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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Simulating secondary organic aerosol from missing diesel-related intermediate-volatility organic compound emissions during the Clean Air for London (ClearfLo) campaign" *by* R. Ots et al.

Anonymous Referee #2

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In this paper, Ots et al. present an interesting method to account for the emissions of intermediate volatile organic compounds (IVOCs). They suggest that VOC emissions can be added proportionally to VOC emissions as opposed to the POA emissions which is the standard method used by current Volatility Basis Set (VBS) models. This approach can potentially pave the way for an accurate representation of IVOCs in the emission inventories which is proved to be a necessity for SOA models during the last decade. Overall, the manuscript is well written and scientifically sound. I recommend this study for publication after taking the following comments into account.

General comment:

1. The authors include additional diesel related IVOC emissions based on the VOC emissions from the transport sector. This resulted in a significant improvement of their model results which brought the predicted SOA close to measurements during winter, spring, and summer while it resulted in an overprediction during autumn (Fig. 17 of the manuscript). However, the transport sector consist only one of the ten sectors that their emission inventory includes. This raise the question of how much their model performance will change (towards overprediction) if they will add the missing IVOC emissions from the rest nine sectors. Do they have indications that the only important source of IVOCs is the transport sector? While I strongly support the suggested approach of deriving the IVOC emissions based on intermediate length alkanes (or naphthalene seen in other studies; Pye and Seinfeld, 2010) I am quite sceptic about the impact shown here by only one sector. I suggest adding a discussion on this matter, probably in section 4.

Specific comments

1. Page 2 line 20: Biomass burning OA (BBOA) is also a usual component that PMF can identify. Does SFOA correspond to BBOA? If so, you should use the latter since it is more commonly used by the AMS community. Furthermore, you should also report the oxygenated organic aerosol (OOA) which then can be split into LV-OOA and SV-OOA.

2. Page 2 lines 22-27: Since the main focus of the manuscript is the simulation of SOA, it would be good to add a sentence regarding the performance of the global models in terms of SOA (e.g.,Spracklen et al., 2011;Jathar et al., 2011;Jo et al., 2013;Mahmud and Barsanti, 2013;Shrivastava et al., 2015;Tsimpidi et al., 2016)

3. Page 3 line 20: Please add recently developed models that follow the same assumption in order to indicate that the factor of 1.5 is widely used up to date (e.g.,Koo et al., 2014;Tsimpidi et al., 2014)

4. Page 4: The line numbers here and in a number of the following pages are not correct. They should either restart in each page or continue throughout the text.

5. Page 5 line 13: Do HOA and SFOA correspond to the fossil fuel combustion and domestic combustion of you emission inventory (EI)? Please clarify since it is not clear if you used these fractions to convert the OC from your EI to OA.

6. Pages 5 line 28: Assuming that POA is treated as non-volatile will result in unrealistically high OA concentrations in the aerosol phase. This will favor the partitioning of your semivolatile compounds (e.g. oxidation products of IVOCs) into the aerosol phase resulting in an overestimation of SOA as well. On the contrary, if you do not assume that POA and SOA participate in the same solution during the phase partitioning, you should expect an underestimation of SOA. Please comment at this point on the implications of your assumption regarding the POA volatility.

7. Page 6 line 2: Tsimpidi et al. (2010) used 4 volatility bins to distribute the oxidation products of VOCs. Can you please report the aerosol yields for the 5th volatility bin that you are using (C*=0.1) and add a reference for them as well?

8. Page 6 line 6: According to Lane et al. (2008) the use of aging reactions improved their results compared to measurements from urban areas but resulted in a strong overprediction over rural areas. They attributed this discrepancy to a potential balancing of decomposition to smaller more volatile products (fragmentation) and production of more substituted less volatile products (functionalization) during the photochemical aging of biogenic SOA. This was also confirmed by laboratory studies (Ng et al., 2006). Therefore they suggested that no ageing of biogenic SOA should be considered. Furthermore, the use of an ageing rate constant of 4.0 x 10-12 cm3 molecule-1 s-1 is kind of conservative compared to what is used lately by most models (i.e. 1.0 x 10-11 cm3 molecule-1 s-1; Fountoukis et al. 2014). According to the above, I suggest either changing your scenarios by using 1.0 x 10-11 cm3 molecule-1 s-1 as a rate constant and assume no ageing of biogenic compounds or to make a sensitivity test and investigate the effect of these assumptions on your results (especially regarding the rate constant).

9. Page 6 line 10: What is the saturation concentration of this “background OA” compound? Is this considered nonvolatile since it is very aged and highly oxygenated? Furthermore, please provide a reference for assigning a value of 0.4 μg m-3 to this compound. Does this value based on measurements?

10. Page 7 line 35 (on the top of the page): What is the SOA mass yield that you used for the 1000 μg m-3 volatility bin? Please provide a reference as well.

11. Page 7 lines 28-29: A more fair comparison between the two approaches would be to add IVOCs proportionally to POA from sector 7 only as you did on your addDiesel scenario. Can you investigate this additional scenario as well?

12. Page 9 Section 3.1: Since POA are assumed to be nonvolatile you would expect to overpredict their concentrations. Are the emissions so severely underestimated? Please report that the presented underprediction will be even more significant if you add the semivolatile character of POA.

13. Page 9 line 20: Please replace the “secondary pollutants” with “secondary inorganic pollutants”

14. Page 10 lines 7-8: Add a reference to Fig. 7

15. Page 13 line 30 (on the top of the page): How you calculated the 40%?

16. Page 15 lines 4-5 (or 33-34): Pye and Seinfeld (2010) have used a naphthalene-like surrogate specie to describe IVOCs instead of the traditional “POA” method. Please refer to this work as well (maybe in the introduction).

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