submicrometer sea-salt). In order to match the CDR-AI relationship over land and ocean separately, we choose to adjust parameters separately for oceanic and continental clouds, and for stratiform and "convective" clouds, where the latter ones are defined as clouds where the convection contributes to the cloud water content.

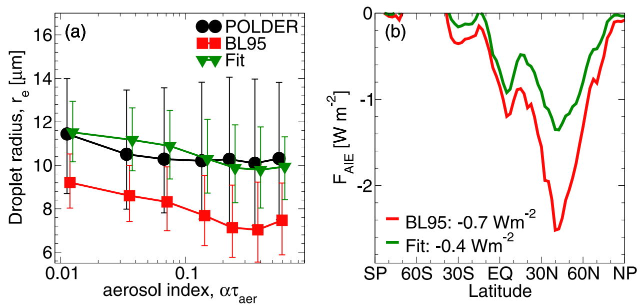

Figure 2. As Figure 1, but using a GCM version with an interactive multi-component aerosol module.

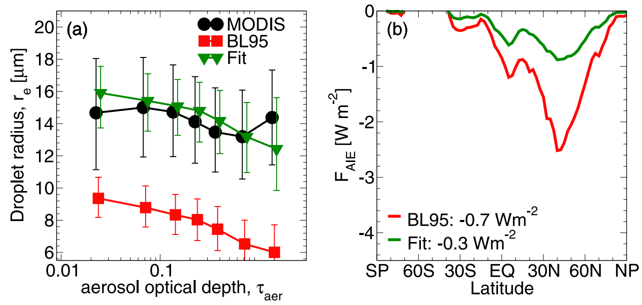

Figure 3. As Figure 2, but with relationships fitted against MODIS satellite data. The measure of aerosol concentration is the aerosol optical depth rather than the aerosol index.

[13] The CDR to AI relationship in the model is adjusted to POLDER-1 retrievals (Figure 2a) yielding the new parameters a_0 = 1.7 (for all clouds) and a_1 = 0.45, 0.30, 0.25, 0.2 for convective clouds over ocean and land, and for stratiform clouds over ocean and land, respectively. This reduces the annual-mean radiative forcing from −0.7 to −0.4 Wm^{-2} (Figure 2b).

[14] We also constrain the model to MODIS retrievals. However, the cloud droplet effective radius is often overestimated by MODIS for optically thick and inhomogeneous clouds (S. Kato, personal communication, 2005). In particular this is the case in the tropics, and thus the fit is done only for mid- and high latitudes (90°S–30°S and 30°N–90°N). Figure 3a shows the CDR to AOD relationships for one year of data, and Figure 3b shows the resulting zonal annual mean radiative forcing. As auxiliary figures[1], we show the fits to the POLDER/MODIS-derived relationships separately over land and ocean. The parameters in equation (1) are a_0 = 1.2 and 1.4 (for all clouds; over ocean and land, respectively) and a_1 = 0.2 and 0.15 (for convective and stratiform clouds, respectively; over both land and ocean). Although the two satellite datasets are rather differ-

[1]Auxiliary material is available at ftp://ftp.agu.org/apend/gl/2005GL023850.

ent, the forcing is reduced by a comparable amount, from -0.7 to -0.3 Wm^{-2} in the global annual mean for MODIS.

4. Discussion and Conclusions

[15] The present study evaluates and improves the representation of the AIE in a general circulation model. To do so, we combine the LMDZ GCM with two different datasets of satellite retrievals (from the POLDER and MODIS instruments). We focus on the "empirical" relationship between cloud droplet number concentration and aerosol mass concentration following *Boucher and Lohmann* [1995]. We fit parameters of the BL95 formula to obtain simulated relationships between cloud top droplet effective radius to aerosol index (or aerosol optical depth, respectively) similar to the ones retrieved by POLDER and MODIS. By doing so, the radiative forcing by the aerosol indirect effect is consistently reduced by roughly 50%, although there are considerable differences in the satellite datasets. The magnitude of the simulated aerosol indirect radiative forcing itself is, however, model-dependent, depending for example on the simulated low-level cloud distribution (e.g., Dufresne et al., submitted manuscript, 2005). In our model, we find a relatively weak aerosol indirect radiative forcing, and when applying the parameters fitted to the satellite data, we get a radiative forcing by the aerosol indirect effect which is consistent with the inverse calculations for climate change of the 20th century [*Anderson et al.*, 2003] and close to the radiative forcing estimated from observations [*Sekiguchi et al.*, 2003].

[16] An obvious advantage of using satellite data as done in the present study in contrast to aircraft measurements as done in the original study by *Boucher and Lohmann* [1995] is that for satellite retrievals, we have a much larger number of data points (of the order of 10^7 as compared to 10^2), well distributed over the globe and over the seasons. Another main point is that the spatial resolution is comparable to that used in current GCMs. This is important as the magnitude of the aerosol indirect effects is to some degree resolution-dependent [*Sekiguchi et al.*, 2003; *Quaas et al.*, 2004]. A potential source of error in our study lies in the assumptions used to establish the relationships between aerosol and cloud quantities from the satellite retrievals. We assume that aerosols retrieved in cloud-free areas are representative of the cloudy-sky areas and that the cloud-base aerosol concentration can be linked to the vertically integrated aerosol concentration as represented by the aerosol index or the aerosol optical depth. A solution to both problems may be found in the forthcoming satellite-based lidar and radar instruments, which will provide vertically resolved datasets. Also, with more computer power and new satellite measurements, more sophisticated parameterizations of the aerosol activation process may be evaluated and integrated into climate models. In particular, future studies need to consider the second aerosol indirect effect, which may affect the relationship between cloud droplet radius and aerosol concentration by its influence on the cloud liquid water content [*Lohmann and Lesins*, 2002; *Quaas et al.*, 2004].

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