## <u>Review on manuscript ar-2025-15</u> "<u>Mass concentration intercomparison of</u> <u>soot generated with Mini-Cast"</u>

This article compares several experimental methods for determining, directly or indirectly, the mass concentration of soot particles produced by a propane/air premixed flame. A mini-CAST generator was used and the total mass concentration of particles produced by this generator was determined by sampling and weighing on filters. The mass concentrations thus obtained for different operating points of the mini-CAST are then used to compare the mass concentrations determined by four methods with very different detection principles (thermo-optical analysis, analysis of the charge carried by the particles, optical analysis and extrapolation from an analysis of the particle size distribution obtained by electrical mobility analysis).

Although the experimental developments appear to have been carried out with great care and the results clearly presented, a number of questions remain concerning the interest of such an article for the scientific community, the generation of the mini-CAST and the <u>comparison of various soot characterisation instruments</u> that have already been the subject of numerous works:

- https://www.tandfonline.com/doi/epdf/10.1080/02786826.2010.482113?needAccess=true
- https://www.tandfonline.com/doi/epdf/10.1080/02786820701197078?needAccess=true
- https://doi.org/10.1089/ees.2014.0038
- https://doi.org/10.1021/es051228v

In addition, the assumptions associated with certain methods, and in particular the extrapolation from data obtained by the SMPS, are questionable, as is the transposition of the conclusions of this study to other sources of soot.

As it stands, this work, while of genuine technical quality, does not seem to me to be truly innovative, as it does not propose any new analytical strategies or corrections to be applied to the technologies investigated. What's more, the number of techniques is limited to the capacities of the two laboratories involved and does not allow us to cover a sufficient number of analysis technologies and instruments of the same technology in order to rule on possible sources of variability inherent in the different methods targeted.

I do not recommend this manuscript as a research article for publication in the journal 'Aerosol Research' and I invite the authors to submit this article in the form of a technical note.

Nevertheless, and in support of the quality of the technical work carried out and presented in this article, here are a number of comments that I feel are important to consider.

## Specific comments

- Abstract: The authors mention that SMPS is an 'offline' method for determining mass concentration. As SMPS performs an on-line analysis of the particle size distribution, I do not think it is appropriate to mention this technique as an 'offline' method. SMPS softwares are also generally capable of directly converting particle size distributions by number into size distributions by mass (assuming spherical particles with a constant density, which is of course not relevant for soot particles), so the measurement is indeed "online".
- 2. Experimental setup: SMPS specifications are missing, please add them;
- Line 68-69: the authors mention that the measurements were carried out 3 times and that the error bars in the graphs correspond to these repetitions, but the uncertainty inherent in the measurement process (in particular the measurement of mass concentration by weighing) is not evaluated, presented or discussed in the context of this comparison of methods;
- Figure 1: the impact of the transport line heated to 180°C, upstream of the filter sampler, on the determination of mass concentration by weighing and thermo-optical analysis was not discussed. One might wonder about a significant effect for samples with high OC/TC values. Have the thermograms been obtained and compared with and without this heated line to ensure that no volatile fraction is desorbed

under these conditions? This point is important as the sample is not heated for the line upstream of the dilution system;

- Still in connection with the impact of this line heated to 180°C, were SMPS size distributions or electron
  microscopy images taken before and after conditioning at 180°C? These questions are intended to shed
  light on the comparability of samples weighed on quartz filters and those characterised downstream of
  the dilution system;
- Has the actual dilution factor been evaluated for the different generation conditions? It is legitimate to wonder about possible particle losses within the dilution system and whether these losses differ according to the miniCAST settings. This point should be discussed and the uncertainty associated with determining this dilution factor should be taken into account when calculating the mass concentrations obtained downstream of the DEKATI diluter.
- Line 77: a heat exchanger is mentioned but not visible on figure 1, please add it;
- Line 77: what methodology (standard, standardised protocol) was used to determine the mass concentration from sampling on quartz filter? Has an assessment of the uncertainties (taking into account the uncertainties inherent in weighing and measuring the volume sampled) been carried out? If so, it should be added to the standard deviation associated with the three repetitions shown in Figures 4 to 8;
- Figures 2 and 3 are not useful, as principles of the PPS and MA 300 instruments have been documented elsewhere and the present article does not introduce any significant changes to the principle of these instruments;
- Lines 114 to 124 seem to me to go into too much detail and could concentrate on the fact that the PPS mass calibration constant was established essentially on automobile emissions with a relatively limited range of size distribution in numbers;
- **Figure 4 right:** the characterisation of the mini-CAST size distribution has already been the subject of numerous publications, so it does not seem useful to illustrate this with a figure (or at least to place this figure in an appendix to the article);
- With regard to **bimodal distributions**, are TEM images of the particles available in order to determine whether they are bimodal?
- Table 1:
  - The first line requires an explanation of the 1% mentioned after the mode at 24.5 nm. If this is a monomodal distribution, shouldn't it be 100%?
  - On what criteria do the authors conclude that the particle size distributions produced within the range of oxidation flow rate 1 to 1.35 L.min-1 are bimodal? Is monomodal smoothing totally unsuitable and on what criteria was bimodal smoothing preferred?
- **Figure 5:** have the measurement uncertainties of the total carbon mass concentration based on thermooptical analysis been determined?
- Line 196 : "we considered a so-called true density for the aggregates that varies depending on the considered point" → clarification of the method used to determine the mass concentration from the particle size determined by SMPS is required. Did the authors consider a single density value for the entire particle size distribution? If so, a discussion appears necessary and must be confronted with the notion of effective density of soot particles. Conversion models, including the fractal morphology of soot, have been developed and are available in the literature. Why not consider them in this article to achieve a conversion from number to mass size distributions?
  - o <u>https://doi.org/10.1016/j.carbon.2024.119197</u>
  - o <u>https://www.tandfonline.com/doi/full/10.1080/02786826.2019.1577949</u>
  - o https://www.sciencedirect.com/science/article/abs/pii/S0010218018304310
  - o https://www.sciencedirect.com/science/article/abs/pii/S0021850215000701
  - o https://www.sciencedirect.com/science/article/pii/S0021850223000769#sec5
- Line 211: the following sentence: "We report in table 2 the values obtained for the OC/TC ratios as determined by thermo-optical analysis and the corresponding evaluated true densities, that were used to evaluate the SMPS mass concentrations reported on Fig. 7" is not clear;
- Lines 213-221: this paragraph questions the capacity of the mini-CAST to be act as a reference generator, the authors stating at the beginning of their article that they wanted to use this generator for its stability.

If the composition of soot varies from one mini-CAST to another, how can the conclusions of this article be generalised to other mini-CASTs

- **Table 2:** It is not easy to know whether the density values given in this table are calculated or determined to give the best agreement with the weighing measurement;
- Lines 224-228: this sentence is not easy to understand;
- Figure 8, left: error on the x-axis legend « Gravimteric »
- **Figure 8, right**: I don't think this figure is useful, as the OC/TC ratio can be mentioned in the left part of the figure.
- **Figure 8 right**: oxydation → oxidation