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Review of Vernocchi et al. MISG characterization

Anonymous Referee #2

Referee comment on "Characterization of soot produced by the mini inverted soot generator with an atmospheric simulation chamber" by Virginia Vernocchi et al., Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2021-345-RC2, 2021

Review of 10.5194/amt-2021-345, Characterization of the MISG soot generator with an atmospheric simulation chamber, by Vernocchi et al.

The authors characterized an Argonaut MISG (Model MISG-2) using measurements of size distribution, elemental carbon, and light absorption. These measurements are thoroughly described. Measurements were repeated for a number of fuel/air flows using ethylene and propane, and also with/without a cyclone to remove large particles. Size distributions were measured using mobility measurements (SMPS; up to about 800nm) and an optical particle sizer (OPS; up to about 8 um). The results are presented clearly and the authors have observed an important supermicron soot mode that has not been reported by the 3 previously published literature studies on the MISG-2, which is by itself a strong reason for publication.

The title and abstract should be modified to emphasize that conclusion, but at the same time the abstract must also mention that this study uses different fuel flow rates than earlier studies. A new title might be "Characterization of supermicron and submicron soot produced by a miniature-inverted soot generator". (Current title uses an acronym with 2 words which are part of the acronym.)

I have a few major comments which should be addressed before publication.

Major comments

The observation of supermicron soot would suggest that future studies should never use ethylene fuel in the MISG (and perhaps also other inverted burners) as a surrogate for

atmospheric soot. This is an important conclusion, and although an earlier study using ethylene in the MISG (Kazemimanesh et al., 2019) noted the supermicron soot, its importance was not emphasized. That study also used a different flow rate. Given this emphasis I would like to request one additional experiment is made before publication. The authors should directly test their hypothesis that "super-aggregates...are likely formed directly in the exhaust line where particles density is very high" (lines 498-499). If this is the case, then could the issue be solved simply by diluting immediately after the MISG? The experiment would be simple. The authors need only to run the MISG with 3 line lengths. Very short, normal (as used previously), and very long. For each line length, measure with the OPS and SMPS. The results should be reported as combined OPS-SMPS size distributions in mass and number weighting.

Second, and continuing from above, a discussion of the physical properties of the supermicron aggregates is missing. For example, if super-aggregates are formed in the exhaust lines, then they should have the same MAC as the particles they are formed from. If they do not, then they must have a different morphology. Chakrabarty et al. (https://doi.org/10.1038/srep05508, 2014, Fig S3) predict a similar MAC for supermicron aggregates as for smaller aggregates. So, the authors might be observing aggregates that are more compacted than expected. This is supported by the trends in Figures 9 and 10. The authors should discuss their data with reference to this and other literature on superaggregates.

Third, I would also request that the authors try harder to reproduce exactly the conditions used in previous studies. As it is, the authors have used higher fuel flow rates (equivalence ratios) than all previous studies (Kazemimanesh et al., 2019; Moallemi et al, 2019; Bischof et al., 2019). It is unclear to me why the authors have not reproduced previous measurements exactly, to allow for comparable results. Is it because the authors used long line lengths and changed the pressure downstream of the flame? Is it because the authors used an "MISG-2" and not an "MISG-1"? Also, as mentioned above, the abstract should emphasize this difference in flow rate.

Fourth, the authors should present SSA from the PAX instrument, to allow for a direct comparison of their measurements with the SSA reported by Moallemi et al. On line 386 the authors write "the comparison with previous literature (Moallemi et al., 2019) ... reported the Single Scattering Albedo instead of the absorption coefficient". The authors cannot change how Moallemi et al. presented their data, but they can match their presentation to Moallemi et al's. The authors have SSA data and should present it.

Minor comments

The absorption coefficient divided by the number concentration is the absorption cross section. Please use this definition in Figure 11.

What exactly is the difference between the MISG-1 and the MISG-2? Previous MISG characterization studies used the MISG-1.

Please specify the line length used between MISG and chamber. Obviously, this is important (see first major comment above). Please also specify the exhaust line length and i.d. (ideally pressure in the line would be reported, if that is not available then reporting these parameters will help).

Table 4. The dark red and dark purple look the same in a black-and-white printout. Use e.g. a lighter red.

The authors first mention Bischof et al. (2019) at line 292. The paper should be mentioned in the introduction; it is a characterization of the same MISG.

Line 79, the air flow is not internally split between combustion and carriage. This makes it sound like there are 2 divided flows. In fact there is one air flow, and some of the air is consumed for combustion.

Line 93, consider writing m^3 air / m^3 fuel (the unit m3/m3 is confusing.)

Line 104, Moore et al. demonstrated the relationship of stoichiometry with particle size for the miniCAST only. The miniCAST is unique from the MISG, because it is a quenched flame. The quenching height is fixed. In the MISG, the open tip can move up and down with fuel flow. Use a difference reference, or change the statement.

Line 248. This is repeatability, not reproducibility.

Line 248, is the repeatability measured day-to-day? Between scans?

Figure 3. Missing error bars. Same for similar figures.

Figure 4 and line 292. The discussion of size vs. stoichiometry compares this work with prior work which was not performed at the same fuel flow rates. The comparison is not fair. It is more reasonable to conclude that the relationship changes at high fuel flow rates (if all data were plotted together, a trend might be observed). The authors need to reproduce earlier measurements to confirm their discussion, or change the discussion.

Line 307-316. The discussion compares "2 um" particles with "4 um" particles but the units are not the same. The 2 um was measured by TEM maximum dimension (or projected area?) and the 4 um was measured optically. Was the optical size corrected for the refractive index and shape of the particles? Please specify "projected diameter in an electron microscope" and "optical equivalent diameter". And please describe the calibration of the TSI OPS 3330 in Methods.

Figure 6. At what MISG flow rates were these data taken?

Figure 7 and 8. Why not use units on the y axis if units are reported in the text? Please change to units. If the authors argue against units, then specify the maximum in the caption.

Figure 9 and 10. Please combine into Panel A and B of the same figure, to avoid repeating a long caption twice. The important point is that one used a cyclone.

Figure 9 and 10. Please change from "Relative EC concentration" to "EC:TC ratio" to make it clear what the EC is 'relative' to. The discussion mentions OC:EC as well, which is confusing. Please always use EC:TC and OC:TC.

Line 366-373. Blank and backing filters should be mentioned in Methods so that the reader is not surprised at the discussion here. The authors should also mention that about 1000 ug/m3 EC was collected on the filters, which means that gas-phase VOCs become less important. Anyway, is the discussion of OC correction relevant if only EC concentrations are reported? Isn't "PC" more relevant, since that is where biases can come in? (In other words, how difficult was it to determine the split point?)

Figure 11. The authors normalized b_abs to N_SMPS. But in Figure 6 the authors showed that N_OPS was important. Why did the authors ignore the particles that the OPS couldn't see? Was a cyclone used? Clarify the text and figure please.

Table 6. Was the AAE calculated using a power-law fit? A fit to 3 points would not be reliable. I recommend reporting 2-wavelength calculations of the AAE, for blue-green and green-IR, (and optionally also blue-IR) which also allows the reader to observe the consistency between the individual PAX instruments.

Figure 12-14. Consider using open/closed symbols to enhance readability in black-and-white printouts.