Response to review comments on acp-2015-586 from reviewer 3

The original comments are provided in black, our response is given below each comment in red.

Thank you for the careful reading of our manuscript and your review.

This manuscript presents the evaluation of high-resolution WRF-Chem simulations over North America. The model skill in reconstructing the Aerosol Optical Depth and Angstrom Exponent is investigated by comparing model results with observations from MODIS Aqua and Terra as well as the ground networks AERONET and EPA. The research topic is certainly within the scope of the ACP. The article is well written and the methodology is clearly described. Moreover, aerosol optical properties are generally poorly constrained in modelling evaluation, especially at high-resolution resolution. For these reasons, I consider that such work should be published in ACP, but only after some revisions. It would have been worth to treat some aspects into more details, and to clarify some points of the discussion. I think that the authors should consider all the corrections of Anonymous Referee #2. In addition to his/her recommendations, I would propose some further corrections in the following.

Thank you for your positive assessment. We have indeed addressed all of the comments of the other reviewer.

General remarks:

- A major concern is that authors never make the connection to aerosol climate forcing, although the title suggests this kind of analysis. A thorough discussion on aerosol climate forcing is necessary. Otherwise the authors should modify the title.

We have modified the title to read:

"Evaluating the skill of high resolution WRF-Chem simulations in describing drivers of aerosol direct climate forcing at the regional scale"

- In many occasions, authors try to explain model biases in AOD estimations with an overestimation/underestimation of aerosol-nitrate and aerosol-sulfate, but no evidence are shown in the text to support this.

We thank the reviewer for raising this issue. In addition to the comparison of nitrate/sulfate ratios presented in the Supplementary Materials we added a further analyses using chemical composition data at 123 IMPROVE sites as explained further in the following points.

Technical corrections and comments:

Page 27316, line 9-14: This sentence is a bit confusing. Please restructure it.

We rephrased as follows:

Prior analyses of Level-3 (1° resolution) MODIS AOD over the eastern half of North America have indicated extreme AOD values (> local 90th percentile) are coherent over regional scales (~ 150 km) (Sullivan et al., 2015). Thus, our evaluation exercise also includes an analysis of the spatio-temporal coherence of extreme events.

Page 27322, line 3: Table 3 shows that MODIS and AERONET data are poorly correlated. In this section it is important to explain the reasons of this disagreement and the effects on the model evaluation.

Thanks for this comment. We clarified in the text and Table 3 that the MFB of MODIS vs. AERONET is strongly affected by some outlier sites and the MFB decreases when we

remove the three most biased sites. Further, the number of co-samples between MODIS is quite limited, thus those MFB may be not very representative. We added the following comment:
"When MODIS is compared to the 22 AERONET stations the MFB is -1.23 suggesting an underestimation of AERONET relative to MODIS. The large bias can be explained noting that the number of co-samples between MODIS is quite small and that MFB is strongly impacted by a few outliers. When we remove the three most biased sites (one land site in the North and two sites along the East coast) the MFB decreases to -0.91."

Page 27323, line 9: What is i?
We changed the paragraph on χ^2 according to recommendation of reviewer 2.

Page 27325, line 6-9: Did your results suggest the same? Did you compare AOD biases with sulfate biases? Did you find a correlation between aerosol-sulfate and AOD estimations?
We added the following details in the Introduction:
"We also include intercomparison with daily mean $PM_{2.5}$ concentrations from 1230 surface stations and near-surface $PM_{2.5}$ composition using data from 123 IMPROVE sites. The $PM_{2.5}$ concentration data for 2008 were obtained from the US Environmental Protection Agency (EPA) AirData web site and represent all available outdoor near-surface 24-hour mean $PM_{2.5}$ measurements in the model domain. Most of these stations report values on a 1 day in 3 schedule. Daily average $PM_{2.5}$ chemical composition are also available on 1 day in 3 and were accessed online through the IMPROVE data wizard."

We added the following analysis and description in section 3.1:
"We further investigated the bias in $PM_{2.5}$ by comparing WRF-Chem simulations with ground-based composition measurements at 123 IMPROVE sites in our domain. We computed the MFB on a seasonal basis between near-surface sulfate and nitrate concentrations in fine mode particles (i.e. Aitken and accumulation mode) (Fig. 4) and found sulfate concentrations are underestimated over almost the entire domain during winter, whereas a positive bias is present in the other seasons. Conversely, $PM_{2.5}$ nitrate tends to be overestimated during winter and fall in the WRF-Chem simulations and underestimated during summer. Thus the positive bias in AOD and $PM_{2.5}$ mass particularly during the summer appears to be associated with excess sulfate concentrations."

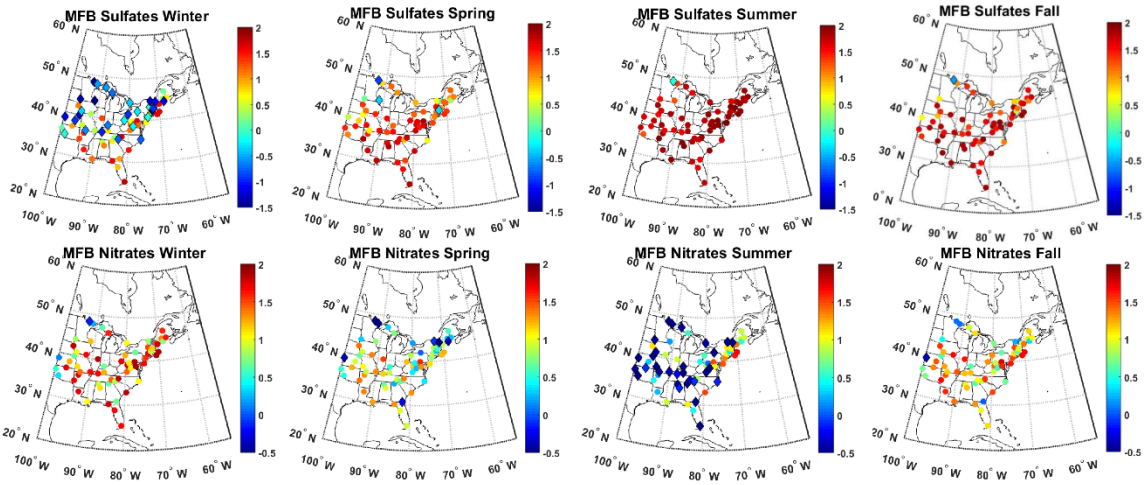

Figure 4. Mean fraction bias (MFB) of near-surface daily mean sulfate (first line) and nitrate (second line) concentrations in fine aerosol particles as simulated by WRF-Chem and observed in $PM_{2.5}$ measurements at 123 IMPROVE sites in different seasons. A positive MFB indicates WRF-Chem overestimates the observations. Note the scales differ between the frames shown for sulfate and nitrate and dots/diamonds refer to positive/negative MFB.

Page 27326, line 5-8: Do you have evidence about this? You should support di statement with some elaborations.
Thanks for pointing this out. We added explicit reference to Figure S3 in the Supplementary Material and the new Figure 4 showing the MFB analysis.

Page 27329, line 17-23: One more time, you only did some hypothesis but no evidence to support these statements. Please, show some elaborations that include particle composition evaluation.
We added explicit references to the new analyses on composition discussed above.

Page 27329, line 23-24: Why higher uncertainties at coastlines? Do you have some previous studies to cite in order to support this?
We added the following reference to support our statement:
Anderson, J. C., Wang, J., Zeng, J., Leptoukh, G., Petrenko, M., Ichoku, C., and Hu, C.: Long-term statistical assessment of Aqua-MODIS aerosol optical depth over coastal regions: bias characteristics and uncertainty sources, Tellus Series B-Chemical and Physical Meteorology, 65, 10.3402/tellusb.v65i0.20805, 2013

Page 27330, line 18: Table 3 suggests that AERONET MFB is 0.5
Thanks, fixed.

Page 27331, line 6: AERONET MFB is -0.59 according to Table 3
Thanks, fixed.