Comment on amt-2021-345

Anonymous Referee #3

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-RC3, 2021

OVERVIEW

The present manuscript aims to characterize soot particles produced with a MISG. Overall the manuscript needs some rewriting and rethinking. Many sections appear to be a list of results which are scarcely interpreted, while many figures are barely described. Hence, some important results are shaded by many non-relevant information and figures. As final results, the conclusions and overall take-home message of the manuscript becomes extremely unclear. As mentioned in the second review, the authors show for the first time that MISG can produce large soot aggregates, which are not well characterized in other previous studies and cannot be generated, up to my knowledge, with the more traditional CASTburner. So, I suggest the authors to focus on this specific aspect of their research. Considering the presence of these large particles, PAX measurements must be corrected for truncation error. In its current form, the manuscript is not suitable for publication.

We have taken seriously the Referee's general comments, which we thank for the useful observations and suggestions for improvement. We have revised the manuscript, trying to put in evidence the new findings, and improving the conclusions with a more clear take-home message. In the following, we reply point-by-point to his notes.

SPECIFIC COMMENTS

ABSTRACT: The abstract is very generic and does not provide any real information on the performances of the burner. I suggest rewriting of the abstract.

Done. In the revised version, we added:

Line18: Significant differences could be observed when the MISG is fuelled with ethylene and propane both in terms of particle size, in particular, the production of sub-micrometric super aggregates was observed for ethylene combustion. With equal combustion conditions, ethylene produced higher number concentration of particles and smaller mode diameters. Soot particles produced by propane combustion resulted in higher EC:TC ratios and they were more light absorbing than particles generated by ethylene combustion.

INTRODUCTION: Introduction does not provide a context and does not present the motivation for this study. At the moment is a list of references without a clear story behind it. It does need some rewriting.

We added some information about the context and motivation of this study.

Line 39: BC is considered one of the most significant radiative forcing agent, second only to CO_2 (Ramanathan and Carmichael, 2008; Bond et al., 2013). Another positive effect on radiative forcing is related to the darkening of glaciers surface due to the deposition of BC (Skiles et al., 2018). Soot contributes to air pollution also via reactions with several gas species, as NO_2, SO_2 and O_3 (Finlayson-Pitts and Pitts, 2000; Nienow and Roberts, 2006).

Line 40: Soot particles are suspected to be particularly hazardous to human health, because they are sufficiently small to penetrate the membranes of the respiratory tract and enter the blood circulation or be transported along olfactory nerves into the brain (Nemmar et al., 2002; Oberdörster et al., 2005).

Line 42: In this context, soot generators are employed as stable sources of soot particles.

Line 59: ASC experiments are the best compromise between laboratory and field experiments, since they simulate real situations but without the uncertaintes and variability of typical field measurements.

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.

L27-34: the definition of "BC" is mostly based on its non-null imaginary part of the refractive index. However, Petzold 2013 made it clear that the term BC is a qualitative definition of BC rather than operational. From your text I have the impression that absorption photometers will directly provide BC concentration. I think that, however, a discussion on soot nomenclature is not needed so early.

We are aware of the paper by Petzold 2013. Our incipit was just to underline that BC and EC quantities depend on the measuring technique used to determine them. In this sense, these quantities are operationally defined. We consider these first lines (and the references therein) as a very brief introduction to the topic which could be useful for some readers.

L43-50: This part needs to be developed further since it will create the right context for your work.

We thank the Referee for the valuable suggestion. We have extended this part to better introduce our work. 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

L51-63: This part provides some sparse technical details of burner and a very generic description of smog chambers. It is not clear what the author wants to communicate here.

In this part, we establish the link between the main components used in our work (i.e. soot generator and ASC) before entering in the material and methods section. We would like to keep this part unmodified.

L74: reference

Done.

L77-80: what are the consequences of the absence of quenching or carrier gas?

Actually, the quenching or carrier gas are present in the MISG too, since a fraction of the feeding air is not used for combustion but used as quenching/carrier of the output flow. The differences between the miniCAST and the MISG are basically two: with the miniCAST, the quenching gas is N2 instead of air, so the quenching effect is reasonably higher than in the case of air. Second, in the case of the miniCAST the quenching gas flow can be selected, thus modifying the flame shape and flow turbulence. In the MISG, the quenching gas is air and the flow can not be selected, since it is just a fixed fraction of the feeding flow.

L84: give number to equation. Recurrent

Done.

L85-91: this occupy more space than needed. Put it as normal text. Recurrent

Done.

155-156: revise indent.

Done.

L166: size distribution measurements

Done.

L181-182: what refractive index was used to derive diameter from OPS?

The default OPS setting was used, i.e. 1.59.

L196-197: Considering the extensive use of PAX measurements in the paper I am genuinely surprised that truncation errors are disregarded. I think it is important to show that truncation is not relevant in these conditions. As recently resumed by Modini et al. (2021) little is known on the dependency of scattering phase function on the particle morphology and how this might impact truncation for highly absorbing aerosol particles. Scattering correction for absorbing aerosol is investigated for the nephelometer instrument by Bond et al., 2009. The argument of the authors is valid, but it should be contextualized if not verified.

We agree with the Referee regarding the opportunity to correct PAX data for truncation errors. By the way, the Modini et al. paper shows how the truncation error can be non-negligible on real aerosol samples when the SSA values are above 0.85. In our work, all the aerosols produced in the chamber have SSA value below 0.3, not surprising considering that they are composed by pure fresh BC particles. Moreover, in the paper by Modini et al., they used a CAPSssa instrument, which principle of operation is completely different from PAXs. It is not clear to us how the truncation error could affect the response of the microphone integrated in the PAX, but in principle we don't expect a significant bias. We didn't know the paper from Bond et al., 2009 and we thank the Referee for bring it to our attention.

Following the suggestion given by the Referee, we will add in the revised text the following sentence

Line 195: "Few papers in literature deal with the correction for truncation errors in nephelometer measurements (Bond et al., 2009, Modini er al, 2021) for highly absorbing particles: little is known on the dependency of scattering phase function on the particle morphology and how this might impact truncation.".

L246-251: If I understand correctly this is simply the relative standard deviation. It is not clear, however, in what conditions these values were calculated.

Yes, it is the relative standard deviation; we added the definition in the text. It was calculated by performing the same experiment many times. This is true for all the combustion conditions listed in Table 1 and 2.

L301-302: does it mean that all size distribution are measured 3 minutes after injection.

It means that the soot injection from the MISG into the chamber lasted 3 minutes and the measurements started just after the mixing time (other 3 minutes). We specified this information in the revised text.

The ageing time in the chamber should be always specified, since concentration and dimeter of particles drastically change due to coagulation, especially at high concentrations.

Specified in Sect. 3.2.1. We added:

Line 302: Data were acquired starting 3 minutes (i.e., the chamber mixing time) after the MISG switching off, for a specific time interval (i.e., 4 to 10 minutes). All the curves are normalized to the same injection time (i.e., 3 min of injection inside the chamber).

L340-372: Describing both EC:TC and OC:EC is redundant. It is also hard to compare and understand the impact of large soot on the OC:EC fraction from the two figures. I would suggest to merge them or focus on the impact of large soot. To be honest, figure 9 could easily go in the supplementary. Is there any correlation between OC:EC and diameter mode, CO2, NO ?

We changed the discussion from OC:EC to OC:TC and merged Fig.9 and 10. These figures emphasize the differences deriving from the use of the cyclone in case the soot is produced by the combustion of propane or ethylene. No correlations were observed between OC:EC and other parameters.

Section 3.2.4 -3.2.5: These sections could be merged. Especially considering the length of Section 3.2.4.

Done.

Figure 11 is barely described or discussed in the text. Hence, it can be moved to supplementary or removed.

We moved it to supplementary.

L412-417: This part is very hard to read and follow. The authors are requested to build a discussion on their result, rather than list numbers in series. This problem appears in almost every section of the paper. As a consequence, Figure 12-13-14 become hard to interpret too. I would suggest move the figures to supplementary, summarise the result in a table and construct a separate discussion for ethylene and propane.

We moved Fig.13 and 14 to the supplementary and Table 5 before Figure 12. We have re-written the discussion.

Line 412: The MWAA analysis at λ = 870 nm (Fig. 10.a) returned compatible MAC values for both propane series (with/without cyclone) and ethylene series with cyclone, while a consistently lower MAC value was found for the ethylene series (worse correlation) without the PM1 cutting. The same picture turned out at the other two wavelengths (in supplementary). By comparing PAX absorption coefficients and EC concentrations at λ = 870 nm (Fig. 10.b), obtained MAC values are more variable with similar values only in the case of propane without cyclone and ethylene with cyclone. At λ = 532, in the case of MWAA, similar MAC values have been found for both the propane series, while, for ethylene series, MAC values were slightly higher when cyclone was used and lower when not. Considering the optical data from PAX, a similar MAC for both fuels was found when the cyclone was present, while it slightly differed in the case of propane without cyclone, and it was much lower in the case of ethylene without cyclone. At λ = 405 nm, the MWAA responses for propane series were still in agreement while the ethylene series showed a higher MAC value when using the cyclone, and a lower MAC value without using it. PAX returned a different MAC value for each of the four conditions. To summarize, if only series with cyclone are considered, MAC values show only small differences depending on the

fuel,larger in the case of PAXs. The ethylene series without cyclone showed the lowest MAC values of the whole data-set: the most likely reason for this difference is the presence of super-micrometric particles (see Sect 3.2.1 and Fig. 6) when the cyclone was not used. With MWAA, the MAC values turned out to be the same in all the runs but the case of the ethylene data collected without the cyclone. With the PAXs analysis, MAC values turned out higher in the series with cyclone, this happened at all the three wavelengths and for both fuels. Since PAXs data showed a higher variability in MAC values, photoacoustic measurements are supposed to be more sensitive to particle size than filter based MWAA analysis.

Figure 15: since you do not correct for truncation error, this comparison between PAX and MWAA is highly questionable, especially for the experiments without cyclone.

As discussed above, we think that the truncation error is not an issue in the case of photoacoustic measurements. This is true especially in the case of experiments with cyclone, where particles are smaller than 1 micron.