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.

E. G. Chapman et al.

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We thank the referee for his/her statement that this work represents a valuable contribution on aerosol-cloud-radiation interactions, and for the detailed comments and questions on the manuscript. Our responses to specific comments are given below. Where our response indicates a change in the manuscript, we are referring to a change in the revised manuscript submitted to ACP relative to the discussion paper published in ACPD.

Comment #1: Aerosol Nucleation. It appears in the present work that the authors adopted the binary H2SO4-H2O scheme by Wexler et al. and the ternary H2SO4-...
NH3-H2O scheme from Napari et al., but indicated a plan to convert to a scheme by Merikanto et al. in the future. It is clear that the classic nucleation approach is problematic in the treatment of binary and ternary nucleation. In addition, it is also realized that organic compounds contribute to aerosol nucleation. Those aspects need to be assessed, particularly in the context of their model performance.

Reply: The referee is correct that simulations presented here are based on the nucleation schemes of Wexler et al. and Napari et al., as this investigation was well underway prior to the publication of the Napari modified scheme as described in Merikanto et al. (2007). He/she is also correct that we are planning to convert to the modified scheme. As discussed on page 14772, Section 2.2, the Napari original scheme is likely to lead to nucleation rates that are higher than observed. In the discussion of Figure 7 on page 14784 we noted that predicted aerosol numbers associated with model Bin 1 (into which freshly nucleated particles would be assigned) are higher than observed, and indicated it could be due to the use of the Napari et al. original scheme. We did not go into a discussion of the fact that coagulation of small, freshly nucleated particles with larger, existing particles are unlikely to move the larger particles out of their current model bin, as the incremental increase in size is small relative to bin spans (Table 1). Coagulation of freshly nucleated particles with larger aerosols also is unlikely to significantly impact overall chemical composition associated with a given (larger) bin. The impact of using the original Napari et al. scheme is thus most likely noticeable for particles associated with Bin 1, an assertion supported by Figure 7. We have expanded the discussion of Figure 7 in Section 4.2 to include these latter points.

In terms of the impact of the choice of nucleation scheme on direct and indirect forcing, we note that classical Mie theory suggests that the largest contribution to aerosol optical depths will be from particles in the range of 0.2-1 micron diameter (our model Bins 3-5, see Table 1). For the fairly polluted conditions in this study (particle concentrations generally exceeding 5000 cm-3), theory suggests that only particles with diameters larger than ~0.1 micron will be activated. Thus particles in the accumulation mode size...
range will be most important for both direct and indirect forcing. Aerosol nucleation should have little effect on the accumulation mode mass, and previous modeling studies suggest that it has moderate effect on accumulation mode number compared to its strong effect on Aitken mode particle number. These points suggest that use of the modified nucleation scheme vs. the Napari et al. original scheme is unlikely to have a significant impact on the overall findings of our work.

Concerning the influence of organic compounds on aerosol nucleation, the referee is correct that there is recent evidence that organics may play a role in new particle formation (e.g., Smith et al., 2008). However, organic nucleation mechanisms that can be used by models are still under development. A sentence to this effect has been added to Section 2.2. Similarly, relative to secondary organic aerosol (SOA) formation, there currently is no SOA mechanism compatible with CBM-Z and MOSAIC, although one is under development. SOA thus is not included in the described simulations. A sentence has been added to Section 2.3 to explicitly state this. The omission of SOA is likely to be more significant on the clear, sunny day of August 9 as opposed to the cloudy, rainy days of August 10 and 11 when rainout/washout of aerosol particles occurred and photochemical activity was suppressed. However, as shown Figure 8 and discussed in Section 4.2 (and covered in the abstract), the model tends to predict more PM2.5 mass than observed, suggesting that uncertainties associated with model emissions and lateral boundary conditions may outweigh the uncertainties associated with neglecting SOA formation for this geographic region for the particular days simulated.

Comment #2: Meteorology Conditions. FDDA will likely reduce the effect of error of winds on the modeling, especially for small-scale or boundary layer simulations, but yet it was not performed.

Reply: As indicated on page 14781, lines 18-21, FDDA for WRF was not publicly available when this investigation was initiated. Both the FDDA modules of Liu et al. (2006) and our cloud-aerosol modules were included in the release of WRF-Chem v3.0, enabling future studies to use the approach advocated by the referee. The wording on
Comment #3: Ice Nucleation. What ice nucleation schemes were adopted?

Reply: Ice nuclei (IN) are not based on predicted particulates for the version of WRF-Chem used in this investigation, nor in the newer officially released routines of WRF-Chem v3.0. Some IN parameterizations have been reported in the literature, but they are mostly theoretical relationships that have not yet been extensively evaluated with measurements and consequently contain large uncertainties. Ice clouds were treated in our investigation using the original ice cloud assumptions inherent in the Lin scheme, which includes a specified ice nuclei distribution. Readers can refer to the original reference for detailed information on this topic. Material has been added to page 14773, Section 2.5 to clarify this point.

Comment 4: Model Description. I suggest simplifying section 2 "Model description", since some parts of this section have been discussed in Fast et al. (J. Geophys. Res., 111, D21305, 2006). This paper should focus primarily on the improvement to the existing WRF-Chem.

Reply: We have edited Section 2, pages 14769-14776, to emphasize the additions/changes to WRF-Chem for this investigation, leaving only brief descriptions of major features covered more extensively by Fast et al. (2006), as both referees suggested.

Minor Comment #1: P14800, Line 31, in "Wesely, M. L.", "16" should be "Vol. 23, No. 6".

Reply: This has been corrected.

Minor Comment# 2: P14812, in Figure 6, the x-axis was not labeled.

Reply: This has been corrected.

Minor Comment #3: P 14773, line 7, water vapor was classified as one of the six
hydrometeors?

Reply: The text has been rephrased to "which treats water vapor and the following classes of hydrometeors: cloud water, rain, cloud ice, snow and graupel"

References


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