

## ***Anonymous Referee #1***

*Tsimpidi et al. quantify the global-scale contributions of combustion emissions to organic aerosols using a global model. Rather than a single value, the authors provide a range utilizing various inputs and parameters reported in the literature for modeling organic aerosols. Those sensitivities include variation in emissions (volatility of emissions, high estimates of IVOCs, an alternative POA emission inventory) alternative OA aging schemes, and alternative OA solubility parameters. The authors then compare results from the various sensitivity simulations against AMS measurements at rural locations.*

*The paper is generally well written and the analysis robust. I recommend the paper for publication but first would like to see a few clarifications and additional points listed below discussed.*

We would like to thank the reviewer for his/her positive response. Please see below our point by point response to reviewer's comments.

### *General Comments:*

- 1. In recognizing there is a computational expense in a more explicit parameterization, are there benefits to utilizing different chemistry/aging schemes for anthropogenic and biogenic OA (e.g. Koo et al., 2014)?*

According to previous modelling studies (Lane et al., 2008; Murphy and Pandis, 2009; Tsimpidi et al., 2014) aging of biogenic SOA may lead to significant over-predictions of OA over rural areas and forests. These findings are confirmed by observational studies that suggest that the aging of biogenic SOA does not result in a large change in its mass concentration (Ng et al., 2006; Donahue et al., 2012). Murphy et al. (2012) attributed this to a balancing of fragmentation and functionalization effects during the photochemical aging of biogenic SOA. On the other hand, the multigenerational chemistry of anthropogenic SOA precursors leads to a net average decrease of their volatility and increase of SOA production (Hildebrandt et al., 2009) and is often parameterized by regional and global models (Koo et al., 2014; Tsimpidi et al., 2016). Therefore, utilizing different chemistry/aging schemes for anthropogenic and biogenic OA gives us the opportunity to account for their different response in photochemical aging. This is now discussed in Section 2.2.

### *Specific Comments:*

- 1. There appears to be some inconsistency as to how the authors define IVOCs. On line 153, IVOCs are defined as having a C\* between  $10^4$  and  $10^6 \mu\text{g m}^{-3}$ . But on line 188-190, when discussing biomass burning emissions, the authors state: "Biomass burning emissions are assumed to cover a range of volatilities from  $10^{-2}$  to  $10^4$  (May et al., 2013a), therefore, no IVOC emissions are assumed from biomass burning sources..." Then, in the low volatility simulations, emissions of IVOCs are assumed to be zero. However, biogenic emissions in the reference simulation, which includes  $10^4$  emissions, and the low volatility simulation are identical ( $28.4 \text{ Tg yr}^{-1}$ ).*

We are sorry for this misunderstanding. The  $10^4 \mu\text{g m}^{-3}$  volatility bin represents IVOCs. Based on the findings of May et al. (2013) for biomass burning emissions, no additional IVOCs were included in these simulations. In the revised manuscript this specific sentence is rephrased as follows: "Biomass burning emissions are assumed to cover a range of volatilities from  $10^{-2}$  to  $10^4$  (May et al., 2013a) and no additional IVOC emissions are assumed from biomass burning sources. Therefore, the sum of their emission factors is unity (Figure 2a)". Furthermore, in the low volatility simulation, the sum of the emission factors is kept equal to unity by distributing the IVOC emissions (with  $C^* = 10^4 \mu\text{g m}^{-3}$ ) to lower volatility bins. Therefore, the biomass burning emission load is identical to the base case simulation, but distributed in lower volatility bins.

2. *In the low volatility simulations, how are the emissions from the  $10^4$  bin that are not considered IVOCs redistributed to the lower bins? e.g. Total biogenic emissions are identical in the reference and low volatility simulations.*

The  $10^4 \mu\text{g m}^{-3}$  volatility bin represents IVOC emissions. In the low volatility case, the 0.3 emission factor that was applied in the  $10^4 \mu\text{g m}^{-3}$  volatility bin of the biomass burning emissions in the base case simulation is equally distributed to the  $10^{-2}$ ,  $10^0$  and  $10^2 \mu\text{g m}^{-3}$  volatility bins by applying an extra 0.1 emission factor in each of these bins. Therefore, the total emission factor for the biomass burning emissions in both scenarios remained unchanged. This information is provided in Figure 2.

3. *Line 219 and 220: The wording here makes it sound as if only emissions in the  $10^4$  and  $10^6$  bins are being increased by a factor of 1.5. Instead, I would recommend rewording this sentence to provide clarity. For example “increased by an additional factor of 1.5 times the POA emissions and then distributed in the volatility bins...”. Also, how are they distributed, equally in the  $10^4$  and  $10^6$  bins? I’d also suggest making it more clear the total emissions in this case, that total anthropogenic emissions are 4x the POA inventory (1x L/SVOCs and 3x IVOCs) and biogenic emissions are 2.5x the POA inventory (1x L/SVOCs and 1.5x IVOCs).*

Following the reviewer’s recommendation, we have rephrased these lines in the revised manuscript as follows: “To estimate an upper limit of the IVOC contribution to the formation of SOA, a sensitivity simulation is conducted in which the emissions of IVOCs are increased by 1.5 times the original POA emissions. These extra emissions are distributed in the volatility bins with  $C^*$  of  $10^4$  and  $10^6 \mu\text{g m}^{-3}$  (Figure 2c) by applying an additional emission factor of 0.5 and 1 respectively. The LVOC and SVOC emissions are the same as in the reference simulation. Overall, the total anthropogenic and biomass burning emissions are 4 and 2.5 times higher respectively than the original POA emission inventory. The decadal average global emission flux of primary organic emissions in this sensitivity test is  $71 \text{ Tg yr}^{-1}$  for both anthropogenic and open biomass burning sources (Table 1)”.

4. *What is the reasoning to perform a model simulation with added IVOC emissions ( $C^*$  of  $10^6$ ) from biomass burning if measurements only support emissions up to a  $C^*$  of  $10^4$ ?*

The May et al. (2013) volatility distribution for biomass burning OA, used in this work, are derived from thermodenuder measurements covering a range of volatilities with  $C^*$  from  $10^{-2}$  to  $10^4 \mu\text{g m}^{-3}$ . However, this range can be extended to even higher volatilities. Agaki et al. (2011) estimated that the unspciated nonmethane organic compound (NMOC) emissions account for 50% of the total observed NMOC. Jathar et al. (2014) reported that 20% of the NMOC emissions are not speciated and are currently misclassified in emission inventories. Given that the unspciated organic emissions are still largely uncertain, we performed a sensitivity simulation by adding organic emissions outside the range of May et al. (2013)’s volatility distribution (i.e., for  $C^*$  equal to  $10^6 \mu\text{g m}^{-3}$ ).

5. *Line 448-451: Underestimates of IVOCs could be one cause of underpredictions, but could it also be other factors like uncertainty in yields (e.g. wall loss) or other missing precursors and/or pathways?*

That is correct. In this study, we have conducted multiple sensitivity scenarios in order to quantify the impact of different parameters on the predicted OA concentration. It is worth mentioning that in all cases tested the model underestimates OA. This suggests that the source of underprediction of OA by atmospheric chemistry models reported in the literature (Tsigaridis et al., 2014) cannot be attributed to only one cause rather to a combination of different factors. Potential causes that

are not explored here (e.g., uncertainties in SOA yields due to wall losses in chambers, missing sources and oxidation pathways, etc.) can also play a role, especially during the winter period. This is already discussed in the conclusions of our manuscript but is now more emphasized in the revised manuscript.

## References

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