This paper describes how an existing aerosol scheme in the WRF-Chem model has been coupled to radiation and clouds to include feedback processes on meteorology. The authors then simulate the atmospheric conditions over a month long period and compare the predicted meteorology, trace gases, aerosols, with measurements. Measurements are from one surface site and a series of aircraft flights. They also examine aerosol-radiation-cloud effects by comparing predicted AOD, COD, and CWP with satellite measurements. Finally a sensitivity simulation is done that removes secondary organic aerosol (SOA) to examine its impact on aerosol-radiation-cloud effects.
The organization and presentation is clear, but I think it is missing some additional description and context, included in my comments below, before the manuscript is suitable for publication. Given that the journal is GMD, the description of the modeling components/approach is presently a little weak.

We thank the reviewer for reading the manuscript with much attention and his/her very useful comments and suggestions that helped us to improve the model evaluation and clarity of the paper. The manuscript has been revised following the reviewer suggestions as described below.

Major Comments

1) NOSOA simulation: Page 814, line 11: Given relatively small size of domain 3, I would assume a large amount of SOA from the outer domains could be transported into and across domain 3 during a 30-h simulation period. Thus, the NOSOA simulation does not necessarily have no SOA, but a reduced amount of SOA. It would have been better to turn off SOA entirely and suggest that this simulation be repeated. In fact, I would prefer if the entire 2 week period was repeated to assess the impact of SOA on AOD, COD, and CWP. Examining only one day, it is difficult to really conclude that including SOA is clearly improved when evaluating predicted AOD, COD, and CWP.

The sensitivity simulation is performed zeroing the arrays pertaining to SOA thus it is not affected by incoming SOA from domain boundaries. SOA concentrations are zero everywhere in the domain. This point has been clarified at the beginning of section 5.
We agree that one day of simulation is too short time to establish if the introduction of SOA really improves the prediction of cloud optical and microphysical properties, therefore we have performed the sensitivity tests also in other days. We have chosen three periods of three days each (with cloudy pixel coverage in MODIS data within the domain 3 larger than 60%) during the scavenging period (17-19 May) and six days during the long range transport period (25-27 and

28-29 May). Moreover, as following another suggestions of the reviewer about the cloud droplet effective radius ("It would be very useful if the comparisons in this section were extended to include droplet effective radius from MODIS. This is a parameter important for radiative forcing, since aerosols will lead to smaller droplets and higher albedo that would alter the radiation budget. The authors would have to screen periods with high clouds (cirrus) so the results focus on warm clouds with liquid cloud droplets"), we have extended our analysis to liquid water clouds, because the microphysics scheme is aerosol aware only for liquid clouds. Therefore, we have rephrased the Sections 4.5 and 5 also adding an analysis ad hoc for liquid water clouds in the three periods chosen, and we have expanded the analysis to all cloud phases in all three periods. Moreover, abstract and conclusions have been modified to make them consistent with the new findings.

2) Context and "connecting the dots": In general there is a lack of discussion on the errors in aerosol concentrations, particle number, and CCN and whether the results are consistent. For example, particle number is somewhat too high, but that should be related to mass that is too high. The authors show that OM is too low but SO4 is too high – but is the total mass too high as well? Is that consistent with the errors in simulated aerosol number and consequently CCN? Also particle number and CCN will be influenced by aerosol components (BC, other inorganics) that are not sampled by existing measurements (AMS instrument). In addition, the discussion of the simulated regional variations of AOT, COT, and CWP should be put in the context of the known biases in the aerosols – which are only evaluated at one point in the domain. It is hard to know how errors in aerosols elsewhere in the domain contribute to the differences between observed and simulated AOT, COT, and CWP.

The aerosol mass biases are discussed for each species analyzed and is related to the gas-phase species biases, thermodynamic equilibrium, model assumptions and emissions. Moreover, the bias of aerosol particles number and CCN is discussed on the basis of the literature. Some of these discussions are speculations that would require several sensitivity tests in order to be verified, but this is beyond the aim of the paper. We agree that there is not enough discussion regarding the link between particle number overestimation and aerosol mass concentration. Since as outlined by reviewer aerosol particles and CCN are influenced by other aerosol species not sampled during the campaign and in order to overcome the lack of these measurements, we have also evaluated the model using the PM_{10} data from AIRBASE network and $PM_{2.5}$ calculated from aerosol size number distribution measurements performed aboard of the ATR42. The

results of the evaluation have been added and discussed in Sections 4.2 and 4.3. The analysis shows that the model underestimates the total aerosol mass concentration in PBL and free troposphere. The aerosol particles that mostly contribute to CCN number are those of accumulation and coarse modes, and accumulation and coarse mode particles are also the major contributor to $PM_{2.5}$ mass concentrations. Being the $PM_{2.5}$ underestimated and CCN overestimated, CCN bias might not depend on model errors in $PM_{2.5}$. One sentence on this has been included in Section 4.4. Moreover in Section 4.4, it has been clarified that "the errors in the CCN prediction arise mainly from the uncertainties in the primary emissions of the aerosol particles and in their distribution in the lognormal modes". The regional variations of cloud properties have been put in the context of aerosol biases with two remarks in Sections 4.5 and Conclusions.

3) Insights from aircraft sampling: An additional figure is needed that show the aircraft flight tracks. It is hard to judge the spatial variability of simulated aerosols. I do not know if the aircraft flew in one particular region or throughout domain three. Readers should not have to go to other papers for this information. I certainly appreciate the summary statistics on all the aircraft flights, but it would have also been useful to include one or two "interesting" flights that shows relatively large SOA predicted by the model that corresponds to observed organic matter and relatively higher CCN.

We agree that the lack of a figure showing the aircraft flight tracks is missing in the paper, therefore we have included a plot with aircraft tracks in Figure 1. The new figure has been discussed in Section 3.2. Indeed, the statistics summary could not be sufficient to characterize fully the model behavior in reproducing aerosol mass and particle concentrations, therefore we have added two Figures showing the vertical profile of the model along the flight tracks on the 14 May. We have chosen this particular day because is a day of high pressure, therefore the interpretation of the results is not affected by cloud processes. New Figures have been discussed in Sections 4.3 and 4.4.

Specific Comments:

Page 793, line 3: change "scheme" to "schemes" >> ***Done***

Page 793, line 3: RACM/MADE/VBS is presented as a new chemistry option, when in fact the gas-phase coupling with aerosols and VBS for SOA was existing in WRFChem. As stated elsewhere the

new part is the coupling with aerosol direct and indirect effects. I suggest a new name that better conveys the differences with the older scheme – perhaps "RACM/MADE/VBS/AE" where AE stands for aerosol effect. >> ***Done. The new scheme has been called RACM/MADE/VBS/AQCHEM, the same name adopted in the code.***

Page 793, line 10: Change "correlation" to "correlation coefficient" >> ***Done.***

Page 793, line 14: Change "observed mass" to "observed concentrations". Mass and concentration is not the same thing. >> ***Done.***

Page 793, lines 14-17: There are many uncertainties in the treatment of VBS that likely contribute to errors as well that should be mentioned, here and where VBS is introduced in the model description. >> **A more detailed description of the VBS treatment of this paper has been added to the Section 2.1 together with key uncertainties that could affect SOA prediction. Some details about the use of some tunable parameters of this VBS treatment have been added to the Section 2.2. Moreover, some remarks about VBS uncertainties have been included in Section 4.3, Abstract and Conclusions.**

Page 795, line 3: In addition to the Grell reference, include Fast et al. (2006) and Chapman et al. (2009) here which presents the first coupling of aerosols to radiation and clouds alluded to in the previous sentence. The Grell paper does not have such coupling. >> ***Done.***

Page 795, line 23: Many readers unfamiliar with WRF-Chem will not know what "traditional" means. Please be more specific. >> ***The sentence has been rephrased: WRF/Chem (Grell et al., 2005) using the Secondary Organic Aerosol Model (SORGAM) (Shell et al., 2001), presents a negative bias of simulated $PM_{2.5}$ mass, mostly attributable to a scarce production of SOA (Grell at al., 2005; McKeen et al. 2007; Tuccella et al., 2012).***

Page 796, beginning of section 2.1. It would be useful to indicate that WRF-Chem is a community model, and such, has many options for trace gas chemistry and aerosols. The authors are using one particular scheme for each. >> ***The sentence has been rephrased: "A pre-release of version 3.4 of Weather Research and Forecasting model with Chemistry model (WRF/Chem) (Grell et al., 2005). WRF/Chem is a community model that has many options for gas chemistry and aerosols.***

***One of these has been updated in order to include a new chemistry option for simulation of direct and indirect effects with an updated parameterization for SOA production.*"**

Page 796, line 23. I know the VBS approach is described in Ahmadov et al. (2012); however, I think some description is needed in this paper. Either here or in the supplemental material that describes its overall approach. There are many treatments of SOA now available, including various VBS approaches that are by no means the same. The main purpose of the paper is to examine the impact of SOA on aerosol radiative forcing parameters, so some description of the SOA treatment is warrented. >> **A more detailed description of the VBS treatment of this paper has been added within Section 2.1.**

Page 798, lines 4-6: This sentence is awkward. Change to "The Lin and Morrison microphysics schemes in WRF/Chem version 3.4 include the prognostic treatment of the cloud droplet number." >> ***Done. The sentence has been rephrased as suggested.***

Section 2.1: the authors should describe some of the cloud-aerosol interactions that are missing in the model that may or may not have an effect on their simulations. This would include aqueous organic chemistry, ice-borne aerosols, etc. >> ***A sentence has been added at the end of the section 2.1: "The reader should note that the contribution to SOA concentration by cloud chemistry is missing and the interaction of aerosol with ice nuclei is not taken into account in this version of the model".***

Page 798, line 26: Please state why this period is chosen. There is no motivation for this period yet. I presume they want to use the measurement set described in the next section, but that has not been described yet. Also why is this period and dataset particularly useful to evaluate the model for their purpose of investigating the impact of SOA on radiative forcing parameters. >> ***A sentence has been added: "We chose this period because aerosol and cloud state-of-art measurements were available to evaluate the model (see Section 3). Moreover, during this period anticyclonic and cyclonic meteorological conditions were observed which allows the evaluation of the model under varying conditions".***

Page 799, line 5: This sentence could be improved. Perhaps "A series of 30-h simulations were performed on each day starting at 00 UTC, with the first 6 h discarded as model spin up." >> ***The***

sentence has been rephrased as suggested and it has been specified that the first 6 h are discarded as model spin up for meteorology.

Page 799, line 10: Here the authors mention a 13-day spin up period. So this contradicts the first sentence of the paragraph. It sounds like the simulation period is for the whole month, but the authors will be focusing on days after May 14 after the chemistry spin up period. The paragraph needs to be rephrased to clarify this point better. >> ***It is not a contradiction with the first sentence of the paragraph because as specified in the text, the first 6 hours of each simulation are discarded as model spin up for meteorology, whereas the first 13 days (as specified in the text) are used as spin up for chemistry.***

Section 3.1: This results section talks about surface aerosol measurements used, but there is no discussion here on the measurements. The authors should include what type of instrumentation was used. >> ***The instrument used is missing only for gas phase species. One sentence describing the instrumentation used at NL0011R EMEP station has been added: "O_3 is measured with an ultraviolet absorbing ozone instrument, NO_x, NO and NO_2 with a chemiluminescence monitor, and NH_3, HNO_3, HONO and SO_2 with an online ion chromatograph".***

Section 3.3: Please state the horizontal spacing of the pixels used in the analysis. >> ***The horizontal spacing of the pixels used in the analysis is 4 km. The information has been added to text.***

Comment: Could have used data assimilation to improve meteorology on domains 1 and 2 to help improve meteorology on domain 3 and thus confidence in aerosol radiation-cloud interactions. >> ***The use of the analysis nudging on coarser domains could improve the meteorology in domain 3, it is a common practice for air quality application. Usually, we prefer to use short simulations instead of analysis nudging.***

Section 4.2: The authors present gas concentrations in terms if ug/m3. It would be better to convert these to ppm or ppb to be more consistent with other comparisons in the literature. >> ***We used ug/m^3 as units for gas concentrations because the measurements are provided in this unit and it is a common unit used in Europe for model evaluations.***

Page 805, lines 6-11: I am assuming the RACM mechanism may be too simplistic to represent HONO well. In addition, errors in simulated HONO may not translate significantly into errors in

simulated aerosols that are the focus of the paper. What would be more important here is some evaluation of VOCs that likely contribute to SOA formation. The authors should provide some context on this subject in this section. >> ***It should be very interesting to compare VOCs that play an important role in SOA formation, but unfortunately VOC measurements are not available.***

Page 805, line 27: Here beings the discussion evaluating the surface aerosol predictions. However, it is not clear how the measurements are compared to the model and additional text is needed to clarify their methods. Are they measurements from an AMS instrument? If so the cut off is normally 1 um, but often it could be as low as 700 nm under certain conditions. The model uses a modal representation of aerosols, so how are the simulated concentrations compared to the measurements. If the authors are using the entire Aiken and accumulation mode, they may artificially introduce a bias in the comparison. >> ***We agree with the reviewer that this a point that should be explained in the text. We added this sentence in the section 4.2: "The reader should consider that aerosol composition measurements performed with the AMS are representative of particles with diameter between roughly 100-700 nm, whereas the model is evaluated with aerosol concentration representative of $PM_{2.5}$. Therefore, a bias could be present in the comparison. This means that the bias found for inorganic aerosols could be smaller than that reported above, conversely the OM bias could be larger of that found".***
". Moreover, a reminder has been also included conclusions.

Page 806, line 1: Please be more specific than just "consistent". Consistent in what way? I presume statistics is meant here. >> ***We mean statistically consistent. This has been specified in the text.***

Page 812, section 4.5: It would be very useful if the comparisons in this section were extended to include droplet effective radius from MODIS. This is a parameter important for radiative forcing, since aerosols will lead to smaller droplets and higher albedo that would alter the radiation budget. The authors would have to screen periods with high clouds (cirrus) so the results focus on warm clouds with liquid cloud droplets. >> ***The comparison has been extended to droplet effective radius. For more details, please refer to point 2 of the major comments.***

Page 814, line 20: I assume "ticker" should be "thicker". >> ***The sentence has been removed because the analysis has been changed. Please see the point 2 of the major comments for more details.***

Figure 7 and 8: Need to define what blue and red denote, and include units for y-axis. >> ***Done***