egusphere 2024-3969

Reply to Referee

Reviewer comments in black

Our replies in red

Changes to the text in blue

My previous review of this manuscript argued that while the technical improvements of the CARIBIC-AMS described therein are very impressive, there was not enough detail provided to actually show that this instrument has promise as a quantitative, autonomous PM sensor in the UT/LS. The revised manuscript has addressed adequately most of these shortcomings but still falls a bit short on the overall quantification front. The authors make a good case that in a lot of ways this is still a case of "COVID-19 hangover", complicated by the general challenges of the CARIBIC platform. Given the overall consistency of the data presented, I am reasonably convinced that the instrument got UT/LS PM right when it was working within a factor of 3 or so if one accounts for the CE, transmission and other uncertainties, which for what the paper aims to show is sufficient. Hence, I recommend publication once a few last comments listed below are addressed. Hopefully after some additional characterizations the next paper will show significant improvements on these fronts, so that the CARIBIC-AMS can truly be a benchmark for CTMs for years to come.

Thank you for this positive rating.

These comments listed below include some responses/clarifications to previously raised points in the discussion which are not germane to the actual review of the revised text, so for simplicity I am going to highlight the actual action items in *italics*:

- *While in the main text it is made clear that the DLs are typical averages under flight conditions, this is not made explicit in the abstract. Also, neither the text nor the abstract make clear that these are CE=1 DLs (I would assume), and that they might change for different ambient conditions. I think both of these points should be added since there is in general a lot of confusion on AMS DLs in general in our community.*

We now state in the abstract that the stated detection limits are typical averages under flight conditions. However, it has to be emphasized that the values refer to CE = 0.5, because they were inferred from in-flight data. We clarified this in the abstract, in section 3.3 and in Table 1.

- I would maintain that instrument blanks are the most accurate way to determine/validate detection limits, provided they are taken under realistic conditions. A 2 hour long blank, as described in Drewnick et al (2009), is certainly not realistic, since as shown in that paper the ionizer basically cleans out after 30 min or so, but that does only partially apply to short blanks like the ones presented by the authors. *So I do think that the figure presented in the response showing agreement between the background variability method and the blanks is valuable, and I would ask the authors to consider including it in the paper.*

We included the Figure as Appendix B, and added some explanation in section 3.3:

Detection limits can also be calculated from the automated blank filters as 3 × σ_{Filter} (σ: standard deviation of the calculated mass concentration during the blank measurement; e.g., Drewnick et al., 2009). Figure C1 shows a comparison of both methods for flight 578. The automated blanks were taken every hour for 10 minutes during this flight. In the beginning of the flight, the background signal method yields higher detection limits than the blank filter method, but after about three hours, both methods agree fairly well.

Also, to clarify, my previous request for "stratospheric" data was a typo, I just meant high altitude data, I apologize for the sloppy wording.

- *I also think the DL discussion would read better if "noise of the background" would be replaced with "short-term variability of the background". While for high m/z it is indeed electronic/counting noise, this is e.g. certainly not true for ions such as SO^+ or CO_2^+, which have a high, highly variable, but not necessarily noisy background.*

We changed to „short-term variability" as suggested

- *Regarding the presentation of the instrument operation, I want to clarify that what I specifically meant in my previous review was a block diagram showing the control boxes/computers and the connected powerboxes. And then ideally (in a different color/style) which piece of software runs on which computer and what the call hierarchy is. I do think this would help the presentation, which is hard to follow at the moment, especially for e.g. a second year student. Such a diagram would also highlight how certain design choices do improve the overall reliability/fail-safe behaviour of the system, which is a bonus for a paper like this.*

We added such a block diagram as Appendix A and refer to it in section 2.2.3.:

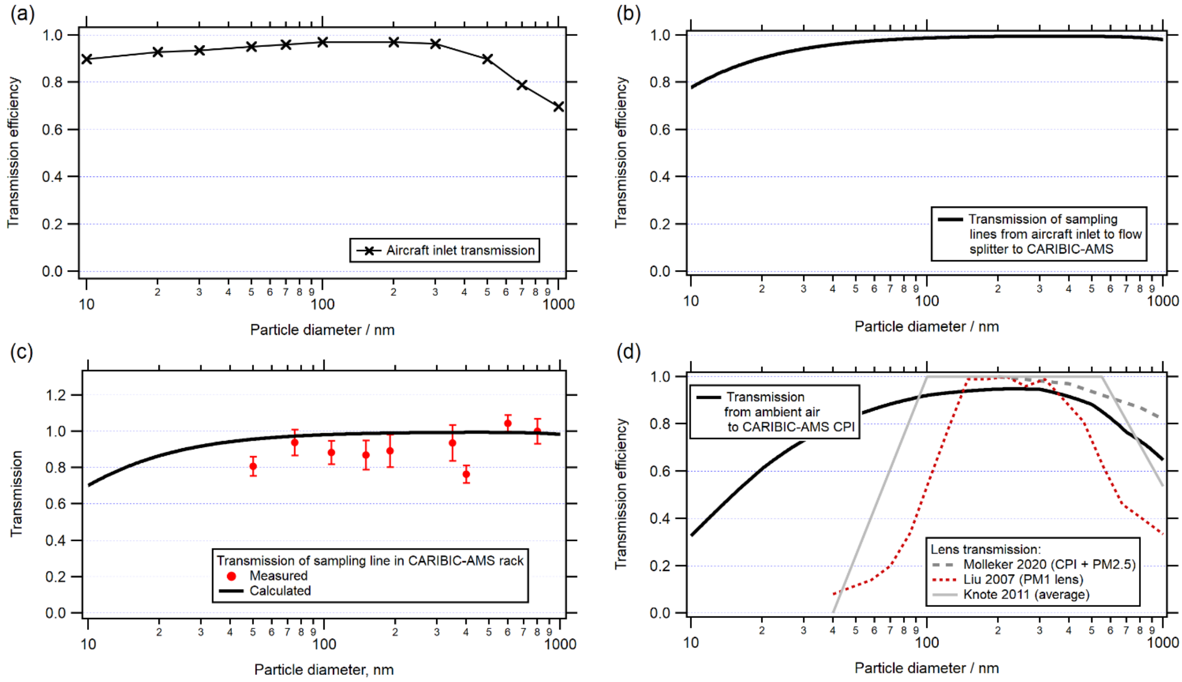
A block diagram of the hardware and software components controlling the CARIBIC-AMS operation is shown in Figure A1.

- Regarding ePToF, a couple of clarifications:
- The time offset for ePToF and PToF is slightly different (see Williams et al, 2016)). Given the different triggering electronics used by the authors it might not be consistent with the Aerodyne findings, but this could be mentioned.
- While the ePToF dutycycle is indeed higher than PToF, both the spread of the signal over 25-50 bins and the noise in the inversion at low signal levels does result in quite high DLs vs MS mode, so I would not be surprised if regular ePToF operations is ultimately not implemented. Best of luck in any case.

Thank you!

- The inclusion of the inlet loss calculations is very much appreciated, although as the authors state themselves the main uncertainty here is in the exact shape of the AMS lens transmission. Which could be included in Figure 4d for reference (potentially along with some others, e.g. the Molleker 2020 curve in case a different AMS inlet was flown).

We included In Fig. 4d the lens transmission curves from Liu et al (2007) and Knote et a (2011), along with the Molleker et al (2020) curve of the CPI + PM2.5 lens, for 250 hPa (UTLS conditions).



We added the following sentences to the main text:
Also shown in Figure 4 (d) are three transmission functions of different ADL used in the AMS: The PM1 lens without CPI after Liu et al. (2007) and Knote et al. (2011), and the measurements at 250 hPa (UTLS conditions) by Molleker et al. (2020) using a CPI and a PM2.5 lens. The overall transmission from outside to the CPI of the CARIBIC-AMS is above 80% in a size range between 40 and 700 nm. This corresponds well to the size ranges of the AMS inlet systems shown in Figure 4 (d).

- I do understand that in-field calibrations are not an option given the CARIBIC way of doing things. *What I was asking for (possibly in a very confusing way) in my previous comments is for the in-between deployment calibration data. It does sound that instrument optimization efforts resulted in very minimal coverage on that front. Still, if there were e.g. calibrations available every 6 months over the 2018-2020 period, that would I think still be more illuminating than the single point in time comparison provided.*

We added a table (Table B1, Appendix B) with IE calibrations performed during the IAGOS-CARIBIC operation phase of the CARIBIC-AMS, along with later calibrations done in the laboratory and after the TPEx campaign in 2024:

Date	IE (ions molecule^{-1})	AB (s^{-1})	IE/AB (ions (molecule s)$^{-1}$)
June 2019	8.44e-8	1.88e6	5.08e-14
August 2019	1.07e-7	1.93e6	5.38e-14
October 2019	1.73e-7	2.06e6	8.39e-14
November 2019	2.01e-7	1.94e6	1.04e-13
November 2020	4.40e-8	1.76e6	2.50e-14
July/Aug 2024	6.23e-8	4.09e5	1.52e-13

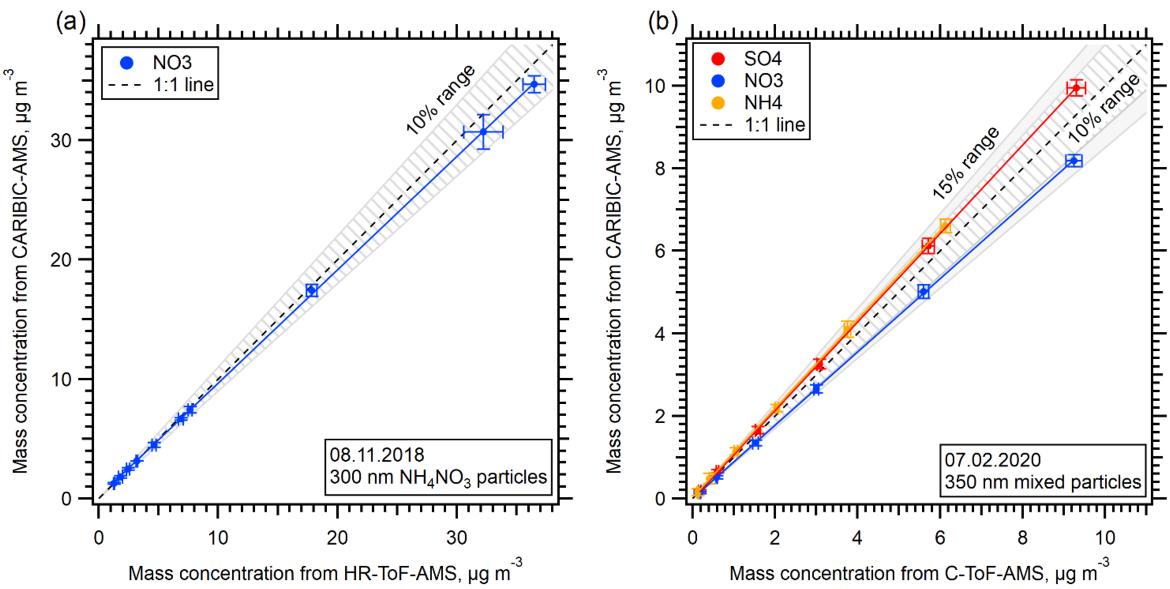
- Regarding the SI discussion, I think we mostly agree and I have no issues with the current text. What is worth pointing out, however, is that the "rapidly changing airbeam over the first hour of flight" most likely has indeed nothing to do with any inherent sensitivity change (hence the lack of change in the SI) but is just CO^+ signal from the high LVOC background in the instrument (if my assessment is correct, the same trend will be observed in the *CO_2^+* background signal). If this checks out it could be mentioned.

Yes, your assessment is correct. The CO_2^+ background also decreases, thus we agree, the (apparent) airbeam decrease is due to the LVOC background. We clarified that in section 232:

This is due to a background of low volatile organic compounds in the vacuum chamber which slowly evaporate after a fresh pump-down and produce ions such as CO^+ (on m/z 28, same as the N_2^+ signal) and CO_2^+.

- Regarding the uncertainty of the nitrate comparison in Fig 7a, I don't think I agree with the 35% uncertainty. If this was ambient nitrate (with the usual uncertainties in terms of mixing state, CE and organic/inorganic contributions) that would certainly be the appropriate uncertainty. But here the authors are putting pure AN particles (their analytical standard) into their respective instruments. *So the uncertainty should really come down to the stability of the AMS and the CPC, and not include either RIE or CE, just the overall IE uncertainty. Per Bahreini, that is 10% (explicitly stated in the SI) and should be used here.*

We agree and corrected this in Figure 7 and in the text.



- Regarding the discussion about Collection efficiency (CE), the terms is typically used with two different meanings:

- Operationally, as in CE = Vchem/Vphys for a given particle sizer. This definition also then includes any transmission/inlet issues and does NOT allow for significant errors on the particle sizer side, since it takes it as ground truth.
- Physically, as a defined, rigoruous vaporizer particle bounce correction independent of the agreement with any other instruments. That is what e.g. the Middlebrook et al, 2012 parametrization addresses.

The authors are clearly talking about the operational definition, which for the purposes of proving adequate quantification severely limits which comparisons can be used (only chemical sensors, basically). *Now, based on the UT acidity measurements reported by Nault et al (2021), the UT is almost as acidic (pH<0) as the lower stratosphere. So it would be very surprising if the CE (in the "particle bounce correction" sense) would be anything but 1, not 0.5 for any of the ambient data presented in this paper.*

Yes, we refer to the operational definition of CE here. We clarified that in the main text in section 3.1:

For the data evaluation, we used an operationally defined constant collection efficiency (CE) of 0.5.

Currently, we have no other available comparison measurement than the aerosol size distribution. We agree that under the high acidity conditions in the UTLS, a CE higher than 0.5 would be expected. For future, more detailed data evaluation we will use the composition dependent CE after Middlebrook et al., 2012, although this may suffer from the high DL for NH4.

Looking at the other plots in Joppe et al (2025), it does appear that using CE=1 would in general improve the median agreement (as expected) while worsening it in the larger plumes (which is likely related to transmission losses/and or mismatch with the UHSAS size range). Given the challenges, this is a nice finding and supports the overall quality of data of the prototype.

- *One item where a clarification would be appreciated is this sentence in the conclusions: "The time resolution of 30 seconds allows for detection of small-scale spatial and temporal structures on the order of 500 m.". Is this meant to be in the vertical, e.g. while climbing? If so please clarify, because at Mach 0.82 or so (typical cruise speed of a jet airliner), 30 s is about 7 km in the horizontal.*

Thank you for spotting this mistake. The 500 m were from another CARIBIC instrument that has 2 sec time resolution. We changed it to 7000 m and rephrased to „detection of spatial structures on the order of 7000 m".

Additionally, we added a newly available refecence for the TPEx campaign (Bozem et al., 2025) and updated the TPEx data reference (Lachnitt, 2025).

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