# Reply to Reviewer 1

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***

## Reply to Reviewer 2

This paper describes a new option for WRF/Chem to allow the simulation of aerosol direct and indirect effects (based on the existing aerosol microphysics scheme (including a VBS treatment of SOA)) and their feedbacks on clouds and meteorology. The paper is interesting and certainly within the scope of GMD, I would recommend publication after clarification on the below issues.

Most of my comments relate to adding extra details/clarification that I think are probably necessary given that this is GMD.

***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 clarity of the paper. The manuscript has been revised following the reviewer suggestions as described below.***

My general thoughts are that it is not entirely clear whether including the new VBS treatment of SOA represents an improvement here or not, since the comparison is made to an equivalent simulation without SOA (not the previous version of WRF/Chem). This is something that could be addressed in the Conclusions and Abstract.

***The difference between the new chemistry option and old mechanism is the SOA treatment. In the old mechanism, the SOA production is based on SORGAM model that, as explained in the Introduction, produces very little SOA mass concentrations (e.g. Tuccella et al., 2012). Therefore, we may assume that simulations with SORGAM and without SOA (in VBS option) are roughly equivalent. The advantage of this assumption is that the model is forced with the same initial and boundary meteorological conditions of the CTRL simulation. The use of SORGAM would require running the model on all three domains, leading to different results on domain 2. This would introduce dependencies on the input data for domain 3 making the comparison not directly comparable to the CTRL run. These observations have been added to first paragraph of the Section 5.***

***Being one day of simulation too short to establish if the introduction of SOA really improves the prediction of cloud optical and microphysical properties, we have performed the sensitivity tests also during different 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 day during the long range transport period (25-27 and 28-29 May).***

***Moreover, following one suggestion of reviewer 1, we have extended the analysis to cloud top effective radius and to liquid water clouds, being the microphysics scheme aerosol-aware only for liquid clouds. This analysis has shown enhanced skills of the model in reproducing clouds when SOA are included in the run. As suggested, these results have been included in the Abstract and Conclusions.***

Specific Comments:

p794, line 18: Specify here that you are talking about the change in aerosol due to anthropogenic emissions, since pre-industrial *times* >> ***The sentence has been rephrased "The change in global median radiative forcing associated to anthropogenic aerosol particles since pre industrial time is highly uncertain and it is estimated to be -0.9 W/m$^2$ within a range from -1.9 W/m$^2$ to -0.1 W/m$^2$ (Boucher et al., 2013)".***

p795, line 22: the range given by Scott et al. (2014) was +0.01 W/m2 to -0.77 W/m2, when accounting for the potential contribution of biogenics to new particle formation. >> ***The values have been corrected.***

p796, line 22-23: given that this is a new aspect that you are examining I think this requires slightly more detail than one sentence. The reader can look up Ahmadov et al 2012 but it would be good to summarise briefly here what this approach involves. >> ***We agree with the reviewer 2 that a better description of VBS could be useful. Therefore we have included a more detailed description of the VBS approach in Section 2.1.***

p799, line 15-17: at the moment this reads slightly like you have the PM2.5 and PM10 emissions and then add extra EC and OC? Might be worth rephrasing to clarify here, is the PM2.5 / PM10 disaggregated into different components? >> ***EC and OC emissions are from another dataset not included in the standard TNO inventory. This point has been clarified: "EC and primary OC emissions are taken from a specific TNO database that is part of the EUCAARI project (Kulmala et al., 2011). These EC and OC emissions are size resolved, they are separated for particles with diameter less than 1 μm, particles with diameter in the range of 1–2.5 μm and 2.5–10 μm".***

p799, line 25: which biogenic emissions do you include? Monoterpenes? Isoprene? Do the NMVOCs contribute anthropogenic SOA? >> ***SOA are formed by oxidation of anthropogenic***

***(alkanes, alkenes, aromatics and cresols) and biogenic (isoprene, monoterpernes and sesquiterpenes) NMVOCs. This point has been included in the VBS description.***

p799, line 26: where do the dust and sea-salt fluxes come from? >> ***With the term "flux" we mean the sea salt and dust emissions from seawater and soil. The sentence has been rephrased: "Dust and sea salt emissions from soil and seawater are calculated on line in the simulations."***

p809, line 24-26: could you expand on this? Or at least clarify the implication, which I think is that if POM is underestimated, the amount of SOA being formed would also be underestimated (according to the VBS approach) >> ***The point has been clarified as suggested "Indeed, the partition between OCV and SOA used in VBS approach depends on the total OM (Equation 1 of Ahmadov et al., 2012), thus if POM is underpredicted the resulting SOA could be underestimated".***

p810, lines 11-15: in this section you refer to some of the specific flights, but other than the date/time of the flights given in Table S1, we don't have any other information about where these flights went; an additional figure to show the flight tracks would be useful, if possible. >> ***We agree that the lack of a figure showing the aircraft flight tracks was 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.***

p811, lines 18-19: how do you actually calculate CCN from the model? Sorry if I've missed this somewhere, Figure 8 just says calculated at 0.2%. Later on you refer to calculating CCN using the particles in the accumulation mode, and mention the hygroscopicity of aerosol, so some clarification on how this calculation is done in WRF/Chem would be beneficial. >> ***WRF/Chem calculates the number and mass of aerosol activated as cloud droplet, and the spectrum of CCN, i.e. the aerosol activated at some specific supersaturation thresholds. We agree that calculation of CCN is not well described in the text, therefore we have included some clarifications in Section 2.1: "The activation of aerosols is based on a maximum supersaturation determined from a gaussian spectrum of updraft velocities and bulk hygroscopicity of each lognormal mode. Bulk hygroscopicity is based on the volume weighted average of the hygroscopicity of each aerosol component. In addition to the activated aerosols at environmental conditions, the CCN spectrum is also determined, i.e. the aerosol particles acting as CCN at some given maximum supersaturation (0.02, 0.05, 0.1, 0.2, 0.5, and 1%) are computed." Moreover it has been also***

***specified in the text that model/observation comparison is done with CCN at 0.2% of supersaturation.***

p811, lines 22-24: this seems like a point that is relevant to your observed v. simulated CN comparisons? if you are missing everything below 15 nm in the observations surely this would account for some of your over prediction, unless you are calculating your modelled CN above 15 nm? Then you say "Therefore the so calculated CCN efficiency is underestimated." But if you are talking about the observations, I would have thought the CCN efficiency is overestimated if your CN value is lower than it should be? Or do you mean relative to the modelled CCN efficiency? Either way this paragraph requires some clarification. >> ***In the comparison between predicted/observed CN we used the measurement of CPC 3010 instrument that measures the number of particles larger than 15 nm because the model does not take into account the ultrafine particle mode, indeed the modelled CN is given by the sum of particles of Aitken, accumulation and coarse modes. Considering that during the campaign Aitken mode had the mean diameter of 30-60 nm (please refer to Crumeyrolle et al., ACP, 2013), we may say that we are not missing anything in the observations below 15 nm.***

***When we say "Therefore the so calculated CCN efficiency is underestimated" we are referring to both observed and modelled CCN, because this should be calculated by using the particles larger than minimum activation diameter. Therefore, the model CCN efficiency is calculated with particles of the accumulation mode (the most favored particles to act as CCN) and it is qualitatively compared to the observed CCN efficiency reported in Crumeyrolle et al., ACP, 2013 that has been computed by using the particles larger than 100 nm. This helps to better characterize the relationship between CCN and the corresponding aerosol populations.***

***For more clarity, the last paragraph of Section 4.4 has been rephrased: "The calculated observed and modelled CCN efficiencies could be underestimated. In general, the CCN efficiency should be computed with the aerosol population with size larger than the minimum activation diameter (Asmi et al., 2012). The latter depends on the aerosol type and ranges from about 50 to 125 nm. We calculated the observed CCN/CN ratio with the measurements of CPC 3010 which gives the total number of particles larger than 15 nm, and modelled CCN fraction is calculated with total particle number given by the sum of the three modes of the lognormal distribution (Aitken, accumulation and coarse). In order to better characterize the relationship between CCN and corresponding aerosol population in the model, predicted CCN efficiency was also calculated with particles of the accumulation and coarse modes (the most favored particles to act as CCN) and it was qualitatively compared to observed efficiency during the IMPACT campaign computed***

***with particles larger than 100 nm. Observed values of CCN efficiency are in the range of 0.28-0.4 and 0.38-0.6 in the PBL and FT (Crumeyrolle et al., 2013), respectively. The simulated CCN fraction calculated with the particles of the accumulation and coarse modes, is always underestimated with respect to the observations, and it is in the range of 0.17-0.3 in PBL and 0.23-0.36 in FT. The model deficiency in simulating the CCN/CN ratio could be attributable to the uncertainties in geometrical diameter and bulk hygroscopicity of the lognormal modes, and updraft velocity that lead to error in the prediction of minimum activation diameter of each mode".***

p812, line 13-14: this is tricky to attribute to anything because we don't have any spatial information on where the number (and mass which is probably more important for AOT at 550 nm?) is over predicted, whereas the AOT is over predicted predominantly in the east of the domain >> ***Unfortunately MODIS data have a good coverage of AOT measurements only for 14 May. This effectively does not allow us to have a general overview of model skill in predicting AOT. Moreover as it is possible to note looking at Figure 1, the aircraft observations of 14 May are over the area to the South-Est of Cabauw that is not covered by satellite data. Therefore we have removed this sentence and we have highlighted that it is not easy characterize the bias of the model in simulating AOT within this study.***

p812, line 14-17: so what is the implication of this? It's probably worth reiterating what your assumptions are regarding mixing state >> ***We think that this is a point to clarify. We have rephrased the end of the first paragraph of the Section 4.5: "In general, model intercomparisons revealed that a large part of the uncertainties in simulating the AOT arises from the assumption on the mixing state. For example, AOT computed with external mixing is larger by 30-35% of that calculated with internal mixing assumption (Curci et al., 2014b). For typical atmospheric particle sizes and in the visible wavelength range, the AOT is then expected to be lower under internal mixing assumption (that is the assumption done in this work). Moreover, a 10% error in predicting AOT may be attributable to the choice of species density, refractive index, and hygroscopic growth factor (Curci et al., 2014b)".***

p813, line 1-2: I think this sentence requires some rephrasing, shift in what sense? >> ***The sentence has been removed after new analysis (please, see the answer to major comments).***

p813, line 8+: it would be useful here to clarify what you are actually turning off, the biogenic emissions? just in this domain? (+ what are the implications of that) It would also be useful to have some domain wide summary statistics (like Table 3) for the NOSOA simulation. Also it would be good to be consistent with how you refer to these simulations i.e., CTRL (sometimes called "reference run") and NOSOA (sometimes called "sensitivity run") >> ***The sensitivity tests have been conducted only in domain 3. The model array of the gas and aerosol concentrations has been set to zero for all anthropogenic and biogenic SOA. Therefore SOA concentrations are zero everywhere in the domain, and simulations are not affected by incoming SOA from boundaries. This point has been included in Section 5. Moreover, the domain summary statistics has been included for both liquid and all phase clouds in Tables 4 and 5.***

p814, line 17: I wouldn't use the word larger (this suggests something about the size of the CCN), do you mean higher concentrations? The increase in droplet concentration does not spatially overlap with the increase in CCN concentration, it would be worth some explanation of this, particularly since in the previous section you are suggesting that overestimating CCN would lead to overestimating CDN which would lead to overestimating CWP. >> ***The aim of maps showing $CCN_{0.2}$ and cloud droplet column was to show that CTRL simulations has more CCN and cloud droplets with respect NOSOA run. It is normal that the increase in droplet column does not overlap with the increase in CCN column, because in these maps CCN are calculated at 0.2% of supersaturation while cloud droplet are calculated from aerosol distribution at environmental supersaturation (please, see our comment about calculation of CCN). If we were supposed to see the overlap between CCN and droplet we should have plotted the columns of activated aerosol particles (cloud born aerosols). However, the sentence has been removed after new analysis.***

p814, line 24-26: Unless I've misunderstood, something is the wrong way around here, which simulation has 10% more optically thin clouds? Or is it just a coincidence that the difference between the amount of the total clouds accounted for by thin clouds in each sim is also 10%? **NOSOA** ***run is the simulation with more optically thin clouds. The sentence has been updated including also the results from other days of simulation.***

p815, lines 3-4: where does the smaller effective radius come from? The previous sentence made sense, perhaps remove (or clarify) this one >> ***The sentence has been removed after new analysis.***

p819, line 5: it would be good to keep the description consistent, if you're using VBS to simulate the partitioning of secondary organics into the particle phase, you're not excluding "SOA particles" as such, would be better to say "SOA is excluded" >> ***The sentence has been rephrased: "As test application of the new chemistry option, we performed a sensitivity simulation where SOA mass concentration is set to zero".***

p819, lines 20-21: these are not the same values as you give on p814, so one set needs updating. >> ***The paragraph has been modified after new analysis.***

Technical Comments:

P793, line 4: insert "the" before WRF/Chem? >> ***Done***
p797, line 7: insert "the" before "new chemistry package"? >> ***Done***
p798, line 17: remove "the" before Table 1 >> ***Done***
p801, line 9: replace "counters" with counter? >> ***Done***
p802, line 4-6: refer the reader to Table 2 here. >> ***Done***
p807, line 10-11: refer the reader to Figure 6 here >> ***Done***
p811, line 4: the end of this sentence doesn't make sense, please rephrase >> ***The sentence has been rephrased "Using several nucleation parameterizations, Pierce and Adams (2009) showed that CCN on average varies by up to 12% within the PBL".***
p811, line 6-7: again doesn't quite make sense, please rephrase (perhaps replace "of predicted CN larger" with "between simulated and observed CN being larger") >> ***We agree that sentence in this form is difficult to understand, it has been rephrased as suggested.***
p811, line 12: "Andrea" should be "Andreae" (it's correct in the Reference list) >> ***Done***
P813, line 20: replace "ticker" with "thicker" >> ***Done***
p835, Table 2: I think it would be useful to specify in the caption what the abbreviations (WS, WD etc) mean so that the reader doesn't need to look elsewhere. Also should the 6th entry in the WD section be WD200 (currently reads WS200)? >> ***The abbreviations have been specified in the caption of the Table 2. WS200 mean the observed/modelled wind speed at 200m. The abbreviations of the variables have been changed for example as follow: WS (m/s) at 10m, WS (m/s) at 20m, etc …***
p844 and p845, Figures 7 and 8: although these are the same as Figure 6 it would be useful to include in the caption what the different colours represent so the reader does not have to keep referring back to Figure 6. >> ***Done***

Supp Info: I think the second “Figure S1” should be called Figure S2. >> ***Done***