

We appreciate the referee's valuable comments and thoughts on our paper. We have carefully revised the manuscript by taking account of the comments. Our responses to the specific comments are given below.

Responses to the comments of Referee#1:

Comment 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.

Reply 1:

We have used the term “photochemically more/less aged” from viewpoints of the degree of atmospheric processing of biogenic precursors. Our measurements indicated that the biogenic precursors were emitted mainly from the local forest, whereas the observed secondary inorganic aerosol (sulfate and nitrate) were transported from regions outside the forest site. This might result in the difference in the degree of photochemical processing between biogenic precursors/SOA and inorganic aerosols observed at the site. Moreover, relative influence of anthropogenic sources on factor 1 and 2 is important to interpret possible factors that control formation of the observed biogenic SOA tracers in this study. In order to clarify this point, a category of “photochemically more/less aged” has been changed to “more/less processed biogenic VOCs.” (Page 13, Lines 11-16)

With regard to the PMF, we have now added a subsection (2.5) that provides the details of our PMF analyses. The new subsection includes descriptions on how we determined the numbers of interpretable factors. (Page 7, Line 26- Page 8 Line 6)

Comment 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?

Reply 2:

Yes, we used a dynamic deadband which was determined from the latest 15-min running mean and standard deviation of the vertical wind velocity. An averaged value of the coefficient b during the whole measurement period was calculated as the slope of $w'T'$ against $\sigma_w(T^+ - T^-)$. We have added descriptions as follows.

“ C^+ and C^- are the VOC concentrations in the upward and downward air, respectively, collected when the absolute value of the measured vertical wind speed is higher than a dynamic deadband value (Mochizuki et al., 2014).” (Page 5, Lines 7-10)

Taking account of the comment, the sentence *“An averaged value of the coefficient b was determined to be 0.38 ($r^2 = 0.98$).”* has been revised to *“The averaged value of the coefficient b during the whole measurement period was determined to be 0.38 ($r^2 = 0.98$), which was calculated as the slope of $w'T'$ against $\sigma_w(T^+ - T^-)$.”* (Page 5, Lines 15-17)

Comment 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 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 NO_2 separately instead of total NO_x , especially when classifying air masses according to photochemical age.

Reply 3:

At our study site, we checked the response time of the ozone and NO_x analyzers including the whole inlet system, and discarded 150-sec data for these gases just after switching the solenoid valves. This is based on our experimental test, which showed that it took approximately 40 sec for the air in the sampling lines to be exchanged. In the revised manuscript, we have added this information to the text in section 2.4: *“On the basis of the response time for O_3 and NO_x analyzers in combination with the sampling lines, we discarded 150-sec data for these gases just after switching the solenoid valves.”* (Page 7, Lines 23-25)

The timescale of interconversion between NO and NO₂ is typically ~100 s during the daytime. This timescale is comparable to that of the turbulent mixing within and out of a forest canopy, but is much faster than that of chemical processing of biogenic precursors to form SOA discussed here (~a few hours). Therefore NO/NO₂ might not be a proper indicator for classifying air masses according to photochemical age in this case.

Comment 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.

Reply 4:

Toluene to benzene (T/B) ratio has been commonly used as an indicator of traffic emissions. The T/B ratios observed during the study period beside July 15 indicates that they are emitted predominantly by motor vehicle-related sources. Previous studies reported that benzene concentration in flue gases increases as a result of incomplete combustion. Although it is not clear from the local wind direction, the lower T/B ratio with higher benzene concentrations on July 15 may be mainly due to the dominant contribution of various fuels such as fuel-oil, diesel, etc.

Because the detailed information about the sources of benzene and toluene is not directly linked with our discussion in this paper, we have not added any statement on the different behavior of benzene and toluene.

Comment 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?

Reply 5:

The lifetime of isoprene is estimated to be ~1.7 h at 25°C assuming the typical OH concentration of 1.5×10^6 molecules cm⁻³. Previous laboratory experiments showed that the timescale for 2-MGA and 2-MTLs production from isoprene oxidation is

several hours (e.g., Surratt et al., 2010), which is comparable to the sampling duration (~3 h) during the daytime in this study.

With regard to the production of 3-MBTCA from α -pinene, the timescale of the production (> ~3 h) shown by laboratory experiments (e.g., Eddingsaas et al., 2012, Kristensen et al., 2013) is longer than the sampling time during the daytime in the current study. This can explain the difference in the diurnal peaks of α -pinene and its oxidation products in aerosols observed in this study.

Taking account of the comment, the following sentences have been added in section 3.2 and 3.3.

section 3.2.: *“Laboratory experiments showed that the timescale for 2-MGA and 2-MTLs production from isoprene oxidation is a few hours (e.g., Surratt et al., 2010). This timescale is comparable to the sampling duration (~3 h) during the daytime in this study, which is consistent with the similar temporal trend of the concentrations of isoprene, 2-MGA and 2-MTLs.”* (Page 11, Lines 24-28)

section 3.3.: *“With regard to the production of 3-MBTCA by OH-initiated oxidation of α -pinene, the timescale of the 3-MBTCA formation (> ~3 h) shown by laboratory experiments (e.g., Eddingsaas et al., 2012, Kristensen et al., 2013) is longer than the sampling time during the daytime in the current study.”* (Page 12, Lines 22-25)

Comment 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!

Reply 6:

According to the comment, we have revised the conclusions section as follows.

*“We measured concentrations and canopy-scale fluxes of isoprene and α -pinene simultaneously with their oxidation products in total suspended particles (TSP) at a *Larix kaempferi* forest site in summer. Isoprene and α -pinene accounted for 23% and 44%, respectively, of the total terpenoids measured in this study. Vertical and diurnal profiles of isoprene and MACR+MVK suggest large emissions of isoprene near the forest floor, which is likely due to *Dryopteris crassirhizoma*, followed by reaction of the isoprene within the *L. kaempferi* canopy. The concentrations of α -pinene also showed peaks near the forest floor with maximums in the early morning and late afternoon, suggesting significant emissions of α -pinene from soil and litter in addition to emissions from leaves at the forest site.”*

*Isoprene and its oxidation products in aerosols exhibited similar diurnal variations within the forest canopy, providing an evidence of SOA formation within a timescale of a few hours via oxidation of isoprene emitted from *Dryopteris crassirhizoma* on the forest floor. PMF analysis indicated that anthropogenic inflow likely contributed to the enhanced concentrations of both isoprene-derived (>64%) and α -pinene-derived (>57%) SOA within the forest canopy. The combined analyses of the fluxes and vertical profiles of BVOCs suggest that the BSOA formation promoted by the anthropogenic inflow was enhanced within the forest canopy even though the BVOC fluxes were relatively low. This study emphasizes an importance of intra-canopy processes for biogenic SOA formation in the presence of significant inflow of oxidants as well as anthropogenic aerosols and their precursors.*

Comment 7: p.10744, line 19: define greek phi symbol

The words “ ϕ 1/4 inch \times 3.5 inch” have been changed to “88.9 mm long, 6.35 mm outer diameter” (Page 4, Lines 25-26)

p. 10745, line 4: define SD

Now “SD” has been defined as “standard deviation.” (Page 5, Line 7)

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

Corrected as suggested. (Page 9, Line 10)

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

Revised as suggested. (Page 10, Line 29)

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

The phrase “atmospheric reactivity for isoprene” has been replaced by “the reactivity of isoprene to form OAs” (Page 12, Lines 2-3)

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

The phrase “atmospheric reactivity for α -pinene” has been replaced by “reactivity of α -pinene to form OAs.” (Page 13, Lines 3-4)

References

Kleindienst, T. E., Lewandowski, M., Offenberg, J. H., Jaoui, M., and Edney, E. O.: Ozone-isoprene reaction: Re-examination of the formation of secondary organic aerosol, *Geophys. Res. Lett.*, 34, L01805, doi:10.1029/2006GL027485, 2007. (Page 19, Line 17-19)

Eddingsaas, N. C., Loza, C. L., Yee, L. D., Chan, M., Schilling, K. A., Chhabra, P. S., Seinfeld, J. H., and Wennberg, P. O.: α -pinene photooxidation under controlled chemical conditions – Part 2: SOA yield and composition in low- and high-NO_x environments, *Atmos. Chem. Phys.*, 12, 7413-7427, 2012. (Page 16, Line 26-29)

Kristensen, K., Enggrob, K. L., King, S. M., Worton, D. R., Platt, S. M., Mortensen, R., Rosenoern, T., Surratt, J. D., Bilde, M., Goldstein, A. H., and Glasius, M.: Formation and occurrence of dimer ester of pinene oxidation products in atmospheric aerosols, *Atmos. Chem. Phys.*, 13, 3763-3776, 2013. (Page 19, Line 22-25)