Comment on acp-2021-367

Anonymous Referee #3

Referee comment on "Evolution of volatility and composition in sesquiterpene-mixed and *a*-pinene secondary organic aerosol particles during isothermal evaporation" by Zijun Li et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-367-RC3, 2021

This work by Li et al. investigates the changes in volatility and composition of SOA from the oxidation of α-pinene and a sesquiterpene mixture as a result of isothermal evaporation. It details laboratory experiments performed with a FIGAERO-CIMS to obtain volatility information and molecular formulas for species in the SOA. Results from the sesquiterpene mixture were compared to that from α-pinene, and also compared between dry (RH <7%) and wet (RH ~80%) conditions. Positive matrix factorization (PMF) was used to investigate the behavior of individual factors before and after evaporation at the two RH extremes. Unsurprisingly, one result is that the sesquiterpene SOA is more resistant to evaporation than α-pinene, implying lower volatility. They also found that under high RH, more of the signal evaporated. This work is novel, will be of interest to the readers of ACP, and provides valuable information to the community. After addressing the minor general and specific comments below, this manuscript will be suitable for publication in ACP.

General comments:

- The use of so many acronyms make the manuscript difficult to follow at times. Some examples include:

- the abbreviation of α-pinene to αpin is unnecessary
- changing STG to ∑TG could make it easier for the reader to remember what this stands for
- RTC for residence time chambers is used to refer to both the physical chamber (lines 94-96, 105, 111) and the experiment type (lines 130, 160, 182, etc) which is confusing. When referring to experiment type does it always refer to a specific

evaporation time length?
- CNerror is only used once (line 155).

- This study could benefit from a more robust discussion of, and comparison to, the several previous studies measuring the evaporation of α-pinene SOA

Specific comments:

Line 78: I think it could be helpful to the reader to 1) specify the mixture of sesquiterpenes, at least with respect to the most dominant species present, and 2) show their structures somewhere.

Paragraph starting line 78: please add your SOA loadings to this paragraph. If there was much more than typical ambient (>~50 ug/m3), can you please comment in the results and discussion section how your results may or may not be affected from partitioning more SVOC to the SOA than would be observed in ambient?

Line 81: Do you think forming the SOA at one RH and evaporating it at a different RH will affect your results? Do you think there is a long or short equilibration time between formation and evaporation RH?

Line 93-96:

- Was there anything in these evaporation chambers to absorb desorbing vapors (for example activated charcoal)? If not, do you think there's a role for re-partitioning?
- The workflow isn't obvious—were all particles sent sequentially through each stage laid out in i-iii, or did particles only go into one evaporation chamber? If particles only went into one type of reaction chamber, was the reasoning behind having 3 types to achieve different lengths of evaporation time?
- Do you expect the same evaporation behavior and results in each chamber (i.e. no impacts from air volume to wall surface area, etc)?

Line 94: "*a* 25 L… chamber*s*"— was one chamber used or multiple? If multiple, were they all identical?

Line 115: saturation vapor *pressure* or *concentration*?

Line 120: instead of "the appearance", "the shape" might be more easily understood, but ok as-is.

Line 145: half the sum thermogram signal or mass?

Line ~190: Can you please discuss why the overall volatility is lower under wet versus dry conditions, yet under these low-volatility wet conditions more signal is lost after evaporation?

Line 200: Couldn't this be tested by comparing the signal to mass concentration under dry versus wet conditions? If the mass concentration is equivalent, you could determine what percent of signal was lost presumably due to evaporation of high volatility material during the SOA collection time. Or if the mass concentration was different but the signal was the same, you'd have a hint that the SOA under the two conditions may have been made via two different pathways (e.g. reactive uptake at high RH vs. vapor-pressure driven condensation at low RH).

Line 239: I think it makes sense to reference some of the papers on thermal decomposition upon heating with a FIGAERO here (or above).

Line 255: Would help to directly reference the figure in place of saying "above"

Line 265: excl = excluding? Please write out complete word for clarity

Line 270: rational should be rationale

Line 285: "decreases with *evolving* isothermal…" or "with *increasing* isothermal…"?

Line 287: if the NCR doesn't decrease with decreasing VFR couldn't this also mean that nothing is happening to the compounds, that they are neither being lost or produced?

line 303: can you speculate on other possible loss mechanisms?

Line 307: Is it possible that there is different gas-phase chemistry or SOA formation pathways (i.e. reactive uptake vs. condensation) at high RH, so these species don't necessarily have to be produced in the particle phase?

Line 319: figure 2, panels a & b only, correct?

Line 331: figure 3, panel a only, correct? And figure 6 panel a?

Line 333: is it possible that there are more compounds grouped into the factor under wet conditions, instead of more signal of the same number of compounds?

Line 393: Should have a reference for α-pinene having largest emissions globally

Line 397: sentence "These findings are generally…" is redundant

Line 740/ Figure 2: Putting the y-axis in panels a & b on the same scale would make these easier to compare.

Key figure: is time on the x-axis for both the left and right figure/schematic?