Author responses to reviews of amt-2021-320: "Aircraft-engine particulate matter emissions from conventional and sustainable aviation fuel combustion: comparison of measurement techniques for mass, number, and size" by J. C. Corbin et al.

2. RC1

General review:

This manuscript compares different measuring techniques for aircraft gas-turbine-emitted nvPM mass number and size, and total number and size from exhaust sampled at 43m of a V2527-A5 and a CFM56- 2C1 aircraft engine burning a range of sustainable and conventional aviation fuels as part of the ECLIF 2 test campaign. The manuscript is well written, and the data presented is novel and relevant to the scientific community. The data processing is of good quality; however, I found that the interpretation of the consistency between nvPM number and mass emission indices was not entirely addressed and sometimes misleading, particularly for mass and size. Please note that I wasn't able to access the supplementary information and table S1 which may answer some of my comments.

We thank the reviewer for their time and comments.

According to an emailed communication with AMT, our references to the "Supplement" should have been references to a "Data Availability" section, and the reviewer was emailed this information as well.

Major comments:

- Loss correction:

o Penetration function 1 measurement: Would you be able to add the size distribution characteristics of the nebulised ammonium sulfate (it could only be in the SI)? Was it representative and the size distributions you typically measured during the test campaign (i.e., GMD ~20-40 nm)?

We added statements of the GMD and GSD to the text:

The ratio of the NASA to NRC PSDs (GMD 30 nm, GSD 1.7) then provided a first estimate of the penetration function.

We note that it is not essential that the GMD be similar to those measured during the campaign, since the penetration function is size-resolved. It is only essential that sufficient number counts are measured in each bin to obtain reasonable uncertainties.

o Penetration function 2: I am unsure of what you mean here. Do you mean you used the UTRC model to predict size-dependent losses in this section (as can be seen in Figure 4) or did you use the full N/M method that outputs a correction factor for nvPM number and mass? If you only used the UTRC model, which particle size distribution did you use? If you used the N/M method, how did you correct for losses to the DMS- 500? The loss correction methodology you used would affect the interpretation of your results, and therefore it should be clearly explained.

The full paragraph starts with Particles may be lost to the walls of sampling lines or to deposits on those walls. The fraction of particles penetrating a given system varies with size, according to a characteristic penetration function. Four penetration functions were applied in this study ... (Figure 4), to clarify that we refer to size-dependent functions as seen in Figure 4. We clarified the subsequent sentence by changing

Function 2 was calculated using the standard loss calculation methodologies provided in SAE documents AIR6504 (SAE, 2017) and ARP6481 (SAE, 2019).

To

Function 2 was calculated using the standard equations for line penetration, as detailed in the loss calculation methodologies provided in SAE documents AIR6504 (SAE, 2017) and ARP6481 (SAE, 2019).

Here the text specifies that the calculations produced Function 2 as shown in Figure 4, which is size-resolved. The following new paragraph was added to state this explicitly:

All reported data are corrected for these penetration functions. Size-resolved data (SMPS) were corrected using the size-resolved penetration functions shown in Figure 4. Size-integrated data (all other instruments) were corrected by weighting the penetration functions by the corresponding measured SMPS PVDs. The correction factors are given in the Data Availability section.

- Figure 5:

o 40% N1 with REF4 graph: It appears the SMPS+TD also measures part of a peak < 10 nm, which appears to be volatile given the SMPS+CS doesn't see it. Does that mean the TD is not 100% efficient at removing volatiles? This should be discussed. Also in line 49, you discuss that an nvPM mode < 10 nm was observed with the CFM56 engine. Can you confirm that it was a nvPM mode and not a volatile mode?

Our data do not allow us to identify whether these small particles were non-volatile or represent an imperfect performance of the CS and TD.

We added the above statement after the description of the <10nm mode. We kept this brief to avoid speculation.

o On multiple PSDs, the start of a large size mode can be observed (~200 nm) which could indicate you were measuring shed particles (unless it came from the engine) or oil. Were you performing regular cleanliness checks? Did you use in-line cyclones to all your analysers? If this was shedding, it could significantly

impact nvPM mass measurements < 10 ug/m3 and would affect EIm estimation from integrated particle size measurement. This is not discussed anywhere in the manuscript; hence discussion regarding this should be added.

There is some evidence for an increase in SMPS-calculated volume at larger particle sizes in Figure 5a, at both 40% and 60% N1. If these large particles indicated the presence of a large aerosol mode which varied independently from the primary mode (e.g. if they were emitted by some other process than the engine itself), they would introduce a EI_m-dependent bias in the ratio of SMPS-based EI_m to other instruments, which was not observed (Section 4.4.2).

We added the text above to the Results. We did not mention shedding explicitly as we feel that a mention requires a citation of a study proving its importance. Shedding is extremely unlikely in our study; our main sampling line was brand new and was baked prior to analysis. There was no evidence of shedding in zero and background air measurements. Also, the large particle mode in Figure 5 represents volatile particles, and is very likely related to oil. Our AMS data indicated the presence of oil-related mass fragments. However, the AMS data are out of scope of the present study.

Additional arguments can be put forward as follows, that we feel are excessive for the manuscript:

Figure 2 shows that the PM mass and number concentrations were close to zero (number is off-scale due to the log scale) for background conditions. Filtered-inlet conditions would therefore be even lower. There is no evidence of shedding here.

Figure 9 shows that there is no change in the ratio of EIm/mean-EIm at lower EIm. Therefore, the bias between SMPS and mass-based instruments was not a function of EIm. If shedding contributed to line concentrations, then its contribution would be larger at lower EIm, and the SMPS would be biased lower at lower EIm (because it would not see all of the shed particles).

- Particle size statistics; GMD and GSD: It is not clear whether you compared the measured particle size distributions or if they were corrected for particle loss to a common sampling point (plenum or probe) using a bin-by-bin approach with measured PSD and the penetration functions (or another method?). Please clarify this in the main text and in the figure titles.

We added a paragraph to Methods:

All reported data are corrected for these penetration functions. Size-resolved data (SMPS) were corrected using the size-resolved penetration functions shown in Figure 4. Size-integrated data (all other instruments) were corrected by weighting the penetration functions by the corresponding measured SMPS PVDs. The correction factors are given in the Data Availability section.

And a comment in Results:

Figure 5 shows selected PSDs [...] The PSDs are corrected for line penetration as described above

And modified Figure 4's caption:

These functions have been used to correct all other presented data.

- Section 4.3: Consistency between number-based emission indices of nvPM and vPM

o APC Vs SMPS number: Have you considered that both SMPSs were just under-reporting due to the large corrections performed within the SMPS software (losses in the DMA, the poor charging efficiency of the bipolar charger, non-linearity of the CPC response)? When were the SMPSs last serviced and calibrated? Were the

SMPSs compared with the APC on the same source prior to the test campaign? As currently written, this section implies that SMPSs are more precise at measuring nvPM number than the standard regulatory compliant APC. I don't agree with your conclusion that the APC was likely overcorrected only because you found the two SMPSs to agree with each-other. What if there was two APC in agreement and only one SMPS?

Here, the Reviewer has helpfully included detailed questions to justify their valid criticism. However, we believe that this criticism results from a miscommunication and not a difference of scientific interpretation. We did not intend to imply that we believed the SMPSs to be more reliable than the APC, nor to use the SMPSs as reference to diagnose problems with the APC.

The old paragraph is:

> Figure 7c compares the measured vPM and nvPM EI_{num} with the mean nvPM EI_{num} (i.e., mean of the NRC CS-SMPS, NASA TD-SMPS, and NARS APC. The grey shading shows that all instruments agreed to within a factor of 2. The APC and DMS500 nvPM EI_{num} were both typically higher than the two similar SMPSs. Substantial variability between the two SMPSs was also observed.
>
> In Figure 7c, the penetration-corrected APC EI_{num} are approximately 50% larger than the SMPS EI_{num} under all conditions. Our measured PSDs rule out the possibility that 50% of particles were not seen by the SMPS. Therefore, we attribute the difference between APC and SMPS results to uncertainties in the APC or SMPS penetration correction functions (Figure 4), i.e., we hypothesize that this difference would not have been observed had the instruments all sampled from the same plenum from comparable sampling lines.

The rewritten paragraph is:

> In Figure 7c, the penetration-corrected APC EI_{num} are approximately 50% larger than the SMPS EI_{num} under all conditions. Our measured PSDs rule out

the possibility that 50% of particles were not seen by the SMPS. Therefore, we attribute the difference between APC and SMPS results to uncertainties in the APC or SMPS penetration correction functions (Figure 4), i.e., we hypothesize that this difference would not have been observed had the instruments all sampled from the same plenum from comparable sampling lines.

We have not specifically addressed the Reviewer's technical comments about SMPS uncertainty because our revisions to the manuscript already cover these details.

Running all counting instruments on the same source, with equivalent lines, would have be an excellent experiment to perform. However, due to the practical limitations of working from separate containers at a field site with limited access, we were not able to perform this experiment.

Note that the only other relevant text in the manuscript is in the abstract and also does not imply a preference for the SMPS data:

[abstract] The commercial instruments used included TSI SMPSs, a Cambustion DMS500, and an AVL APC, and the data also fell within approximately 50 % of their geometric mean.

o L560: The DMS-500 was measuring unstripped aerosol and therefore could be picking up volatiles in comparison with the other nvPM EInum analysers, which could also explain why it was reporting higher values.

We agree and had made this statement. Now, we have further clarified (see previous point of response).

o Were all the size/number analysers within the recommended 12 months service and calibration period? If not, that could explain some of the disparities observed between the different size analysers. For example, the DMS-500 is calibrated for number and size to a traceable standard but drifts over time and Cambustion only

certifies measurement precision of 10% for size and 20% for number within 12-months. I suggest you add discussion on the calibration uncertainty associated with all analysers.

Please see two responses above. All instruments for used for measurement of particulates in this manuscript have larger uncertainties than experienced with measuring gas phase properties, for instance. It is not unusual for 20% uncertainty with these instruments. This is well known in aerosol science (Kulkarni, Willeke, and Baron, 2011), and adding a discussion on this topic to this manuscript would not represent a contribution to the literature.

Kulkarni, P., Baron, P. A., & Willeke, K. (2011). Aerosol measurement: principles, techniques, and applications. John Wiley & Sons.

o Figure 7: It is biased that you only used the SMPSs to calculate the mean and then you compared that mean to the DMS GMD and GSD given it wasn't included in the mean calculation. Why didn't you apply the same methodology as for the nvPM mass analyser, calculating the mean using all the different analysers?

This is a misunderstanding. We used all available data. Figure 7c's caption states

"In panel (a) and (b), mean is defined from the CS-SMPS (NRC) and TD-SMPS (NASA) data. In panel (c), the mean additionally includes the APC (NARS) data"

- Section 4.4: Consistency between mass-based emission indices

o Scatter < 100 mg/kg fuel: Are you sure this reflects the noise levels of the instruments? Analysers like the LII and MSS are, to my knowledge, capable of

precisely measuring down to 1 ug/m3. Is the difference between the LIIs bias or scatter? Did you consider shedding from your system could impact nvPM mass measurement as mentioned in a comment above or that potentially inaccurate calibration caused this difference (see comment below)? Please discuss this in the manuscript.

The reviewer's theoretical noise level for LII and MSS is correct, but in practice LIIs sometimes have higher detection limits (unpublished data from NRC) and MSSs suffer from background-gas absorption as does the CAPS (Elser et al., 2019).

We addressed shedding in our response above. Shedding is one of multiple potential reasons for the scatter, but we have no evidence for it in our work. In fact, we have evidence against it (above). Discussing only this hypothesis would suggest to the reader that it is more likely than the alternatives; we wish to avoid that implication.

o I would expect the scatter between the three LIIs to be lower than reported given they are the same analyser, particularly the two NRC LIIs given they are next to each-other and presumably calibrated in the same manner. This is not addressed or discussed in the text, which is surprising given the detail that goes into the fluence sector. Could it have to do with the calibration performed for these analysers? Did the laboratory diffusion flame show ICAO annex16 applicability? If not, it could well explain some of the scatter you observe for EIm. This is something worth discussing in the manuscript.

Here the reviewer has correctly focussed on the scatter between the three LIIs, rather than the ratio between them. One reason why the apparent scatter is higher is that the two NRC LIIs (0331 and 0574) were not operated continuously for the entire campaign. Consequently, there are only 5 points in Figures 7 and 8 where the two NRC LIIs can be directly compared with one another. Figure 13a shows that the scatter between these two LIIs is minimal over short time periods (compare the thick teal line with the dark red

squares). Therefore, calibration is not a likely cause of the scatter. We speculate that some of the scatter between LIIs is also due to the penetration corrections, which were applied on a point-by-point basis and which were discussed in Section 4.3.

To minimize speculation, and because we have no direct evidence for penetration corrections causing the differences between LIIs, we hesitate to discuss this one hypothesis in detail. We consider it very likely that other unknown hypotheses may play a role.

o SMPS based EIm: I find the interpretation of this section misleading, as it suggests the SMPS is nearly as good as an LII or MSS at measuring nvPM mass. First, the SMPSs generally do not capture the full VSD (as can be seen in figure 5).

This is true, but:

Since the spread of nvPM EI_m reported by the two SMPS systems was smaller than the bias, their difference relative to the reference EI_m cannot be attributed to measurement biases (such as the limited size range detected by the instruments).

(underlined text is new)

Secondly, you assumed unit density for the CS-SMPS but the particle density could well be below 1 g/cm3, particularly given particle effective density decreases with increasing size and the mass is carried by the larger particles. Using integrated particle size measurement to derive mass is strongly influenced by the density you select and should be discussed.

It is generally true that the effective density function has a strong influenced on SMPS-based mass estimates. However, for the specific size and effective density functions expected for aviation soot, larger uncertainties may arise due to line penetration corrections than effective density. This has been shown nicely by Durdina et al. (2014) and cited on line 470 (Methods). The reason is that the line penetration correction for the small sizes of

aircraft soot particles tends to be very large, relative to other soot sources. This point does bear repeating in Results, so we added:

(We reiterate that our assumption of constant effective density is expected to introduce negligible uncertainty for the small soot particles emitted by aircraft turbine engines; Durdina et al., 2014).

Furthermore, I disagree with L613-L618; I believe the main reasons for the higher SMPS predictions is density assumption, the measurement uncertainty for the size bins >100 nm where the number count is very low, and potential shedding interference (see comments above). It is also surprising to me that in all figure 5b there is excellent agreement between the 2 stripped SMPS VSD's however it is then observed in figure 8b that there is no agreement. I am unsure how this is consistent if the same assumption regarding density is made?

In response to the previous comment, we pointed out the systematic study by Durdina et al. (2014) which proves that the density assumption incurs negligible error.

The reviewer also hypothesizes that low number counts at large size bins caused the SMPS errors. This would imply a uniquely higher scatter in the SMPS data at small concentrations (since the upper edge of the distribution, which contains most of the mass, would become 'small' first), which is not observed in Figure 9b.

To the reviewer's comment that agreement between the SMPS PVDs in Figure 5b appears better than Figure 8b, we point out that there are a few experiments where the SMPS data overlap in 8b. We did manually choose the example in Figure 5b and undoubtedly introduced some bias, but all SMPS data have been presented using summary statistics in other figures and the raw data for which are included in the Data Availability section. Also, the validity of the summary statistics such as GMD and GSD in Figure 7, which are more appropriate for lognormal distributions, was checked for all presented data.

o L645: if the TAP and PSAP require a filter change at each test point to operate optimally, doesn't it make them not suitable for aircraft nvPM mass measurement? Particularly given the mass loading you've experienced were typically lower than certification measurements as you were sampling 43m downstream of the engine. If that's the case, then I suggest re-writing the abstract and conclusion to highlight this.

Indeed, this is the least convenient aspect of the TAP and PSAP. But these instruments are tiny (can be operated handheld) and the sacrifice is necessary for aircraft measurements.

We made no relevant statements in the Conclusions, and the only relevant Abstract statement is:

The commercial instruments used were one TAP, one PSAP, and two SMPSs. These techniques are used in specific applications, such as on-board research aircraft to determine PM emissions at cruise.

It remains true that these instruments "are used" and, since the abstract is already very long, we have decided not to modify the sentence.

- Conclusion: I suggest re-writing the conclusion considering the comments above. For example, L574, I disagree that 10 ug/m3 is the noise level of the instruments. I suggest replacing "instrument" by "instrument calibration and sampling methodology".

We changed to:

the noise level of these instruments in our sampling setup

We cannot ascribe the observations to calibration issues, which would instead cause systematic bias. However, the calibrations could be due to background interferences, which become more important at low concentrations.

Minor comments: - General: The manuscript could do with more cross-referencing for the reader to find information more easily.

We have added a couple of cross-references in the process of this review.

- L39 & L169: replace “sampling” by “measurement”

Done

- L117: add “minimum” before 50%.

Fixed

- L122: I don’t believe it’s true that the APC, MSS and LII are the only commercial instruments that satisfy the SARP. For example, a Dekati DEED and a Grimm or TSI CPC is a commercial system that satisfies the SARP. Please clarify.

The reviewer is correct. Rather than attempt to complete our list of commercial instruments, we have removed this statement, to allow for future instruments which may enter the market.

- L152 & L183: I don’t think you can reference something that hasn’t been published yet or that that hasn’t been peer reviewed yet. As a reviewer to this paper it is hard to critically appraise the statements and conclusions without being able to see the detailed experimental set-up and graphs addressing fuel effects etc.

This is a fair comment, but the two papers are “companion” papers. We should have provided a draft manuscript. At the reviewer’s request we are happy to provide a copy of the companion paper, which is now submitted to Fuel and cited as such (Schripp et al.)

- L219: Why was the plenum only maintained at 33°C? It seems odd to me that you first sampled via a 60°C heated line, then a 33°C plenum and then other 60°C heated lines. This would promote thermophoretic loss (although very small) and could cause water to condense. Please justify.

This was a practical limitation and will be corrected in future work.

- L230: Why did you use a 25m line between the Dekati diluter and the NARS instruments? Was it because you couldn't get container 2 any closer to container 1? Wouldn't it have been better to use a shorter line to minimise diffusional losses and reduce your loss correction uncertainty which accounts for some of the discrepancies observed in your data? Given the 4:1 dilution ratio and sampling position the NARS system was not in compliance anyway hence could have been further optimised.

This was done because the goal of the experiments was to have the NARS use its standardized sampling line. The penetration function of this sampling line has been characterized in detail.

- L244: Please quantify what you mean by "good agreement".

This statement was used to justify the selection of one instrument over another:

"The CO_2 measurements from the NASA LI-COR 7000 were in good agreement with those taken by DLR [...] but had a faster response time and were therefore used as the reference for instruments in Container 1."

We acknowledge that "good agreement" is not a scientific assessment, but given that the response times differed, the data would need to be deconvoluted before any point-by-point statistics are used. The difference in response time was related to a difference in flow rates. Since we have therefore not sought to systematically compare these two instruments we have avoided any quantitative statements here.

- L255: The DMS-500 measures from 5 nm not 10 nm. Also, what is the size range of the EEPS?

DMS500: fixed

EEPS: 5.6 to 560 nm. Added.

- L317: drift not drifted.

fixed

- L371: Typo CO2 not CO2

fixed

- L376: remove “from”.

fixed

- L518: What inversion matrix was used for processing the DMS-500 data? Please add to manuscript.

Log-normal inversion with a bimodal calibration matrix. Added.

- L550: It’s not true the SMPS measures at 10 nm with a 100% efficiency as lots of corrections are applied (charging efficiency, loss through DMA and tubing). Please add “corrected” before 100%.

Text changed (see above)

- Figure 7: The x-axis is labelled “mean nvPM xxx”, however some total PM is also shown (as labelled in the legend). Please clarify and correct x-axis.

The total PM is not included in the X axis. The X axis is correct.

- L783: therefore not therefor

Thereof not therefore. Unchanged. Thank you for the comments.

3. Other changes

We also made the following changes to the manuscript.

1. We realized that the reported aromatic concentrations for SAF1 and SAF2 in Table 1 were incorrect. They are now fixed.
2. In Section 4.1.1 we clarified that only the ‘ordinate data’ and not ‘the measurements’ were normalized by the mean.
3. We removed the citation to the manuscript by Anderson et al. (in prep.).