

Interactive comment on “Improving the representation of secondary organic aerosol (SOA) in the MOZART-4 global chemical transport model” by A. Mahmud and K. C. Barsanti

The authors would like to thank both reviewers for their comments on this manuscript. We appreciate the time and effort put into these reviews. We have worked diligently to address the concerns raised and incorporate the suggestions made. Please see our detailed responses below.

Reviewer #1

Major

R1C1. It is unclear how MZ4-v1 compares to the baseline simulation and indeed why exactly this update was implemented. Are the new 2p yields based on new experimental results? It would be useful to compare 2p yield parameters for the different precursors in the baseline and MZ4-v1 scheme. How do the volatilities compare?

Response: The parameters used for the baseline (base-case) simulation are the default MOZART parameters normalized to the same temperature and density. The default parameters include parameter values for experimental temperatures as high as 308K, which do not represent SOA formation at ambient temperatures. The base-case values are now included in Table 1 so that the yields and volatilities of the parameters for each of the simulations can be compared. The motivation and derivation of the 2p-VBS parameters are now described in more detail in a supplementary section.

R1C2. The comparison with observations (3.2) is uneven and inadequate. The authors are largely citing a random handful of previous studies in the literature. The paper cites no surface observations over North America or Europe! I suggest that the authors start by taking the global OA observations reported by Zhang et al., 2007 and quantitatively compare those with their simulation (scatter plot?). They could then add to these values the measurements from Asia and South America that are cited in the manuscript (regions not necessarily well represented in Zhang et al., 2007).

Response: We thank this reviewer for suggesting the use of the observational data from Zhang et al., 2007. A new figure (Fig. XX) has been added to the revised manuscript that shows a scatter plot of modeled (y-axis) vs measured (x-axis) organic aerosol (OA) mass concentrations at different locations around the world. Measured data were obtained primarily from Table SI-1 of Zhang et al. (2007) for North America, Europe and Asia. The figure shows that the updated model performed relatively better for sites in North America compared to the sites in Asia and Europe in predicting OA concentrations; over-predicted at most locations in Europe and under-predicted at all locations in Asia. A discussion of the measured dataset and model results is provided in section 3.1 of the revised manuscript.

R1C3. High NO_x yields were used in this work and some discussion of the impact of this assumption should be included; particularly in tropical regions, where high NO_x is generally not appropriate.

Response: The assumption of high NO_x was retained to be consistent with the default MOZART SOA parameters. The default parameters for both biogenic and anthropogenic precursors are from “high NO_x” chamber studies. In addition the SOA module is not coded to allow branching between high and low NO_x conditions, which currently is considered the appropriate approach

for treating the competition for RO₂ between NO and HO₂ under “high” and “low” NO_x conditions, respectively. With the current SOA module, one could apply high NO_x parameters and then low NO_x parameters to get a sensitivity range; however, this was determined not to be necessary because of the overwhelming contribution of isoprene to SOA on the global scale (and high and low NO_x parameters for SOA currently do not exist). Thus, for the tropical regions, and other regions dominated by isoprene emissions, there is no significance to the assumption of high vs. low NO_x conditions, from the perspective of SOA formation (i.e., not considering changes in the gas-phase chemical mechanism, such as radical recycling). Future work will include application of a revised SOA module that does allow branching and thus consideration of different NO_x regimes.

Minor

R1C1. Abstract, line 6: clarify that these VOCs were ADDED to the new scheme and are not the only VOCs under consideration in the MZA-v2 scheme.

Response: This has been clarified in the abstract. The line now reads “...and by treating SOA formation from the following additional volatile organic compounds (VOCs): isoprene, propene and lumped alkenes in the model.”

R1C2. Page 4189, line 20: “double estimated” is confusingly phrased. The 16.4 Tg/yr number cited from Henze and Seinfeld is clearly not double the production from Chung and Seinfeld of 11.2 Tg/yr. Suggest you re-phrase to something like “Henze and Seinfeld : : : isoprene makes up over half of total SOA production (16.4 Tg/yr)”.

Response: We thank the reviewer for pointing out this potentially confusing phrasing. Henze and Seinfeld (2006) estimated nearly double the amount of global SOA production in their model when they considered isoprene as an SOA precursor (not as compared to Chung and Seinfeld). The revised manuscript has been reworded as follows: “Henze and Seinfeld estimated that isoprene, which had been ignored previously, could nearly double estimated global SOA production (from 8.7 Tgyr⁻¹ to 16.4 Tgyr⁻¹).”

R1C3. Page 4189, lines 20-22: It should be clarified that the estimates from Spracklen et al. are based on observations and are not bottom-up as in the previous cited studies. Indeed, the sentence that follows in lines 22-24 incorrectly implies that the difference between Chung & Seinfeld and Spracklen et al. has something to do with improved estimates of VOC fluxes and SOA parameters. This is not the case.

Response: SOA production from anthropogenic and biogenic sources estimated by Spracklen et al. (2011) is constrained by measured dataset – this has been clarified in the revised manuscript. The authors intended to emphasize that the differences among previously “modeled” estimates are largely due to changes in the identities and fluxes of the VOC precursors. This has also been clarified on page 4189 in the revised manuscript.

R1C4. Page 4192, lines 5-14: References for aerosol modeling in MOZART-4 required.

Response: We regret this inadvertent omission. The following references have now been added in regard to the different components of aerosol model: Barth et al., 2000; Chin et al., 2002; Jacob, 2000; Lamarque et al., 2005; Tie et al. 2001,2005 and Tie et al., 2003.

R1C5. Page 4192, line 26: “C > 3” is technical, would be helpful if you spelled out “more than 3 carbons”

Response: In the revised manuscript, C>3 has been spelled out in line 26 on page 4192, but the abbreviation is retained in later occurrences.

R1C6. Page 4193, lines 11-12: Is the baseline scheme that described by Lack et al., 2004? If so, clearly cite at the beginning of section 2.2. (If not, indicate the origin of baseline simulation). Also clarify if an iterative solution was obtained for SOA when SOA is included in Mo, and whether irreversibility was assumed.

Response: This paragraph has been rewritten to clarify that the SOA module is based on gas/particle partitioning theory (Pankow) in the form of the two-product model (Odum), with some specific assumptions based on Lack et al. (2004). It has been noted that partitioning is not calculated iteratively, and SOA formation is assumed to be irreversible.

R1C7. Page 4194, lines 3-5: Reference experimental data used to obtain these fits.

Response: The parameters used in the simulations were either previously published, or in the case of the 2p-VBS from a manuscript that is to be submitted, thus no data fitting was done for this manuscript. A description of the motivation and derivation of the 2p-VBS parameters is now provided as a supplemental section.

R1C8. Page 4195, line 1: what are “major stable species”??

Response: while levels of the reactive gas-phase species came from emissions inventories (anthropogenic) or models (biogenic), a number of long-lived species (i.e., stable species) came from observed concentrations. These species included CH₄, H₂, and N₂O. In the revised manuscript, this description has been moved closer to the description of the emission inventories and model and now reads: As described in Emmons et al. (2010), concentrations of long-lived species (e.g., CH₄ and N₂O) are obtained from ground- and satellite-based measurements.

R1C9. Page 4195, lines 15-16: This comparison is perhaps limited by using only monthly output, but this is not an inherent characteristic/limitation of global CTMs (output timescales limited only by model time step).

Response: The authors agree with the reviewer’s comments and have revised the sentences in the results and discussion as follows: “For example, most of the measurements are taken at specific locations that might be influenced by local emissions. This makes the comparison with global chemical transport model output, which is typically saved for grid cells in the order of degrees, quite difficult. ”

R1C10. Page 4195, lines 17-20: This is unclear and poorly worded. It appears that the authors are discussing limitations associated with estimating SOA using OC-EC tracer methods. This is not the only method for estimating SOA, so a more complete discussion is required with respect to challenges in reporting SOA. In fact one could also point out here some of the inherent measurement errors for OA (detection limits, artefacts, size cuts, etc) that are also relevant.

Response: The authors will rewrite this section with expanded discussion of SOA measurement techniques, and their challenges in light of the reviewer suggestions.

R1C11. Page 4200, lines 12-14: Heald et al., 2011 actual shows that their model reproduces the vertical profile of OA in most locations.

Response: While Heald et al. (2011) stated that “ the standard GEOS-Chem simulation reproduced the observed vertical profile” in reference to the shapes of vertical profiles Heald et al. (2011) also stated that “observations are underestimated in 13 of the 17 field campaigns (the median observed to simulated ratio ranges from 0.4 to 4.2)”.

R1C12. Page 4200, lines 20-24: Henze et al., 2006 should be cited here – they first showed that including isoprene SOA increases OA loading aloft (and discuss why).

*Response: Henze and Seinfeld (2006) has been cited in the revised manuscript in regard to simulated increase in SOA aloft due (in part) to the significant emissions of isoprene. Henze and Seinfeld also attribute the increase in SOA aloft to the fact that isoprene SOA products are shifted less toward the particle phase than that of the other VOCs (i.e., are of higher volatility), however, that is not what is represented by the model parameters. The gas/particle partitioning constant of the lower volatility product (product 2 in Henze and Seinfeld, product 1 in this work) is an order of magnitude higher (i.e., partitioning shifted to the particle phase) than that of the other precursor hydrocarbons (see Chung and Seinfeld, 2002). Thus it is our conclusion, as stated in this paper, that instead the isoprene oxidation products are *less volatile* and thus able to stay in the particle phase even under dilute conditions. The revised text now reads: “..had a significant effect on vertical profiles. The reason for this increase in SOA aloft is likely twofold. First, as noted by Henze and Seinfeld (2006), is the magnitude of isoprene emissions; and second, is the relatively...” on page 4200 in the revised manuscript.*

R1C13. Page 4201, lines 17-27: The authors could (and should) verify their hypothesis for the change in lifetime of total SOA by calculating the lifetimes of the anthropogenic and biogenic SOA separately.

Response: Model simulations were re-run with only anthropogenic and biogenic SOA precursors separately. As expected, a simulation with only anthropogenic precursors predicted SOA lifetime of 17.61 days which is higher than the predicted lifetime of 11.21 days by the simulation incorporating biogenic precursors only. Contributions from anthropogenic precursors to the total SAO is significantly low compared to the biogenic precursors, and much of the SOA is formed in the lower atmosphere where the SOA removal processes are most effective. . This finding is consistent with the discussion on Page 4201 lines 17-27. It will be added in the revised manuscript to strengthen our assumption about the reported decrease in SOA lifetime from the base-case prediction.

R1C14. Page 4205, line9-12: It might be worth noting that this model simulation (and that of Lin et al.) still considerably underestimate the ACE-Asia observations, despite an overestimate of surface observations in SE Asia that is noted in Section 3.2.1. It's not clear how this model performs at the surface in East Asia (but perhaps after adding comparisons with Zhang et al., 2007 this could be addressed).

Response: Yes. This is now addressed in the revised manuscript by the addition of comparisons with Zhang et al. (2007), as suggested in major comment R1C2.

R1C15. Section 3.2.3: There are several other global models studies to which this one could be compared, including Hoyle et al., 2007 and several of the Tsigaridis et al. papers.

Response: Hoyle et al. (2007), and Tsigaradis and Kanakidou (2007) are already cited in the introduction section of the manuscript. Additionally, findings on global budget and lifetime of SOA from Hoyle et al. (2007), and Tsigaridis et al (2003) are also discussed in section 3.2.3 of the revised manuscript.

R1C16. Page 4207, line 11: “Figure 7 shows a measure” is incorrect. The figure shows a simulation, not a measurement.

Response: This has been corrected in the revised manuscript and now reads: “Figure 7 shows the spatial distribution of total aerosol optical depth (AOD) for the base-case MOZART-4 simulations”.

R1C17. Section 2.1/3.3: The model description needs to include discussion of the optical and size assumptions and formulation of the AOD calculation in MOZART-4 with appropriate references.

Response: The following discussion and reference for the AOD calculation is now provided in the revised manuscript. In MOZART-4, the AOD is calculated only when the photolysis rates are calculated (zenith angle, SZA is less than 90 °), so the monthly average DTTOTAL (which holds calculated total AOD) variable was scaled by the fraction of daylight hours per day. Variable FRACDAY holds the daylight hours information in the model. Total AOD reflects contributions from aerosols originating from primary and secondary organics, sea-salt, sulfate, nitrate, and dust. The FTUV module in MOZART-4 generates 17 wavelength bins for each of which optical properties of those aerosol types are utilized. The optical and physical property data for each of those types of aerosol were obtained mostly from OPAC dataset by Hess et al., 1998. Except dust, other aerosol types including sulfate, nitrate, organic carbon, carbon black, SOA and sea salt are treated for water uptake in the AOD calculation in the model. In the absences of adequate data SOA is treated similarly as the OC in AOD calculations.

R1C18. Section 3.3: Given the relatively small role that SOA plays in the global AOD simulation, I suggest that the authors trim this section. Table 4 and one of Figure 7/8 could be removed.

Response: We have adopted the reviewer’s suggestions in part by removing Fig. 8, but have retained Table 4 because we believe that the table contains an important analysis of SOA contribution to the total AOD for various regions in the world.

R1C19. Section 4: The conclusions largely repeat numbers given in the Results section. I suggest that these be summarized more succinctly.

Response: The authors agree with the reviewer and have rewritten the conclusion section to be more succinct, while retaining the quantitative discussion of the model results.

R1C20. Figure 2: The comment on mass of C is confusing. Is the figure actually showing total mass or mass of C only? If the later, the label on the color bar should be changed.

Response: The authors agree that this comment is indeed confusing and has been omitted in the revised manuscript. All figures are now in units of $\mu\text{g m}^{-3}$.

Reviewer #2

Summary Comments

Are the authors using the VBS to calculate the 2-product model parameters, and then they use the 2-product model formulation? If yes, they should not name their method VBS, since it is not. Maybe they indeed use a simplified VBS approach with less (probably 2) bins? This has to be described in greater detail. How many bins do they use? What assumptions do they make for the aging of the semi-volatile gases and conversion to less volatile species? How do the volatility bins correlate with the 2-product model values in Table 1?

Response: The discussion of the 2p-VBS parameters has been significantly expanded and added as a supplemental section. The expanded section addresses the questions and concerns raised by this reviewer regarding the derivation of the 2p-VBS parameters, aging assumptions, etc. In short, the 2p-VBS parameters are derived by generating pseudo-data from VBS fits (parameters from Tsimpidi et al., 2010) and then fitting the pseudo-data using the Odum 2p (two product) approach. The result is a reduced number of parameters per precursor (that can be used with any 2p model framework) that reproduces calculated SOA yields obtained with the full set of VBS parameters, including their temperature dependence. "Aging" or continued oxidation of volatile/semi-volatile species is not considered. The full derivation, testing, and application of the 2p-VBS parameters will be presented in a forthcoming manuscript by Barsanti et al.

Specific comments

R2C1) P. 4192, l. 5-7. The model is not having dust? In figure 7 it appears that it does.

Response: The aerosol model in MOZART-4 does include dust. It is treated as four size distributions: 0.05–0.5, 0.5–1.25, 1.25–2.5, and 2.5–5.0 μm . Dust is not assumed to take up water in the model. The optical properties of dusts are taken from Hess et al. (1998) that are used in aerosol optical depth (AOD) calculations in the model.

R2C2) Eq 1: There are no gas-phase products from this reaction?

Response: Eq. 1 does not distinguish phase. The oxidation reaction happens in the gas phase and products are then allowed to partition into the particle phase (as stated in the text); the products then will exist in both the gas and particle phases depending on their volatilities.

R2C3) P. 4193, l. 14: How is "small amount of Mo" defined? How small? How does the choice of this amount affects results, if at all?

Response: M_o is calculated from both hydrophilic and hydrophobic components of organic carbon (OC1+OC2) in the model at each time step. M_o directly affects the SOA yield calculations through Equation 2 in the manuscript. Following the method of Lack et al. (2004), the model utilizes SOA "bulk yield" values if the amount of M_o is less than or equal to $0.2 \mu\text{g m}^{-3}$, otherwise SOA yield is calculated based on partitioning theory (Equation 2). In the revised manuscript, "small amount" has been replaced with the specific value of $\leq 0.2 \mu\text{g m}^{-3}$.

R2C4) P. 4193, l. 20: Why these two reactions are excluded from the 2p-VBS?

Response: As noted previously, the discussion of the 2p-VBS parameters has been expanded, including the motivation for deriving the parameters and the approach. VBS parameters from Tsimpidi et al. (2010), which serve as the basis for the 2p-VBS fits are not available for monoterpenes+NO₃. In the assessment of the 2p-VBS fits it was determined that the VBS parameters for isoprene (and thus the 2p-VBS parameters) did not represent the bulk of available chamber data and thus more widely used parameters, such as those from Henze and Seinfeld, were more appropriate.

R2C5) P. 4194, first half: This discussion must be moved earlier.

Response: The SOA model revisions, as described in the first half of 4194, have been paraphrased and included in the introduction. We believe that the discussion of the model revisions should not be moved earlier in the section, as it follows the general description of the SOA model itself.

R2C6) What is the spinup time of the model?

Response: Each simulation had a model spin-up time of one month in this study (this is stated on page 4195 in the revised manuscript).

R2C7) P. 4197, l. 22-23: temperature is also greatly important.

Response: The authors agree with the reviewer and appreciate the attention brought to this oversight. The discussion on page 4197 in the revised manuscript now includes temperature as an additional variable influencing the SOA production.

R2C8) P. 4198, l. 1: Do BIGENE and C3H6 play any role, even minor, compared to isoprene? I would expect them to be negligible, given their low emissions and not very high aerosol yields.
Response: This reviewer comment is consistent with the results presented on page 4200 line 3-7 in the manuscript that ~99% of the enhanced SOA mass was attributable to isoprene (ISOP).

R2C9) P.4198, middle: What happens downwind Australia in December?

Response: SOA production nearly mimics the pattern of emissions i.e. SOA is predominantly formed where the emission sources are. Monoterpenes in the southeast coastal regions of Australia are emitted at a rate of $\sim 1.0 \text{ mg m}^{-2} \text{ day}^{-1}$ in the summer month of December, which resulted in SOA mass concentration of up to $0.23 \text{ } \mu\text{g m}^{-3}$. SOA mass enhancement (as shown in Fig. 4) in the vicinity of this region might have been resulted from the updates to the SOA parameters in the model. However, apparent SOA enhancement farther away from this region could be attributed to model artifacts, more like the enhancements in far north of the northern hemisphere (Fig. 4) where the base-case SOA concentrations are generally negligible.

R2C10) P. 4199, l. 15: the 1-sigma includes both temporal and spatial variability?

Response: Correct. The $1-\sigma$ standard deviation included both spatial and temporal variability based on average SOA mass concentrations that were saved on a monthly basis for all simulations.

R2C11) The text from p.4200 l-6-10 should be moved in the middle of p. 4199.

Response: Done.

R2C12) P. 4200, l. 12-18: what is the temperature effect on the adopted K_p ?

Response: The adopted K_p values were for a temperature of 298K and thus likely underestimate SOA formation at lower temperatures aloft, since K_p values are not corrected as a function of ambient temperature in the MOZART SOA model.

R2C13) P. 4201, l. 4: this is valid only if there was enough time for the model to spinup.

Response: Correct. The authors believe that the one month model spin-up time was sufficient to support this assumption. For a comparable study using the GISS GCM II model, Farina et al. (2010) also utilized a one month spin-up time and also assumed the net SOA production equals to the net deposition flux in the model. .

R2C14) P. 4201, l. 16: These are pretty long lifetimes. Are there a lot of SOA above clouds? What is the boundary layer vs. free troposphere lifetime? How about the load?

Response: A range of SOA lifetime between 6.2 (Chung and Seinfeld, 2002) and 15.6 days (O'Donnell, 2011) can be found in literature. Modeled lifetimes of ~11.2 - 13.6 days in the current study are within the reported values. It was assumed that the net production of SOA is equal to total dry and wet depositions in the model. So the lifetime of SOA at different model heights could be affected by dominant removal processes at respective heights. Figure 6 shows an apparent increase of SOA concentration above cloud, and in the free troposphere, where the lifetime of SOA is generally higher as the removal processes are less effective at these heights. We believe that the lifetime of SOA at lower troposphere of ~11.2 days, where removal processes are the most effective would be less than the lifetime in the free troposphere. Consequently, the loading of SOA in the boundary layer should also be higher compared to the free troposphere.

R2C15) P. 4202, l. 2: “little impact” is not correct, since figure 4 shows a factor of two difference in many places.

Response: We thank the reviewer for identifying this potential source of confusion. “Little impact” was only in reference to the change in SOA production from the first set of simulations as compared with the second set of simulations in which isoprene was added as a precursor. The increase in absolute SOA concentration from the base-case due to parameter updates ((not shown) in the model is not as significant as it is for the case (also not shown) where additional species combined with updated parameters were considered. This has been explained clearly in the revised manuscript.

R2C16) P. 4202, l. 5: It is ok to compare only one simulation, if this is considered to be the best. If it is, it has to be stated.

Response: We have compared model predicted SOA concentrations with available observations for all simulations, but we only reported the results from the simulation with additional species and updated parameters (MZ4-v2) since the predicted SOA concentrations were comparable to measurement data at different location in the world. It has now been clearly presented in the revised manuscript.

R2C17) P. 4203, l. 6: There are data available: Zhang, X. Y., Wang, Y. Q., Niu, T., Zhang, X. C., Gong, S. L., Zhang, Y. M., and Sun, J. Y.: Atmospheric aerosol compositions in China: spatial/temporal variability, chemical signature, regional haze distribution and comparisons with global aerosols, *Atmos. Chem. Phys.*, 12, 779-799, doi:10.5194/acp-12-779-2012, 2012.

Response: The authors would like to thank the reviewer for providing the reference for measurement data. Based on the measured data presented in the paper, a new Fig. SI-XX was generated, which is provided in the supporting information section. The figure shows annual average modeled vs measured SOC (secondary organic carbon) concentrations for 14 rural and

urban sites in China. The error bars represent 1- σ of the monthly averaged data points. The updated model captured the measured SOC concentrations relatively well at most sites, except for one urban site and three rural sites in China. This will be added in the model evaluation section of the revised manuscript.

R2C18) P. 4203, l. 13-15 and p. 4204, l. 8: The OM/OC ratio is also an issue. In addition, the parameterization itself can be a major source of uncertainty that can contribute to the discrepancy.

Response: The revised manuscript now includes a discussion of the uncertainty that may be attributed to the OM/OC ratio and the parameterization of the 2p SOA model.

R2C19) Why section 3.2.3 is different from section 3.1.3?

Response: In section 3.1.3 the global budget estimates are presented and in section 3.2.3 these model estimates are compared with other modeling studies. The authors believe that better clarity is achieved in the manuscript by separating the results and model evaluation sections; however, as a means of connecting these sections, the discussion is presented in the same chronology in both sections.

R2C20) P. 4206, l. 10-15: This is a very strict statement. There is no reason for models to be exactly identical in order to be compared with each other. There are many useful comparisons that can be extracted from different models, when their differences are known and understood. A great example is the several AeroCom intercomparisons.

Response: The authors agree with the reviewer's comment and the relevant section has been revised in the manuscript.

R2C21) P. 4207, l. 1-2: The removal does not influence the production, at least not directly. Unless the authors claim that the removal affects the pre-existing aerosols that new SOA can condense on, this statement is incorrect, no matter how the aerosol lifetime is being calculated in the model (p. 4201, l. 4).

Response: The authors agree with this reviewer comment. The SOA module in ECHAM5/HAM model (O'Donnell et al., 2011) allows partitioning of semi-volatile products on to pre-existing organic material, which is also subject to removal processes. This has now been clarified in the revised manuscript.

R2C22) P. 4209, l. 1-2: How about other regions?

Response: Total AOD data from literature including Remer et al. (2008), Lee and Chung (2013), and Hsu et al. (2012) were obtained and compared with those from the current study. The model predicted AOD in the current study is comparable with those found in literature, particularly over land regions including Indonesia (0.15-0.2), South America (0.15-0.3), North Africa (0.3-0.8), and Australia (0.1 – 0.15). A discussion will be added in the revised manuscript.

R2C23) Table 1: Are these numbers the original ones, or the updated ones, before adding the new species?

Response: Table 1 and associated text have been revised to include all three sets of model parameters and better illustrate the differences between the model runs.

R2C24) Figure 5: maybe it is better to show v2-v1?

Response: Figures 4 & 5 show relative changes in SOA concentrations at surface from the base-case simulation due to revisions to (i) the existing 2p parameters, and (ii) treatment of additional species combined with updated parameters, respectively. We believe both figures are important to show the relative effects of revisions made to the SOA model, and would like to keep Fig. 5 in the revised manuscript. The reviewer suggested evaluation can still be made by comparing Fig.5 with Fig. 4 in combination with base-case SOA presented in Fig. 2.

R2C25) Figure 6: since the authors have measurements, why not show them here?

Response: Available measurement data may not well represent the regions as well as the model heights that are shown in Fig.6. This figure shows regionally averaged annual SOA concentrations, and indicates how revisions to the SOA module affected the results from the base-case. A discussion on modeled vs measured vertical profiles with more representative regions and time of the year is presented in section 3.2.2 in the manuscript.

Technical corrections

1) P. 4196, l. 18: “SOA formation” should be “SOA concentration”.

Response: Corrected.

2) P. 4199, l. 18: “global land mask” should be “model’s land mask”.

Response: Done.

3) P. 4207, l. 14: “and sulfate” should be “sulfate”.

Response: Extra ‘and’ was omitted.

4) Table 4 appears out of order in the discussion.

Response: The numbers between tables 4 & 5 have been switched to follow the order in the discussion.

5) Figure 1 is not needed.

Response: Agreed, Figure 1 has been removed. A discussion about relevant changes in MOZART source code has been added in section 2.1 in the revised manuscript.

6) The color scale of Figure 2 is not very good. Bby eyeballing it, a maximum of 1ugC/m3 might be better.

Response: The color scheme of Figure 2 is now set to maximum SOA concentration of 1 μgm^{-3} in the revised manuscript.

7) Add a/b in Figure 3.

Response: Done.

Additional Figures

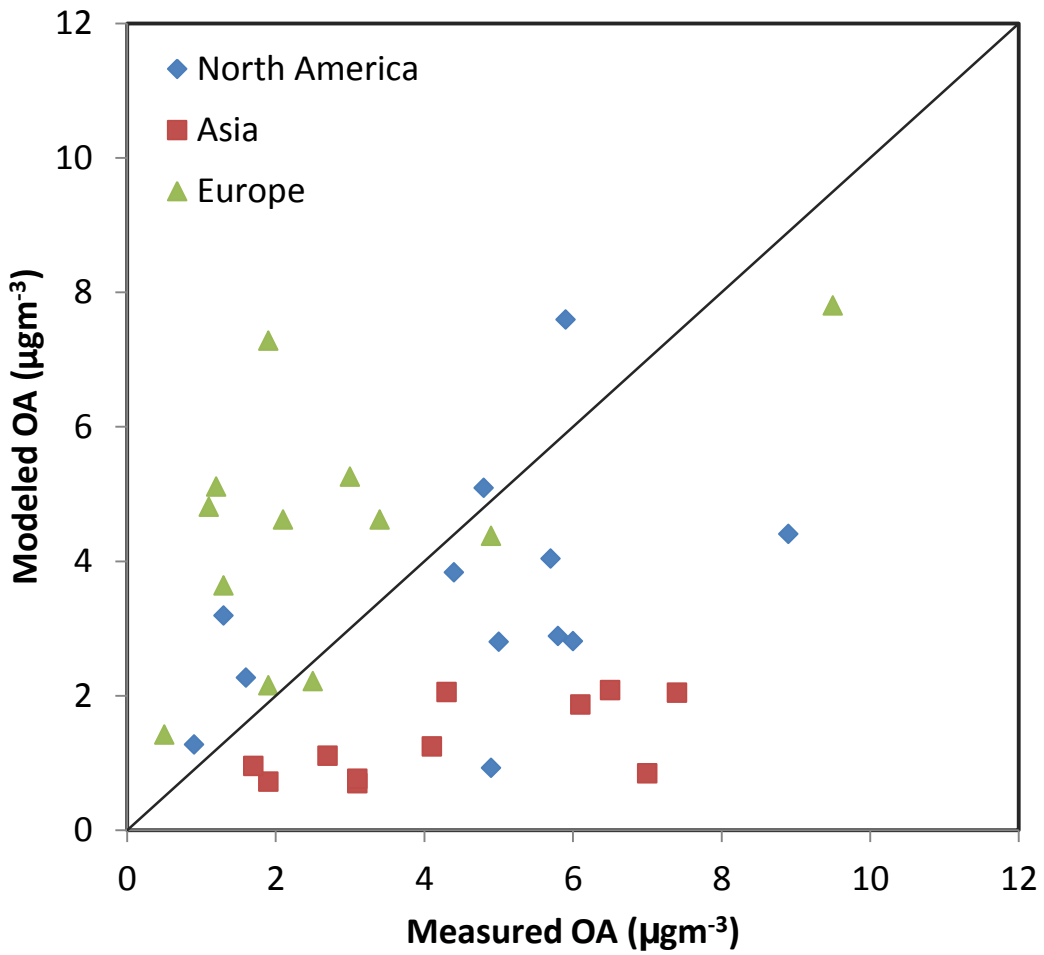


Fig. XX: Modeled vs measured organic aerosol (OA) mass concentration for different sites around the world. Data were adapted from Zhang et al. (2007).

Supporting Information

1. SDescription and Derivation of 2p-VBS parameters

The 2p-VBS parameters were conceived in order to take advantage of the robustness of the volatility basis set (VBS) fitting approach (e.g, see Presto and Donahue, 2006), while allowing the computationally-efficient and widely-used two product (2p)-modeling framework to be retained. The parameters were derived by: 1) using VBS fits (Tsimpidi et al., 2010) to generate pseudo-data, and 2) fitting the pseudo-data using the 2p approach (Odum et al., 1996). Each of the VBS parameters of Tsimpidi et al. (2010) at $T = 298$ K were used to generate 263 pseudo-data points for $M_o = 0$ to $200 \mu\text{g m}^{-3}$ at each of three temperatures (272, 298, and 324 K) using an effective $\Delta H_{\text{vap}} = 30 \text{ kJ mol}^{-1}$ (see Pathak et al. 2007b); those 789 pseudo-data points were then fit to generate one set of 2p parameters (per set of VBS parameters), thus labeled “2p-VBS”. For each of the SOA precursors, the 2p-VBS parameters were able to represent SOA formation with the same degree of uncertainty as the VBS parameters (i.e., no additional uncertainty is introduced by the 2p-VBS fit). It therefore can be assumed that the SOA yield and mass predictions using the Tsimpidi et al. (2010) VBS parameters and the 2p-VBS parameters produce equivalent results (in the absence of any “aging”), including temperature dependent SOA yields. The 2p-VBS fits result in a reduction from 4 “bins” (8 parameters, typical for VBS) to 2 “bins” (4 parameters) which can be utilized in existing 2p model frameworks, such as MOZART. The MOZART SOA module does not allow for aging or processing of SOA, thus the gas-phase oxidation (beyond the initial oxidation of the parent VOC) that is often represented in applications of the VBS is not considered in this work. For the precursors included in the MOZART simulations, it was determined that the 2p-VBS parameters represented available chamber data well, with the exception of isoprene. Therefore, the parameters of Henze and Seinfeld (2006) were used. In addition, the MOZART SOA module includes oxidation of monoterpenes by NO_3 for which Tsimpidi et al. (2010) VBS parameters, and thus 2p-VBS parameters, are not available. The monoterpene+ NO_3 parameters were based on Chung and Seinfeld (2002). The development, testing, and application of 2p-VBS parameters will be presented in a forthcoming manuscript by Barsanti et al.

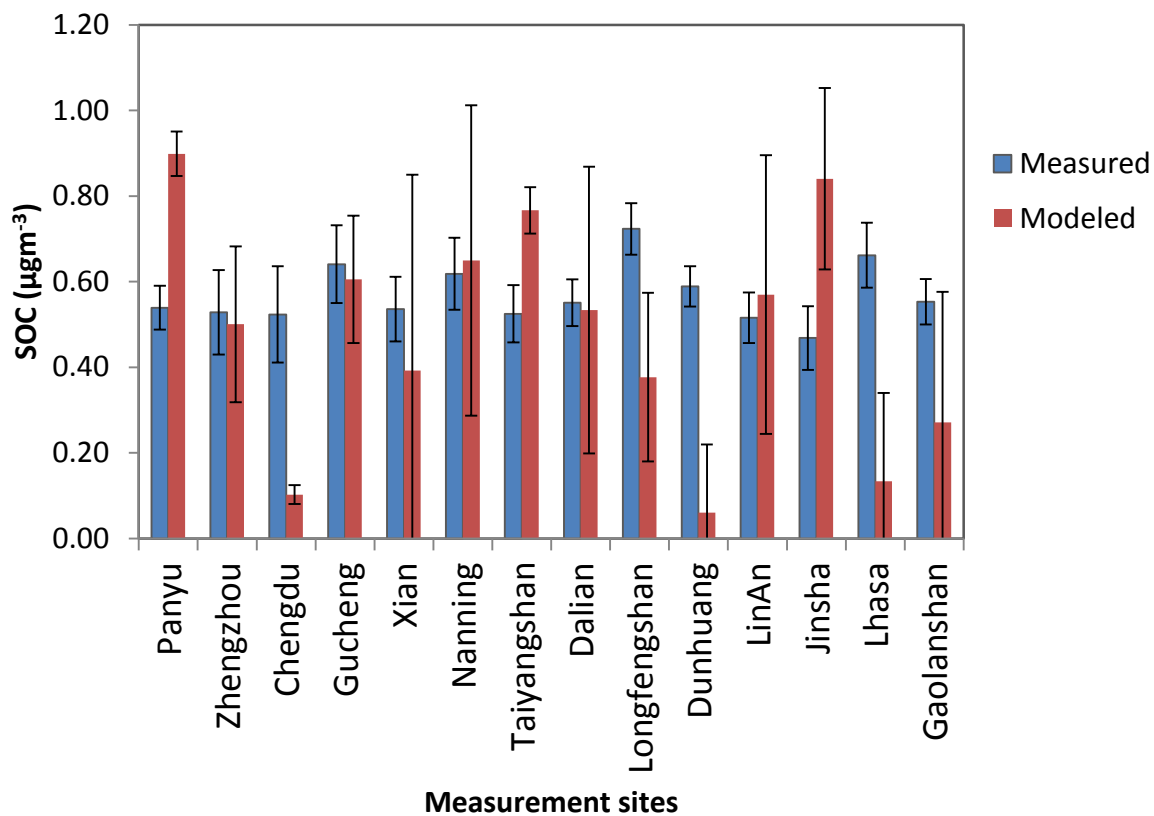


Fig. SI-1 Measured vs Observed comparison of secondary organic carbon at various locations in China. Measured data were adopted from Zhang et al. (2012).