Tropospheric sulfur simulation and sulfate direct radiative forcing in the GISS GCM

Koch, D., D.J. Jacob, I. Tegen, D. Rind, and M. Chin

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Abstract

Global simulations of tropospheric sulfate are performed on-line in the Goddard Institute for Space Studies (GISS) general circulation model (GCM) and used to calculate anthropogenic sulfate direct radiative forcing. Our model includes prognostic in-cloud oxidant H_2O_2, as well as DMS, MSA, SO_2, and sulfate. Compared to most previous models (except others with prognostic H_2O_2), this model has relatively high anthropogenic SO_2 and sulfate burdens. We show that this is due partly to the depletion of the prognostic H_2O_2 and partly to the lack of complete SO_2 oxidation in moist convective updrafts over polluted regions. Model agreement with surface observations is not remarkably different from previous studies. And, like some previous studies, we propose that an additional in-cloud or heterogeneous oxidant is likely to improve the simulation near the surface, particularly over Europe during winter. We find that the use of a new DMS source results in low S values in remote regions and discuss the longstanding difficulties of properly simulating DMS source and oxidants. Because of the relatively low natural source and high flux of SO_2 to the free troposphere, our model indicates a larger global anthropogenic contribution to the sulfate burden (77%) than estimated by previous global models. Additional high altitude observations of the sulfur species are needed for model validation and resolution of this issue. Direct radiative forcing calculations give an annual average anthropogenic sulfate forcing of -0.67 W m^{-2}. This value is within the range but near the high end of previous estimates, partly because of our large anthropogenic sulfate burden. We compare the radiative forcings due to online vs. offline sulfate and find little difference on a global average, but do find differences as great as 10% in some regions. Thus, for example, over some polluted continental regions, the forcing due to offline sulfate exceeds that of online sulfate, while over some oceanic regions the online sulfate radiative forcing is larger. We show that these patterns are probably related to the correlation between clouds and sulfate, with positive correlations occurring over some continental regions and negative correlations over northern oceanic regions.