Response to comments from J. Fast

This is a very interesting paper that compares differences in PM mass, size distribution, and radiative effects of aerosols over regional scales associated with external and internal mixing approaches. It is also clear that a lot of work was needed to implement the source-oriented external mixing approach into the WRF-Chem model. As one of the contributors to WRF-Chem community model, I have several comments that would be useful for the authors to address.

Response: The authors thank Dr. Fast for comprehensive suggestions to improve the manuscript. Changes and response are made according to all the comments. Specific responses are listed with each comment.

Comment: Page 16458, line 11: I do not think the authors have shown that the approach "more accurately predicts radiative feedbacks". While their approach is more realistic than assuming only internal mixing, they have not compared simulated aerosol optical properties with any observations. What they have shown are differences resulting from external and internal mixing assumptions. The approach and results are better stated in lines 26 and 27.

Response: Line 11 was rephrased and comparison of particle scattering coefficient between observation and internal and external mixing models was added.

Comment: Page 16459, line 5. Figure 7 shows the absolute differences, so it is difficult to infer percent differences as stated in this sentence. It would seem that a maximum difference of 20% may not occur in many grid cells.

Response: The averages over land and the SJV within 4km domain were added and the manuscript was modified accordingly.

Page 16459, lines 7-8: The authors mention increases in atmospheric mixing; however, the changes in radiation as small and may not significantly affect vertical mixing and the paper never shows how differences in the mixing state of aerosols affect boundary layer structure and mixing.

Response: The differences in predicted planetary boundary layer height were added in Figure 6. It shows that SOWC increased PBL compared to the internally mixed version.

Page 16460, line 25. Matsui et al. JGR (2013) is a recent paper that simulates the BC mixing state in the WRF-Chem model. While this recent development is not yet available in the public release of the model, it is an example of a more detailed representation of the aerosol mixing state in a regional model.

Response: Discussion of Matsui et al. JGR (2013) was added.

Section 3: Are particles from different sources permitted to mix together? This is not clear in the description. If not, what is the impact of neglecting coagulation between different particulate types? Lines 1-9 on page 16466 suggest that coagulation happens only within a single source type.

Response: The coagulation between different particulate types is allowed and the source-origin of the larger particles is preserved. In other words, when two particles from different sources coagulate, the smaller one from type A attaches to the larger one from type B, and concentration of type B increases while the concentration of type A decreases. This approximation is efficient and relatively accurate since the fastest coagulation rates occur between very small particles and very large particles. The text was modified to make this clear.

Page 16465, lines 4-7: Regarding the initial and boundary conditions, were there observations available that provide the mixing state of particles for use in WRF-Chem? This information seems to be defined in the papers cited, but it seems significant enough that some description is needed in this paper.

Response: There was no available observation to provide the mixing states of initial and boundary condition particles for this study. In the cited papers, the initial and boundary conditions were tracked separately. More description was added to the text.

Page 16466, lines 18-20: In the public version of WRF-Chem, the ACM2 scheme is not compatible with chemistry. The developer of ACM2, has been modifying the boundary layer scheme to include mixing of trace gas and aerosol species that will likely be available in a future release of WRF-Chem. Therefore, some discussion is warranted to describe how the non-local and eddy diffusion is included in the present version so that confusion is avoided for future users of the code.

Response: The authors thank the reviewer for pointing out this. Detailed information about adding ACM2 was added to the text.

Page 16467 lines 1-2: I believe the standard WRF code (without aerosols from WRFChem) assumes no aerosols for those radiation calculations. The latest version (3.5) does include climatological aerosols for some of the radiation schemes.

Response: The text was modified to correctly present it.

Page 16467 lines 3 – 13: The authors describe how aerosol optical properties are calculated, which is very similar to the existing aerosol optical property module in WRFChem as described by Fast et al. (2006) and Barnard et al. (2010). Was this module modified to account for the new aerosol model, or was a new optical property module developed? Based on the description, the basic difference between the authors treatment of aerosol optical properties and the default treatment of aerosol optical properties is simply due to differences in the aerosol model mixing state.

Response: The aerosol size bins implemented in the SOWC model are not fixed as described in Fast et al. (2006). The SOWC model employs moving size bins that change in size depending on the condensation / evaporation of semi-volatile materials (most notably ammonium nitrate in the current simulations). For this reason, it was not possible to employ the Chebyshev economization in the current study as was done by Fast et al. (2006). A full set of Mie scattering calculations was performed during each radiative forcing time step. This treatment of the optical properties of model aerosols has been clarified in the revised text.

Page 16471, line 12. The use of the word “feedbacks” here does not seem technically correct. The aerosols directly affect the shortwave radiation, but feedbacks are usually assumed to be processes that close the loop, i.e. changes in radiation that subsequently affect meteorology and then back to aerosols.

Response: This is an important point. The text was corrected to use the term “feedbacks” correctly.

Page 16471, line 25. It is true the maximum difference is 2 K, but this is not occurring of over a large region (it is hard to differentiate the pink and red regions in the figure).

Response: The averages over land and the SJV within 4km domain were added. The text was modified to better present the differences.

Page 16472, lines 20-29: There is not much explanation for the differences in total BC between the external and internal mixing states. I assume that most of these differences are due to differences in dry deposition?

Response: The differences in meteorological conditions and the particles properties all contribute to the differences in concentrations of BC. The relative importance of each factor can be quantified in a series of sensitivity runs, but these were not undertaken in the current study. The manuscript was modified to clarify this point.

Comment: Page 16474, figure 12: There are large differences in PM concentrations, but it is not clear what are the factors contributing to the differences. The large differences imply large differences in mass in general but that might not be the case. Are some of the differences between the two simulations simply due to small meteorological differences that result in small spatial shifts in the PM plume? It would be useful to compute the total mass as a function of time over the San Joaquin Valley between the two simulations to get a better idea of the real differences in mass.

Response: Total $PM_{2.5}$ mass in the ground level of the SJV covered by the 4km domain predicted by the external case and internal case was compared as shown in Figure A4. It shows that the total mass predicted by the two cases is different, which is consistent with the differences shown in Figure 12 (Figure 10 in new version).

Page 16475, line 7: The phrase “more accurate optical properties” does not seem right. I agree that the source-oriented external mixing approach is more realistic than internal mixing; however the authors have not shown that the present optical properties are more accurate in terms of observations.

Response: Comparison of particle scattering coefficient between observation and internal and external mixing models was added in Table 3. It does show that source-oriented external mixing predicts better particle scattering coefficient than the internal mixing. However, lack of absorption observation does not show the comparison of particle absorption coefficient. The text was modified to describe these new comparisons.

Page 16476, lines 6-9: I agree that the model is more realistic, but in the future it would be interesting to compare the BC mixing approach of Matsui et al. (2013) with the present source-oriented external mixing model.

Response: The authors thank the suggestion from the reviewer. Definitely we want to continue development of the source-oriented WRF/Chem model and compare to other similar calculations.