

## ***Interactive comment on “Semi-volatile and highly oxygenated gaseous and particulate organic compounds observed above a boreal forest canopy” by Ben H. Lee et al.***

### **Anonymous Referee #2**

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Lee et al. describe aerosol and gas-phase measurements of organic compounds from tall tower located above a boreal forest. The measurements show the diurnal patterns of gas-phase species, measured using an I-CIMS, and particle-phase species, measured using a FIGAERO inlet. The authors find that most gas and particle-phase species exhibit either a morning, daytime, or nighttime enhancement. In the gas-phase, smaller molecules dominated the organic distributions, though highly oxygenated molecules (or HOMs) were observed during the morning and daytime. In the particle phase, HOMs were observed in each diurnal subgroup. Of these compounds, the organic nitrates constituted a significant fraction of the detected organic species, with highest contributions at night. A non-negligible amount of nitrate dimers were ob-

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served, which were suspected to be formed by the reaction between  $\text{NO}_3\text{RO}_2 + \text{RO}_2$  radicals.

The results from this study contributes to the evidence that organic nitrate species formed from biogenic VOC oxidation significantly contribute to organic aerosol, especially at night. The results are interesting and well-interpreted, the paper is well written, and the figures are nice and descriptive. I recommend the manuscript for publication provided that the authors address the following very minor comments.

Page 4, lines 26 - It's not clear why NNMF was not applied to raw concentration counts. Is this to give equal weight to all species (i.e., the assumption is that changes in concentrations will be approximately equal across species)? Furthermore, how were the errors estimated? Please clarify.

Page, Lines 8 -12 - I really like this approach for resolving factors, especially as the authors are not trying to over-interpret the data. Can the authors mention how well the variability was explained by the resolved subgroups? Also, what type of residual was left over not explained by NNMF?

Page 5, Lines 21-22 - I'm confused by what the authors are trying to say here. Do the authors mean to say that high abundance masses observed in the gas phase were also observed in the particle phase, but that the presence of these species was unexpected based on volatility? Can the authors give some examples to help orient the reader? This would be useful when interpreting the results in Fig 1.

Page 6, lines 1 - 3. Couldn't the variability also be explained, in part, due to higher emission rates of monoterpenes as a function of temperature?

Page 7, lines 9-11. Do the authors have other data that could show whether the breakup of the nocturnal boundary layer contributed to the trends observed here? Were there vertically resolved measurements (e.g. temperature, RH, etc) that support the presented of a nocturnal layer below the tower? I realize that this will not change

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the interpretation of gas and particle phase correlations, but it would be interesting to know if the morning diel pattern is dominated by sudden burst of species produced during the night time, or by a sudden burst in oxygenated species once photochemistry kicked in.

Page 9, Lines 12-23. Is it reasonable to infer that the agreement between the AMS (located below the forest canopy) and FIGAERO CIMS (located above the forest canopy) in pON provides evidence that that the tall tower was within the nocturnal boundary layer?

Figure 3: This figure is great and conveys a lot of information. Can the authors comment on what appears to be a bi-modal distribution in the C11-C20 compounds? There appears to be two peaks in the nO distributions, with one peaking around 5-6 oxygens, and the other peaking at 8-10 oxygens. Is this related to carbon number, or is this explained more readily by other processes (auto-oxidation of dimers)?

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