**Atmospheric
Chemistry
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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.

E. G. Chapman et al.

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We thank the referee for his/her statement that this work represents a useful contribution to an emerging area of interest, and for the extensive comments and suggestions related to improving and extending our work. As indicated below, we have incorporated many of these suggestions into our revised manuscript. However, some comments addressed areas outside the scope of the current investigation. 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.

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Interactive Discussion

Discussion Paper

Comment #1: Model Description. A lot of the background information in the model description section could be deleted as it is covered in Fast et al. (2006). 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?

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.

Concerning Section 2.8 on the Advection Scheme, our experiences answering inquiries from other groups and as instructors at WRF-Chem user meetings clearly indicate that the reasons for using a positive definite advection scheme are not as well known throughout the modeling community as one would expect. We specifically included this section with its simple, easily understood tests to emphasize why it is important to select an appropriate advection scheme. We think this section is especially relevant since (a) the positive definite advection scheme was not publicly available in WRF-Chem prior to the release of version 3.0; (b) positive definite currently still is not the default advection scheme but must be chosen by the user (based on a WRF v3.0.1.1 repository checkout dated 22 September 2008); and (c) its use is particularly important in modeling regions with isolated point sources. For these reasons, we respectfully disagree with the referee's suggestion that Section 2.8 be reduced significantly. We have, however, tightened the language and reduced the space required.

Concerning the question about aerosol activation and resuspension, these processes are generally associated with vertical transport, and from a numerical standpoint it is desirable to calculate activation/resuspension and vertical transport simultaneously. The splitting of vertical advection and vertical turbulent mixing in WRF-Chem (and many other models) complicates this. In WRF-Chem, the activation/resuspension are calculated simultaneously with turbulent vertical mixing, as in Ghan et al. (2001). With the grid resolutions employed in this study, most of the vertical transport in boundary-layer clouds is represented by vertical turbulent mixing. We have revised Section 2.5 to include this information.

Comment #2: 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?

Reply: Within the 6 and 2 km resolution domains (Domains 2 and 3), aerosol-cloud interactions are handled solely by the microphysics scheme. As is standard practice, both the microphysics scheme and a convective parameterization are invoked for coarse grids in WRF-Chem. Thus clouds are invoked for the 18 km resolution domain (Domain 1) via the Lin microphysics scheme and the Kain-Fritsch cumulus option, as indicated in Table 2, page 14803. Convective parameterizations are not typically valid for grid spacings less than about 10 km. Note that we did not present Domain 1 results in the manuscript and, as indicated on page 14790, excluded cells within 5 nodes of domain boundaries from all Domain 2 and 3 analyses. The results presented for Domains 2 and 3 therefore represent the indirect effects arising from the new parameterizations and modules incorporated into the model.

As to whether the model adequately captured observed cloud fields during the study period, we believe it did. Locations of simulated clouds were compared with GOES-12 visible and infrared satellite images over Domains 2 and 3. The comparisons sug-

gest the model generally produced clouds at the right time in approximately the right locations, just as comparison of the simulated and observed cloud optical depths at Indiana, PA (Figure 9) also indicated. The comparison was best on 10 August when strong large-scale forcing produced synoptic-scale cloud bands. Even on the relatively sunny day of 9 August, when the clouds were driven by more local forcings, the simulated/observed cloud characteristics were similar and any errors tended to be in exact cloud location. A brief discussion of the comparison of observed GOES-12 and simulated cloud locations was added to page 14787, as part of the discussion of Figure 9 CODs. For completeness, we note here that comparison with cloud observations derived from MODIS satellite data are problematic due to the location and (infrequent) timing of satellite overpasses relative to model Domains 2 and 3.

Concerning the question on the implications of model configuration choices on simulated aerosol-cloud interactions, both the selected microphysics scheme and the cumulus parameterization choice will affect the distribution of simulated cloud properties, and potentially influence the magnitude of the impact of aerosols on clouds. A complete examination of all possible combinations of options and their impact on simulated quantities was not the objective of this investigation. However, we have added a sentence to our discussion, page 14793, first paragraph to indicate that additional studies to explore the effects of various parameterizations on model results are needed.

Comment #3: Cloud Formation. 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?

Reply: There are switches in the model to turn on/off the effect of aerosols on radiation, and the aerosol model (MOSAIC) can be run with or without the treatment of

cloud-aerosol interactions. One simulation could be performed with cloud-aerosol interactions turned on and aerosol direct forcing set to zero. Another simulation could be performed with cloud-aerosol interactions turned off and aerosol direct forcing activated. Differences in the simulations could be compared to qualitatively examine the magnitude of the impact of aerosols on cloud formation; however, such comparisons are not a true quantification of the aerosol direct and indirect effect since each simulation ignores a process that will have cumulative non-linear effects over time. To quantify the instantaneous aerosol direct and indirect forcing requires additional code to save the forcing associated with each aerosol effect. This was not done in this study, since the objective was to examine the effect of point emissions on the cumulative direct and indirect effect in the atmosphere, as would occur during real-world conditions.

Comment #4: Particulate Emissions. Adams and Seinfeld (Geophys. Res. Letts., 2003) suggest that primary emissions 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.

Reply: PM2.5 emissions used in this study, both ground level and from elevated stacks, included particulate sulfate, as noted on page 14779, lines 13-15. We did not distinguish in our model runs between primary sulfate and sulfate formed from SO2 oxidation, and cannot comment on the relative radiative impacts of the two sulfate sources.

As explained on page 14779, last paragraph of Section 3.2, the NEI99 emission inventory used in this study does not chemically speciate PM10, reporting it instead as total mass within a given NEI99 grid cell (area sources) or as a total mass from a specific stack (point sources). Inventory PM2.5 emissions are chemically speciated. Without any guidance from the U.S. EPA on what coarse particulate chemical speciation profiles are appropriate, it seems reasonable to use the PM2.5 chemical speciation as

a first cut approximation, as in many cases PM10 sources in a given grid cell will be related to the PM2.5 sources.

Comment #5: SO2 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?

Reply: As explained on pages 14778-14779, CEMS data replaced NEI99 inventory data for SO2 and NOx for Domain 2 and 3 stacks emitting >24 tons SO2 day-1, while stacks >100m in height that were not CEMS-replaced were adjusted by factors recommended by Frost et al. (2005). Under these criteria, all Domain 1 point sources outside of the nested domain regions and <100 m height and all Domain 2 and Domain 3 point sources <100m height with emissions <=24 tons SO2 day-1, along with all area sources of SO2 and NOx, are not adjusted. We are not emissions inventory developers; we used what we believed to be the best available emission estimates for the spatial and temporal extent of our simulations, but do not expect the estimates to perfectly reflect actual emissions. We examined the predicted boundary layer depth during the afternoon, but concluded that errors in the simulated depth could not fully account for errors in the simulated dilution of SO2. We have not been able to identify any other model process that could be significantly contributing to the over-estimate.

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

Reply: Close inspection of Figure 8b reveals that peak simulated PM2.5 values occur near 1200 Z, or 0700 local standard time (LST). Sunrise at the center of our modeling domain (i.e., near Indiana, PA) for August 9-11 occurs almost two hours earlier, circa 0520 LST. Sufficient OH radical can form and react in this two-hour period as there is an "explosion" of photolytic activity near sunrise (c.f., Finlayson-Pitts and Pitts, 2000).

Additionally, at 0700 LST the boundary layer should be relatively shallow. It is possible that our simulated boundary layer depths at this time in cells corresponding to the U. S. EPA AQS sites are too shallow. This would cause material contained within the boundary layer to be mixed through too small a volume, yielding higher simulated concentrations relative to observations. We do not have measurements of boundary layer depths at this time everywhere in the domain to compare against, but have revised the discussion of Figure 8 to include mention of the possible impact of errors in simulated boundary layer depths on modeled PM2.5 concentrations.

Comment #7: Uncertainty. 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? How does one ascertain that the noted magnitudes are reasonable and that the additional modules implemented in the model are working correctly?

Reply: As noted on in the discussion on pages 14792-14793, large spatial variations in the effects of aerosols were noted in our simulations, and statistically verifying model trends (including influences on cloud formation and cloud properties) will be difficult because necessary observations are not yet routinely made. Short-term field experiments such as the June 2007 Cumulus Humilis Aerosol Processing Study (CHAPS) and the upcoming VAMOS Ocean-Cloud-Atmosphere-Land Study (VOCALS) represent efforts to measure the quantities necessary to begin addressing the individual process-related uncertainties associated with the overall indirect effect. Until such measurements are routinely available on numerous geographic and temporal scales, true uncertainty analyses cannot be conducted. Other than standard numerical tests to ensure correct coding and a "reality check" to ensure model behavior is consistent with our current understanding of atmospheric processes, we are constrained by available observations to the types of comparisons presented in the manuscript. Additional numerical experiments were outside the scope of investigation reported here, and would be of limited use in addressing the referee's comment.

Comment #8: Simulation Length. 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 [Authors' Note: We believe the referee means Figure 10], 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.

Reply: In modeling emissions scenarios, questions invariably arise as to how far down-wind impacts from given sources may be felt. Figure 10 attempts to address this issue, albeit in a limited way. This analysis also represents part of the "reality check" described above: was the overall behavior of the model consistent with expectations when point source emissions were shut off? Do aerosols gradually exit the domain and do AODs eventually drop? We agree with the referee that our results suggest the need for longer simulation times and specifically stated this in our discussion (page 14793, lines 2-4) and in the Summary (page 14795, lines 24-28). We also clearly stated in the Summary that computational constraints limited the length of our simulations and that our results should be viewed as demonstrative.

Comment #9: Two-way nesting. 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?

Reply: Yes, two-way nesting is used in the chemistry/transport calculations. The topic of consistency/mass conservation in nesting is addressed extensively in Fast et al. (2006). As noted there, the monotone interpolation technique used with nesting in WRF-Chem does not conserve mass, but the errors are expected to be small. Fast et al. (2006) describe tests where the mass conservation error in the region including and adjacent to a nested domain was 0.12% after a 24-hour simulation period, equivalent to a timescale (i.e., mass burden divided by the rate of change) of ˜830 days. The timescales of processes of interest in this investigation are much shorter,

and any errors associated with nesting should have a negligible impact on the results and conclusions presented here. We again note that cells within five nodes of domain boundaries were excluded from the analyses presented in our work (page 14790, lines 10-11), further minimizing any potential influences.

The referee may also be alluding to the known behavior that higher resolution simulations can produce different chemical results than coarse simulations for certain species that are highly affected by nonlinearities. This is one reason why we use a high resolution nest over our region of particular interest. No special treatment was attempted to account for such nonlinearities when interpolating between nests. The default monotone interpolation technique built into WRF (see the WRF documentation [http://www.mmm.ucar.edu/wrf/users/docs/arw_v3.pdf] and Smolarkiewicz and Grell [1992]) was used to regrid the fine domain results back to its parent nest. Since the nested child domain overwrites the contents of the parent domain at every parent time step, even if the solutions differ between the parent and child domains, the parent values within the collocated region of the domains are not ultimately used. This maintains consistency as much as reasonably possible given the highest resolution present at each location. Parameterization of the impact of sub-grid scale processes on uncertainty in aerosol concentrations and resulting climate forcing is an area of active research in our group, but is a topic considerably outside the scope of the present investigation.

References

Fast, J. D., Gustafson, Jr., W. I., Easter, R. C., Zaveri, R. A., Barnard, J. C., Chapman, E. G., Grell, G. A., and Peckham, S. E.: Evolution of Ozone, Particulates and Aerosol Direct Radiative Forcing in the Vicinity of Houston Using a Fully Coupled Meteorology-Chemistry-Aerosol Model. J. Geophys. Res., 111, D21305, doi:10.1029/2005JD006721, 2006.

Finlayson-Pitts, B.J. and Pitts, J. N., Jr.: Chemistry of the Upper and Lower Atmo-

sphere, Academic Press, San Diego, CA, 2000.

Frost, G. J., McKenn, S. A., Trainer, M., Ryerson, T. B., Holloway, J. S., Sueper, D. T., Fortin, T., Parish, D. D., and Fehsenfeld, F. C.: Effects of Changing Power Plant NOx Emissions on Ozone in the Eastern United States: Proof of Concept. J. Geophys. Res-Atmos.,111, D12306, doi:10.1029/2005JD006354, 2006.

Ghan, S. J., Easter, R. C., Hudson, J., and Breon, F.-M.: Evaluation of Aerosol Indirect Radiative Forcing in MIRAGE. J. Geophys. Res-Atmos., 106, 5317-5334, 2001.

Smolarkiewicz, P. K., and Grell, G. A.: A class of monotone interpolation schemes, J. Comp. Phys., 101, 431-440, 1990.

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