Atmospheric Measurement Techniques Discussions

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Review of Vernocchi et al. MISG characterization

Anonymous Referee #2

Referee comment on "Characterization of the MISG 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.

We thank the Referee for his valuable comments. In the following, we reply point-by-point to his notes.

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.)

We modified the title and abstract as suggested.

The revised title is: Characterization of soot produced by the Mini Inverted Soot Generator with an atmospheric simulation chamber

We added in Line 13: "This work deepens and expands the existing characterization of this soot generator that is also coupled with an atmospheric simulation chamber. Differently from previous works, MISG performance has been also tested at different fuel flows and higher global equivalence ratios. MISG exhausts were investigated after their injection inside the atmospheric simulation chamber: this is another novelty of this work".

Line 24: The soot characterization opens to various kinds of experiments in ASCs. Particles with well-known properties can be used, for example, to investigate the possible interactions between soot and other atmospheric pollutants, the effects of meteorological variables on soot properties and the oxidative and toxicological potential of soot particles.

We have also added some statements about the scope and novelty of our work in the last part of the introduction.

Line 63: "Differently from previous works (Bischof et al., 2019; Kazemimanesh et al., 2019; Moallemi et al., 2019), the MISG has been connected directly to an atmospheric simulation chamber; performance has been tested also at different fuel flows and higher global equivalence ratios. The present characterization deepens and expands the existing knowledge on particles and gases produced by this soot generator. The comprehensive characterization of the MISG soot particles is an important piece of information to design the subsequent experiments. Well-characterized soot particles could be used to investigate the effects that atmospheric parameters can have on soot particles, and also to study the interactions between soot particles and other pollutants".

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.

In the revised text, we will emphasize this issue in the abstract as well as in the conclusions.

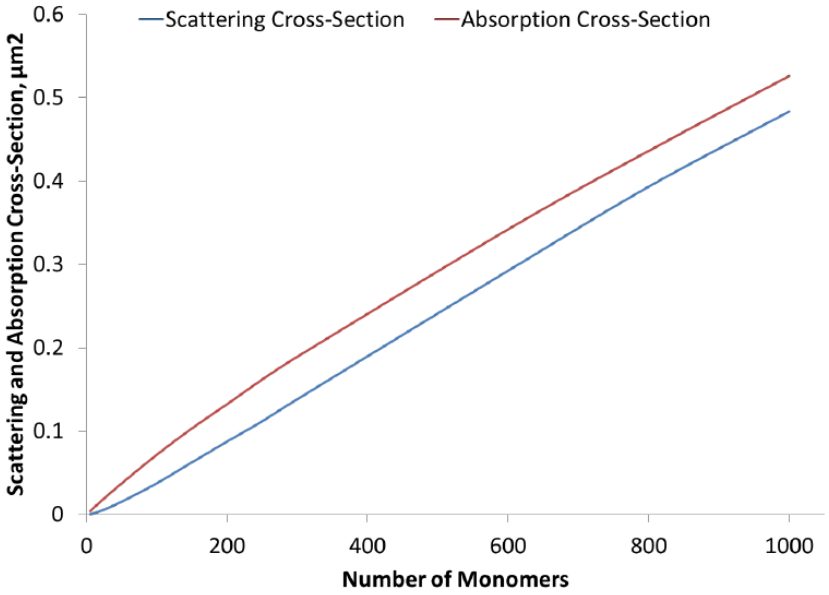
That study also used a different flow rate. Given this emphasis I would like to request one additional experiment is made before publication. The authors should directly test their hypothesis that "super-aggregates...are likely formed directly in the exhaust line where particles density is very high" (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.

This point has been raised by more than one Referee, and we agree that it is an interesting point to investigate. We will be able to answer to this question after some extra experiments, 1) by modifying the line length as suggested, 2) by inserting a dilution system just after the SG exhaust. Anyway, we have just a doubt about the effect produced by the modification of line length. After the small quartz cell where the flame burns, the exhaust is carried outside the SG after passing through a copper serpentine, with length roughly 40 cm long. If coagulation happens in this section, no way to understand if super-aggregates are formed in the flame or after.
Since our atmospheric chamber is currently engaged full time in non-postponable experiments, we will perform the experiment as soon as possible, in agreement with the editor.

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.

We thank the Referee for the interesting paper; however, it investigated soot emission by wildfires, not by laboratory sources. Moreover, we are confused about the indication of Figs. 9 and 10 about the interpretation of global equivalence ratios vs. EC concentration. Why should these Figs suggest that observed aggregates are more compacted than expected? About S3 figure by Chakrabarty et al.: it seems to us that the MAC is quite far from constant with respect to the size of the super-aggregates. If we interpret correctly this figure, the MAC is constant in respect to the single monomer.



We agree with the Referee that a full characterization of super-aggregates, independently if they are formed directly at the flame or after in the exhaust line, would be of great interest. At the same time, we think that such deep characterization is outside the scope and the possibilities of the present work. Anyway, we will try to answer to this point with the experiments discussed in the addendum.

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.

The opportunity to reproduce exactly the condition used in previous works is for sure a good scientific procedure. However, we did not reproduce previous measurements for several reasons. First, our intention was to explore new operation conditions in order to expand the knowledge of the SG. Secondary, we wanted to compare the soot produced by propane and ethylene, since all the previous works focused on one fuel only at a time. So, we considered mandatory to use combustion conditions directly comparable between the two fuels and, at the same time, to maximize the possible comparisons (i.e., same air flow with different fuel flows, same fuel flow with different air flows, same global equivalence ratio and air flow with different fuels). In the extra-experiments we are planning we will try to reproduce the best way possible some of the already experimented burning conditions to have comparable results. Anyway, the set-up will not be exactly the same, since our experiments make use of the simulation chamber, and its exclusion can not be considered. Since our atmospheric chamber is currently engaged full time in non-postponable experiments, we will perform the request experiments as soon as possible, in agreement with the editor. We will be able to answer after the extra experiments requested by the referee.

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.

Done.

Minor comments ----------

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

Modified and moved to Supplementary as requested by RC3.

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

We did not know the answer so we asked to the manufacturer. He explained us that there are just a few minor changes between MISG-1 and MISG-2. The main difference is that MISG-1 had a 1/2" exhaust tube while MISG-2 has a 3/4" exhaust tube. We thank Jason Olfert for this information.

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).

We added the line length used between MISG and chamber in sect. 2.2. Since the exhaust line does not pass through the chamber volume, we believe that the only useful length is of the line between MISG and chamber.

We added in Line 159: The connection between MISG and ChAMBRe was made by Swagelok adaptors (size ¾") and ISO-K flanges (16 mm diameter) to avoid any possible leak; the length of the line was 65 cm.

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

Done.

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.

Done.

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.

Corrected.

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

To avoid confusion, we removed units as suggested by RC5.

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.

We deleted the statement.

Line 248. This is repeatability, not reproducibility.

Corrected.

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

We added details in the text. The repeatability is measured between identical repeated experiments.

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

Done.

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.

The Referee is right, the direct comparison is not possible since the feeding flows are different. Anyway, keeping in mind this information, we can discuss the trend of different particle properties by varying flows and compare them to the previous works. We added this statement (Line 292): "Even if the direct comparison between our findings and results from previous works are not directly comparable (since feeding flows are different), some similarities can be identified".

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.

We used the default TSI refractive index. The OPS had been calibrated by the manufacturer. We appreciate the suggestion to uniform the quantities, but in our discussion we are not interested in giving precise numbers but just give the order of magnitude of the particle dimensions. Of course this parameter has been measured with very different techniques (TEM vs. OPS) and we are not even trying to match them. So we added in the text the specification of kind of diameter as suggested.

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

We added this information in the caption: "MISG was fuelled with 7 lpm of air and 75 mlpm of fuel during propane experiment or 127 mlpm of fuel during ethylene experiment. No cyclone."

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.

Done.

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.

Done.

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.

Since it is not the EC:TC ratio but the EC concentration normalized to the highest concentration of the whole data set, we changed "Relative EC concentration" to "Normalized

EC concentration".
We modified the discussion by using 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.

We added this mention in Sect. 2.5, line 214: "We also performed some tests adding a backup filter during the sampling to determine the volatile fraction of OC."

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.

Added.

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?)

We reported only EC concentration values because we observed that OC concentration values were negligible. Since sample contained almost only EC, thermograms were easy to analyze with split points clearly identifiable.

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.

The figure refers to experiments performed without the cyclone, we added the information in the text. We consider only SMPS data because the number concentration of super micrometric particles is negligible compared to the total number concentration. Anyway, when the cyclone was inserted between ChAMBRe and PAXs, particles generated from propane combustion were even more absorbent than the ethylene generated, even if with a small gap. As suggested by other referees, we moved this figure in the supplementary and we added the Figure with cyclone too.

Table 6. Was the AAE calculated using a power-law fit?

Yes

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.

We added a Table in the Supplementary with the results of AAE from the 2-wavelenght calculations.

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

Done.