

Interactive
Comment

Interactive comment on “Emissions of biogenic volatile organic compounds and subsequent formation of secondary organic aerosols in a *Larix kaempferi* forest” by T. Mochizuki et al.

Anonymous Referee #2

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Overall Comment and Recommendation:

These authors conducted collocated measurements of isoprene and alpha-pinene fluxes as well as their oxidation products in aerosol collected at a mixed-forest site during summer 2012. This site was close to an urban area and suggested to be influenced by urban emissions. The flux measurements seem appropriate, but I cannot comment on the quality of this dataset. With regards to the SOA measurements, they are rather limited in scope and also many important details are missing in the manuscript that describe how exactly these were done (please see specific major comments below). In addition, there are many important details about how the PMF was conducted miss-

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Discussion Paper



ing from the manuscript (please also see specific major comments below). One of the major conclusions of the manuscript is that the biogenic SOA formation seems to be controlled by anthropogenic infow to the forest canopy. From the data presented, I'm not sure this conclusion is fully supported by the data. For example, considering there was inorganic aerosol and met data availability why wasn't aerosol acidity considered? More importantly, there have been some major recent advances, as I describe below, in our understanding of both isoprene and alpha-pinene SOA chemistry. Discussion of how the authors data fits into the recent lab and field findings is warranted, especially considering their conclusions. In the current form, it appears that the paper is more or less reporting the data but not really providing strong context for their data and detailed analyses to more fully confirm their conclusions. In the current form, I'm borderline if this should be rejected. However, if the authors can overcome some of the major issues pointed out here then I suggested accept with major revisions noted.

Specific Major Comments:

1) Sampling Size: I would argue that this sampling period is rather limited. Do the authors worry at all that this is providing a very narrow window into what occurs at this site? How do you know this time is optimal for studying this chemistry at this site? It is unclear why this period was selected. This seems to be a major weakness of this study.

2.) Experimental Section: There clearly needs to be a subsection within the experimental section and likely corresponding SI section that provides the details of your PMF analyses. There is no way for the reader to know how well you conducted your PMF and why you ended up selecting 3 factors. Why not 4 or 5 or more? This absolutely needs to be included in a revised submission. How do we know if this PMF means anything, considering this is a statistical solution?

3.) SOA tracer analyses: How were the isoprene- and alpha-pinene-derived SOA tracers quantified? Ideally, authentic standards are preferable, as is starting to be done

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by other groups (Surratt, Claeys, Keutsch or Geiger groups). If ketopinic acid was used based on the Kleindienst et al. (2007, Atmos. Environ.), I don't think this is a good standard to use for all compounds and could affect the quantification reported here. The authors need to clarify. In a revised manuscript details of identification, quantification and how this was done is absolutely needed.

4.) Aerosol Sampling:

I have several questions about the aerosol sampling:

a.) TSP is not ideal, especially in terms of size cuts. Most of the BSOA will be in the fine mode, so it isn't clear to me why TSP sampling was selected here?

b.) Precombustion of filters at 450 C for 6 hours may not be long enough. Why did the authors not consider higher temps (550 C) for 12 hours or longer? In addition, with TSP sampling, are the authors worried about any potential artifacts from gas-phase absorption or evaporation of semivolatiles? This issue needs to be addressed here. Why wasn't PM_{2.5} considered?

5.) Discussion of known mechanism of SOA formation and how this relates to acidity and NO_x is lacking:

I'm surprised there was no mention of the likely importance of isoprene epoxydiols (IEPOX) formed under HO₂-dominant conditions (Surratt et al., 2010, PNAS; Lin et al., 2012, ES&T; Lin et al., 2014, ES&T; Nguyen et al., 2014, PNAS; Gaston et al., 2014, ES&T; Riedel et al., 2015, ES&T Letters). It has been shown that the presence of wet acidic sulfate seed aerosol controls the uptake of IEPOX in forming SOA, including the 2-MTs. I'm curious to know why the authors didn't consider calculating aerosol acidity using their inorganic aerosol data and met data using one of the available thermodynamic models, such as ISOROPPIA II (Guo et al., 2014, ACP) from Thanos Nenes group at GA Tech. You could explore how acidity correlates to these compounds. In addition, how do these compounds correlate to sulfate levels?

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6.) Discussion of relevant new pathways related to alpha-pinene SOA:

I suspect ozone chemistry might explain your higher levels of alpha-pinene SOA. If so, how do your results fit into the context of recent work on ELVOC chemistry (Ehn et al., 2014, Nature)? It is now thought that ELVOC chemistry explains most of the SOA mass from alpha-pinene + O₃.

Related to this, based on your ozone measurements at the site, do you expect the lifetime of alpha-pinene to be shorter with O₃ than with OH? You should consider at least doing a back-of-the-envelope calculation.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 10739, 2015.

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