Interactive comment on "Organic aerosol in the summertime Southeastern United States: Components and their link to volatility distribution, oxidation state and hygroscopicity" *by* Evangelia Kostenidou et al.

Anonymous Referee #2

The manuscript 'Organic aerosol in the summertime SEUS: Components and their link to volatility distribution, oxidation and hygroscopicity' by Kostenidou reports on ambient aerosol measurements using a HR-TOF-AMS coupled to a thermodenuder inlet. Total submicron non-refractory OA was split into four main factors: BBOA, Isoprene-OA, MO-OOA and LO-OOA. The observed thermogram profiles of OA and of each of its four factors are shown, along with the modeled volatility distribution of the components of the sum and the four factors. Lastly, an attempt to reconcile hygroscopicity, O:C and volatility is made. The results presented will make an important contribution to the

existing body of knowledge on the composition of OA, specifically, the OA present in a moderately-polluted biogenic-rich region of the SEUS. One potential issue needs to be addressed, and a few other clarifications need to be made for this work to be published.

An interesting observation using this TD-AMS setup from SOAS was that the MO-OOA exhibited a lower MFR subsequent to heating compared to the LO-OOA component of OA. That is, more of the 'more oxygenated' component had evaporated or been destroyed by heating relative to the 'less oxygenated' component. A more detailed explanation for this phenomenon than what is currently provided (essentially referencing the work of Karnezi et al.) is needed. How does the model predict that MO-OOA was composed of less volatile material compared to those of LO-OOA? What observation or information was fed into the Karnezi model, etc.? For instance, MO-OOA and Isoprene-OA appear to exhibit distinct MFR thermogram profiles (figure 2), which the Karnezi model is able to reproduce well. Yet, MO-OOA and Isoprene-OA possess nearly indistinguishable C^* versus mass fraction distributions (figure 3). How is the model able to come up with essentially the same composition for two OA factors that exhibit distinct thermogram profiles? One conclusion the authors draw from this MO vs LO thermogram observation is that MFR alone can be misleading. So I assume the authors relied on the model to be the arbiter. What exactly was the contribution of TD?

Are you able to calculate a C^* versus mass fraction distribution at each thermogram temperature setting? Determining how the C^* of the material that compose a given OA factor evolves as it is incrementally heated would provide an important constraint.

Comparison to previous work on OA measured during SOAS needs to be more focused. A thorough comparison (and explanation of why there seems to be a discrepancy) to the work of Hu et al. is more appropriate here as the two utilized the same approach (TD-AMS). For instance, how do the factor assignment (BBOA, Isoprene-OA, MO-OOA, LO-OOA) compare to that of the Jimenez group? That data is publicly available and comparison to it should be included in the analysis. In comparing against the works of Saha and Lopez-Hilfiker, a discussion of how different techniques can yield

different observations or interpretations is more suitable. For instance, the thermogram profiles obtained from shown by Lopez-Hilfiker et al. show residual IEPOX signal desorbing off the FIGAREO-CIMS inlet well above 100C whereas the TD-AMS saw none above 100C. Suggesting artifact in another method without supporting evidence is not justified.

Why do the abundance distributions predicted for the OA factors (figure 3a-3d) look the way the do? That is, why do those for MO-OOA, LO-OOA and isoprene-OA appear to be bi-modal, whereas that of BBOA is not? Perhaps it is this arbitrary designation into three log base-10 bins that obscures the real distribution? Can the distributions be shown on finer C* scales? What is the lower limit of C* that can be detected with this approach? But basically, what is the source of the low volatility material as opposed to the high volatility material? OH versus O3 versus NO3? Aging? Is there an atmospheric chemistry explanation for the calculated volatility distribution?

The error bars shown in figures 1 and 2 indicate there is little variability in the thermogram profiles throughout the SOAS campaign. Have the authors looked for any dependence of the steepness of the thermogram profiles on time of day, high/low OA loading, RH, temperature, particle acidity, high/low NOx, etc, all of which varied widely through the duration of SOAS?

A strong point was made that the findings of Jimenez et al. (2009) had been contradicted, but no thorough explanation for the possible reason for the discrepancy.

References made in some of the SI figures are not in the SI citation list.