

Interactive comment on “Formation of secondary organic aerosol in the Paris pollution plume and its impact on surrounding regions” by Q. J. Zhang et al.

Anonymous Referee #3

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This study presents quite useful results from an excellent dataset. Overall the quality of the data and analysis is sufficient for eventual for publication in ACP, but several issues must be addressed.

Major comments: 1. There should be more discussion of the impact of using OA/Ox instead of OOA/Ox as done for the other studies quoted. By how much does OA/Ox exceed OOA/Ox? I am guessing about 50% based on the reported ~30% contribution of HOA to OA, but the comparison of the measured OA/Ox ratios to the modeled ratios and to other studies' measured ratios would be much clearer if OOA were also used. The rationale for not using LV-OOA and SV-OOA as stated is that HOA contains some

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oxidized POA, and as a result “Thus use of OA in this study avoids these attribution problems.” – but it leads to the questions just listed above. How much is the comparison to modeled SOA/Ox affected when OOA vs. OA is used?

2. More discussion is needed regarding the (measured) NO_x and BC plume not being spatially coincident with the (measured) secondary OA and Ox plume on the 29th. This seems to suggest that the origin of the Ox and OA on the 29th is not the Paris plume. What are the ramifications for the comparison of model vs. measured OA/Ox? Are the modeling inputs for this day simply not appropriate? On a similar note, are there problems w/the spatial distribution of emissions in the model?

Related comment: What is the ratio of gasoline to diesel consumption in the greater Paris region, especially compared to the other locations where the OOA/Ox method has been used? (Tokyo, Mexico City, etc.)? I do not suggest adding a detailed analysis of the differences in emissions, but a short discussion of the major differences in emissions between these locations would inform the comparison of the slopes and their modeled values, especially given the importance of aromatic VOCs in the modeled results. How much of the overestimate of OOA/Ox by the model is explained by the SOA formulation in the model vs. the accuracy of the emissions (both quantity, speciation, and spatial distribution)? Is the ratio of aromatic VOC emissions to other Ox and SOA precursors particularly high in Paris?!

3. Contrary to most other studies, this one has modeled OOA/Ox actually exceeding measured OA/Ox. More discussion is warranted. Is this due to the yields and version of the volatility basis set used, or does this model work better for the Paris region for some reason, and if so why? As presented it is difficult to assess why the model actually overestimates OOA/Ox- does this represent the “current” state of SOA modeling, or just one particular way of doing it?

Minor comments: 1. pg 8075 line 20 and several other locations: While this study is certainly not the first to use the term “Oxidant” to refer to Ox (= O₃ + NO₂), as in

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“...as a function of oxidant formation”, it is very confusing, since the word oxidant also refers to OH, O₃ and NO₃. Indeed later in this manuscript the term “oxidant agents” are used, as well as “oxidant levels”. I highly recommend simply referring to Ox as Ox and ozone as ozone but NOT as “oxidant”. Note that the term Ox is not an abbreviation for “Oxidant”! Ox was initially defined as O₃ + O for stratospheric chemistry...

2. Abstract: Rather than state that there is “good agreement”, state that the model actually overestimates the observations, by ___%.

3. Description of NO_x measurements- very little info is given. I had to look in the Freney et al paper, and from there to the supplement to find adequate description of the measurements – important given the importance of NO₂ to Ox in urban plumes.

4. Figure 4 – would be better to plot Ox rather than O₃

5. pg 8077, line 16: “flight legs perpendicular to the plume time” – what does this mean? Pg 8078, line 2, “... were privileged” – please clarify/edit.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 8073, 2015.