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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Modeling secondary organic aerosol in an urban area: application to Paris, France" *by* F. Couvidat et al.

Anonymous Referee #2

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This paper describes a modeling study of organic aerosol levels in Paris measured during the MEGAPOLI study. It describes the development and application of an updated scheme to simulate secondary organic aerosol (SOA) formation from semivolatile and intermediate volatility organic compounds (SVOC and IVOC) based on surrogate compounds. The predictions of the updated model are compared to a previous version which used a more empirical approach to simulate SOA from SVOC and IVOC. The performance of both models is also evaluated against field data. Both models do a reasonable job reproducing the measurements.

Overall I liked the paper. Given the lack of speciation data for SVOC and IVOC emissions, there is value to exploring different surrogate based approaches. I recommend that the paper be published after the authors have addressed the following comments.

The proposed approach is significantly more complicated than a purely empirical scheme such as Grieshop et al. (which appears to be the basis of the reference model). In the end, both approaches appear to predict similar amounts of SOA (e.g. Figure 2). Is this agreement simply an artifact of fitting the rate constants for the non-alkanes surrogates to match the results (page 23477 line 17)? If you are fitting key model parameters of the surrogate model, what is the value of what is the value of this additional complexity? I am not arguing for the Grieshop approach (which has its own problems) but as a community we need to struggle with and justify the addition of complexity to these models. The paper would benefit from a discussion of these issues. What is the essential complexity that the model needs?

Assigning surrogates –This paper used the speciation data from Schauer to assign the surrogates. However, Schauer only speciated a small fraction of the low-volatile (SVOC and IVOC) emissions. The paper should explicitly state the percentage of emissions that were speciated. The paper should also do a better job justifying the assignments. Implicit in the approach is that the composition of the unspeciated is the same as the speciated component. That is unlikely to be true. For example recent work by Goldstein group suggests that most of the unresolved complex mixture is branched alkanes (Isaacman et al. Analytical Chemistry 2012). How would that alter the model predictions? It may not if key parameters have simply been fit to experimental data. However, branched alkanes have lower yields than aromatics.

Model Evaluation – The paper only uses organic and elemental carbon measurements made using filters or an in situ filter based approach. These data (provide relatively little constraints for the model. Did MEGAPOLI collect AMS, thermodenuder or other data that would help differentiate between the different modeling approaches. E.g. AMS data might help differentiate between models based on O/C ratio.

Minor comments

Fitting emissions with measured data (page 23483, line 23). A challenge with this

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approach is that it assumes the model has captures the effects of meteorology and concentrations. That should be pointed out. Given the uncertainty in meteorology it seems like the fitted profile is likely no better than the TPM approach.

Effects of temperature on gas-particle partitioning (page 23484 line 17) – The paper hypothesizes that problems with temperature predictions may explain some of the problems with model performance during the morning rush hour. Recent data suggest that the sensitivity of POA to temperature is on the order of a few percent mass loss per K (Ranjan et al. AST 2012), which comparable to data for SOA. How much would the model predictions of temperature need to be off for this to be a plausible explanation. It seems like problems with emissions and/or boundary layer height are more likely than temperature problems.

Page 23480 line 40 – typo "?Sciare"

Page 23480 Last paragraph beginning with "During the this Megapoli" Were the dynamic blanks run with the denuder? If so, it seems like the denuder is not operating very efficiently?

Page 23485 line 23 –Phase mixing of SOA and POA. It seems strange to invoke another modeling study to "prove" that there is not phase mixing. The published experimental data on this subject are mixed. (Song et al. GRL 2007 versus Asa-Awuku et al. GRL 2009)

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 23471, 2012.