

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 #1

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Mochizuki et al. present field measurements of anthropogenic and biogenic VOCs as well as VOC oxidation products both in the gas and aerosol phases in a coniferous forest. In particular, the authors show concentration profiles and above-canopy fluxes of isoprene and alpha-pinene, and concentrations of benzene, toluene, methacrolein, methyl vinyl ketone, and secondary organic aerosol compounds derived from isoprene and alpha-pinene. After discussing vertical profiles and diurnal cycles, the authors apply positive matrix factorization and interpret the PMF factors with respect to photochemical age and anthropogenic influence. From this analysis, they attempt to derive the controlling factors of biogenic secondary organic aerosol formation and finally con-

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clude that anthropogenically influenced air masses contribute to elevated concentrations of SOA derived from isoprene and alpha-pinene.

The manuscript is well written and clearly structured. A detailed investigation of intracanalopy processes that affect biogenic VOC oxidation and secondary aerosol formation, and the interaction of anthropogenically vs. biogenically influenced air masses, is timely and very interesting and important. Thus, the manuscript is well suited for ACP. However, in my opinion, some of the ideas put forward in the manuscript must be revised by the authors. In particular, the authors may consider the following comments:

1) My main concern is with the interpretation of the PMF analysis in section 3.4 of the manuscript. I do not fully agree with the classification of the three factors extracted from this analysis. For example, factor 3 is described as anthropogenically more influenced/photochemically less aged. However, the biogenic markers isoprene and pinene are dominant contributions to this factor. While the lack of oxidation products of biogenic precursors indicate photochemically less aged air, there is also a substantial contribution of secondary inorganic aerosol (sulfate and nitrate). Overall, I do not agree that the PMF analysis allows an interpretation of the three factors in a two-dimensional space of anthropogenic influence and photochemical age. This challenges the following discussions in section 3.4 and Figure 9, and thus, one of the main conclusions of the manuscript - the enhanced formation of biogenic SOA due to the inflow of anthropogenic precursors and aerosols.

2) With respect to relaxed eddy accumulation (p. 10745): Did the authors apply a wind deadband for REA sampling? Did the authors use the averaged b value mentioned in the manuscript, or the instant b value calculated from equation (2) in each individual 1 hour interval?

3) With respect to the ozone and NO_x profile measurements (p. 10747): Did the authors check the response time of the trace gas analyzers in combination with the sampling lines of the profile system, and discard data just after switching the valves? How

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fast is the air in the sampling lines exchanged? In a similar setup with the same type of analyzers, I would expect that data at least within 60 to 90 s after switching the valves must be discarded. In addition, it would be very interesting to discuss the concentrations of NO and NO₂ separately instead of total NO_x, especially when classifying air masses according to photochemical age.

4) Can the authors explain the clear change in benzene and toluene concentrations between 7/15 and 7/16 in Figure 3c? On 7/15, benzene concentrations are clearly higher than toluene, while toluene concentrations are typically higher on other days.

5) The authors suggest that the diurnal cycles of 2-MGA and 2-MTLs follow the diurnal cycle of isoprene, thus indicating local production of isoprene-derived organic aerosol (p. 10751). Is the estimated timescale for 2-MGA and 2-MTLs production from isoprene oxidation consistent with this interpretation? What is the estimated timescale for 3-MBTCA production from alpha-pinene oxidation? Is it sufficiently long to expect a difference in the diurnal peaks of alpha-pinene and its oxidation products, as stated on p. 10752?

6) The content and structure of the Abstract and the Conclusions section are basically identical. Please revise the Conclusions section and put the main results and conclusions in a broader context!

Technical comments:

p.10744, line 19: define greek phi symbol

p. 10745, line 4: define SD

p. 10748, line 23: replace "suggsted" by "suggested"

p. 10750, line 23: add "a" between "reported for" and "Pinus sylvestris": "...reported for a Pinus sylvestris forest..."

p. 10751, line 26: What exactly do you mean by "atmospheric reactivity for isoprene

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oxidation"

p. 10752, line 26: What exactly do you mean by "atmospheric reactivity of alpha-pinene"

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