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Comment on amt-2021-345

Anonymous Referee #4

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-RC5, 2021

Review of "Characterization of the MISG soot generator with an atmospheric simulation chamber"

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

General comments:

This paper discusses the physical, chemical, and optical properties of soot produced by burning propane and ethylene in a miniature inverted soot generator (MISG). Although some aspects of this work (such as flame shape vs. fuel and air flows) have been discussed in previous studies, there are some novel aspects to the paper: combining the MISG with an atmospheric simulation chamber and studying the optical properties of soot in depth. The methodology used in the paper is sound and valid but the paper itself is cluttered and poor in terms of readability. I suggest that the authors streamline the paper by omitting the discussion on flame shape with combustion conditions or moving it to the supplementary material, and instead focus on aspects that have not been covered in similar other studies.

We have followed the Referee suggestion, moving tables 3 and 4 to Supplementary and improved the general quality of the text.

The whole manuscript should be edited for grammar and proper academic writing too. There are also some discrepancies between the results presented in this paper and previous papers that characterized the MISG soot, which need to be discussed in more details by the authors (see my comments below). Overall, the paper is not acceptable in its current form and needs major revisions before it can be published.

We improved the text as suggested.

Specific comments:

Article title: Avoid using an acronym in the title without fully defining it first.

Done. We have modified the title following both RC2 and RC5 comments.

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

Abstract should be written as one paragraph.

Done.

Abstract: MAC stands for mass absorption cross-section, not mass absorption coefficient.

Done.

Section 1: Combine paragraphs 2 and 3.

Done.

Line 45: List the "several other purposes" in those references more specifically.

Done. In the revised version we added:

Line 45: "such as studies on atmospheric processing of soot particles, characterization of uncoated/coated and fresh/denuded of soot particles"

Section 1: The introduction is written poorly and needs to be improved in terms of readability and transition between paragraphs. It should also clearly state the objective(s) and novelty of the study near the end of the introduction.

We added some information and clarified better the purpose of the work:

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 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.

Line 69: Change "air and fuel flow in an opposite way to the buoyancy force" to "… in opposite direction to the …"

Done.

Line 70: Change to "The resulting diffusion flame is more stable by reduced flickering of flame tip"

Done.

Line 77: lpm and mlpm should be defined (it is better to use L/min or mL/min as units of flow rate).

Done.

Line 79: This statement is not correct. Kazemimanesh et al. (2019) states that part of the air flow is used in combustion and the rest is used to dilute the exhaust products.

Corrected.

Lines 81-101: The definition of equivalence ratio is based on fuel-to-air ratio, thus the reader would not be confused if you define the fuel-to-air ratio (instead of AFR) first. Also, all equations should be numbered.

Corrected.

Line 93: The units used for AFR are not clear to me. AFR is a unitless parameter, so just get rid of any units.

Done.

Line 103: Many of the in-text citations in this article should be in format of Author (Date). Please consider this whenever suitable during revision. For example: Moore et al. (2014) demonstrated that fuel-lean flames produce soot particles …

For in-text citations, we followed the AMT guidelines that say "If the author's name is part of the sentence structure only the year is put in parentheses ("As we can see in the work of Smith (2009) the precipitation has increased"). If the author's name is not part of the sentence, name and year are put in parentheses ("Precipitation increase was observed (Smith, 2009)")"

Line 144: Consider changing to "at the fuel tube nozzle"

Done.

Line 125: I cannot find Section 2.1.2 in the paper.

This section was not present and has been deleted. We thank the Referee for noticing this material mistake.

Line 179: It is known that the multiple charge correction algorithm in the TSI AIM software breaks when the median mobility diameter is relatively large (>200 nm). Can the authors show the uncorrected and corrected size distributions for 2-3 cases in the supplementary material?

While checking the figures, we noticed that data were not corrected for the multiple charge correction algorithm. We apologize for this mistake in our text. We added a pair of uncorrected and corrected size distributions in the supplementary material (Fig. S1).

Line 218: Change peculiar to a better adjective.

Done.

Line 236: "To our knowledge, no literature information is available for the ethylene in the flow range of Table 4." This statement is not true. Kazemimanesh et al. (2019) studied the MISG and its flame shape with ethylene and air flow rates (80-130 mL/min and 4.0-10.0 L/min, respectively) that partly cover Table 4.

Modified in "A similar characterization with ethylene also exists but it only partly covers the flow ranges explored in the present work. We got some differences especially in the transition range to Open tip flames, probably due to the different setups. Also the subjectivity of the visual determination, that is user dependent, can lead to differences."

Lines 241-251: The authors talk about various experiments that they did and the calculated repeatability (mistakenly noted as "reproducibility") in mode diameter and concentration. However, it is not clear what conditions were tested and the results are not shown in the paper or the supplementary material.

Corrected and specified. The conditions tested are reported in Table 1 and 2, while results are discussed in the text.

Page 10 – Fig. 4 and the discussion around it: The particle mode diameter reported for ethylene flames is constant at ~175 nm. This is inconsistent with previously reported values of ~240 nm and up to 270 nm (Kazemimanesh et al., 2019). The same reference also reported an initial sharp increase in particle size and concentration with increasing ethylene flow rate, which eventually levelled off to a relatively constant value. This is in contrast to the trend seen in this paper. These differences must be noted and discussed in the paper.

We added the discussion about these differences, that probably depended on the different combustion conditions.

Line 292: Even if the direct comparison between our findings and results from previous works (Bischof et al., 2019; Kazemimanesh et al., 2019; and Moallemi et al., 2019) are not directly comparable (since feeding flows and global equivalence ratios are different), some similarities can be identified. Previous works observed that by increasing the fuel flow, the particle number concentration increases too, that is what we observed for propane. In addition, Bischof (2019) also reported that the particle mode diameter, with propane, did not depend on the global equivalence ratio, as we also observed, but for ethylene. Kazemimanesh (2019) showed a clear increase in mode diameter, corresponding to an increase of fuel flow rate, that reached a quite constant value (i.e., around 240-270 nm) for ethylene. This trend differs from our observations, since the mode diameter in our case turned out to be quite stable at about 175 nm independently on feeding flows. This difference is probably due to the global equivalence ratios used: while in (Kazemimanesh et al., 2019) global equivalence ratios are lower than 0.206, in our case they are higher than 0.213. In (Moallemi et al., 2019), instead, they observed an opposite behaviour for mode diameters: they retrieved that at fixed fuel flow, a higher air flow produced a slight decrease of the mode diameter. Both (Moallemi et al., 2019) and (Bischof et al., 2019) measured mode diameters < 200 nm, but they used different combustion conditions (i.e., lower global equivalence ratios resulting from higher air flow or lower fuel flow). We can conclude that, as expected, global equivalence ratio is the principal parameter affecting size distributions of soot particles.

Anyway, as request by RC2, we will perform experiments that replicate the conditions used in the previous works, so we will able to compare the same operative conditions used by (Kazemimanesh et al., 2019).

Fig. 3 and 4: The authors should consider adding error bars to the data points. In addition, it is not clear why a linear fit is shown for the data points when the paper does not offer any evidence or support for trend.

Done and specified ("Lines aim to facilitate the reader eye."). The same changes were applied to Fig. 7, 8, 9 and 10 too.

Fig. 5: I suggest that the authors show and discuss figure 5 before figures 3 and 4, as this will enhance the readability and flow of the paper. I was completely lost about the results shown in figures 3 and 4 when I first read the paper until I saw figure 5. Figures 3 and 4 are essentially the size distribution parameters extracted from figure 5 and shown with respect to equivalence ratio.

Modified as suggested.

Lines 307-316: Can you show the number and volume distributions side by side in Fig. 6? What is meant by "relative particle number concentration" in Fig. 6? [dN/dlog dp]/N_tot?

We noticed some inaccuracies in the text about the kind of discussed distributions and we corrected them. Since in the text we discussed number and mass distribution, we modified Fig.6 by showing number and mass distribution side by side. In addition, the caption was modified by changing "relative concentration" to "normalized concentration"

Section 3.2.3 (EC-OC analysis): The authors did not elaborate how they calculated TC (total carbon). OC can exist in gas-phase or as condensed semi-volatile particles and the authors need to distinguish between the two when calculating TC. The authors briefly mention the use of a second filter, which should help in determining the mass concentration of OC existing as semi-volatile particles.

Total Carbon was calculated by the thermal-optical analysis as the sum of the whole evolved carbon during the analysis. The instrument was calibrated by using a standard solution.
We used backup filters to estimate the semi volatile/volatile fraction of OC, which resulted compatible with the organic contamination on blank filters. Hence, the total OC is given by the concentration value measured on the main filters. We are not able to evaluate OC in gas-phase.

Fig. 9 and 10: I do not quite understand why normalized EC concentrations are shown rather than the absolute values of EC concentration or the EC/TC ratio. The latter two parameters are more important for researchers when using a soot generator.

We have shown the normalized concentrations to emphasize the differences deriving from the use of the cyclone in case the soot is produced by the combustion of propane or ethylene. Anyway, the EC values are those measured on the filter and deriving from the concentration in the chamber and not directly produced by the soot generator.

Line 384-385: Why is propane soot more light absorbing than ethylene soot at all three wavelengths?

This is a very good question. Optical properties such as absorption depend on several parameters, mainly composition, mixing state, aging, and size. Considering all the experiments reported in this work, no differences in composition can be expected, since only EC particles are present: this means that differences in absorption cannot depend on particle composition. Also mixing state and aging can not explain this difference: soot inside the chamber is fresh and only EC is present. We can explain the higher light absorbing capability of propane by considering differences in: size distributions (see Figs. 3-5) and morphology/density of the particle produced by the burning of the two different fuels. We have added in the revised text these considerations.

Line 498: The formation of superaggregates is related to high particle concentration in the exhaust line. This means that by diluting the MISG exhaust, the formation of these large aggregates can be alleviated. Kazemimanesh et al. (2019) and Chakrabarty et al. (2012) suggest that these superaggregates are formed at the stagnation plane of the flame tip, which seems more plausible. The authors should note and discuss these differences in the paper (not in the conclusions section).

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 and add the results in the revised text after some extra experiments, by inserting a diluter between MISG and ChAMBRe as suggested. We will also try to modify the injection line length, as suggested by RC2. 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.