Reply to Referee #2.

First of all, we'd like to thank anonymous referee #2 for the detailed comments, which help improve the manuscript. All the reviewer's points have been carefully considered and our responses are given below after each review comment (given in italics).

This paper is interesting in that it makes use of a chemistry-based scheme to explore the possible importance of different pathways to SOA. It is very useful that the community explores different approaches, and such chemical mechanisms do offer new insights compared to those arising from the popular VBS approach. Many of the results are indeed interesting, and useful to the community, but I am less sure that the results are convincing. This isn't helped by the fact that the text is often confusing and makes statements that aren't backed up by results presented in this paper.

Reply: we will check through the full paper again and do as much as we can to clarify the paper.

Firstly, I think that what the authors present is an interesting sensitivity study. They claim though (most prominently in the abstract, line 18) that their model "realistically" predicts the organic carbon mass observed in the Northern hemisphere. A quick look at the Figures would give a rather different impression. (Even if the scatter plots were perfect I would assume this was largely fortuitous or tuned, as I still doubt that today's knowledge would allow any group to "realistically" simulate OC!). The paper then has a tendency to interpret any problems in terms of just two or three specific processes, for which actually little or no evidence is presented (again, see abstract for the example of having too high update rates of two compounds).

Reply: Our scatter plots for the Northern Hemisphere may be fortuitous, but they are certainly not tuned. The word of "realistically" may not an appropriate word to describe the simulations for the Northern Hemisphere, but the model does a relatively good job compared to other models. We will make specific comparisons to other model results in the revised paper and drop the term "realistic".

Further, the model comparison with observations is limited in some important ways, and I think more should be done here (see below).

As a final general comment, the paper needs a thorough read through by the authors, to make sure that every sentence makes sense in itself, and to likely readers. All statements should be either supported by the material presented, from other literature, or omitted. I give examples below.

Specific Comments (major)

* Firstly, I thank the authors for their response to my query on the terminology concerning non-evaporative other SOA (ne_oSOA). As this term did seem to cover both real ne_oSOA (the oligomers) and "traditional" SOA (let's call it sv_SOA for semi-vol) which is in dynamic

equilibrium, an interesting follow-up question is then why the real ne_oSOA and the sv_oSOA are not documented separately? This would be consistent with the other species treated here e.g. from glyoxal, and would allow the reader to see the importance of the oligomorization assumptions.

Reply: The burden of sv_SOA is much smaller of that of ne_oSOA, e.g. 0.06 Tg of sv_SOA vs. 0.63 Tg of ne_oSOA for August in Simulation A. So we did not report them in the initial submission. This will be corrected in the revision.

* On a similar issue, how do the oligomer products fit into the results shown in Table S1?

Reply: For each semi-volatile species in the table S1, there is a corresponding non-evaporative oligomer product. All of these oligomers are lumped into ne_oSOA. This will be explained in the revised manuscript. Specifically, we will delete the explanation of ne_oSOA starting on line 25 on page 26356, i.e., "we call these SOA as ne_oSOA hereafter in this paper (see Table 1). "Ne" stands for "non-evaporative", and "oSOA" means "other oxidative SOA" to differentiate from SOA formed from the uptake of glyoxal, methylglyoxal and epoxide which will be described in Section 2.2.2 ", and move it to below the line 18 on page 26362 to add one more paragraph of explanation as below:
"For convenience, we refer to the SOA formed through the mechanism above as ne_oSOA hereafter in this paper (see Table 1). "Ne" stands for "non-evaporative", and "oSOA" means "other oxidative SOA" to differentiate from SOA formed from the uptake of glyoxal, methylglyoxal and epoxide which will be described in Section 2.2.2 below. It should be noted that this "non-evaporative" SOA is formed from condensed semi-volatile organic compounds, which are partitioned into the aerosol phase through gas-particle partitioning. These semi-volatile species are listed in Table S1."

* Page 26359. The issue of vapor pressure is critical to SOA schemes (shown in several papers), but there is hardly any mention of its importance here. How sensitive are your estimates to this parameter?

Reply: You are correct in that estimates of vapor pressure estimation are still very uncertain, which we discussed in the paragraph starting on line 14 on page 26359. Barley and McFiggans (2010) and other papers have already demonstrated the sensitivity of the predicted SOA mass to various vapor pressure estimation methods. In the revised paper, we will expand the discussion as follows: .

"The Myrdal and Yalkowsky method was shown to provide good estimates of vapor pressure for a set of organic species of importance for SOA formation (Camredon and Aumont, 2006). There are, however, many methods in the literature that have been developed to estimate vapor pressure. Barley and McFiggans (2010) assessed the ability of different vapor pressure methods to predict vapor pressures of lower volatility compounds. As noted by Barley and McFiggans (2010), the Joback method for estimating boiling points tends to underestimate the amount of material that should partition to the aerosol phase. Nevertheless, Booth et al. (2010, 2011) used the new room temperature low vapor

pressure data of polyfunctional compounds especially acids and diacids and showed that other methods (e.g. the method of Nannoolal et al. (2008)) do not predict the data as well. Thus, we should keep in mind uncertainties in the vapor-pressure estimates when drawing the conclusions regarding the ability of this mechanism to reproduce observations. For example, Simpson et al. (2007) showed that the total modeled carbonaceous aerosol over Europe was very sensitivity to the choice of vapor pressures. Valorso et al. (2011) tested the sensitivity of SOA formation from α-pinene photooxidation to three different vapor pressure estimation methods (i.e., Myrdal and Yalkowsky (1997), Nannoolal et al. (2008), and the SIMPOL-1 method from Pankow and Asher (2008)), and they found that the predicted SOA mass concentrations varied significantly from 8 to 28 μg/m^3 in "high NO$_x$" experiments. Despite the sensitivity of SOA formation to different vapor pressure estimation methods, Camredon et al. (2010) found the best agreement with experimental aerosol yields of **α**-pinene dark ozonolysis experiments using the Myrdal and Yalkowsky method. Thus, we use this method here."

* *It would have been useful to show or discuss what happens if you assume 0.5 or 2 day lifetimes for oligomer formation.*

Reply: You are correct. We had performed box-model sensitivity studies (not reported), but we agree that it is important to note this sensitivity. We performed two extra one-month (August) runs with one-month (July) of spin up assuming 0.5-day and 2-day lifetimes for oligomer formation, for simulation A. The global burden of ne_oSOA increases from 0.63 Tg for the 1-day-lifetime run to 0.95 Tg for the 0.5-day-lifetime run and decreases to 0.40 Tg for 2-day-lifetime run. We will continue these two simulations for an additional set of full one-year sensitivity tests and discuss these results in the final paper.

* *This chemical mechanism makes specific predictions of which pathways contribute to SOA formation in the atmosphere, e.g. (p. 26368) that 66% of the production rate is from methylglyoxal and epoxide. What is the observational evidence that these specific products account for such a large fraction of ambient OC?*

Reply: There are very few measurements that can quantify the contribution of the uptake the glyoxal, methylglyoxal and epoxide on the SOA formation for different locations. Volkamer et al. (2007) used the difference between the modeled and measured gas-phase glyoxal concentrations to suggest that the contribution of glyoxal could explain 15% of the SOA formation in Mexico City. Froyd et al., (2010) reported airborne measurements of organic sulfates, and found that the contribution of the epoxide sulfate ester to the tropospheric aerosol mass could be as high as 20% in regions downwind of isoprene emissions. There are no measurements that discern the importance of the uptake of these species on SOA formation on a global basis. Given the uncertainty in the SOA formation from glyoxal, methylglyoxal and epoxides (see the detailed discussion below) the high production rates of ne_MGLY and ne_IEPOX may need to be re-evaluated when more measurements are available. We will emphasize this uncertainty in the discussion section in the revised manuscript as follows:
"Trainic et al. (2011) conducted experiments to study the uptake of glyoxal on ammonium sulfate seed aerosols under hydrated conditions over a wide range of relative humidities, (RH

from 35% to 90%) and found that the reactive uptake rate decreased with increasing RH. The ratio of the final organic aerosol mass to the seed mass at the 50% RH condition was similar to that found by Liggio et al. (2005a) (who conducted their studies at 49% RH), but decreased by 57% when the RH increased from 50% to 90%. This trend was attributed to the slower glyoxal oligomerization rate caused by the dilution of the ammonium sulfate aerosol at the higher RH values (Liggio et al., 2005a; Trainic et al., 2011). Thus, the work of Trainic et al. (2011) may indicate that the uptake coefficient adopted here (which was based on Liggio et al. (2005a)) is too high in much of the tropics where typical RH values are around 90%. In addition, the simple treatment of irreversible surface controlled uptake might be misleading if there is competition between reversible vs. irreversible uptake and of bulk reactions vs. surface processes (Ervens and Volkamer 2010) for SOA formation. Furthermore, the same uptake coefficient for both cloud droplets and aqueous aerosols can not account for the difference in the chemistry of carbonyl compounds that occurs in cloud water and aerosol water (Lim et al., 2010; Ervens and Volkamer 2010) Therefore it may be that improvements to the processes responsible for uptake of glyoxal, methylglyoxal and epoxide would improve the model. Given the sparse amount of data, however, the conclusion that the model overestimates of surface OM concentrations observed at 3 sites in tropical forest regions may not necessarily lead to the conclusion that the model is in error. It would be valuable to have more measurements and model studies to examine the properties of organic aerosols in the tropics."

Other Comments

* Page 26349, 1st paragraph. It might be good to mention already here the IVOC and OPOA concepts briefly (Robinson et al, 2007). The simple distinction between POA and SOA is fading rather quickly these days.

Reply: We will add: " The distinction line between these two categories, however, is changing as a result of recent studies which show that the volatility of emitted particles can change as a result of the oxidation of primary emissions and that previously unrecognized semi-volatile and intermediate volatility compounds (S/IVOC) provide an extra source of SOA (Robinson et al, 2007; Jimenez et al., 2009)".

* Page 26349, line 24. The Simpson et al study seemed to do quite well for Nordic sites, so I didn't intrepret that as a general failure to model SOA? Actually, that study demonstrated how the SOA results were very sensitive to vapor pressure assumptions.

Reply: Thank you. The Simpson study was able to do well in Northern Europe, but not in Southern Europe. We will correct this in the revision, and also mention their sensitivity to vapor pressure assumptions. Specifically, we will change this text as follows: "Simpson et al. (2007) found that their SOA modeling framework under-predicted SOA concentrations at Southern European sites, but predicted that SOA levels were within the range of observations in Northern Europe."

* Page 26350, The Slowik paper year is wrong

Reply: Thank you, we will correct it to 2011.

* *Page 26350, line 27. The text states that the problem "requires" a rigorous and thorough gas-phase chemical mechanism. Donahue has argued in several papers (e.g. Donahue et al 2009) that the complexity of SOA requires the opposite - an approach where we know that we cannot know the individual species and reactions. A short discussion or rephrasing might be appropriate here.*

Reply: You are correct. There are lots of species and reactions that we still do not know, so the development of a "rigorous" mechanism may not be completely possible. The method of "fitting" through the use of a volatility basis set, may be desirable for some applications. However, we prefer to tie the formation of SOA to an explicit chemical mechanism, so that the contribution the specific reaction mechanism and the specific individual species to SOA formation is known. Without this, it may not be possible to compare specific species with measurements (though, admittedly few are available now). Also, we think our approach can be updated as chemical mechanisms are further developed. Furthermore, The explicit mechanism provides a prediction for sensitivity to precursor emissions (e.g. NO_x) based on a more detailed chemical reaction sequence. It is true there is a lot of uncertainty in this - but a simpler representation such as traditional 2-products model has the same uncertainties, and a weaker basis for predicting the impact of NO_x on SOA in particular. We will add a discussion of these issues to the revised manuscript as follows: "The complexity of the emitted VOC mixture and the degradation chemistry requires a rigorous and thorough gas-phase chemical mechanism that describes SOA formation. Donahue et al (2009) argues that the complexity of SOA cannot be followed in detail and requires an approach where species are lumped into individual "volatility basis sets". Although the development of a " rigorous" mechanism may not be completely possible given that there are many species and reactions that we still do not know, we prefer to tie the formation of SOA to an explicit chemical mechanism, so that the contribution of the specific reaction mechanism and the specific individual species to SOA formation is known. Without this, it may not be possible to compare specific species with measurements (though, admittedly few are available now). Also, we think this approach can be updated as chemical mechanisms are further developed."

* *Page 26351, 1st line. What does "As a consequence" mean here? The previous sentence mentioned that isoprene was 1/3 of VOC emissions, nothing to directly support the consequence stated here.*

Reply: we will change this to say: " In addition, SOA derived from biogenic VOCs dominate the predicted global atmospheric SOA burden…"

* *Page 26351, line 3, "some measured data". Actually, there is a wealth of data on this now, not just some.*

Reply: we will change this to say, "many measured data".

* *Page 26352, line 9 grammar: better "has been" than "was also".*

Reply: This will be changed.

Page 26353, line 4 grammar: what do you mean, "we also"? ("we" might do?)

Reply: this will be changed to: "In this work, we use 1997 meteorological fields...".

Page 26354 - explain where the short names used here (e.g. ACO2) are explained

Reply: We will add the explanations in the end of this page as follows: "The short names used here (e.g., HAC) are adopted from Ito et al. (2007). The full species and chemical formula for them can be found at http://www-personal.umich.edu/~sillman/web-publications/Ito_2007_TableA1.pdf".

Page 26356. It was confusing to read the sentence starting "Therefore ", when the previous sentence seems to have nothing to do with the text following "Therefore".
Please re-phrase.

Reply: we will modify this to say, "In this paper, we carried out three different simulations…"

Page 26358. The formatting of the list at the end needs to be improved

Reply: done.

Page 26358, line 17. Add "and properties" to distribution. Many things can go wrong even if the products were captured perfectly, and we seem to be very far from that stage at the present time.

Reply: We agree and will add this. We agree that we still have many uncertainties in the explicit chemistry mechanism. But it is useful to explore an explicit mechanism even before all the chemistry is fully understand.

Page 26365, line 13 - reference for terpene emissions?

Reply: We will add the Guenther et al. 1995 reference, so this sentence will be changed to "The biogenic terpene source is 117.6 Tg C/year, which is based on the work of Guenther et al. (1995) as modified by Wang et al. (1998)".

Page 26366, lines 5-7 - formatting of rate coefficients mangled.

Reply: We will correct this, thank you.

Page 26369 - it is important that the vapor pressure be sub-cooled if necessary.

Reply: Do you mean page 26359? Yes, we consider the sub-cooling if necessary when calculating the vapor pressure, so we will clarify this on line 13 of page 26359 by adding "(sub-cooled if necessary)" after the "compound vapor pressure".

Page 26366 - how was the dry deposition of semivolatiles treated? Bessagnet et al. (2010) showed it to be quite important, as also noted by Hallquist et al., 2009.

Reply: For the semi-volatile gas phase species, we used the dry deposition algorithm of Wang et al. (1998), which follows the methodology of Wesely et al. (1985). We will clarify this in the revised version and mention the study of Bessagnet et al.(2010). Specifically, we will describe it on line 19 on page 26366 as follows: " Bessagnet et al. (2010) demonstrated the importance of dry deposition of semi-volatile organic compounds on estimating SOA concentrations. In this paper, we treat dry deposition using the same algorithm as that of Wang et al. (1998)."

* *Page 26367, paragraph starting on line 18 - this paragraph started a distracting discussion in the section entitled results. Move later in the text.*

Reply: we will move this after line 19 on page 26368, right after " Simulation B".

* *Page 26369. I found the chemistry description here confusing, partly as the authors do not distinguish between RO2 in general, and the specific RO2 comnpounds that form their products. I suggest a more explicit approach using more numbered equations, and greater clarity.*

Reply: we will specify the specific RO2 compounds (i.e. the sources) that produce PRN2. So the lines between line 11and line 12 will be changed to : "Globally, PRN2 mainly originates from the reaction of RIO_2 (RO2 from isoprene) with NO, although it also comes from a few other reactions (e.g. the reaction of RO2 from monoterpenes with NO). The reaction of RIO_2 with NO competes with isomerization through the 1,5-H shift or 1,6-H shift. This competition reduces the PRN2 formation rate from the reaction of RIO_2 with NO."

* *Page 26369, what does "may" originate ... "potentially" mean. A little over kill on the qualifiers I think!*

Reply: we will clarify this as stated above: "Globally, PRN2 mainly originates from the reaction of RIO_2 (RO2 from isoprene) with NO, although it also comes from a few other reactions (e.g. the reaction of RO2 from monoterpenes with NO)."

* *Page 26370 suffers similar problems. The last 2 sentences say that the only pathway to form epoxides is through the RO2 + HO2 reaction (R3), but RO2 includes CH3O2 as well as any more complex radicals.*

Reply: We will change the paragraph starting on line 28 on page 16370 as follows:
"The only pathway to produce epoxides is the reaction of RIO_2 with HO_2. The introduction of the Peeters et al. (2009) HO_x recycling pathway decreases the production of epoxides in spite of the increase in RIO_2 and HO_2 concentrations. Most of the epoxides are lost by reaction with OH,

and about 20% reacts on sulfate aerosol to form organic aerosols."

Page 26373. As this model has explicit chemistry, I was confused to read that it only predicts OM. Don't you have the full chemical formula of each product, and hence both OC and OM?

Reply: Yes, we have. These are listed in table S1 for every chemical formula of each product. We will make this clearer in the revised manuscript.

Page 26374. You say the larger particles are not captured very well by the model. Do you have any large particles at all?

Reply: We do not have large organic particles (> 2.5 micro meter) in our model, but the EMEP observations include large particles (2.5-10 micro meter). We will explain this in the revised version as follows: "These large particles are not captured very well by our model, since the model only accounts for PM2.5 particles.".

Page 26374, line 13 ... since isoprene emissions are low .... I would have thought it just as relevant that POA sources are high (e.g. biomass burning). (The Simpson et al. CARBOSOL paper mentioned there used levoglucosan to demonstrate that accounting for wood-burning very well "fixed" the wintertime OC concentrations.)

Reply: Thank you. High POA sources are another possible reason. We will change this as follows: "While SOA dominates OM in the summer, in the winter POA contributes most to OM since POA sources are high (e.g. biomass burning) and isoprene emissions are low.".

Page 26374. Many Szidat papers have shown similar results, e.g. Szidat et al., 2007

Reply: We will cite the Szidat papers also. Thank you. In particular, we will add the above on line 13 on page 26374: "While SOA dominates OM in the summer, in the winter POA contributes most to OM since POA sources are high (e.g. biomass burning) and isoprene emissions are low, which is consistent with other studies (Szidat et al., 2007; Simpson et al., 2007)."

Page 26374. I wonder why the one Gilardoni paper was used for this comparison. The site involved is rather subject to local emissions within the Po Valley, and GEOSCHEM cannot resolve such areas I think. The CARBOSOL sites would have provided a more regional data set for comparison to GEO-CHEM. In addition, the referees had some concerns about the summertime results of the Gilardoni et al. paper that I do not think were adequately addressed. Relying on only one such study in one location is rather dangerous though.

Reply: We will add a comparison to the CARBOSOL sites to Table 8 (only surface sites: Aveiro and K-Puszta) (see below). Here, we only pick up two surface sites (i.e., Aveiro and K-Puszta) as did Simpson et al. (2007). It may be interesting to include the other two mountain sites, but surface sites reflect the boundary layer sources of OA (e.g. biomass burning) more directly. In addition, the comparison with these surface sites may supply more comparability to the comparison with the EMEP network for which most

observations are near the surface. From the table below, the primary biomass burning organic carbon in the winter is significantly under-presented in the model. This may be due to the coarse resolution of the model, which cannot resolve the local emissions. Another possible reason for the winter-time discrepancy is that the emissions from domestic wood combustion are not fully represented in our emission database. As an aside, we are using the IMPACT model, not GEOS-CHEM.

Station Name	Source	Winter				Summer			
		Observation	Simulation A	Simulation B	Simulation C	Observations	Simulation A	Simulation B	Simulation C
Aveiro	POC_bb	8.96	0.02	0.02	0.02	0.28	0.06	0.06	0.06
	POC_ff	0.18	0.08	0.08	0.08	0.30	0.02	0.02	0.02
	SOC_bio	0.70	0.08	0.08	0.09	2.54	0.28	0.21	0.27
	SOC_ff	2.27	0.04	0.04	0.04	0.30	0.01	0.01	0.01
K-Puszta	POC_bb	4.30	0.01	0.01	0.01	0.32	0.03	0.03	0.03
	POC_ff	0.59	1.75	1.75	1.75	0.27	0.56	0.56	0.56
	SOC_bio	2.24	0.04	0.03	0.03	3.48	2.06	1.68	2.10
	SOC_ff	1.63	0.25	0.23	0.23	0.19	0.18	0.13	0.14
Ispra	POC_bb	11.9	0.02	0.02	0.02	0.5	0.04	0.04	0.04
	POC_ff	1.2	0.82	0.82	0.82	0.6	0.34	0.34	0.34
	SOC_bio	2.0	0.13	0.12	0.13	3.1	1.41	1.19	1.43
	SOC_ff	2.3	0.25	0.25	0.25	1.3	0.22	0.18	0.20

Page 26375. On a similar theme, why compare with urban sites from Zhang et al. 2007? GEOS-CHEM isn't designed for that, and these sites just confuse the results. I think this should be removed.

Reply: Given that we have so little data, we wanted to include as much comparison as possible. This is also in keeping with your statement "*the model comparison with observations is limited in some important ways, and I think more should be done here (see below).* We will mention the fact that the model resolution is too poor to fully capture urban concentrations.

Page 26376, line 15. Clarify where this 10% came from? Do you have observed POA?

Reply: We will clarify this in the following manner: "Therefore it seems likely that either the production rate of SOA in the model is too large or the biogenic sources are too strong, although the prediction of POA at this site explains about 10% of the total predicted OM."

Page 26376-26377, general. I am not convinced that comparing a model with 4x5 degrees resolution with local isoprene data can provide much information on the reasons for over or underestimation of SOA. This paragraph is attempting too much detail. Some of the speculation could be moved to the discussion section, some could be omitted.

Reply: We agree with that it is difficult to determine the cause of underestimation of SOA from local data on isoprene from a 4x5 model. But part of our reason for using an explicit mechanism is to see whether different oxidation schemes can lead to better results, and, as stated above, there are very few measurements that include both a suite of measured VOC's as well as SOA, so even "local" measurements are of interest. As shown, when the isoprene

is fit better (in Simulation B, for example), the SOA only decreases by a small amount. And it is too high for all mechanisms, so something still needs to be improved (BTW, we would not conclude this were it not for the availability of the 3 tropical measurements). This most likely will involve the glyoxal and methylglyoxal part of mechanism, but could be all parts. We will move some of this discussion to the discussion section.

* Page 26378, line 9. Looking at Fig. 7a I don't see support for the statement that the results "compare favorably with the INTEX-B results". The most interesting part of the figure is that the observations cover a very wide range, and GEOS-CHEM fits within that, closer to INTEX-B at high altitude.

Reply: We should have said " compare favorably with the INTEX-B results at latitudes higher than 3 km".

* In general, why do all simulations give the same results for Fig. 7a? They are hardly distinguishable here. Over North America (Fig 7b) there are modest differences at least.

Reply: This is due to the different contributions of POA to total OA. Over East Asia, POA accounts for most of the OA (e.g. about 88% are POA at the surface), while POA only explains a small portion of OA in the North America region (e.g. POA only account around 12% at the surface). And POA changes very little from Simulation A to Simulation B and C. We will state this in the revision.

* Page 26378, same paragraph. I don't think 3 significant figures are needed when comparing OC data.

Reply: We will correct this in the revision to 1 significant figure in Fig. 7a and to 2 significant figures in Fig. 7b.

* Page 26378. It would be good with a summary of the model performance for SO4.
One cannot draw many conclusions about SOA predictions without knowing how well a model performs for the easier compounds.

Reply: Thank you. We used the same mechanism and model for SO4 as used by Liu et al. (2005) and they did a thorough evaluation. We will note the need to compare to SO4 and refer readers to that paper. Specifically, we will add the following on line 22 on page 16378: " The IMPACT model was used to predict sulfate aerosol by Liu et al. (2005) based on the aerosol module developed by Herzog et al. (2004). They did a thorough evaluation of its capability to predict sulfate, and we use the same mechanism and model for sulfate here."

* Page 26380, line 22. I didn't see much evidence that you underpredict POA in winter.
I agree it is likely, but not shown.

Reply: We will add the comparisons with CARBOSOL measurements to support this.

* Page 26380, line 28. Did the model "significantly overestimate OM". The model has coarse resolution, and the data are sparse.

Reply: We will state the magnitude of the overestimate here (about a factor of 3). We think this is significant, but it is better to be quantitative. While the data are sparse and the resolution of the model is coarse, we think a factor of 3 at 3 places in the tropics is an important overestimate.

Page 26380, last line. I disagree.

Reply: Do you refer to the last full sentence on this page? We will be more specific and say " the model simulates Asian pollution layers during the INTEX-B campaign very well above 3 km"

Figures:

Fig. 1 has no units on y-axis

Reply: We will add it.

Add year of modeled and observed data to all plots

Reply: We will add the years for the observed data in the caption of all plots. However, it would be tedious to explain that the model run uses emissions for year 2000, and meteorology for 1997 in each plot. We will add this to Table 1, which describes all three runs.

Tables:

S1 - is this the global total? Which temperature is K valid for? Format numbers using decimals where possible: it is hard to spot the large contributers when all are in scientific notation.

Reply: Yes, it is the global total. K is for 298 K. We will state this in the revised manuscript and change the numbers to decimals where possible.

S3, S4: number reactions please.

Reply: done.

Add year of modeled and observed data to all plots

Reply: We will add the years for the observed data in the caption of all plots. However, it would be tedious to explain that the model run uses emissions for year 2000, and meteorology for 1997 in each plot. We will add this to Table 1, which describes all three runs.

Extra Refs:

Bessagnet, B.; Seigneur, C. & Menut, L. Impact of dry deposition of semi-volatile organic compounds on secondary organic aerosols Atmos. Environ., 2010, 44, 1781 -1787

Szidat, S.; Prévôt, A. S. H.; Sandradewi, J.; Alfarra, M. R.; Synal, H.-A.; Wacker, L. & Baltensperger, U. Dominant impact of residential wood burning on particulate matter in Alpine valleys during winter Geophys. Res. Lett., 2007, 34, L05820

Reference:

Booth, A. M., Barley, M. H., Topping, D. O., McFiggans, G., Garforth, A., and Percival, C. J.: Solid state and sub-cooled liquid vapor pressures of substituted dicarboxylic acids using Knudsen Effusion Mass Spectrometry (KEMS) and Differential Scanning Calorimetry, Atmos. Chem. Phys., 10, 4879–4892, doi:10.5194/acp-10-4879-2010, 2010.

Booth, A. M., Montague, W. J., Barley, M. H., Topping, D. O., McFiggans, G., Garforth, A., and Percival, C. J.: Solid state and sub-cooled liquid vapor pressures of cyclic aliphatic dicarboxylic acids, Atmos. Chem. Phys., 11, 655–665, doi:10.5194/acp-11-655-2011, 2011.

Camredon, M., Hamilton, J. F., Alam, M. S., Wyche, K. P., Carr, T., White, I. R., Monks, P. S., Rickard, A. R., and Bloss, W. J.: Distribution of gaseous and particulate organic composition during dark α-pinene ozonolysis, Atmos. Chem. Phys., 10, 2893–2917, doi:10.5194/acp-10-2893-2010, 2010.

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