Interactive comment on “Functionalization and fragmentation during ambient organic aerosol aging: application of the 2-D volatility basis set to field studies” by B. N. Murphy et al.

B. N. Murphy et al.
spyros@chemeng.upatras.gr
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Responses to Referee #2

This manuscript represents continued efforts to use a model to describe mass concentrations and the chemical nature of organic aerosol. While the work itself is not terribly novel (uses mostly pre-existing modeling techniques and data sets), it is important and well within the relevance of ACP. It is well written, and all texts, tables, and figures are necessary. The title, abstract, and citations are all appropriate. The science method used is mostly sound; please see below for specific minor issues/comments.

1) On page 9871, lines 4-7, why is the assumption about biogenic aging relaxed here? (Also on page 9876, line 1). Please defend and explain.

This main issue here is deconvolution of mass enhancement due to aging and that due to first generation yield parameterizations. In previous work with PMCAMx-2008, a 3-D CTM, we concluded that mass enhancement due to biogenic aging was probably already accounted for in the first-generation parameterization (Murphy and Pandis, 2009; 2010). The major evidence for this came from overprediction of OA mass at forested sites when enhancement from biogenic SOA aging was assumed to be zero, yet anthropogenic SOA was allowed to age. Of all the model configurations, this setup performed the best. Due to new evidence from chamber studies that biogenic SOA compounds do age (Ng et al., 2006; Tritscher et al., 2011), as well as field observations from radiocarbon analyses that find high contributions from modern (biogenic) carbon to the total OA mass, we relax here our assumption and explore scenarios where biogenic SOA compounds age similarly to anthropogenic SOA compounds. An important finding here, then, is that fragmentation pathways may be necessary to implement if one is to avoid large overprediction by organic aerosol models.

2) On page 9872, lines 24-26, why is a rate constant increased by a factor of 3 used here? (Also on page 9878, lines 24-25). As above, please defend and explain.

The rate constant is increased by a factor of 3 simply to try to reveal a signal in the model. The same effort in Murphy et al. (2011) found little change in OA mass or O:C, and increasing the rate constant to the upper bound helps determine whether or not the process might be important. This is now explained in the revised manuscript.

3) On page 9878, line 13, I would suggest removing the words "seem to" - the factors all affect the organic aerosol characteristics.
4) On page 9900, figure 3, the gray line is difficult to see. Please correct/improve.

Corrected.

Extra Refs


Interactive comment on Atmos. Chem. Phys. Discuss., 12, 9857, 2012.