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Coupling aerosol-cloud-radiative processes in the WRF-Chem model: Investigating the radiative impact of elevated point sources

E. G. Chapman, W. I. Gustafson Jr., R. C. Easter, J. C. Barnard, S. J. Ghan, M. S. Pekour, and J. D. Fast
Pacific Northwest National Laboratory, P.O. Box 999, Richland, 99352 Washington, USA

Abstract. The local and regional influence of elevated point sources on summertime aerosol forcing and cloud-aerosol interactions in northeastern North America was investigated using the WRF-Chem community model. The direct effects of aerosols on incoming solar radiation were simulated using existing modules to relate aerosol sizes and chemical composition to aerosol optical properties. Indirect effects were simulated by adding a prognostic treatment of cloud droplet number and adding modules that activate aerosol particles to form cloud droplets, simulate aqueous-phase chemistry, and tie a two-moment treatment of cloud water (cloud water mass and cloud droplet number) to precipitation and an existing radiation scheme. Fully interactive feedbacks thus were created within the modified model, with aerosols affecting cloud droplet number and cloud radiative properties, and clouds altering aerosol size and composition via aqueous processes, wet scavenging, and gas-phase-related photolytic processes. Comparisons of a baseline simulation with observations show that the model captured the general temporal cycle of aerosol optical depths (AODs) and produced clouds of comparable thickness to observations at approximately the proper times and places. The model overpredicted SO_2 mixing ratios and $PM_{2.5}$ mass, but reproduced the range of observed SO_2 to sulfate aerosol ratios, suggesting that atmospheric oxidation processes leading to aerosol sulfate formation are captured in the model. The baseline simulation was compared to a sensitivity simulation in which all emissions at model levels above the surface layer were set to zero, thus removing stack emissions. Instantaneous, site-specific differences for aerosol and cloud related properties between the two simulations could be quite large, as removing above-surface emission sources influenced when and where clouds formed within the modeling domain. When summed spatially over the finest resolution model domain (the extent of which corresponds to the typical size of a single global climate model grid cell) and temporally over a three day analysis period, total rainfall in the sensitivity simulation increased by 31% over that in the baseline simulation. Fewer optically thin clouds, arbitrarily defined as a cloud exhibiting an optical depth less than 1, formed in the sensitivity simulation. Domain-averaged AODs dropped from 0.46 in the baseline simulation to 0.38 in the sensitivity simulation. The overall net effect of additional aerosols attributable to primary particulates and aerosol precursors from point source emissions above the surface was a domain-averaged

reduction of 5 W m^{-2} in mean daytime downwelling shortwave radiation.

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