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Atmospheric Measurement Techniques Discussions
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Interactive comment on "Measurement of non-volatile particle number size distribution" *by* G. I. Gkatzelis et al.

Anonymous Referee #2

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Review of AMT-2015-93

This paper describes data collected through a thermodenuder (TD) operated at high temperature (∼400 C) during several laboratory and field measurements of aerosol size distribution, chemical speciation (with a HR-AMS) and black carbon/aerosol light absorption (with a MAAP). The results show that SOA particles formed in chamber experiments from single precursor compounds essentially completely evaporate during passage through the TD – evidenced by a complete loss of aerosol mass and number. On the other hand, ambient measurements, and measurements of oxidation of ambient aerosols, indicate that a substantial fraction of the particle number contains what are termed 'non-volatile' cores. The study quantifies the number fraction remaining via the particle size distributions measured before and after heating, and uses these measurements to gain insight into the sources of ‘non-volatile’ cores.

This is an interesting set of studies, but I question whether what is presented is really a new experimental methodology, and thus whether AMT is the appropriate venue. Thermodenuders have been operated for decades, including at elevated temperatures (Clarke et al. 2007; Clarke 1991; Wehner et al. 2002), and number-weighted size distributions have been compared in many of these studies. At the most, the paper presents a slightly different way of presenting data collected using what is now a fairly standard technique. Therefore, my feeling is that this work does not belong in a journal the focus of which is new method and tool development because there is quite sparse information on the actual data analysis (e.g. PMF on denuded aerosol spectra) and many of the insights are very particular to the campaigns in which the measurements were collected. Apart from this issue, my main concerns pertain to issues with data analysis and presentation of results. Below I list several of these. One other general issues is that the tables and figures are excessive and not very ‘information-dense’ – there are 14 figures and many of them are essentially displays of raw data time series. I would suggest that the authors work to reduce the number and select figures that more strongly support the point of the paper. Figures 1 through 8 could easily be moved to a supplement. It would be helpful to see a figure like Fig. 11 showing NFR vs BC mass fraction, rather than just BC mass. Figures showing size distributions should indicate the number of scans or time span shown, and should ideally include confidence intervals. Focusing the presentation of results would help make the paper more cohesive.

Title: The title is not representative, as a good amount of the discussion and supporting evidence comes from mass (and absorption) measurements. “Measurement of non-volatile sub-micron particle fraction” might be a more appropriate choice. I also question the characterization of the material measured under these conditions as ‘non-volatile’. This is ‘operationally defined’ here based on the measurement parameters, but the potential impacts of changes in these measurements is not give. For example, TD residence time can have a controlling effect on particle evaporation (Saha et al. 2015; Saleh et al. 2011) and organics may form char or form oligomers at high temperatures.(Denkenberger et al. 2007). ‘Non-volatile’ may be a short-form name for what was measured, but these potentially influences on measurements are mostly not discussed. The use of the chamber-generated SOA evaporation to assess the method is interesting, but these aerosol are relatively homogenous and don’t contain non-volatile cores, to which materials might, for example, adsorb. Chamber experiments incorporating nonvolatile seeds (e.g. fullerene soot, or even NaCl) would be more relevant to assessing the method.

P6357, L 15-17 – ‘Could have more significant health impacts’ – this is vague and not sufficiently justified. The latter part of this sentence is not clear why can’t the impacts be determined via mass measurements? That is what you purport to do with BC measurements – what additional information does this approach give you? If this statement is true, it could be true for both semi-volatile and non-volatile insoluble aerosols. Health effects of the semi-volatile fraction (substantially available in the ultrafine range) could be also complicated because of its dynamic nature and ability to partition into different phases.

P6359, L24 – This design has a room temperature centerline RT of ~15 seconds, which means particles in this study actually had a substantially shorter residence time (298K/673K *15 s). It is not relevant that the RT is longer than other designs if the aerosol system does not equilibrate – was it run at other residence times to assess whether the aerosol had equilibrated??

P6360, L16 – This CPC doesn’t have a 1 LPM flow mode. If the flow rate was modified somehow this should be clarified.

Eq. 1 – This represents transmission, not loss. Particle loss at higher TD temperature is an important, but uncertain step for interpreting TD data.

Eq. 2 – Application of empirically estimated loss parameterization using a non-volatile tracer (e.g., Eq.2 in this study using NaCl) to a wide range of laboratory and atmospheric conditions is challenging should be critically evaluted. For example, evaluation of the non-volatile particle size distribution measurement with a direct measurement of lab-generated pure BC particle (e.g., fullerene soot) could be used to evaluate Eq. 2 with a more atmospherically prevalent species. In addition, it is not clear how mass loss in the AMS was addressed. Was equation 2 applied? If so, how was this done?

P6362, L6-9: No details are given on the operation or configuration of the AMS. Details on SMPS operation are repeated. I recommend that instrumentation description be combined into one section.

P6366, L9-L10: The MAAP does not directly measure aerosol mass, rather measures aerosol absorption at a specific wavelength and applies an assumed mass absorption cross-section (MAC) to estimate mass. This MAC can vary by a factor of 2-3 between locations. Therefore, reporting and using BC mass is not advised unless these values were compared with another, mass-based instrument. If mass data (e.g. from OCEC analysis) is not available, the impact of potential variation in the MAC should be assessed and discussed. Where BC is only used in a relative sense, this is less a concern, but where BC 'mass' is compared to e.g. AMS mass – this needs to be done with caution and uncertainties discussed.

P6367, L14-20 – The use of PMF on aerosol that has been heated to such an extent needs more explanation/discussion. The assumption here seems to be that the factors extracted from PMF (e.g. OOA, BBOA) are consistent at ambient temperature and 400 C. It seems questionable that all of the species that contribute to these factors evaporate at exactly the same rate, and also that some of these species wouldn't be modified by such vigorous heating. However, no assumptions are discussed and the only mention of the potential chemical transformation is organic pyrolysis and soot oxidation. Examining the spectra of the residual OA and the factors extracted using only denuded spectra and only ambient spectra is called for. The assumption of 'constant' OA factors has been made and assessed in previous AMS-TD studies (Huffman et al.

2009a; b), it needs to be re-examined here.

P6368, L15: what are detection limits?

P6368, L17-18: this statement calls for a reference to worth that have shown this to be true.

P6369, L1-3: This is an estimate of an error in total mass, but the method discussed here refers to aerosol number. A small amount of mass may correspond to an enormous number fraction of particles, especially in the ultrafine range. Extending this uncertainty estimate to number seems to be required if this justification is applied to develop this method. See also comment above about use of AMS spectral information (and potentially aerosol-time-of-flight) to assess changes in organics with heating. AMS aerosol-TOF data could also be used to assess the fractal dimension of the aerosol under heated/unheated conditions to assess whether aerosol shape changes, which could potentially effect sizing in the SMPS.

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