

Review of: “Determining the link between hygroscopicity and composition for semi-volatile aerosol species” by Alroe et al., submitted to *Atmospheric Measurement Techniques*

This manuscript (amt-2018-17) reports an approach that couples the Aerosol Mass Spectrometer (AMS) to a volatility and hygroscopicity tandem differential mobility analyser (VH-TDMA) setup. This approach allows separation of the semi-volatile and low volatility components and comparison to chemical composition. The main novel advance of this approach over other similar approaches is the incorporation of size dependent aerosol chemical composition from the AMS, which allows investigation of aerosol chemical composition in the size range most relevant to the VH-TDMA experiments and to cloud droplet activation.

The manuscript is well written and within the scope of *Atmospheric Measurement Techniques*. The manuscript may be publishable if the below major comments associated with data quality and the use of the thermodenuder are addressed in revision.

Major Comments:

1. The first major comment relates to data quality and echoes many of the comments from Anonymous Reviewer 1. The authors assert that the main advantage of their approach over previous similar approaches is the incorporation of size dependent aerosol chemical composition measurements. However, the authors also state that, for the chamber measurements, which had a much higher mass concentration than the ambient measurements, “the observed signal was quite unstable...often falling below the detection limit. For this reason, data below the detection limit was not removed as it would have excluded a large proportion of the data” (page 7, lines 13-15). If the signal is so unstable and a meaningful measurement so difficult to obtain, it is unclear how this approach represents an advance over previous versions, which is the key argument of the paper. The robustness of this approach must be discussed in substantial detail in any revision. Included in that discussion must be details concerning the experiments (e.g. whether the AMS measurements in Fig. 3 represent the fraction around 100 nm in the smog chamber and, if not, what the mass concentrations around 100 nm were during that experiment; what time resolution was used in the AMS measurements and how that compares to the time dependent composition changes; what is, and what factors are governing, the AMS limit of detection; what mass concentrations are required for this approach to be viable; etc.).
2. The second major comment relates to the use of the thermodenuder approach. It is known that many chemical components of secondary organic aerosol (e.g. oligomers) can thermally decompose when passed through a thermodenuder at temperatures as low as 100°C (Hall and Johnston, *Aerosol Sci. Technol.*, **2012**, 46, 983-989). This observation may have a significant impact on the interpretation of the VHTDMA measurements, especially since the thermodenuder used in this manuscript is ramped up to 500°C. In the revised manuscript, the authors should include a discussion of the limitations of the thermodenuder approach with respect to separation of semi-volatile and low volatility components against likely changes to aerosol chemical composition resulting from thermal decomposition within the thermodenuder.

Additional Comments:

1. In their revised manuscript, the authors need to better clarify the temperature threshold that separates semi-volatile from low volatility. Is the cut-off at 120°C? This is inferred in the text (page 7, lines 23-25) but is not stated in a clear and direct manner. The authors should more clearly define what is meant (functionally) by SVOC and LVOC.
2. Page 4, line 10: Do the authors mean “If the aerosol chemical composition is strongly size-dependent....”?
3. The authors should ensure all references are accurate. For example, Cerully et al. (2017) and Huldebrandt Ruiz et al. (2015) were both published in *Atmos. Chem. Phys.* but their references indicate *Atmos. Meas. Tech.* as journal in which they were published.