Authors' response to the review of the manuscript titled "Modification, Calibration, and Performance of the Ultra-High Sensitivity Aerosol Spectrometer for Particle Size Distribution and Volatility Measurements During the Atmospheric Tomography (ATom) Airborne Campaign"; submitted to AMT on August, 9, 2017

The authors would like to thank the reviewers of the manuscript for their careful and positive evaluations. Our responses are listed below in blue, while reviewers' comments are in black.

Apart from the minor changes suggested by reviewers we have updated data presented in Fig. 11. We have noticed that an incorrect dlogd values were used. The corresponding volatile and non-volatile fractions mentioned in Section 6.2 were also updated accordingly. These changed by  $\sim$ 1-2 % as compared to the initially reported values. This has not impacted integrated aerosol surface and volume concentration data reported here.

## Reviewer #1 (received and published: 29 September 2017)

This is a very thorough and well-written manuscript describing the evaluation and performance of the UHSAS instruments deployed during the ATom field campaign. The authors have investigated every aspect of the instrument performance and carefully quantified uncertainties. They have provided detailed descriptions of their methods and experimental details. The instruments performed with close agreement under similar operating conditions. The initial results with one thermodenuded instrument suggested a large fraction of the aerosols were secondary in nature. As this field campaign continues, these results should be very enlightening and help to further our understanding and constrain uncertainties regarding the ability of aerosols to act as CCN. My main concern/question is regarding the sensitivity of the instruments to particle refractive index and instrument calibration while operating in the thermodenuded mode. With the exception of sea salt which has a refractive index similar to ammonium sulfate or organic carbon (more volatile fractions), other non-volatile species such as dust or soot have a higher refractive index then was tested as part of the calibration (not to mention effects of complex refractive index). Are the authors concerned about different uncertainties between the thermodenuded and non-thermodenuded instrument when comparing the two measurements? I recommend publication after addressing very minor comments below.

We thank reviewer #2 for positive evaluation.

Because the refractive index of the atmospheric particles is not know a priori, we decided to use ammonium sulfate-based UHSAS sizing calibration and evaluated uncertainties with a reasonable range of scattering aerosols. Ammonium sulfate has a refractive index (m=1.527) that is representative for the remote marine aerosol sampled during ATom. We emphasize in the Section 3.2 that soot is not sampled quantitatively because of incandescence and/or refractive index. For dust we are concerned about both shape and refractive index. Uncertainties in both instruments for such cases must be evaluated on a case-by-case basis using best estimates of the refractive index based on other measurements, coupled with optical simulations of instrument response. We have added a remark to this effect in Section 3.2.

The fact that the two UHSAS instruments are measuring aerosol with different refractive indices is something we haven't considered quantitatively. As the reviewer points out, this is a particular problem above the MBL, where dust is likely the dominant component. Frankly, the counting statistics are sufficiently poor that this error is likely to dominate any uncertainties during comparisons between the two instruments in the free troposphere except in specific dust and dust + biomass burning cases. We have added

a remark cautioning about this sublety in Section 3.2, but believe this will have to be addressed on a caseby-case basis.

Figure 3: Do the solid lines represent fits to the data? Please note in caption.

Yes. Figure caption has been updated.

Page 18, line 25: I assume the "agreement" values correspond to slopes?

Yes. This sentence has been updated and now reads: "The corresponding slopes for aerosol surface and volume concentration varied between 0.97-1.02 and 0.95-1.08, respectively."

Page 19, line 25: Add "number" before "concentration"

Corrected.

Page 20, line 2: I'm not sure "coarse-mode" is typically used for particles less than 1 um?

The sentence reads now: "In the FT, ~3 % of the particle volume was non-volatile, dominated by a few particles with with  $Dp > 0.3 \mu m$  and uncertain due to poor counting statistics."

Page 20, line 7, Figure 11 Caption: typo for "isn" before "the MBL case" and "UT" instead of "FT".

Thanks for finding these subtle errors. We have fixed these two typos.