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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Measurement of the ambient organic aerosol volatility distribution: application during the Finokalia Aerosol Measurement Experiment (FAME-2008)" *by* B. H. Lee et al.

Anonymous Referee #1

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The authors explain the analysis method used for combining TD-SMPS and TD-AMS data sets from FAME-2008 to construct a volatility distribution of organic aerosol.

The introduction including the review of denuder history is quite nice, succinct, and informative. The overall manuscript is easy to follow and well written.

The overall balance between science and technical is heavy on technical and light on atmospheric science. AMTD rather than ACPD could be the more appropriate venue for publication. In this regard, the title and content of the manuscript seem not to match.

There is a major omission in this manuscript in that the fundamental assumption of the absence of reaction inside the particle is not stated. Several thermodenuder studies have emphasized the occurrence of particle phase reactions at the elevated temperatures in thermodenuders, both for ambient as well as laboratory aerosols. Examples include:

Denkenberger, K. A., R. C. Moffet, J. C. Holecek, T. P. Rebotier, and K. A. Prather (2007), Real-time, single-particle measurements of oligomers in aged ambient aerosol particles, Environ. Sci. Technol., 41, 5439-5446.

Wu, Z. J., L. Poulain, B. Wehner, A. Wiedensohler, and H. Herrmann (2009), Characterization of the volatile fraction of laboratory-generated aerosol particles by thermodenuder-aerosol mass spectrometer coupling experiments, J. Aerosol Sci., 40, 603-612.

This omission is critical. For example, Figure 8 that goes with the title of the manuscript is especially problematic. The figures before Figure 8 show MFR vs. temperature, so these are observations, even if underlying effects are not fully known. But Figure 8 places a framework around these observations by using an enthalpy to estimate room temperature volatility from observations at elevated temperatures. This is the process of translating the observations into the representative bins for panels B and D. This representation is potentially very wrong if there are considerable particle phase reactions at elevated temperatures, as the recent literature suggests. The authors make no mention of this complicating and perhaps even dominant phenomenon that would potentially completely invalidate Figure 8.

Equation 1 for calculation of density is not correct because densities add as inverse quantities for mass fraction. This could just be a typo, i.e., the stated equation would be correct for volume fraction. But if the written equation was the one used in the analysis then this could be a point of error.

A seemingly important citation to Epstein et al. mentioned in several figures is not

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found.

The claim in the abstract of ambient aerosol being two orders of magnitude less volatile than laboratory SOA is an overstretch because in point of fact the manuscript compares only to one laboratory system, which is the dark ozonolysis of alpha-pinene.

Page 17438, line 18, phrasing and citations could be revised because present version suggests that the citations on line 19 are related to the use of thermodunders. In fact, they relate only to the AMS.

Page 17442, line 21, this sentence does not make too much sense, i.e., slope needs to be stated to support the argument. Right now, the statement is that R2 = 0.95 without a statement of slope so "agreed well" cannot be concluded from provided information.

In Fig 7 and Fig 9, there are only 3 data points for 105 sec but 10's of data points for 14 sec. What implication does this difference in the quantity and quality of empirical information have for conclusions? Similarly, the temperature range is 100 to 140 C but the model is applied with extrapolation from 30 to 100 C. The analyzed data sets seem sparse and not well balanced.

Would the authors confirm that the data points for 14 sec and 105 sec in Figs 7 and 9 were collected nearly simultaneously from the same air mass? That is, if the data were collected on different days and the mass spectra are different, then the comparison is problematic.

The summary comment is that the content and tone of manuscript are more appropriate to AMTD rather than ACPD. If transferred to AMTD, some of the questions above indicate significant re-working of the manuscript is necessary. The aspects related to the sparseness of the data set might not be able to be overcome as a standalone manuscript. A workaround could be that the content of this manuscript should not appear separately but instead as a section within a more comprehensive manuscript that includes other aspects related to FAME-2008.

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