Reply to Anon Review #5

We thank the reviewer for their helpful and insightful comments. Please see responses to specific comments below.

1. My main concern is the fact that the authors seem to state that the MOZAIC module is the only way in WRF-Chem able to reproduce complex processes (P6068L20-23:"Of these, only MOSAIC (Zaveri et al., 2008) uses the more rigorous sectional representation of aerosol size distribution, enabling detailed aerosol interactions with radiation and clouds (Chapman et al., 2009)."). While I don't question the ability of MOZAIC to have good performance, I believe the authors should put some perspectives on their choice in regard on other available aerosol modules present in WRF-Chem. Moreover, I think it would be very helpful if the authors could justify the reason why they use a sectional model and not a lognormal model to represent the aerosol dynamics.

We agree we have perhaps been too strong in the wording supporting the sectional approach and underrepresented the modal approach. The lines:

"There are several aerosol mechanisms available in WRF-Chem. Of these, only MOSAIC (Zaveri et al., 2008) uses the more rigorous sectional representation of aerosol size distribution, enabling detailed aerosol interactions with radiation and clouds (Chapman et al., 2009). MOSAIC is only compatible with a subset of chemical mechanisms in WRF-Chem. For this study, the gas-phase mechanism used is CBM-Z (Zaveri and Peters, 1999)."

Have been replaced with:

"For this study the MOSAIC aerosol (Fast et al., 2006; Zaveri et al., 2008) and CBM-Z gas-phase (Zaveri and Peters, 1999) mechanisms are used. MOSAIC uses a sectional representation of aerosol size distribution, with detailed aerosol interactions with radiation and clouds described by (Chapman et al., 2009). MODAL approaches have also been successfully used with WRF-Chem to investigate aerosol–radiation–cloud interactions using the Modal Aerosol Dynamics model for Europe (MADE) scheme (Ackermann et al., 1998, Grell et al. 2011). The sectional approach gives a more nuanced representation of compositional variation across 8 size bins as opposed to 3 modes, and does not a-priori assume that the aerosol size distributions are log-normal. However, it comes at the cost of being significantly more expensive to run than the modal scheme."

2. P6069L27-30: The authors enumerate the different mixing rules present in WRF-Chem to compute the aerosol optical properties. They also rule out the volume-averaging rule. However in the rest of the manuscript they only consider the Maxwell- Garnett mixing rule, except in the conclusions where they say the shell-core rule should be tested. For more constitency, I think it would be better to state

from the beginning that you chose the Maxwell-Garnett mixing rule (and justify it), or to test the results of the 2 other mixing rules and show some results (as it is only an easy chosable option in WRF-chem).

A line stating the use of the Maxwell-Garnett mixing rule has been added earlier in the manuscript, in response to a previous reviewers comment. During the development and testing period for this work, the shell-core method in WRF-Chem was found to be unstable and resulted in the model crashing. Given more time, we may have tried to trouble-shoot this but as it is not a primary focus of the study, the Maxwell-Garnet was used as the most realistic stable method. Comparisons of different mixing rules has been the subject of other papers (e.g. Bond et al., 2006); so, given the stability issues, we considered a similar analysis to be both impractical, and beyond the scope of this study.

3. While I understand it is a very complex aspect, it would be very interesting to have a more detailed information about the aerosol composition, especially with the large set of accurate instruments presented by the authors. For example, I think it would be a great challenge to present more detailed results of the cToF-AMS as the secondary organic component and the secondary inorganic ions, and compare them with the model results. It is so helpful and rare to have such a various set of equipment at disposal that the potential of a more detailed analysis based on in-situ measurements and not only satellite observations as it is usually the case should be at least mentioned for further studies.

For this paper the focus is, as a development paper, the evaluation of the plume-rise parameterisation and its impacts on the modeled aerosol fields. Because of this we have focused on primary aerosol, as this makes up the greatest portion of the aerosol mass. We agree with the reviewer that detailed comparisons with inorganic and material with the AMS would be valuable, and these comparisons are planned for future studies. However, given the state of development of this analysis, we plan for this to be presented in future papers. Detailed analysis of the cToF-AMS data is to be presented in Darbyshire et al. (in prep); followed by further comparisons with WRF-Chem (incorporating a VBS representation of secondary organic material). A note that these further investigations will be the focus of future study has been added to the summary and conclusions section of the manuscript.

4/ Finally, again it would have been great to present time series of comparisons of aerosol modelled (composition and concentration) and observed along the flight path. Indeed, while most studies rely on AOD data and often have to boost their aerosol emission to match the AOD observations, you have the opportunity to bring new perspectives on aerosol emissions from fires by having a complete set of in situ observational data. A comparison between aerosol concentration/composition observed and modelled with the original and modified emissions would have been very helpful for the community .

While in an ideal world the authors agree timeseries would be valuable, in practice we have found such comparisons between flights measurements and model can be easily misinterpreted, distracting, and show the model in an overly harsh light. Lowe et al. (2014) gives an in-depth discussion on the difficulties of comparing aircraft data with high resolution WRF-Chem over the UK in the RONOCO campaign, where the dominant emissions sources are from (relatively) well-quantified large cities or point sources (e.g. power plants or oil refineries). For SAMBBA, these comparisons are even more challenging because the emissions are from fires smaller than the 1-km resolution of the MODIS fire count data, let alone the 25km resolution of the model, and there is considerable uncertainty in the emissions from fires and variation between different fires (as discussed in the paper). This means there is significant subgrid variation picked out by the flights, even when plumes have been removed, that would be unreasonable to expect the model to reproduce. The width of the box-plots in Figures 9 and 10 give an indication of just how much more varied the flight data is compared to the model. To present the model in a fairer light, we have therefore chosen to show horizontally averaged data, or the spread of data across the whole run, as opposed to direct point-by-point comparisons.

Lowe, D., Archer-Nicholls, S., Morgan, W., Allan, J., Utembe, S., Ouyang, B., Aruffo, E., Le Breton, M., Zaveri, R. A., Di Carlo, P., Percival, C., Coe, H., Jones, R., McFiggans, G.: WRF-chem model predictions of the regional impacts of N_2O_5 heterogeneous processes on nighttime chemistry over north-western Europe, Atmos. Chem. Phys. Discuss., 14, 20883–20943, doi:10.5194/acpd-14-20883-2014, 2014.