

Interactive comment on “The Importance of Size Ranges in Aerosol Instrument Intercomparisons: A Case Study for the ATom Mission” by Hongyu Guo et al.

Anonymous Referee #2

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The paper from Guo et al. reports on the importance of the size ranges of both inlets and instruments in aerosol intercomparison with a special focus on airborne measurements during ATom Mission. Such a systematic analysis is essential for better understanding the difference that can be observed when comparing different instruments or inlets to each other, especially during aircraft measurements where altitude can strongly affect the size range of the aerosol inlet and/or instruments. This work is, therefore, suitable for a publication to AMT. However, several issues first must be clarified.

Major comments:

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- First of all, the authors must reexamine what is the real focus of the manuscript. Although the title mentions “size range in aerosol instrument intercomparison”, abstract and introduction only refer to AMS issues, while only a few words are made on the other inlets and instruments on the conclusion. This does not reflect both manuscript and supplementary information, which are deeply discussing the effect of altitude on the size-range of different inlets and instruments without directly comparing them to AMS results. Consequently, if the manuscript aimed to investigate the effect of the AMS aerodynamic lenses transmission efficiency during intercomparison exercises during ATom Missions, discussions on MOUDI-PM1, URG-PM1 inlets, SAGA and PALM-AMP are not needed on the manuscript since the AMS was not connected to such inlet or no direct compared to them. On the other hand, if the target of the authors was to discuss how the size-range of inlets and individual instruments are affected by altitude during the ATom Missions, then the authors may need to reconsider the discussion on the AMS and the comparison V_{chem} vs. V_{phys} or $V_{phys-TC}$.

- According to the title, I would expect more direct intercomparison between the different results providing by the instruments, similarly than for the AMS (V_{chem} vs $V_{phys-TC}$) rather than simply comparing numbers or transmission profiles. For example, why the authors did not compare their AMS results with SAGA, and PALMS-AMP? how the change in transmission efficiency of each instrument affect the comparison?

- Only very sparse information is done on the different methods used to estimate the change of the size-range of the instrument with the altitude. It would be great to have, for example, a dedicated section on it detailing the different hypotheses and models used for the calculation.

- Focusing on the AMS, the authors did not convince me of the importance of transmission efficiency. Although the focus of the manuscript (based on abstract and conclusion) is to demonstrate the importance of the transmission efficiency of the aerodynamic lenses of the AMS during intercomparison, it is first mandatory to ensure, before any comparison that all the reference instruments are properly calibrated. Here,

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the calibration and quality assurance of the different optical size spectrometers are not mentioned. How do the 3 instruments compare on their overlapping size-range? The manuscript will certainly gain consistency if the authors can also discuss these points as well as their uncertainties rather than only citing literature (lines 301-307) similarly than for the AMS. In such an exercise, not only the AMS had to be carefully calibrated.

- Another critical aspect is the absence of discussion when comparing correlations with and without applying the size-dependent correction method. Figure S4 shows a very good correlation between the instruments without applying it. The discussion on the effect or not of the size-dependent correction should be supported by quantitative numbers as well to demonstrate that transmission efficiency of the AMS plays an important role (which was up to now never considered on the literature) and properly quantify how much V_{chem} can be affected by the transmission efficiency of the AMS lenses. The reason is certainly on the definition of the V_{phys} itself. Why V_{phys} has a volume concentration ranging up to $4.8 \mu\text{m}$? This is quite surprising since there is a long discussion on the change of the size-range of the AMS between the missions (section 3.1 and 3.2). Moreover, the large fraction of super-micrometer particles on the volume size distribution is controlling most of the total volume mass concentration on V_{phys} . Why the authors did not cut V_{phys} to $1 \mu\text{m}$ for all missions? Or more simply using $V_{phys-0.75}$ or $V_{phys-0.95}$, corresponding to their own estimated AMS upper-size-cutting (PM0.75 for ATom 2 and PM0.95 for ATom2, for example). It would also be possible to consider a volume size distribution up to 800 nm (equivalent diameter similarly to most of the SMPS system). Applying such kind of approach provides i-/ a comparable size-range between the 2 instruments and ii-/ the possibility to extrapolate the results to most of the AMS work that was done up to now, which are mostly using SMPS for comparison. Only then, the authors can properly discuss the importance of considering or not the aerodynamic lens transmission efficiency on the comparison with collocated measurements, and how much it can influence the correlation. It would be also interesting to do a sensitivity test based on the influence of the coarse mode concentration (low and high coarse mode periods). For this purpose, Table S2 is certainly one of the most

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important for the comparison between the different instruments. However, I don't fully understand why only $V_{phys-TC}$ is used and not a systematic V_{chem} and $V_{phys-TC}$ on a similar size range to quantify the effect of the AMS transmission efficiency correction on different systems.

- Does it make sense to apply the transmission curve on V_{phys} ? This approach makes V_{chem} and V_{phys} dependent variables. Furthermore, assuming the size-spectrometers have perfect transmission efficiency and particle detection up to $4.8 \mu\text{m}$, it is not completely surprising that after applying the AMS transmission efficiency correction, a perfect correlation is obtained. Finally, it does not say that the two instruments better correlate than without applying the correction. Instead, it would be better to correct the AMS mass or volume concentrations by the corresponding transmission efficiency. This means first ensure that ePTOF and MS mass concentrations agree, then correct the ePTOF mass concentration from transmission efficiency to get the final AMS mass concentration. Then, it would be possible to compare to parallel measurements of the same size cutting. However, I have to recognize that I am not sure whether this is feasible on this dataset or not.

- Although, the authors carefully investigate the transmission efficiency of their instrument; key information on the calibration protocol itself is not mentioned. For example, how the particles were generated and selected (especially for mono-dispersed super- μm ammonium nitrate particles)? Which instrument was used as a reference in parallel to the AMS? The authors used 400 nm (mobility) ammonium nitrate particles to performed their IE calibrations, however, looking at Figure 2, this diameter seems to be already out of the 100 % transmission efficiency of their aerodynamic lenses having rather a transmission ranging from 80-95%. How did the authors deal with that? Does it mean that their RIE (and the resulting AMS mass concentration) is underestimated?

- Last but not least, the reported discussion regarding PALMS and PALMS-AMP analysis must be clarified and solved on the revised version of the manuscript.

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Minor comments:

Abstract:

- The abstract is rather unspecific and gave poor information on the “critical evaluation of the size-related factors”, the “good agreement” when comparing AMS with “size spectrometers” and “standard PM1 volume”. Without any indication of the transmission efficiency, the size range of the reference used, and results before and after applying the correction, how can we conclude from an “absence of significant unknown biases”? More details on the “upper end of the AMS size-dependent transmission” as well as the upper size cutting of the size-spectrometer must be provided here.

- Why results from the other instruments (SAGA, PALM) and inlets (MODI-PM1 and URG-PM1) are not mentioned in the abstract?

- line 28: What is a good agreement? Please provide number

- line 29: Please precise from which size-spectrometer (and its size-range) were used.

- line 31: What is a “standard PM1 volume”?

Introduction: Over the last two decades, there is a lot of works reporting comparison between AMS and collocated instruments either for individual chemical species and/or total mass/volume. Most of the time the correlations are within the different instrumental uncertainties. Therefore, the authors should provide more clear justifications for their motivations and the need for such correction on the introduction.

- line 53: ACTRIS and not ACTRIS

- line 65: Could you clarify the sentence “Volume comparisons probe the ability of the AMS to quantify total mass and predict aerosol density based on fractional composition accurately, and hence it the most germane comparison for total quantification”

- line 73-75: Does it make sense to repeat this information. This was already mentioned in the introduction.

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- line 112: Details on the modifications made on the AMS compared to a “standard” one would be appreciated.

- line 112 – 284: I think a detailed description of the AMS is not needed. AMS is not a new instrument and it was already described in the cited papers. Moreover, I don't follow how section 2.3 is splitting. For example, the authors first discuss the factor influencing the collection efficiency (line 125-145) and then with the impact of the RH and the CDCE (line 194-199). A similar comment can be made for the IE and RIE. The current splitting leads to unnecessary repetitions. For example line 143-144: “CE is estimated to contribute substantially to the overall uncertainty of the AMS concentration measurements (Bahreini et al., 200), line 169-170 “For the AMS reported mass concentration, uncertainties (i.e. accuracies) in CE (30%)”, and line 211 “[...] are similar to those from Bahreini et al. (2009), due to the dominate uncertainties contribution from CE (30%)”. Similarly, the uncertainties of the RIE is discussed on line 157-172 and again online 204-211. A single AMS section will improve it.

- line 269: density calculation (eq. 5): why not directly include sea-salt since i-/ you quantified it on your AMS measurements (line 256) and ii-/ you include it anyway later (line 382-386). A similar comment can be made for rBC. Then you can discuss once how total mass concentration and density were estimated. Here again, it will improve the clarity of the manuscript.

- line 272: Density of OA was calculated based on the Elemental Analysis of the organic mass spectra ranging from 1.51 – 1.59 g cm⁻³ (line 280) but the default OA density is set to 1.7 g cm⁻³. Does the default value make sense? Could it be that the value of 1.7 is artificially high due to low concentration and overall strong contribution of CO₂+ to the total mass spectra?

- line 301: Vphys is a combination of 3 optical particle size-spectrometer ranging from 2.7 nm to 4.8 μm. How these 3 instruments are comparing together on their overlapping size range?

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- line 358: Do you need to include the SAGA on the manuscript since it is no use for the comparison with the AMS?
- line 441: Is this section necessary, since none of the instruments used for the discussion were connected to such inlets?
- line 515: the two following sentences are unclear and need to be rephrased “AMS observes a fraction of V_{phys} (Figs. 1 & 3). AMS transmission vs. dp was calculated based on the calibrated transmission vs. dva (Eq. 1) and the AMS estimated time-dependent pm (Eq. 5), and applied to the AMP size distributions used to derive V_{phys} .”
- line 520: How is the correlation without corrected V_{phys} by AMS transmission?
- line 523: “slightly worse”? Could we say that this correlation is slightly worse according to all instrumental uncertainties? Both are within 10%. Instrumental uncertainties on the comparison should be discussed. Which type of regression was used (classical linear fit or least orthogonal distance fit)?
- Figure 1: Please use a log-scale for $dN/dlogd$ to better see the particle number > 100 nm.
- Figure 4: Can the strong increase of V_{phys} compare to V_{chem} at the lowest altitude simply be associated with marine sea-salt coarse mode?
- Figure S4: It is unclear if the UHSAS volume was corrected or not from the transmission efficiency of the AMS. If not, the scatter plot should certainly provide a similar slope than the ones obtained for V_{chem} vs $V_{phys-TC}$. Why converting UHSAS in mass concentration while all the discussion is made in volume? How is the size distribution after applying transmission efficiency correction?

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