

Interactive comment on “Evolution of organic aerosol mass spectra upon heating: implications for OA phase and partitioning behavior” by C. D. Cappa and K. R. Wilson

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This interesting paper is well written and presents important results of great interest to the research community. Experiments with lubricating oil (LO) and alpha pinene ozonolysis (α P) aerosols demonstrate some unexpected behaviors in these important proxy systems for atmospheric aerosols and also introduce a range of questions for future experimentation. The lack of compositional change in the α P aerosol during

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evaporation and the hypothesis that this is due to its glassy state (as supported by other recent observational and theoretical work) are especially interesting.

We look forward to this work being published in ACP when comments from the referees are addressed. Here we present a few points, focusing on relevant work we did at CMU, that might also be considered when the manuscript is being revised.

1) Our α P aerosol dilution experiments with a unit-resolution AMS showed variation (though not necessarily reversible) in the relative contribution at various m/z during both formation and later evaporation of the aerosols by dilution (Grieshop et al. 2007). For example, the contribution at m/z 44 decreased during two stages of aerosol formation and increased during dilution, indicating a variation in particle composition with aerosol loading consistent with the partitioning of semivolatile products with different volatilities and levels of oxygenation (Figure 3 in Grieshop et al., 2007), similar to what has been observed in some TD studies.

2) Discussion in Section 3.5.3 contrasts evaporation coefficients (γ_e) determined using a model of particle evaporation in the TD to those implied by the slow evaporation observed in our chamber-dilution experiments with α P (Grieshop et al. 2007) and LO (Grieshop et al. 2009) aerosols. The results for α P particles seem broadly consistent, while those for LO are quite different. The slow evaporation of both aerosols in our experiments was a surprise for us and certainly could be consistent with glass-like aerosol. Loss of semivolatile vapors to the Teflon walls of the smog chamber (Loza et al. 2010; Matsunaga and Ziemann 2010) does complicate interpretation of the formation and evaporation of semivolatile organic aerosols. For example, a ‘buffering’ of vapor-phase concentrations in the chamber during dilution by slow release of vapors lost to the walls could retard the evaporation of particles in our experiments. However, one must also be concerned about condensed phase reactions at elevated temperatures inside a TD.

However, there are likely significant differences between the aerosols in these and our

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experiments that should be noted and complicate a direct comparison. For example, the LO aerosols were generated in different ways. In this study, they were produced for introduction into the TD by heating liquid oil at 80 °C to generate particles via homogeneous nucleation in air, followed by dilution to control aerosol concentrations. This approach will likely create aerosols that are comprised of the more volatile fraction of the LO. Although LO aerosol may completely evaporate inside a TD at 80 °C the concentration of lube oil is very low. In an engine, LO does not evaporate at 80 °C because concentrations are much higher. In contrast, our study was based on aerosols generated with used LO flash vaporized at 425 °C directly into the chamber, also forming aerosol via homogenous nucleation. Figure S.1 in the Supplemental Information accompanying the paper showing the LO data (Grieshop et al. 2009) indicates that our LO aerosols were quite similar in composition to the source oil, though perhaps slightly biased towards higher volatility material. The LO thermogram in this manuscript shows an aerosol that is substantially more volatile than that in our work.

Minor points:

P. 28438, L23-25 – This statement is unclear

P. 28454, L15 – Grieshop is misspelled

References:

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