

Anonymous Referee #1

Overall, the article is well written and provides a useful comparison of a global model to a very large AMS data set. The agreement between fresh OA and SV-OOA and aged OA and LV-OOA is an interesting result. I recommend publication after addressing these comments.

We would like to thank the reviewer for his/her thoughtful revision. We have incorporated all his/her suggestions in the revised manuscript. Below is our response to his/her specific comments.

My main comment is to include some caveats about how comparisons based on total mass (or even 3 components of mass: POA, LV-OOA, SV-OOA) may not be able to resolve source sectors or governing pathways very well. As an example, IEPOX-SOA has been shown to be a major contributor to ambient OA around the world via more recent AMS PMF analysis (Hu et al. 2015). This type of SOA is likely formed via processing of later generation isoprene products in aqueous acidic aerosol, a process not considered in the model used here. How does neglecting this type of SOA bias comparisons? In addition, cooking aerosol (17% of total OA in Pasadena, Hayes et al. 2013) is not specifically evaluated. Is it included in your emission inventory? Should the model expect to reproduce observed OA if it is missing known pathways?

This is a valid point raised by the reviewer. We have already identified the vehicle cold starts and residential heating sources as a possible cause of model underperformance during winter since the amount of the OA emissions from these sources are probably severely underestimated in our emission inventory. As the reviewer pointed out, residential and commercial cooking is another important source of OA that can contribute significantly to measured POA (around 50%) and total OA (15%-20%) over urban areas (Sun et al., 2011; Mohr et al., 2012; Ge et al., 2012; Hayes et al., 2013). However, these emissions are not included in emission inventories and global models (Tsigaridis et al., 2014), including the one used in this study. While we have excluded this type of OA from our model evaluation (in cases where cooking OA (COA) has been resolved by the PMF analysis) in order to eliminate any model bias resulting from the omission of cooking emissions in our inventory, organic compounds emitted from cooking sources can be oxidized and form SOA. Therefore, the lack of cooking emissions can be considered as a possible source of OOA underestimation by the model over urban and urban downwind areas. Furthermore, the model does not simulate SOA formation from aqueous-phase reactions, therefore cannot produce IEPOX-SOA that has been recently resolved as an important source of SOA close to isoprene sources (Hu et al., 2015). However, IEPOX-SOA has been identified in only a few of the AMS datasets used in this study in areas strongly influenced by isoprene emissions (e.g. Duke Forest, NC). Therefore, IEPOX-SOA cannot be considered as a major source of model/measurement discrepancy in most of the areas investigated here. A discussion concerning the implications of COA and IEPOX-SOA on model performance has been added in section 5.2.2 of the revised manuscript.

Minor comments:

- 1. This is a personal preference, but I encourage use of Tg C/yr instead of Tg/yr when referring to OC*

Following the reviewer's recommendation we changed the units of OC emissions to $Tg\ C\ yr^{-1}$.

2. *Line 64: point out that the magnitude of missing IVOCs is likely source specific*

We have added to the text that the magnitude of the missing IVOC emissions is estimated to be between 0.25 and 2.8 times POA emissions, depending on the type of the source.

3. *Line 121: Heald et al. 2005 and de Gouw et al. 2005 were performed before isoprene was even considered a source of SOA via traditional semivolatile pathways (e.g. Henze and Seinfeld 2006). These outdated references should be replaced with more recent references to assert that global models currently underestimate OA.*

We have replaced these references with Tsigaridis et al. (2014) who evaluated the current performance of 32 global models in simulating OA mass.

4. *Line 257: Do Aiken et al. 2008 OM/OC values need to be updated in light of new AMS calibrations by Canagaratna et al. 2015?*

Canagaratna et al. (2015) estimated an OM:OC ratio of 1.34 for HOA and 1.64 for BBOA which are very similar to the values used in our study (1.3 and 1.6 respectively). Therefore, in the revised manuscript we have replaced the reference to Aitken et al. (2008) with the Canagaratna et al. (2015).

5. *Lines 310-315: was cooking OA resolved in any data sets or is it likely part of HOA and BBOA?*

Cooking OA (COA) has been resolved in some datasets (e.g., Barcelona by Mohr et al., 2012; Fresno, CA by Ge et al., 2012; New York, NY by Sun et al., 2011; Pasadena, CA by Hayes et al., 2013 etc.). However, cooking emissions are not included in our emission inventory; therefore COA was not used in the model evaluation.

6. *How is deposition of SOA/SOG handled?*

The removal of SOA and SOG species through dry deposition is calculated within the DRYDEP submodel (Kerkweg et al., 2006a) based on the big-leaf approach and the dry deposition velocities depend on physical and chemical properties of the surface cover (e.g., the roughness length, soil pH, leaf stomatal exchange, etc.). The sedimentation of aerosols is calculated within the SEDI submodel (Kerkweg et al., 2006a) using a first-order trapezoid scheme. In cloud scavenging and rainout of gas and aerosol species are treated by the SCAV submodel (Tost et al., 2006). The effective Henry's law coefficient used for calculating the scavenging of all SOG species is equal to $10^5\ M\ atm^{-1}$. This discussion has been added in section 2.1 of the revised manuscript.

7. *A table of the yield parameters for the traditional VOCs and IVOCs should be reproduced from Tsimpidi et al. 2014 in the supporting information (or in text)*

Following the reviewer's recommendation we have added a table in the supporting material which summarizes the aerosol mass yields used to distribute the oxidation products of VOCs in four volatility bins. The oxidation of IVOCs results in only one product with lower saturation concentration by a factor of 10^2 and a stoichiometric coefficient of 1.15 to account for two added oxygen atoms. This information is reported in the text.

8. *Line 392 and thereafter: How much does the seasonality of emissions drive the higher POA in winter? Cold temperatures favor partitioning to the particle as indicated, but higher residential wood combustion could be expected as well.*

This is a good point. The wintertime emissions of fPOA used in this study are indeed 12% higher than the annual average emissions. Therefore the seasonality of the emissions can explain part of the higher POA concentrations calculated during winter (36% on average). We have included this interpretation in our discussion in 4.1.2 of the revised manuscript.

9. *Paragraph starting on line 408: The annual tropospheric burden vs year plot could be made more interesting by including regional trends. Do the emissions capture the expected increase in emissions from China vs decrease in the US?*

We have included the continental tropospheric burdens in Figure 6b. Indeed, the Asian anthropogenic OC emissions have increased during the simulated decade by 12%. On the other hand, anthropogenic OA emissions over North America have decreased by 15% during the same period. Over Europe, anthropogenic OA emissions increased until the year 2005 (5%) and then started to decrease reaching 4% lower emissions (compared to 2001) by the end of the decade. However, the simulated fOA tropospheric burdens over the continents (Figure 6b of the revised manuscript) do not reflect this clear trend of emissions since other factors (i.e., meteorology) play an important role on the calculated tropospheric burden as well. This information has been added to the text. We have also performed the same analysis for the simulated bbOA tropospheric burden by including the results from the three main rainforests (Amazon, Congo, SE Asia).

10. *Line 428: Clarify the IVOCs from biomass burning. Line 274 indicates IVOCs from biomass burning were not added to the existing POA, yet IVOCs account for 40% of the emissions?*

We have rephrased this sentence to avoid confusion. May et al. (2013) quantified the biomass burning organic emissions in the volatility range from 10^{-2} to 10^4 $\mu\text{g m}^{-3}$. We did not add any IVOC emissions in the $C^* > 10^4$ $\mu\text{g m}^{-3}$ bins for open biomass burning. As a result 40% of the traditional biomass burning OA emissions are assumed to be IVOCs with $C^* = 10^4$ $\mu\text{g m}^{-3}$ (Table 2 of the manuscript). This information has been added to the revised text.

11. *Section 5.2.1: How do you separately diagnose errors in the magnitude of POA emissions from the volatility profile?*

We have not explicitly diagnosed any errors in POA emissions. Any reference to possible errors in the magnitude of POA emissions from specific source sectors are just hypotheses that should be considered and further explored in future studies. Errors in the POA volatility distributions can also explain parts of the discrepancy. This is explained in the revised manuscript.

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