

Evaluation of aerosol distribution and optical depth in the Geophysical Fluid Dynamics Laboratory coupled model CM2.1 for present climate

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[1] This study evaluates the strengths and weaknesses of aerosol distributions and optical depths that are used to force the GFDL coupled climate model CM2.1. The concentrations of sulfate, organic carbon, black carbon, and dust are simulated using the MOZART model (Horowitz, 2006), while sea-salt concentrations are obtained from a previous study by Haywood et al. (1999). These aerosol distributions and precalculated relative-humidity-dependent specific extinction are utilized in the CM2.1 radiative scheme to calculate the aerosol optical depth. Our evaluation of the mean values (1996–2000) of simulated aerosols is based on comparisons with long-term mean climatological data from ground-based and remote sensing observations as well as previous modeling studies. Overall, the predicted concentrations of aerosol are within a factor 2 of the observed values and have a tendency to be overestimated. Comparison with satellite data shows an agreement within 10% of global mean optical depth. This agreement masks regional differences of opposite signs in the optical depth. Essentially, the excessive optical depth from sulfate aerosols compensates for the underestimated contribution from organic and sea-salt aerosols. The largest discrepancies are over the northeastern United States (predicted optical depths are too high) and over biomass burning regions and southern oceans (predicted optical depths are too low). This analysis indicates that the aerosol properties are very sensitive to humidity, and major improvements could be achieved by properly taking into account their hygroscopic growth together with corresponding modifications of their optical properties.

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1. Introduction

[2] Aerosols scatter and absorb short-wave and longwave radiation, thereby perturbing the energy budget of the Earth-atmosphere system. Such effects from anthropogenic aerosols exert a direct radiative forcing of climate, but its quantification is difficult due to the large spatial and temporal variability of both the composition and distribution of aerosol [*Ramaswamy et al.*, 2001]. In that regard, the global coverage of atmospheric in situ measurements is still not sufficient for a proper evaluation of the role of aerosols

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on climate. Consequently, climate and aerosol models have been the primary instruments utilized for the Third Assessment Report (TAR) of the Intergovernmental Panel on Climate Change (IPCC) to assess aerosol forcing [*Penner et al.*, 2001]. The wide range of results from different aerosol models indicates that significant uncertainties remain, particularly concerning the role of organic and black carbon aerosols [*Penner et al.*, 2001].

[3] Because of computer limitations, most coupled climate models cannot afford to solve prognostic equations for aerosol concentrations. Instead, their distributions are simulated off-line with chemical transport models (CTMs), which are driven by meteorological fields either from reanalysis (for simulation of the last few decades) or GCM (for past and future simulations). Climatological monthly mean distributions calculated from the CTM simulation are then used as an input for the radiative scheme of the coupled climate models. Since the IPCC TAR, the characterization of aerosols on regional and global scales has been improved considerably with developments that include new parameterizations in CTM, new satellite instruments, longer data records of monitoring stations, and

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recent field campaigns. Such recent improvements now allow models to simulate multiple species of aerosol, as well as their sources, optical properties, and hygroscopic growth. Evaluation of these latest developments with available data sets constitutes a crucial part of any assessment.

[4] In the framework of the 2007 IPCC Fourth Assessment Report (AR4), aerosol distributions have been simulated over the period 1860-2100 with the MOZART 2 (Model for Ozone and Related chemical Tracers, version 2) chemical transport model [Horowitz et al., 2003]. These aerosol distributions are part of the set of historical, present and future short-lived forcing agents in the Geophysical Fluid Dynamics Laboratory (GFDL) coupled climate model CM2.1 simulations for AR4 [Delworth et al., 2006]. Among these agents, the tropospheric aerosols include: sulfate (SO_4^{-}) , black carbon (BC), organic carbon (OC), mineral dust and sea salt. The present paper aims to evaluate the "present-day" (1996-2000) mean aerosol distributions and optical depths used in CM2.1. From the historical run (1860-2000), we select the period 1996-2000 as representative of the present climate for two reasons. First, as shown in section 4.2.1, the influence of volcanic aerosols on AOD is negligible after 1996. Second, the CM2.1 simulations after 2000 use different IPCC scenarios, and any analysis of the results as a function of the scenarios is beyond the scope of this article. The evaluation is based on comparisons with climatological values of ground-based and remote sensing observations, as well as with results from other aerosol modeling studies. In nearly all cases, the data sets cover several years of records and their mean values are considered as climatological observations.

[5] After providing a brief description of the models and the aerosol fields in section 2 and 3, respectively, we first compare the annual mean values of surface concentration and aerosol optical depth with individual data sets in section 4. Then, in section 5 we combine the data sets to compare simultaneously the seasonal variation of surface concentration and AOD for various environments. In our conclusions, given in section 6, we highlight the strengths and weaknesses of the model results and provide suggestions for improvements.

[6] This paper is the second in a series of three. In the first one, *Horowitz* [2006] describes the MOZART simulations of ozone and aerosol distribution, evaluates the ozone concentrations, and describes the sensitivity of model results to wet removal rates for aerosols. The third paper (in preparation) presents and evaluates the instantaneous radiative forcing resulting from natural and anthropogenic agents, including the short-lived ones (aerosols and ozone).

6. Conclusions

[34] This study evaluates the strength and weakness of aerosol distributions and AOD used to simulate climate change with the GFDL coupled model CM2.1. The concentrations of sulfate, organic carbon, black carbon and dust were simulated with the MOZART 2 model [*Horowitz*, 2006], while the sea-salt concentration is from *Haywood et al.* [1999]. Our evaluation is based on comparisons at the global and regional scales with ground-based and remote sensing observations dating from 1980. The comparisons include aerosol surface concentrations measured over islands by the University of Miami, over the United States by the IMPROVE monitoring network, and over Europe by the EMEP monitoring instruments. The aerosol optical depth is compared with AVHRR and MODIS satellite data, and ground-based Sun photometer data from AERONET. We also compare our results with previous modeling studies.

[35] The individual evaluation of each aerosol component shows the following strengths and weakness.

[36] 1. The first component is sulfate: The annual mean surface concentration is reproduced within a factor 2 with values ranging from 0.05 μ g m⁻³ in remote marine atmosphere to 13 μg m⁻³ in polluted regions. In general, the simulated concentrations are overpredicted in summer and underpredicted in winter. Sulfate mass column and zonal mean profiles are comparable to other studies, although the global mean burden is about 15% higher. The major discrepancy compare with observations is in the amplitude and seasonal variation of sulfate AOD. In some regions where sulfate dominates the aerosol extinction, the simulated optical depth is a factor of 2 or more higher than the observations during some months. In Europe and North America the aerosol optical depth is overestimated by up to a factor of 5 in April. In maritime environments, there is no apparent discrepancy of AOD, but this is because the dominant SO_4^{-} contribution to AOD compensates for the underprediction of sea-salt burden. The global mean sulfate AOD is twice the value from other model studies. As the optical parameters are similar to these studies, we find that this discrepancy is due to the treatment of hygroscopic growth and the occurrence of very moist conditions. Sulfate is allowed to grow in our study up to 100% relative humidity while other models impose a somewhat arbitrary cutoff from 95% to 99% RH to distinguish haze particles from cloud droplets. In situ measurements by Wulfmeyer and Feingold [2000] indicate that maritime aerosols continue to grow at least to 98.5%. Laboratory measurements were also unable to measure growth beyond 98% RH. As the models have to extrapolate values up to 100% RH, large discrepancies may arise. As no statistics have been archived on the occurrence of model RH values greater than 95%, it is not possible to determine the exact impact of the simulated RH on the AOD. We suggest that the occurrence of very high RH values in the model and the hygroscopic growth of sulfate at these RH values ought to be further investigated.

[37] The second component is organic and black carbon: The annual mean concentration is generally overestimated in polluted regions by up to a factor of 2. Other model studies indicate that carbonaceous aerosols are systematically underestimated, particularly for organic carbon. An exception is West Africa where other models show significant loadings of carbonaceous aerosols associated with biomass burning activities during the dry season while our results do not show any perturbation arising from such activities. The source of this discrepancy seems to be caused in part by the emission inventory in West Africa. Although the surface concentrations does not seem to systematically underestimate the observations, the global mean optical depth due to carbonaceous aerosols is a factor of 2 to 3 lower than reported by other studies. This is because of the error of specific extinction of OC, which differs from that in other modeling studies. Our value was inadvertently converted to organic matter. An additional effect could be the lack of hygroscopic growth of organic aerosols which could improve the comparisons under moist conditions.