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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Coupling aerosol-cloud-radiative processes in the WRF-Chem model: investigating the radiative impact of elevated point sources" *by* E. G. Chapman et al.

Anonymous Referee #3

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This study reports on the further development of the WRF-Chem modeling system through the addition of a prognostic treatment of cloud droplet number and activation of aerosols to form cloud droplets. The indirect radiative effects of aerosol loading in the northeastern U.S. are investigated through a model sensitivity simulation in which emissions from elevated point sources were removed. This work is timely and represents a useful contribution to an emerging area of interest. However, in its current form, I find several important omissions in the manuscript discussions and feel that the manuscript and the analyses presented could benefit from some additional work. The following comments/suggestions are offered which may help improve the usefulness of this manuscript.

1. A lot of the background information in the model description section could be deleted as it is covered in Fast et al. (2006). Instead, it would benefit the readers if Section 2.5 is expanded to provide additional details on the formulation and implementation of aerosol-cloud interactions in the model. The section on advection scheme and the discussion on the use of a positive definite advection scheme do not add much new information. Mass consistency requirements are well acknowledged in tracer transport calculation in CTMs. While it is good to see conservative advection in WRF-Chem, the tests shown in Figure 1 are quite standard. This discussion could also be reduced quite significantly. Instead it would be useful to elaborate on the implementation of the aerosol-cloud interactions and its testing. For instance, it is mentioned that aerosol activation and resuspension are calculated simultaneously with turbulent mixing - what is the rationale for this - is it for physical or numerical stability reasons?

2. An important aspect missing in the discussion of the WRF model configuration is the choice of the convective cloud scheme. Was a convective parameterization invoked for the 6 and 2 km resolution domains or were all clouds assumed to be resolved? Are these schemes able to adequately capture the observed cloud fields during the study period? What are the implications of these model configuration choices on simulated aerosol-cloud interactions?

3. In this reviewer's experience, the mechanism that triggers cloud formation in models such WRF and its predecessor MM5, is quite sensitive even to slight perturbations in the radiation calculation. Inclusion of aerosol direct forcing alone was found to result in relatively large simulated instantaneous changes in cloud and radiation fields. Have the authors noted similar effects and if so, how does one differentiate those effects from the ones associated with activation of aerosols to form cloud droplets?

4. Adams and Seinfeld (Geophys. Res. Letts., 2003) suggest that primary emissions

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are more efficient per unit mass than gas-phase emissions at increasing CCN concentrations. Were sulfate emissions from elevated stacks considered in this study and what were the relative impacts SO4 emissions versus that produced from SO2 oxidation on the noted radiative impacts? Also, the rationale of speciating coarse PM emissions using the PM2.5 speciation profiles needs to be explained.

5. It is curious that the SO4 and SO2 overestimates are attributed to possible overestimation of the emissions. Using the CEM data in a grid model one would expect SO2 mixing ratios to be underestimated relative to in-plume aircraft measurements due to artificial dilution over the model grid volume. Could any other model process be contributing to the noted discrepancies?

6. The peak values of PM2.5 in figure 8b appear to occur during the night-time. Can sulfate over-prediction (which is attributed primarily to SO2 oxidation by OH) contribute to these night time peaks as suggested?

7. Based on analysis of COD, 1-4% changes in clouds due to elevated point sources, is suggested. Is this significant relative to other uncertainties in predicting COD? Some discussion on the significance of the noted magnitude should be provided to help put the results in perspective. How does one ascertain that the noted magnitudes are reasonable and that the additional modules implemented in the model are working correctly? Some analysis along that direction would go a long way in strengthening the manuscript.

8. It is not clear what the intended message is from the analysis presented in Figure 10. A three day simulation appears to be too short to study the indirect effects and the robustness of the additional modules. The noted divergence in AOD values illustrated in Figure 3, perhaps arise from not having a long-enough "spin-up" as also suggested in the discussion ("by August 11 these starting aerosols have exited the model domain"). A longer simulation period would help strengthen the analysis and the inferences drawn from the sensitivity runs.

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9. The use of two-way nesting is mentioned. Was it also used in the chemistry-transport calculations? If so, what method is employed to maintain consistency in non-linearly evolving chemistry on the different grid resolutions?

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