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Interactive comment on "Estimates of the organic aerosol volatility in a boreal forest using two independent methods" *by* Juan Hong et al.

Anonymous Referee #1

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General Comments

The authors have estimated volatility distribution of ambient SOA in a boreal forest by interpreting Volatility Tandem Differential Mobility Analyzer (VTDMA) measurements with a kinetic evaporation model. The modeling results show that 40% of the SOA was semi-volatile, 34% low-volatile, and 26% extremely low-volatile, with an effective enthalpy of vaporization value of 80 kJ/mole for all organics. They also independently estimated semi-volatile and low-volatile organic mass fractions by applying Positive Matrix Factorization (PMF) to the High Resolution Aerosol Mass Spectrometer data, with the factor separation based on the oxygenation levels of organics (relative abundance of mass ions at m/z 43 and m/z 44). The manuscript is well written, but the usefulness of the results is questionable. The study could become publishable if the following comments are satisfactorily addressed.

Detailed Comments

1) It should be made clear in the abstract, body, and conclusion that this study estimates the effective volatility distribution of SOA after it has formed and undergone aging (particle-phase reactions) and not the volatility distribution of the condensing organic gases that produced the SOA. While the authors do not imply the latter, some readers may inadvertently misinterpret the results and incorrectly apply them for modeling purposes if the caveat is not explicitly stated.

This then brings up the question of the usefulness of the results. If the estimated volatility distribution of SOA does not represent the volatility distribution of the condensing organics then where and how would one use this information? What is it that we have learned from this exercise that is of value? To drive this point further, it is possible that the semi-volatile fraction of the condensing organics may be more than 40%, such that some then undergoes particle-phase reactions to form low- and extremely low-volatile compounds. Upon heating the SOA in the VTDMA, these newly formed compounds (e.g., dimers, oligomers) may partially decompose back to the original species or may even fragment (especially at higher temperatures) to something completely different before evaporating. However, these processes are not examined in the present study. The evaporation model simply assumes that the three lumped species (with different volatilities) do not chemically interact. Thus, it is difficult to draw any useful or meaningful conclusions out of the present analysis.

2) Furthermore, since the VTDMA experiments were carried out under dry conditions, the boreal forest SOA is expected to be viscous (Virtanen et al. 2010 Nature), especially at 25 C, such that there would be significant particle-phase diffusion limitation for the evaporating species. Perhaps the evidence for this limitation is present in Figure 3, which shows that the model tends to be way more evaporative than observed at the lower temperatures–in fact, the first data point indicates no evaporation (MFR = 1). This can potentially skew the effective volatility distribution estimated by the model quite a bit.

3) It is stated that the residence time inside the thermodenuder was around 2.5 s. Does this mean that the evaporation model was run for just 2.5 s to simulate each data point? Also, does the model assume that the aerosol instantly reaches the targeted temperature the moment it enters the thermodenuder? Can the authors estimate the time it takes the aerosol to reach the target temperature? The model should account for it if it's comparable to the residence time.

4) Please show error bars on the VTDMA measurements displayed in Figures 2 and 3.

5) The authors state that the agreement between the VTDMA results and the PMF-derived results as reasonable when it is quite the opposite. The linear correlation coefficient (r) of 0.4 indicates there is not a good correlation between the VTDMA- and PMF-based results (Figure 5c,d). The coefficient of determination (r^2) is only 0.16, which means only 16% of the variation can be explained by the linear relationship between the two methods. It is clear that the comparison of VTDMA and PMF results was not quite successful. I suggest that the authors revise the text at all the appropriate places and describe the results of correlation as they are.