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 and https://doi.org/10.5194/amt-2021-345-RC4, 2021

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

We performed the additional experiments requested by the Referee.

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.

2) K2018 discussed superaggregate formation in a stagnation plane, citing literature by Chakrabarty et al. different to the citation I gave earlier. The stagnation plane hypothesis is inconsistent with the present manuscript's hypothesis that coagulation occurred in the sampling lines. The stagnation plane hypothesis may also better explain the difference in EC:TC of the superaggregates. Regardless, I still recommend that the authors test different sampling line lengths directly since that test is simple. (This comment extends my original 2nd major comment.)

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.

We tested different line lengths: the short line was 30 cm, the normal line was 65 cm long and the long line was about 5 m. We also diluted MISG exhaust just after the outlet of the generator maintaining the normal length of the exhaust line, by adding an extra air flow, the ratio between dilution air (dry) and MISG generator was 4:1.

Only the experiment with the longest line showed a significant decrease in particle concentration, probably due to the losses inside the pipeline. These results suggest that superaggregates were formed at the stagnation plane of the flame tip, as correctly reported by the Referee and references he cited (Kazemimanesh et al. (2019) and Chakrabarty et al. (2012)).

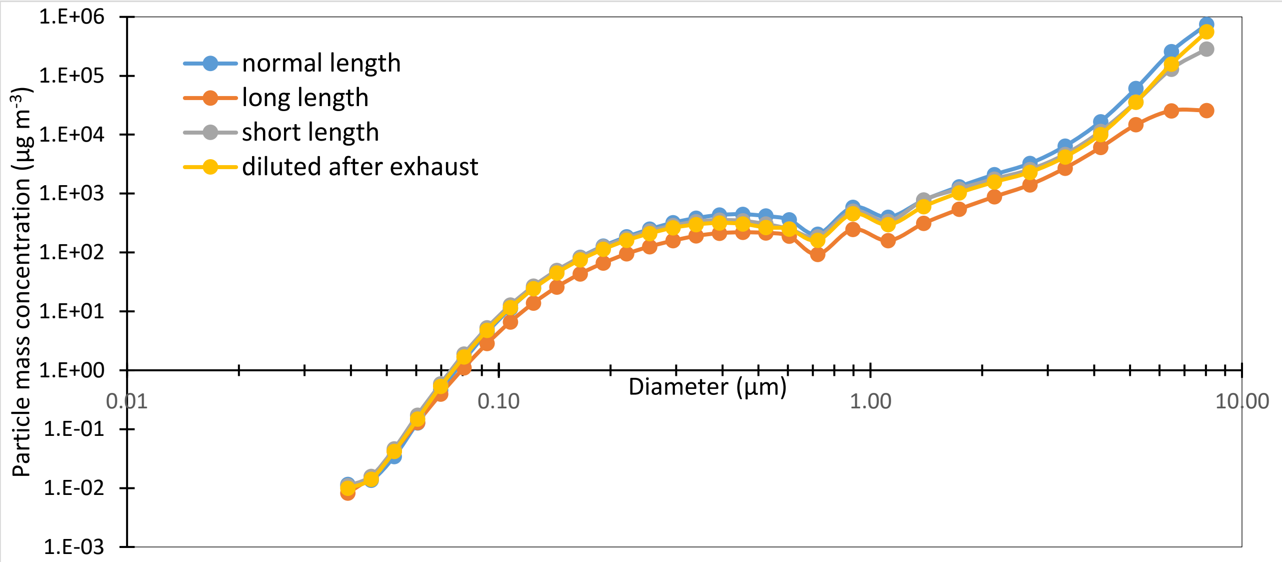

Figure 1: Comparison between mass size distributions measured by SMPS and OPS. The MISG was fuelled with 7 lpm of air and 127 mlpm of ethylene.

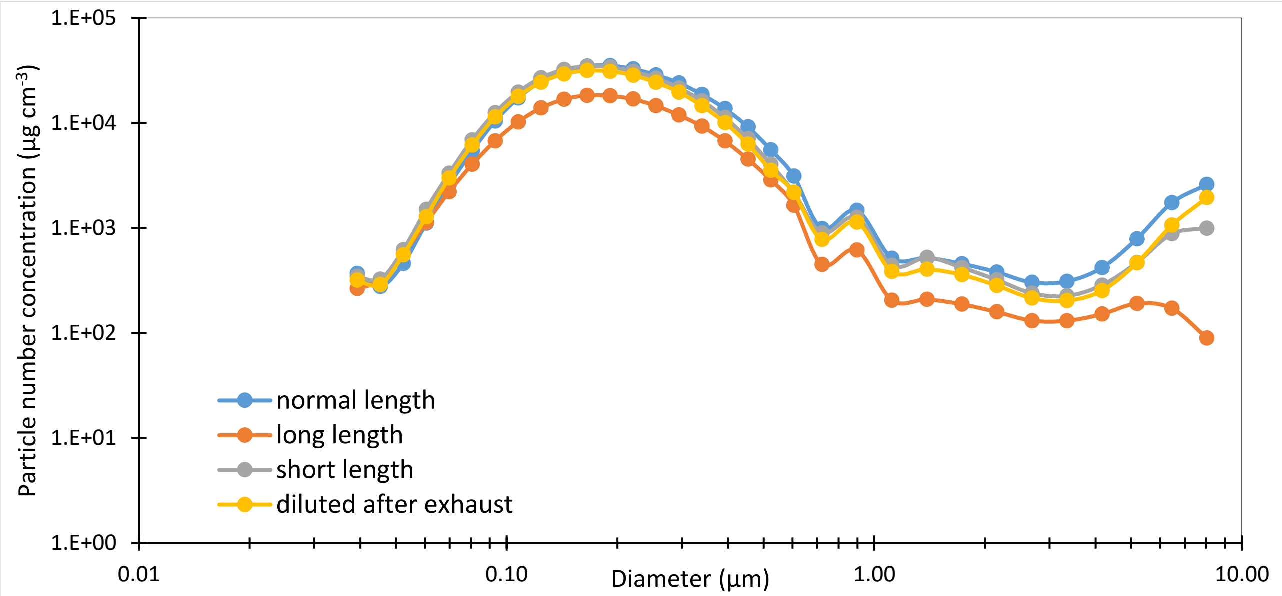

Figure 2: Comparison between number size distributions measured by SMPS and OPS. The MISG was fuelled with 7 lpm of air and 127 mlpm of ethylene.

In the revised text, we added:

Line 309: ethylene combustion produced a limited number of big particles, likely super-aggregates, probably formed at the stagnation plane (Chakrabarty et al., 2012). This hypothesis was confirmed by dedicated experiments with the setup specifically modified in respect to the basic one (see Supplementary Fig. S.2).

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.

We replicated some of the combustion conditions reported in the previous literature works. We explored 9 lpm of air - 100 mlpm of fuel and 10 lpm of air - 100 mlpm of fuel for ethylene and 8 lpm of air - 61 mlpm of fuel and 9 lpm of air - 61 mlpm of fuel for propane.

Table 1: Comparison between results of previous literature work and our replicated experiments.

	Kazemimanesh et al., 2019		**This work**	
	Mode diameter (nm)		Mode diameter (nm)	
Ethylene: 9 - 100	242		191 ± 8	
Ethylene: 10-100	250		220 ± 9	
	Moallemi et al., 2019		**This work**	
	Mode diameter (nm)	SSA	Mode diameter (nm)	SSA
Propane: 8 - 61	150 - 190	0.17 – 0.22	202 ± 12	0.16
Propane: 9 -61	130 - 160	0.16 – 0.20	165 ± 10	0.14

In the revised text, we added:

Line 267: In addition, we reproduced some of the conditions investigated in the previous works obtaining a good agreement for the mode diameter and SSA figures (see Supplementary for details §3).

1) My original review stated that K2018 reported TEM size distibutions up to 2 um. This is true, but K2018 also reported aerodynamic size distibutions. (Moallemi et al. 2018 reported only TEM.) The physical interpretation of aerodynamic and optical size distributions should be discussed in detail (see e.g. https://dx.doi.org/10.1080/027868290903907). What is the optical equivalent diameter of 2 um aerodynamic diameter soot aggregates in the supermicron regime, considering morphology? Calculating the answer to this question is difficult, but measuring it is simple: the authors can compare OPS size distributions with/without the cyclone. (This comment extends one of my original minor comments.)

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'll try our best to characterize these super-aggregates, using the instruments we have in our lab and within the scope of the present work.

We measured SMPS+OPS distributions both without and with the cyclone as suggested, for 7 lpm of air and 127 mlpm of ethylene.

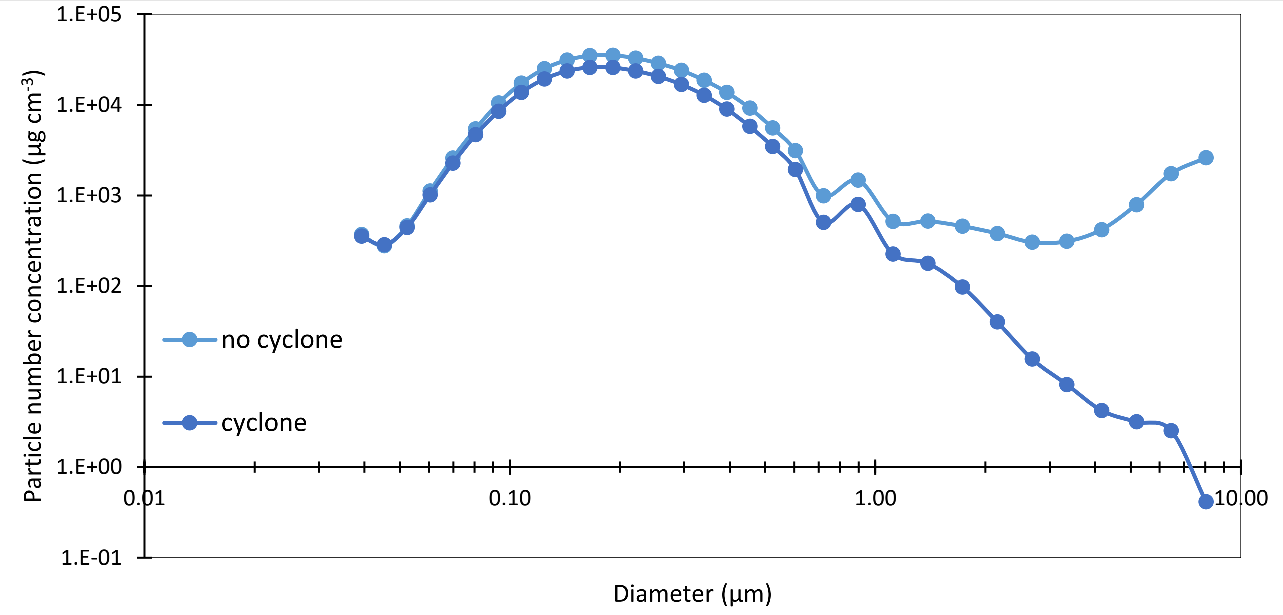

Figure 3: Comparison between number size distributions measured by SMPS and OPS. The MISG was fuelled with 7 lpm of air and 127 mlpm of ethylene.

3) K2018 also showed that superaggregate formation depends on fuel flow rate, with negligible superaggregates observed at the lowest flow rate (which also produced a lower number concentration). So did the authors observe 'larger' superaggregates because they used a higher fuel flow rate, or because they used an optical particle sizer instead of an aerodynamic one? (This comment extends my original 1st and third comments.)

We thank the Referee for the thorough speculation on the super-aggregates origin. We don't have the answer to this question, but we can try a simple experiment: we will use lower flow rates to see how their dimension change with them.

We performed an experiment using 6 lpm of air and 80 mlpm of ethylene. The formation of super-aggregates larger than 4 µm decreased consistently. Answering to the Referee's question, we observed larger superaggregates because we used a higher fuel flow rate.

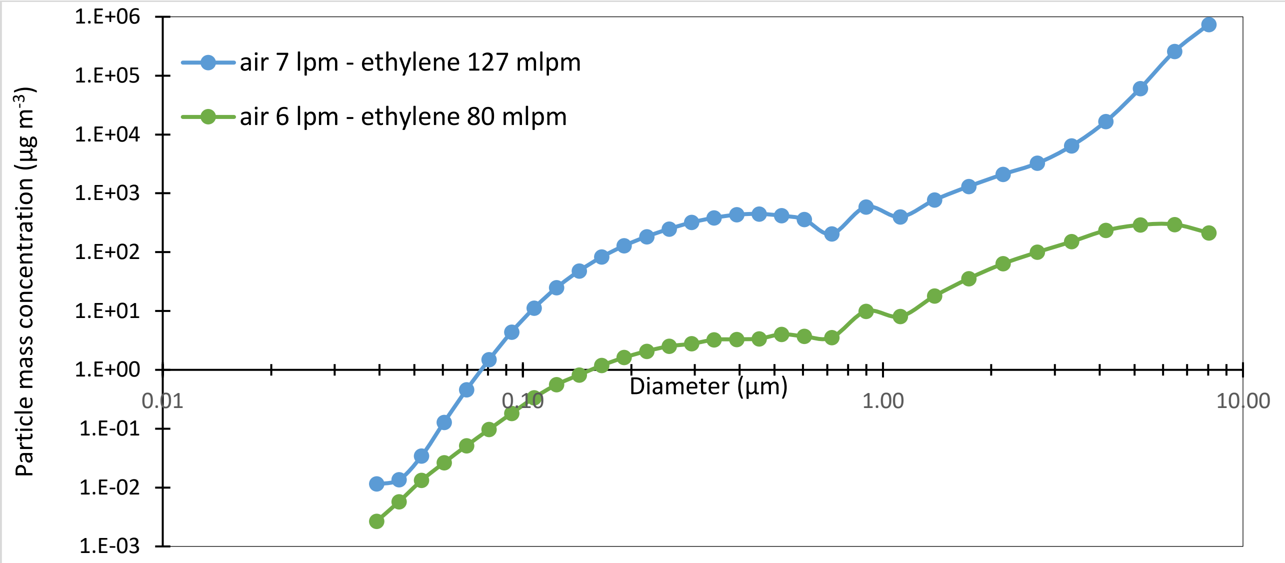

Figure 4: Comparison between mass size distributions measured by SMPS and OPS.

In the revised text, we added:

Line 317: Anyway, super-aggregates formation by ethylene combustion can be partly reduced by using lower air and fuel flow rates (see Supplementary Fig. S.3 for example).