doi:10.5194/acpd-13-10561-2013 ("Does the POA-SOA split matter for global CCN formation?" by W. Trivitayanurak and P. J. Adams)

Revision and response to reviewer comments

This document describes revisions made to the manuscript listed above in response to comments by anonymous referees. The most significant changes are in the comparisons with observations, which have been enhanced significantly based on the referee comments.

The evaluation of EC/OC mass, which had been based only on European (EMEP) observations, is now considerably enhanced by the inclusion of North American (IMPROVE) data as well. Similar to many other global models of organic aerosol, we tend to underpredict OC concentrations by a factor of ~2 on average. The model predictions actually look significantly better with the IMPROVE data since the model underpredictions versus IMPROVE are more modest than those for EMEP.

Evaluation of aerosol number concentrations has been enhanced by evaluating CCN concentrations in addition to the CN10 evaluation that was in the original manuscript. The CCN evaluation is quite encouraging since the model has only a modest (20%) low bias after including carbonaceous aerosols, whereas without them, it was biased low by a factor of ~2.6.

As a result of these extensions, Section 3.3. on model evaluation has been revised considerably. Figures have been added, revised, and re-ordered accordingly (the current Figures 2-4). The abstract and conclusions have also been updated to reflect these changes.

In what follows, we detail specific responses to the reviewer comments. Referee comments are in normal text, and our responses are in *italics*.

Anonymous Referee #3

General comments

The paper summarizes a new treatment of carbonaceous aerosol in the TOMAS microphysics module and presents an application of the model to simulations of relative impacts of POA and SOA emissions on CCN formation. The treatment of microphysical processes is compelling and provides a good basis for modeling effects of POA and SOA emissions on CCN formation.

However, a substantial concern is that the objectives of the study are not sufficiently clear. The authors state in very broad terms that results of their study address the relative importance of POA and SOA emissions for CCN. This is misleading given that they make very specific and highly idealized model assumptions about emissions and processes. The rather brief review of previous studies in the introduction lacks specific context and clear motivation of this approach. See comments below for details.

We will respond to the detailed comments below.

Specific comments

Throughout the text: It should be clarified whether the model simulates elemental carbon (EC) or black carbon (BC). BC is the non-volatile and refractory component of the carbonaceous aerosol whereas EC is normally determined by thermal measurements. Emissions data sets for models are typically for BC.

We agree that the original manuscript was too loose in its distinctions between EC and BC. The situation is difficult because emissions inventories are indeed typically for BC whereas major networks measuring mass concentrations (EMEP and IMPROVE) report EC concentrations. Tami Bond, in her 2004 carbonaceous inventory (Bond et al., 2004), converted EC measurements to BC on a 1:1 basis, albeit with a higher uncertainty to account for ambiguities from the different measurements and definitions. We are unaware of any better way to make a conversion at this time.

For the sake of consistency with the emissions inventories used and with earlier aerosol modeling in GEOS-Chem, we have decided that the model simulates BC rather than EC. Therefore, in general, the paper has been revised to say BC rather than EC in almost every case.

The EMEP and IMPROVE data used for comparison were EC measurements, so we compare model-predicted BC versus the EC measurements of those networks directly. We have added the following text to Section 3.3.1 to acknowledge the EC vs BC issue and make this assumption explicit:

"For both networks, we compare our model predictions to measurements of EC as opposed to BC, which are not precisely equivalent (Bond and Bergstrom, 2006), but the relationship between BC and EC is still problematic. Noting this issue and potential uncertainties it entails, we assume that our model predictions of BC are equivalent to EC and can be compared to EC measurements on a 1:1 basis. In fact, BC emissions inventories have sometimes used EC measurements as equivalent to BC, albeit with higher assigned uncertainties (Bond et al., 2004)."

P. 10563, l. 15 and p. 10567, l. 22 and elsewhere: If OA and OM are considered to be equivalent in this study it is not clear why different terms are used for the same type of aerosol?

We frequently use organic matter (OM) to make it clear when a numerical value (e.g. an emissions rate) is OM as opposed to OC. Additionally, since the numbers tracked in the model are actually OM, we also refer to the model species as "hydrophobic OM" and "hydrophilic OM". When we discuss processes implemented in the model that affect these OM species, we use *OM.*

We have made this clearer by revising Section 2.3 ("Carbonaceous aerosols implementation") to read:

"Size-resolved carbonaceous aerosols, namely organic matter (OM) and black carbon (BC), are the new aerosol species implemented in this work. The model, therefore, tracks the OM mass

concentration rather than the OC mass concentration, including the contributions of oxygen, hydrogen, and other elements to the aerosol mass."

The original manuscript (Section 2.3.1 Emissions) already had the following text:

"To convert from OC mass normally reported in emission inventories to OM mass presented in our model, we assume a globally uniform and constant OM:OC ratio of 1.8 (El-Zanan et al., 2005;Yu et al., 2005;Zhang et al., 2005)."

In Section 3.3.1 (the evaluation of carbonaceous aerosol mass), we have added the following to make the OC evaluation clear:

"We compare our model-predicted OM concentrations to OC measurements by dividing by a uniform OM:OC ratio of 1.8."

Since organic aerosol (OA) is a more common term, we continue to use it when we refer to particle-phase organics generically. For example, from the Introduction, "Organic aerosol (OA) is found to be a significant fraction of aerosol mass in the accumulation mode that accounts for most CCN (Hitzenberger et al., 1999)." We also always use POA and SOA rather than POM or SOM since these acronyms are by far more commonly used. We have searched through the manuscript to ensure that our usage conforms to these patterns and made a few edits of cases that deviated.

Table 1: OM emissions in the current study are about 60% higher than the more recent CMIP5/RCP emissions. This difference reflects fundamental biases and uncertainties in emissions which are relevant to the current study. Uncertainties in emissions should be considered.

It appears that the reviewer has confused OC and OM emissions since our emissions are in fact ~20% lower than the newer RCP scenarios. Nevertheless, we have noted the more recent values in Section 2.3.1 on emissions:

"Primary emissions of OM and BC in Table 1 total 55 Tg yr-1 and 7.7 Tg yr-1, respectively. After this work began, the Representative Concentration Pathways scenarios (Lamarque et al., 2011) were finalized but present very similar emissions for the year 2000: 35.9 Tg OC yr-1 (or 64.6 Tg yr-1 when converted to OM) and 7.8 Tg yr-1 of BC."

It is our opinion that current uncertainties in emissions are >20%, so the older, slightly lower, organic emissions we are using may still be considered reasonable.

P. 10568, l. 13-17: References need to be added. The assumed sizes of primarily emitted aerosol particles are rather small (a brief review is available from Bond et al., 2013, for instance). Model results can be expected to be sensitive to the assumed sizes of the emitted particles. Given the choices made here, one may expect the model to produce CCN concentrations that are too high. Therefore, the sensitivity of CCN to relative POA emissions is perhaps unrealistically high in the model. What is the impact of the assumed size on these sensitivities?

We have searched Section 3 (regarding emissions) of Bond et al. (2013) for information about the emitted sizes of primary particles but do not find information there. Measurements of (processed) ambient size distributions are not good constraints; due to the short lifetime of ultrafine particles with respect to coagulation, the average size changes quickly. Moreover, it should be remembered that most of the mass is OC rather than BC, so the chosen size distribution must account for the observed size distribution of OC. While BC tends to occur in the "soot mode" at larger sizes, combustion sources typically exhibit an organic-rich second "nucleation mode" mode at smaller sizes (~30 nm or even smaller).

Section 2.3.1 on emissions has been enhanced to include a fuller discussion of these issues. First, we note the importance of the issue:

"In general, size-resolved emissions inventories of primary particles are not available for global modeling, so assumptions about the size distributions are required. However, recent studies have emphasized the sensitivity of aerosol number and CCN predictions to either the number or size of primary particles (Pierce and Adams, 2009;Spracklen et al., 2011a;Reddington et al., 2011)."

and also some discussion and support for the values used here:

"Although subject to significant uncertainties, these values are similar to those assumed in other global modeling studies: median diameters of 30-60 nm for fossil fuel combustion and 80-150 nm for biofuel combustion and biomass burning (Reddington et al., 2011). Numerous studies have shown that the number distributions from fresh vehicle exhaust have median mode diameters of 30 nm or even smaller (Putaud et al., 2004;Van Dingenen et al., 2004;Ban-Weiss et al., 2010). Although other choices of primary size distributions are plausible, these are consistent with emissions measurements, other global models and, as will be shown later, result in predicted CN10 and CCN number concentrations that show little or no bias with respect to ambient observations."

P. 10569, l. 17-18: Please clarify whether the surface area of the hydrophilic aerosol component is used here?

This section now reads:

"…we assume that the produced SOA irreversibly condenses onto the size distribution in proportion to the pre-existing aerosol surface area under ambient (hydrated) conditions, an assumption that has produced results in reasonable agreement with observations (Riipinen et al., 2011). We use total aerosol surface area from all species, <u>both hydrophobic and hydrophilic, inorganic and organic</u>,…" (the underlined portion is newly added)

P. 10570, l. 16-17: Are the same assumptions are applied to all types of emissions, including open fires? The original approach proposed by Cooke et al. (1999) only applies to fossil fuel emissions. It seems unlikely that these assumptions are generally applicable to all types of emissions.

Although the assumptions originally made by Cooke et al. (1999) were applied only to fossil fuel emissions, it has been common practice in global modeling to apply these same assumptions to all emissions categories. We are not aware of any models that do otherwise, nor are we aware of data to support other assumptions, nor did Cooke et al. (1999) provide quantitative support for these assumptions. We have clarified the history of these assumptions and mentioned the need for more systematic evaluation:

"Our emissions of OM and BC use hygroscopicity and mixing state assumptions following those originally made by Cooke at el. (1999) for fossil fuel sources and subsequently applied to all source categories by most subsequent global aerosol modeling. Very few measurements have been made to test these assumptions."

P. 10570, l. 29-P. 10571, l. 1: It seems that there will almost certainly be a systematic effect on aerosol burdens and the level of significance depends on circumstances such as the length of the simulation etc., even if the difference is small. Perhaps the sentence should be reworded?

We have deleted the original, vague sentence saying the effect was "small" and replaced it with the following summary of earlier work that sought to quantify this very effect:

"Park et al. (2005) explored a range of aging timescales in the GEOS-CHEM model and found that timescales of 0-2 days were consistent with TRACE-P observations of BC scavenging. The corresponding range for the global BC burden was 0.11 +/- 0.03 Tg C."

P. 10571, l. 20-23: A rather problematic assumption is to take the POA emission rate as the total (POA+SOA) aerosol source and to artificially vary the contributions of POA and SOA emissions. This implies that the SOA emissions can be represented by POA, which is a poor assumption for various reasons. Fundamental sources of uncertainty are very different for POA and SOA emissions. Spatial patterns and diurnal cycle of SOA emissions differ markedly from emissions of POA. Furthermore, the conversion of organic precursors gases to SOA mainly occurs in the troposphere above the surface. This increases the lifetime of the aerosol relative to aerosol that is emitted at the surface. For instance, Zhang et al. (2012) attribute an increase in POM burden between ECHAM-HAM1 and ECHAM-HAM2 by about 50% to vertical transport of condensable gases by convection and SOA formation in the upper troposphere in HAM2, which has been omitted in HAM1. Consequently, effects of POA emissions on CCN concentrations can be expected to be underestimated.

Although sources of POA and SOA are generally different, primary POM emissions in current inventories are likely to go through a cycle of evaporation, oxidation, and recondensation as oxidized, lower-volatility products (Hallquist et al., 2009). The author should clarify the purpose of their sensitivity study by identifying specific processes that produce POA and SOA in the atmosphere. Information about causes of uncertainty for these processes needs to be included so that the reader can understand why such a very wide range of SOA/OA source ratios (0%, 10%, 50%, 90%, and 100%) needs to be considered.

These points are well taken, but current knowledge of SOA formation still limits the confidence we can have in any model prediction, even one with greater process sophistication. Therefore, we choose to address these issues by adding a fuller discussion and including caveats. For example, we now elaborate on the limitations of forming SOA immediately at the surface in Section 2.3.2 (on the model SOA mechanism):

"In the model, SOA products are assumed to be produced immediately after emission of monoterpenes and are thereafter treated as non-volatile. Neglecting the time lag between emission of an SOA precursor and formation of SOA is a simplification used in much previous work, but it is worth considering some of its implications. Perhaps the most important is that the model SOA will be formed immediately in the boundary layer, where it is more subject to dry and wet deposition processes while, in reality, some SOA may form in the free troposphere and be longer-lived. For example, when transitioning from a simple SOA scheme similar to this one to a more physically based one, Zhang et al. (2012) found a 49% increase in OA burden, largely due to the long lifetime of SOA aloft. It is still unclear, however, whether missing sources of OA, probably SOA, occur mostly in the boundary layer or free troposphere. Measurements and box modeling suggest that substantial underprediction is already apparent in the polluted boundary layer (Volkamer et al., 2006). Model underestimates of OA in the free troposphere are severe in some instances but not observed in others (Heald et al., 2011;Heald et al., 2005). The simple approach used here and in other global models may underestimate the contribution of SOA to CCN to the extent that long-lived SOA is indeed formed in the free troposphere."

Given the mixed results of model-measurement comparisons, uncertainty over multi-generational organic aging, and uncertainty over the heats of vaporization of SOA compounds, we do not think it can be said with certainty that SOA formation occurs "above the surface" if this means in the free troposphere.

Some of these issues are also discussed in Section 2.4, where we lay out the sensitivity scenarios.

"The 5 experimental simulations have SOA/OA source ratios of 0%, 10%, 50%, 90%, and 100%. The reason for the wide range of scenarios is to include the full range from traditional OA models that are dominated by POA to AMS measurements that suggest a world dominated by OOA and SOA. Given the status of scientific knowledge and current representation of OA processes in the model, these scenarios capture some, but not all, of the differences between POA and SOA. For example, the fact that POA emissions contribute a large number of particles to the atmosphere (Spracklen et al., 2011a) whereas SOA formation simply grows existing particles is represented; however, the assumption of instantaneous SOA formation (see Section 2.3.2) does not capture the potentially different SOA lifetime if it is largely formed in the free troposphere. Moreover, the likelihood that POA emissions shrink by evaporation and then regrow by SOA formation (Hallquist et al., 2009; Robinson et al., 2007) is not yet represented in the microphysics used here. Therefore, the sensitivity experiments should be viewed as partly idealized representations of the differences caused by different POA and SOA formation pathways."

P. 10573, l. 24: Missing "emissions"?

Clarified to now read, "…but it should be remembered that SOA production in the BASE case is only 18 Tg yr-1." (We inserted "production" rather than "emissions").

Table 3: Why is the nucleation rate greater for BASE? This seems counterintuitive given that the aerosol burden is higher for this simulation. This should lead to less efficient nucleation of sulfate aerosol owing to increased condensation of sulfuric acid on pre-existing aerosol.

Unfortunately, we do not have a definitive answer at this time, and we lack the detailed diagnostics to answer this question without re-running the simulations. As a partial answer, we will note several things. First, in our group's 10+ years of modeling aerosol microphysics, it is frequently the case that aerosol perturbations lead to some surprising feedbacks, especially where nucleation is involved. Second, since the tropospheric-average nucleation rates tend to have large contributions in the upper troposphere (see, for example, very high number concentrations in the upper troposphere in Figure 7 of Trivitayanurak et al. (2008) from an earlier version of this model as well as GLOMAP; Pierce and Adams (2009) is similar as are all of the global microphysical models I am aware of). Third, the change in nucleation rate (~10%) is quite small and will have even smaller impacts on CN10 and CCN concentrations. For example, Pierce and Adams (2009) (see Table 2 in that paper, comparing the BINARY and REDBINARY simulations) found that a globally averaged change in the J10 nucleation rate of a factor of 2 led to a ~20% change in CN10 concentrations and a few percent change in CCN(0.2). Therefore, it is highly possible that the PBL carbonaceous aerosol induce a very small feedback in nucleation rates aloft. Given the ~10% change in J10 here, we expect this to have at most a few percent change in CN10 and a negligible change in CCN.

P. 10575, l. 5 and following: Why are only comparisons for Europe included? Data from other networks should be added, e.g. from IMPROVE. Comparisons for Europe are unlikely to be representative of results in other regions, which makes these comparisons rather pointless.

As discussed at the beginning of this reviewer response, a major revision of the paper has been to include IMPROVE measurements of OC to the model evaluation section.

P. 10575, l. 22-23: This speculation seems vague. The relationship between CCN concentrations and aerosol mass is highly nontrivial. It can be argued that a skillful representation of POM particle sizes is more important for CCN concentrations than an accurate simulation of aerosol mass yet little attention is given to aerosol size in this study. Furthermore, comparisons for mass are based only on a very small number of model grid points, which does not provide any useful constraints for global results.

The original manuscript speculated that we may underpredict the carbonaceous contribution to CCN because our OC masses were low compared to EMEP. We agree with the reviewer comments. This section required revising anyway since the revised manuscript now includes a CCN evaluation (see first page of this response document). This section is now more cautious:

"Overall, these comparisons suggest that our model has approximately the correct number of primary carbonaceous particles of both ultrafine and CCN sizes but insufficient carbonaceous mass at the polluted boundary layer sites measured by the EMEP and IMPROVE networks. Because the ultimate contribution of carbonaceous aerosol to CCN depends on both number and (soluble) mass, the model's good CCN predictions may result from some compensating errors. Nevertheless, the comparison demonstrates skill similar to other global models."

P. 10576, l. 23: Do the authors rule out other effects? It is somewhat surprising that only three different effects of changes in emissions on CCN concentrations can be identified. For instance, as shown in Table 3, OA aerosol emissions generally affect nucleation rates, the growth of the aerosol through coagulation, and the deposition. All of these changes clearly have important consequences for aerosol number and CCN concentrations. It would be useful to include results similar to Table 3 here.

We do not rule out other effects/feedbacks. Rather than say there are three effects, we identify three direct impacts and recognize potential for feedbacks:

"There are three direct impacts on the CCN(0.2%) population when we increase SOA in the split: 1) direct decrease of CCN(0.2%) by reducing CCN-mode primary emissions and 2) reduction of UF-mode primary emission that might grow to CCN, and 3) additional condensation of SOA that may or may not contribute to CCN production, depending on what size of pre-existing particles that SOA condenses onto. These direct impacts may induce other feedbacks including changes in nucleation and deposition rates."

P. 10580, l. 15: Please provide mean values instead of a range of values.

We have done so. It now reads:

"As a result, comparison of predicted EC and OC to measurements shows that the model underpredicts carbonaceous mass concentrations in the North American and European boundary layer by a factor of 2.1 (LMNB of -0.33), similar to several other "traditional" global models."

(The original manuscript said a "factor of 2-4", and this is now more specific. The log mean bias turns out to be closer to a factor of 2 than 4 due to the addition of the North American IMPROVE sites to the comparison.)

Anonymous Referee #4

The manuscript by Trivitayanurak and Adams describes two rather distinct scientific topics. The first, which does not appear in the title of the paper, is the implementation of carbonaceous aerosols in the TOMAS microphysics, hosted by the CEOS-Chem chemistry/transport model. The second is a set of experiments that try to quantify the primary-secondary organic aerosol split from the microphysical point of view, assuming that only primary aerosols affect aerosol number. The study is of potential interest, but has some significant problems and limitations that do not make it suitable for publication in ACP in its present form. In addition, given the two different topics covered, I would suggest to split the paper in two; one will properly describe the carbonaceous aerosol implementation and will do a thorough comparison with measurements (probably a GMD paper), and the other will study the POA/SOA split.

As discussed already, the comparison has been substantially enhanced by including IMPROVE OC observations and an evaluation of the model-predicted CCN. We prefer not to split the paper in two and note that this was not recommended by the other reviewer.

One important aspect is that the manuscript is not citing all relevant literature. In addition, the few references that are used are mostly old, with only about 10 of them being from the last 5 years. The same applies to the data used for the model evaluation; they are very few and rather old. Numerous datasets have become available from more recent campaigns and data compilations. The authors should seriously consider updating their datasets for a more detailed model evaluation. Comparing only a dozen European stations for carbonaceous aerosols and about that many for CCN mostly over Europe and the USA is far from sufficient.

We have expanded the comparison to observations as already noted. Comparison to recent field campaign data is highly problematic given the variability in aerosol concentrations and short sampling times. We have a strong preference for long-term, climatological observations. Moreover, a meaningful comparison against these field campaign data would require much longer simulations using meteorological fields corresponding to the various years of the various field campaigns.

In the process of revision, references have been updated with ~20 now being from the last 5 years.

Specific comments
CCN(0.2%) is quite long. I suggest using CCN, while mentioning that for the whole manuscript a 0.2% supersaturation is implied, unless otherwise noted.

We are afraid this would cause more confusion just to save a bit of space. Quantitative results will differ greatly based on the supersaturation chosen. Interpretation of results can be different for CCN at fixed supersaturation vs CCN at fixed meteorological conditions (e.g. updraft velocity). To avoid confusion (or people seeking for that one crucial line indicating the supersaturation value), we think it's better to have the metric be clear whenever we give a CCN concentration value at a given supersaturation (rather than CCN in the generic sense).

p. 10568, l. 14-17: Is this a guess, or there are references that support it? How sensitive is the model on the choice of these numbers?

The assumed size distributions are educated guesses with data to support them, but they are by no means definitive. The model is sensitive to these choice of values. For full details, see our response to Reviewer 3 on assumed size distributions of primary particles, their importance and limitations.

p. 10568, l. 20: I do not understand what the point (1) means.

The original manuscript was needlessly wordy here. We have streamlined this portion to read:

"This work incorporates a simple treatment of SOA because, given rapidly changing knowledge of SOA and TOMAS's focus on aerosol microphysics, a simple treatment is more flexible and allows for easier sensitivity studies."

p. 10568, l. 26: anthropogenic aromatic organic compounds have a small contribution;
IVOC have been shown to produce much more.

This is what we meant but failed to make clear by saying only "traditionally". We are now much more explicit:

"Traditional global models that have included SOA produced from "traditional" anthropogenic organic precursors (i.e. single compounds with well-characterized smog chamber yields such as aromatics) have predicted that these make a small contribution (about 10%) to global OA sources (Tsigaridis and Kanakidou, 2003;Farina et al., 2010), so they are not considered here. Potentially much larger sources of anthropogenic SOA from IVOC oxidation (Jathar et al., 2011;Pye and Seinfeld, 2010) are considered in the sensitivity studies (see Section 2.4)."

p. 10570, l. 1: There is no externally mixed OC? If not, why? Externally mixed primary
OC can evaporate and reduce the total aerosol number. What fraction of EC is
externally mixed, on average?

We are not aware of any global model that currently accounts for both aerosol size/microphysics <u>and</u> the semi-volatile nature of POA. A rigorous coupling of the organic partitioning to aerosol microphysics requires detailed mass transfer calculations and a large increase in the number of tracked species that would increase computational times by probably more than one order of magnitude. Doing so with a treatment of mixing state would require another, similar increase in computing time. We have added an explicit caveat that this process is not accounted for in the model (see response to similar comment above from the other reviewer).

We have revised the section on aging to address EC: "Because the aging timescale is relatively short, hydrophilic OM tends to dominate the organic aerosol composition. For the same reason, internally mixed EC constitutes 85% of the EC burden."

p. 10570, l. 11: How sensitive is the model on the choice of 140nm for activation?

To be clear, we do not assume that particles simply activate at 140 nm. This value simply measures the hygroscopicity of the pure "hydrophilic OM" species. This has been clarified:

"Pure hydrophilic OM is assumed to have a critical dry diameter for activation of 140 nm at 0.2% supersaturation, but activation calculations are performed with the composition of the "internally mixed" population as discussed above."

To answer the reviewer's question, we have performed sensitivity calculations where we vary the kappa value of organics from 0.1 to 0.3, covering the range of ambient observations. For internally mixed aerosols, an assumption we feel is reasonable for predicting global CCN, impact on CCN(0.2%) concentrations are ~5%, one of the smallest sensitivities we have examined. Unfortunately, we have not published these results, so there is no easy way to address this in the manuscript.

p. 10570, l. 11-15: where do these numbers come from? Are they best-guess, or found in the literature? What do other models use?

The assumed hygroscopicity values come from the Raymond and Pandis laboratory measurements already cited in the paper, but they are quite consistent with a wide range of other measurements. We have added:

"In terms of the k parameter (Petters and Kreidenweis, 2007), hydrophilic OM has k=0.18 and hydrophobic OM has k=0 at 273 K. The assumed density of hydrophilic OM is 1.4 g cm−3 and hydrophobic OM is 1.8 g cm−3. Kappa values inferred for ambient organics, laboratory SOA, and many single organic compounds found in OA are quite consistent with these values, mostly falling in the range of 0.1-0.3 (Suda et al., 2012; Petters and Kreidenweis, 2007; Gunthe et al., 2009)."

p. 10571, l. 20: Why neglect SOA distribution? The different POA/SOA splits can be justified due to different POA aging timescales, but monoterpenes SOA should always be present. Neglecting this source will strongly affect the aerosol microphysics, hence aerosol lifetime and CCN. In the idealized experiments the biogenic SOA should always be present. How about SOA from isoprene?

Elsewhere, we have added the caveat (as discussed in regards to similar concerns from the other reviewer):

"Therefore, the sensitivity experiments should be viewed as partly idealized representations of the differences caused by different POA and SOA formation pathways."

p. 10574, l. 12-15: Why not use the data mentioned at p. 10572, l. 15-17 as well?

The data mentioned is problematic for model evaluation either because they are very short field campaign data (therefore requiring a detailed simulation of the period in question) and/or they are relatively old compared to the emissions and meteorology used here. Since they are not directly helpful to this paper, we have deleted this section.

p. 10576, l. 5: "predict" is a very strong word, since the experiments presented in the manuscript are idealized and are not based on real SOA calculations.

This has been changed to "suggest".

Table 3: Why does nucleation (J10) change? You use organics in the binary nucleation?

The nucleation parameterization depends only on the sulfuric acid vapor concentration, so the change in J10 is due to some small, but undiagnosed, feedback. See our response to a similar question from the other reviewer.

Also, the microphysical growth is in reality the amount of aerosols that cross the 80nm size due to growth alone, right?

Correct. To avoid any confusion, we have added the following sentence to the discussion of Table 3: "Microphysical growth process terms in Table 3, including condensation of sulfuric acid, condensation of SOA, and aqueous production of sulfate by cloud processing, denote the rates at which particles cross the 80 nm size threshold chosen here."

References

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