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 ~1-2 % as compared to the initially reported values. This has not impacted integrated aerosol surface and volume concentration data reported here.

Reviewer #2 (received and published: 13 October 2017)

The manuscript presents a detailed description of modifications to and the subsequent evaluation of two optical particle counters (UHSAS) for use during a multi-year aircraft campaign. It also describes and characterizes a thermal denuder for measuring the non-volatile fraction of aerosol. The subject is appropriate for AMT and the manuscript is well written and clear. I recommend publication once the following minor points have been addressed.

We thank reviewer #2 for positive evaluation.

Section 3.1.1, line 10: Recommend changing "This bias" to be more specific, e.g., "The DMA sizing bias". Also, line 14 I believe the authors mean the biases are propagated to the aerosol and volume concentration uncertainties, correct? I assume no adjustment was made to the DMA diameters based on the PSL offset?

Line 10: The sentence reads now: "This DMA sizing bias is estimated to be about 7 % at sizes below 0.07 µm and decreases to 1% for sizes above 0.13 µm."

Line 14: The sentence reads now: "Still, these potential biases are propagated through to the aerosol surface and volume concentration uncertainties discussed below."

Concerning the latter comment above. That is correct. No adjustment has been made to the DMA diameters based on the PSL offset. The sentence reads now: "No adjustments were made to the DMA diameters, but the potential biases when compared to the PSL sizes are propagated through to the aerosol surface and volume concentration uncertainties discussed below."

Section 3., lines 3-9: The section should also mention that the complex RI of BC will also affect the sizing in addition to LII impacts. I think it is also helpful to the reader to clarify how the incandescence of BC would affect UHSAS sizing (e.g., BC cores heat up and vaporize coatings and affect scattered light signals as the particle moves across the beam, other effects?).

We agree. We have added the following text (page 8):

Line 3: "The refractive index of soil dust may exceed the range of real refractive indices considered here. In addition, dust can be both absorbing and aspherical. When dust is an important component of the atmospheric aerosol, uncertainties in both the denuded and thermodenuded UHSAS instruments should be evaluated on a case-by-case basis using best estimates of refractive index and shape based on other measurements, coupled with optical simulations of instrument response. Also, because the thermodenuded UHSAS instrument volatilizes non-refractory particles, the refractive indices in the aerosol measured by the two instruments will differ. This problem is probably minor in the MBL because sea-salt aerosol has a refractive index within the range of the calibrants. For the free troposphere, however, there may be substantial sizing biases between the two instruments that should be considered case by case using additional information on aerosol composition."

Line 13: "Even without incandescing, the complex refractive index of BC particles (n=2.26-1.26i at λ=1064 nm; Moteki et al. 2010) substantially alters UHSAS sizing compared with the calibration aerosol."

Reference added:

Moteki, N., Y. Kondo, and S. Nakamura, Method to measure refractive indices of small nonspherical particles: Application to black carbon particles, J. Aerosol Sci., 41, 513–521, 2010

Section 3.3: It appears the detection efficiency was only performed for UHSAS-2? Is there a reason for this? Even if results were similar it would be helpful to report diameters for 50% detection efficiency for both instruments for ammonium sulfate.

The detection efficiency was performed for both UHSAS-1 and -2 and results are reported in Table 1 (attached below for convenience). We did not include data for both instruments in Fig. 4 for clarity. We now refer specifically to Table 1 in Section 3.3 and comment that the detection efficiencies differ. The UHSAS-2 instrument is substantially older (serial #7) than the other, and has a lower laser power probably due to older, more contaminated optics.

We have added the following sentence: "Detection efficiencies for both UHSASs are provided in Table 1. The thermodenuded UHSAS begins detecting particles at a larger diameter than the other instrument."

Table 1. Detection efficiency of UHSAS-1 and UHSAS-2

Particle	Real refractive index, n	Wavelength, λ (nm)	Reference	Dp_{50} (nm)	
				UHSAS-1	UHSAS-2
PSL	1.58	780	Yoo et al. (1996)	n/a	n/a
$(NH_4)_2SO_4$	1.527	1054	Hand and Kreidenweis (2002)	$72.8\ ^{+1.2}/_{-5.9}$	$62.8\ ^{+1.0}/_{-5.9}$
DOS	1.44	532	Pettersson et al. (2004)	$75.9\ ^{+1.2}/_{-6.0}$	$68.2\ ^{+1.1}/_{-5.9}$
Limonene oxidation products	unknown	n/a	n/a	$78.9\ ^{+1.3}/_{-6.0}$	$69.7\ ^{+1.1}/_{-5.9}$

Section 4.2, lines 2-4: The particle losses through the TD are only reported down to 150 nm, but the both UHSAS systems measure down to about 70 nm. More information regarding particle losses through the TD should be provided between 70-150 nm. Also, will pressure in the TD affect the losses? I assume this varied over a similar range as described for the UHSAS? I do not think more experiments are needed, but some brief discussion of potential impacts would be useful.

We do not currently understand the loss mechanism in the TD. The loss is larger than we expect for flow through the unheated TD. Based on the particle size and residence time in the TD, diffusion to the walls is one possible loss mechanism and may be larger than expected due to the activated-carbon fabric in the TD. Another possible loss mechanism would be if a portion of the flow in the TD occurred in activated-carbon fabric region and the fabic acted as a filter leading to losses in the unheated TD. Further laboratory measurements are planned to understand these losses.

However, for this manuscript, without a clear mechanism for the losses, we cannot extrapolate to smaller size and lower pressures and can only report our measurements at 150 nm and 835 hPa. We have added a sentence to manuscript stating that the particle loss mechanisms in the TD are not clear: "The mechanism and size-dependence of this particle loss is currently unclear and requires further investigation."

Figure 6: What are the grey, circular "cloud" shapes on each side of the heated section at the top of the figure?

We are not sure what the reference is to. This is what we see on our screen and printouts.

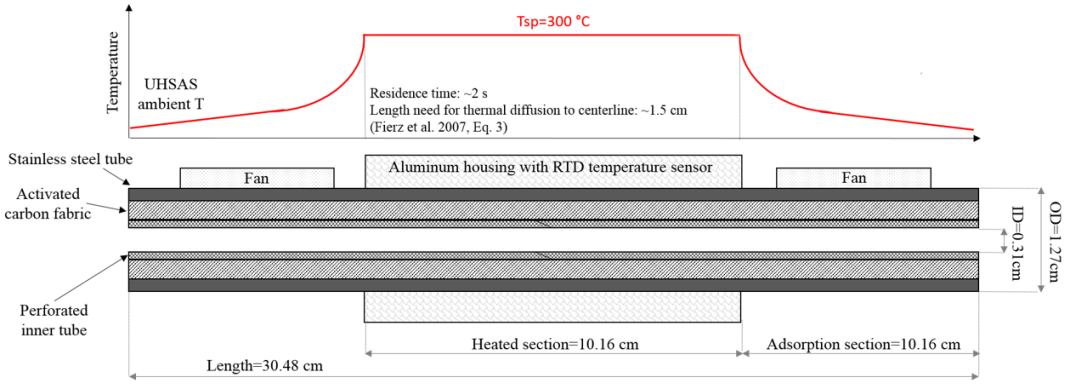

Fig.6. Schematic cross-section of the thermodenuder and conceptual temperature profile. Temperature is measured at a single point with a platinum RTD sensor inside the aluminium housing around the heated section. The thermal diffusion length estimate assumes standard pressure and temperature and typical flow in thermodenuder, small perturbations in temperature, and is use only for qualitative understanding of heat flow in the thermodenuder.

Section 5.3: Are sample flow rates changed during flights? A minor point, but it would be interesting to know if UHSAS response is affected by sample flow rate within a reasonable range. Assuming the residence time in the denuder could be maintained the count rate could be increased in the FT to improve the statistics and reduce uncertainties.

The sample flow rate changes in flight. As described in section 3.6, the sample flow rate drops when the UHSAS operates at lowest pressures. This effect does not affect UHSAS sizing characteristics (Fig.S1).

We agree that maintaining the flow rate constant in the free troposphere would improve the counting statistics, and reduce uncertainties.

Section 6.1, line 17: I assume the reason to only compare 100-900 nm and not the full UHSAS size range is to avoid slight variations in detection efficiency and saturation? I am curious how well the instruments compare over the full range given by the manufacturer.

Yes, we wanted to compare over a range where both instruments measure with 100% efficiency. As indicated in Table 1, the UHSAS-1 (thermodenuded instrument) is not as sensitive as the newer unit, and 100 nm is safely above the roll-off in detection efficiency to ensure a fair comparison. The diameter of 900 nm has been chosen as the upper limit of the UHSAS sizing calibration curve based on measured ammonium sulfate particles (Fig.3). Above 900 nm the calibration is based on fitted parameters rather than the actual measured calibration particles.