The importance of organic compounds for the first aerosol indirect effect: sensitivity to cloud formation parameterizations and meteorological fields.

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Atmospheric aerosols are a focus of attention because of their important impacts on clouds and climate change. Organic aerosols represent an important fraction of tropospheric aerosol; in polluted regions organic aerosols are the second most abundant component by mass of the aerosol burden (Ramanathan et al., 2001). A number of studies have shown that organics play an important role in the aerosol indirect effect (AIE) which is one of the greatest sources of uncertainty in the assessment of anthropogenic climate change. Given that ambient organic aerosols may consist of hundreds of species, their representation in global climate models is typically limited to a few classes; this leads to further challenges in simulating AIE. In addition, much uncertainty also arises from the relationship used to link aerosol with cloud droplet number concentration (CDNC) and the variability in the predicted meteorology that contribute significantly to the differences in predicted AIE between GCM studies. Here it is assessed the importance of organic aerosols for the first AIE under different cloud droplet formation parameterizations and meteorological fields.

For this, the 3D NASA Global Modeling Initiative (GMI) chemical-transport model is used. GMI allows easy interchange of different model components while maintaining all others identical allowing a direct intercomparison of results obtained between alternate representations of aerosol, chemistry and transport processes. CDNC is computed using both empirical correlations (i.e., Menon et al., - LB (2002)), and physically-based parameterizations (i.e., Abdul-Razzak and Ghan - AG (2000), and Fountoukis and Nenes - FN (2005)). Emissions from the IPCC CMIP5 (CMIP) are used. Sensitivities are examined under two meteorological fields (i.e., NASA GEOS4 finite volume GCM (FVGCM) and NASA GEOS1-STRAT (GEOS)) for the same time period. Computed CDNC is used to calculate the effective radius (Re). The CLIRAD-SW solar radiative transfer model is used online to calculate the cloud optical depth (COD) and the shortwave fluxes from the surface to the top of the atmosphere (TOA). COD is calculated as a function of the effective radius. Evaluation of modeling results (i.e., Re, COD) is performed against satellite products from Moderate Resolution Imaging Spectroradiometer (MODIS) platform.

Depending on the combination of meteorological field and cloud scheme used the annual mean CDNC ranges between 63 and 189 cm⁻³ (Figure 1) with larger differences seen over heavily polluted regions (e.g., Europe, China, NE USA), regions affected by long range

transport of pollution (e.g., Atlantic Ocean) or by biomass burning (e.g., S. Africa, S. America). Simulations without taking into account organics lead to a reduction in CDNC over S. America and S. Africa that are affected by biomass burning; lower CDNC values are also seen in eastern Asia because of the use of fossil fuels. The impact of organics on the first AIE is mixed: in pristine regions organics increase CCN increasing AIE, while in polluted areas particles grow larger, decreasing CCN and therefore counteracting AIE.



Figure 1. Simulated annual mean CDNC (cm⁻³) for all droplet formation schemes used under the GEOS meteorological field (right panel) and relative change in CDNC when neglecting organic aerosol, (i.e., CDNC_{tot} – CDNC_{OC}) (left panel). Global averages are shown in the upper right hand corner of each panel

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