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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "The influences of mass loading and rapid dilution of secondary organic aerosol on particle volatility" *by* K. R. Kolesar et al.

Anonymous Referee #1

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General comments:

The ACPD paper 'The influence of mass loading and rapid dilution of secondary organic aerosol on particle volatility' by Kolesar et al. reports a study on thermally induced evaporation of a-pinene secondary organic aerosol (SOA) formed by ozonolysis. Mass thermograms were measured and these were compared to a kinetic model with three different scenarios: previously measured volatility basis set (VBS) including semi-volatile organic compounds (SVOCs), assumption of dimer formation and including low-volatile organic compounds (LVOCs). The authors find that the SVOC version does not capture the observed evaporation, and conclude that the SOA contained substantial fraction of dimers and/or LVOCs. The modeling method includes several assumptions - which are needed due to the several unknown properties - and thus a large

uncertainty is associated with the quantitative results. However, qualitatively the results provide useful information. The manuscript is well written, the methods are in general valid and the topic fits well in the scope of ACP. Therefore I recommend publication in ACP after the below comments have been addressed.

Specific comments:

1) The conclusion of dimer formation being important relies on the kinetic modelling with dimers included in the model. This requires an assumption of initial particle composition which in this work is approximated by first calculating the particle composition based on partitioning theory and previously measured VBS and then calculating the monomer/dimer equilibrium (p. 10006, l. 16-21). However, if the SOA contains dimers, these compounds would be accounted for in the lower volatility bins when VBS was determined from the growth experiments, not by the VBS bins corresponding to the SVOC monomers. The authors estimate the dimer formation time scale to be only a fraction of a second (p. 10014, l. 4-5) which would suggest that the dimers would have been formed also in the growth experiments where VBS was determined and would therefore contribute to the lowest VBS bins. As most of the particle mass is estimated to consist of dimers (p. 10013, l. 8) the way the dimers are treated in the calculation of initial composition could make a large difference on results. Could the authors comment on how big error this could cause on their results and what would be the possible consequences of this regarding their conclusions?

2) It is said that the dimers were assumed non-volatile. Were all the VBS bins treated the same way regarding dimer formation? Is it justified to assume that the dimers formed from the compounds in the most volatile VBS bin would also be non-volatile? How would it change the results if the dimers formed from the most volatile SVOCs would evaporate (even though much slower than the monomers)?

3) p. 10004, eq. 2: The VFR was calculated based on volume-weighted average diameters. However, based on Fig. 1 the size distributions were rather wide. Did

the model consider polydisperse particle population or did it assume monodisperse population? How large uncertainty does the use of average diameter for VFR cause in respect the comparison of measurement and model?

4) p. 10010, l. 2-6: Evaporation upon isothermal dilution can be rapid for the most volatile SVOCs. Therefore, the composition of diluted SOA can be different, especially if large fraction of initial mass was of the most volatile SVOCs.

5) The assumption of initial composition of particles determines to a large extend the modeled mass thermograms. In the low-volatility version of the model the total concentration of each compound C_i,tot was calculated with the exponential equation (p. 10018, l. 11). Could the authors clarify the use of this equation a bit? It is not clear why such exponential equation is used and if it is physically justified. Did the a1, a2, a3 and a4 have same values for each C*? It is said (p. 10018, l. 12-16) that the a values were determined for one certain C_OA value, however later the same set of a values is used for varying C_OA values. Is this consistent use of the a values and the equation for C_i,tot.

Technical comments:

p. 10009, l. 23: There should probably be ‘high’ before ‘C_OA’.

Figure 1: I find the x-axis numbers confusing as one would by quickly looking think that the average diameter was 2-4 nm, instead of 20-40 nm. I would thus recommend modifying the x-axis.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 9997, 2015.