Answer to Referee 1

Review on manuscript ar-2025-15 "Mass concentration intercomparison of soot generated with Mini-Cast" > Reviewer comments to answer from authors on the revision of their article

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 comparison of various soot characterisation instruments 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.

Thank you for your deep and pertinent review. In the following answers, we have tried to enhance the clarity and impact of the demonstration of the scientific relevance of the paper. We have modified the title of the paper and brought some improvements. The new proposed title is "Evaluation of mass measurement techniques for soot with different size distributions and OC/TC contents"

REVIEWER ANSWER > see my comments hereafter

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".

It has been modified in the abstract.

REVIEWER ANSWER > perfect

- **2. Experimental setup:** SMPS specifications are missing, please add them; It has been added in the text.

REVIEWER ANSWER > perfect

- **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;

A paragraph has been added to the text to evaluate the gravimetric weighing based on the Iso 15767 standard.

REVIEWER ANSWER > thank for this addition

- **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;

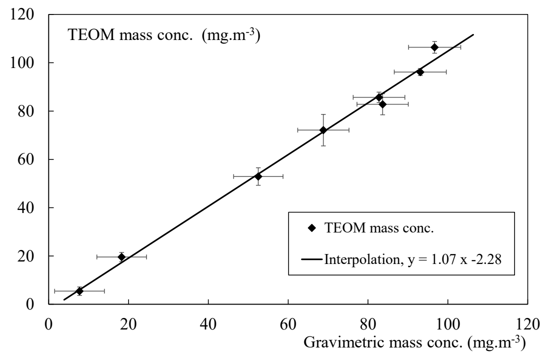
Indeed, this is an important point, and the aim of this experiment was to test the filter sampling procedure that is applied in direct exhaust measurements, particularly for ship engines. In such exhaust measurements where temperatures are over 200 °C depending on the sampling location, the main issue is to avoid condensation of moisture and semi-volatile species. The procedure follows the ISO 8178 standard, which specifies a target temperature of 180 °C for the sampling line and a maximum residence time of 3s. With a sampling flow rate of 7.7 L/min, a line length of 1 m, and an inner diameter of 6 mm, the aerosol residence time is approximately 0.22 s. This duration is extremely short compared to the 173 s of the first temperature plateau at 140 °C on the thermogram shown below, which is required to vaporize only 1.3% of the total carbon mass.

In the revised presentation of the results from this study, the TEOM measurement is considered the reference. All measurements are performed downstream of the dilution system (TEOM, SMPS, PPS, MA300), except for the gravimetric measurement, whose results are validated by comparison with the TEOM measurements, as discussed later in the article.

REVIEWER ANSWER > thank you for this convincing answer, **nevertheless uncertainties remain regarding the dilution rate and possible particle losses, thereby calling into question the relevance of comparing the mass concentrations** determined by **filter weighing** with the mass concentrations obtained using **characterisation method downstream of the dilution system**.

Answer: Dilution systems can indeed introduce losses through various mechanisms. However, many instruments are unable to handle high concentrations, and only dilution systems make their use possible.

The goal of this study is to compare different experimental techniques downstream the diluter. According to EN16450, the reference method for PM mass concentration can only be offline filter-based gravimetric measurement (filter weighting). That's the reason why we used a non-diluted filter-based approach, and to access the OC/TC information. Then we compared the mass concentration obtained by both this gravimetric method and the TEOM measurement system, after dilution, to establish the equivalence, as illustrated in figure 5 of the submitted paper, below. This demonstrates that the two sampling branches in our experiment setup are equivalent, and that the TEOM is equivalent to the reference offline gravimetric method.



- 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;

All instruments are positioned behind the dilution system, so at iso sampling conditions, except the filter mass sampling. We provided elements on the previous question/remark, and believe they clarify the comparability.

REVIEWER ANSWER > see my previous comment

Answer: we believe that we have answered this point above.

- 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.

All the instruments used in this study are positioned downstream of the dilution system and are therefore all potentially affected by biases introduced by the dilution process, with the exception of the gravimetric filterbased sampling, as previously mentioned.

REVIEWER ANSWER > the authors' reponse confirms my questions about the **lack of relevance of comparing the mass concentrations** determined by **filter weighing** with the mass concentrations obtained using **characterisation methods downstream of the dilution system**.

Answer: we believe that we have answered this point above.

- **Line 77:** a heat exchanger is mentioned but not visible on figure 1, please add it;

Thank you. It has been added to the article. REVIEWER ANSWER > perfect

- **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;

Indeed, the standard procedure implemented is already described in the manuscript, but we now provide a more detailed explanation of the uncertainty calculation, which is based on the combined uncertainties associated with weighing and sampled volume measurement. For this uncertainty evaluation, we followed ISO 15767 to assess the uncertainty related to weighing, evaluated the uncertainty on the sampled volume, and applied uncertainty propagation to estimate the overall uncertainty, which combines measurement uncertainty with the uncertainty associated with repeatability based on the three repetitions. The error bars shown in Figure 5 for the "Gravimetric mass conc." result from this analysis, while the uncertainty in the determination of "Total Carbon (TC) mass conc." determined through the thermo-optical analysis is estimated at 13%, as established through the literature review.

REVIEWER ANSWER > thank for this valuable addition to the manuscript

- **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.

We wanted the reader to find all the operating principles of the devices used in this article without having to resort to additional bibliography.

REVIEWER ANSWER > I reiterate my comment on the lack of interest in explaining yet again the principle behind these instruments, which have been detailed many times in the past. In my opinion, a short, concise article always has more impact.

The two figures also help the reader understand that these are mass measurements obtained using very different principles—optical attenuation for the MA300 and surface electrical charge for the PPS. From this perspective, this seems important to us as well.

- **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; It has been modified in the text. REVIEWER ANSWER > perfect

- **Figure 4 right:** the characterization 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);

In the bibliography, the intermediate points have not been investigated. It is interesting to show the intermediate size distributions.

REVIEWER ANSWER > Indeed, these intermediate points could justify the interest of this figure if TEM images were associated (see my next comment).

- With regard to **bimodal distributions**, are TEM images of the particles available in order to determine whether they are bimodal?

No TEM images have been done. The study was conducted at eight different oxidation air flow settings and involved several instruments or analytical protocols requiring significant analysis time. The analysis and statistical processing of the large number of TEM images that would have been required did not seem reasonable to us, although such an approach would indeed have enriched the experimental investigation.

REVIEWER ANSWER > I was not suggesting characterising all of the MiniCAST's operating points using TEM, but rather those operating points that give rise to a bimodal particle size distribution, as I believe this is important for understanding the origin of this bimodality.

Indeed, taking TEM images would make it possible to observe the coexistence of two particle populations with different size distributions. This would require a large amount of picture analysis to produce statistically relevant information. However, the aim of this study was not to investigate or explain this bimodality—already observed in the analysis of the SMPS distributions—but rather to focus on the mass concentration evaluations of the instruments, for different particle size distributions and different OC/TC ratios, while exploring a wide operating range of a soot generator.

- 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%?

Thank you. It has been corrected in the article REVIEWER ANSWER > you're welcome

- 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?

The procedure implemented to perform a least-squares minimization aimed at representing the measured distribution as the sum of two log-normal distributions is presented on the left-hand side. On the right, it is clearly visible that a single log-normal distribution fails to accurately represent the measured distribution produced by the miniCAST at the 1.25 L/min oxidation air flow condition.

REVIEWER ANSWER > thank you for this convincing answer

- Figure 5: have the measurement uncertainties of the total carbon mass concentration based on thermooptical analysis been determined?

For the Total Carbon (TC) mass concentration based on thermo-optical analysis, Sipkens et al. (2024) evaluated the TC uncertainties to 13%, based on bibliography. Brown et al. (2017) reported combined standard error below 13% for a reproducibility between four laboratories and Schmid et al. (2001) reported an uncertainty on their TC measurement between 6.7% and 11%. We therefore decided to take the conservative value of 13%, as reported in Figure 5.

REVIEWER ANSWER > I still do not understand what the authors mean by an uncertainty estimate based on bibliography.

This part of the paper has been removed in this last submitted version of the manuscript, based on the third reviewer's remarks. The OC/TC analyzer is used only for the evaluation of the OC/TC ratio. The measurement of uncertainties on the Total Carbone concentration TC has no longer to be discussed in the paper.

- 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?

- https://doi.org/10.1016/j.carbon.2024.119197
- https://www.tandfonline.com/doi/full/10.1080/02786826.2019.1577949
- https://www.sciencedirect.com/science/article/abs/pii/S0010218018304310
- https://www.sciencedirect.com/science/article/abs/pii/S0021850215000701
- https://www.sciencedirect.com/science/article/pii/S0021850223000769#sec5

Indeed, other approaches have been developed to estimate an "effective density" of soot particles, considering their fractal nature. And based on an analysis of various data sets available in the literature, Olfert and Rogak proposed a model that expresses the effective density as a function of mobility diameter. This model allows the evaluation of the decrease in effective density with the mobility diameter of soot aggregates and is expressed using two parameters: a negative exponent and a reference density defined at a mobility diameter of 100 nanometers. The data used to develop this model originates primarily from studies of particles produced by internal combustion engines or gas turbines, and a value of 510 kg.m^{-3} for the effective density at 100 nm was proposed for this model. However, Yon et al. reported higher effective density values at 100 nm for the miniCAST and reported values ranging from 1100 to 765 kg.m^{-3} for oxidation air flow rates between 1 and 1.5 L/min. Although only three operating points were investigated, the decrease in effective density with increasing oxidation air flow rate was found to be linear. And we observed that the decay in OC/TC with oxidation air flowrate is also linear. We therefore conducted a 'best fit' identification procedure to obtain the values of $_{eff,100}$, the density at 100 nm that would allow us to recover the TEOM reference mass concentrations from the SMPS analysis together with the Olfert and Rogak model. We obtained a set of values $_{eff,100}$ "Best fit analysis" (see Fig. 7 right) and observed that the change in this density at 100 nm with the OC/TC ratio that can be represented by a linear interpolation, expressed as:

$\rho_{eff,100}(x) = 932 + 661 \cdot x$ (4)

with x the OC/TC ratio.

Figure 7 – Evolution of the OC/TC ratio as a function of the oxidation air flowrate (left). Identification of $_{eff,100}$ as a function of OC/TC ratio.

From this Equ. (4), interpolated values of $_{eff,100}$ can be evaluated. These effective densities at 100 nanometers determined through this procedure are reported in Table 2, and the corresponding mass concentrations, labeled "SMPS effective density," are shown in Fig. 8: the use of this effective density model combined with the measured size distributions results in a good estimation of the mass concentration.

Figure 8 – Comparison of gravimetric mass concentrations with evaluations based on PPS and SMPS measurements.

REVIEWER ANSWER > I reiterate my incomprehension regarding the authors' decision not to consider in their approach the fact that the effective density is a function of both the OC/TC ratio **and the electrical mobility diameter of the particles**. Either I have not yet understood their approach, or as it stands, even if the agreement between the measurements seems acceptable, this approach only proposes a conversion factor that is of very limited interest to readers because it is too dependent on the experimental conditions considered in this study. I don’t see the interest to develop a so complex approach, trying to include OC/TC ratio in this conversion if the fractal morphology of soot particles is not considered.

This comment indeed indicates a misunderstanding. The approach developed here does in fact take into account both the OC/TC ratio and the mobility diameter for the calculation of the effective density, by applying the model proposed by Olfert and Rogak (Olfert and Rogak, 2019), with the equation (5), the effective density is a function of the mobility diameter: $\rho_{eff} = \rho_{eff,100}\left(\frac{d_m}{100}\right)^{-0.52}$ and with $m = \rho_{eff} \cdot \pi d_m^3/6$, with dm the aggregate mobility diameter.

In this model by Olfert and Rogak, $\rho_{eff,100}$ is the effective density for aggregates with a 100 nm reference scale. We proposed to consider that this effective density could be a function of the OC/TC ratio, and the analysis we conducted shows that a linear interpolation (Equation 6) provides good results.

We have modified the conclusion to take this comment into account and to better highlight the dual dependence on particle morphology and the OC/TC ratio in the assessment of mass concentration based on size distributions measured by the SMPS.

- **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;

Indeed we clarified this sentence. But first we mentioned earlier in the text that the density is considered as constant for a given point of the mini cast: “For this evaluation, we initially considered a constant so-called “true density”, independent of the particle mobility diameter, but varying with the OC/TC ratio and therefore dependent on the selected operating point of the miniCAST”

Then the sentence itself is clearer and we explain that the true density is calculated.

“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 calculated with the Park mixing model (Eq. 2), that were used to evaluate the “SMPS true density” mass concentrations reported on Fig. 7”

- **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

Indeed, the “S” in miniCAST was introduced by its designer to indicate "standard." It is therefore a generator that is intended to be reproducible. However, the setup installed downstream of the generator can lead to variations in the soot size distributions, due to mechanisms such as agglomeration and deposition. Moreover, to our knowledge, no intercomparison between different miniCAST generators is available, so strict equivalence from one generator to another cannot be guaranteed. Nevertheless, we emphasize here the generator’s stability, which is well established: it can produce soot with stable concentration and size distribution over relatively long periods, thus enabling gravimetric measurements and downstream characterization under dilution, using a steady source.

REVIEWER ANSWER > I had noticed the subtlety of the ‘S’ in the name MiniCAST, and I would even add that it is not just the S but also the T. I am not questioning the commercial arguments, but rather the fact that the variability inherent in this system (demonstrated and mentioned several times by the authors) may call into question the applicability of the results obtained for other systems of the same type (in particular the OC/TC ratio, which seems to vary significantly).

Indeed, the focus of the study is not on the generator itself, but on the quantification techniques. We chose this generator because it has been the subject of several studies in literature showing that it produces soot similar to that emitted in fire situations or from engine exhaust, both of which are of interest. However, the conclusions of the study indicate that the mass concentration quantification techniques can also be applied to other carbonaceous aerosols that can be produced by other generators.

- **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.

Indeed, we now detail that "true densities calculated with the Park mixing model (Eq. 2)". And it is also mentioned in table 2 itself. Since an effective density is also used now, it is also mentioned that it is calculated with Equ. 4.

REVIEWER ANSWER > see my previous comment on Line 196

We responded to this comment, that indicated a misunderstanding. The approach developed here does in fact consider both the OC/TC ratio and the mobility diameter for the calculation of the effective density, by applying the model proposed by Olfert and Rogak (Eq. 5 for the mobility diameter, and Eq. 6 for x=0C/TC ratio dependance).

- **Lines 224-228**: this sentence is not easy to understand;

Indeed, we tried to clarify, and modified the sentence, as proposed below

The measured values of the OC/TC organic fraction contained in soot compare well with those obtained in the previous study by (Marhaba et al., 2019a) for equivalent operating conditions of the Mini-CAST. For the considered points named CAST1, CAST2 and CAST3 in Marhaba et al. study (CAST3 corresponds to 1 L/min, CAST2 to 1.15 L/min and CAST1 to 1.5 L/min of oxidation airflow), Marhaba et al. reported OC/TC ratio values of 87%, 46.8% and 4.1%, respectively. In the same operating conditions, we measured corresponding OC/TC values of respectively 56.7%, 46 % and 6.2%. For the point CAST3, our measured value is significantly lower, while the two other points are coherent. However, as highlighted by (Moore et al., 2014), soot production conditions can vary with parameters other than the overall carbon/oxygen ratio of the flame, and variations in mode size or OC/TC ratio have already been observed between different studies using different Cast or Mini-CAST generators.

REVIEWER ANSWER > this point supports my previous comment on the reproducibility of the MiniCAST (see my previous comment regarding the reference aspect of the MiniCAST).

We responded to this comment. We mentioned that the focus of the study is not on the generator itself, but on the quantification techniques.

- **Figure 8, left**: error on the x-axis legend « Gravimteric »

Thank you. It has been modified. REVIEWER ANSWER > you're welcome

- **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.

We understand your recommendation to limit the number of figures. However, to maintain the clarity, we were not able to modify the manuscript in this direction. REVIEWER ANSWER > In my opinion, a short, concise article always has more impact.

We have indeed removed the corresponding figure in the newly submitted version. The OC/TC values for each study point are provided in Table 2.

- **Figure 8 right**: oxydation ·oxidation :

Thank you. It has been corrected in the article. REVIEWER ANSWER > you're welcome

In conclusion, I would like to thank the authors for their hard work in responding to all of my comments. Their responses confirm their expertise, but I remain very sceptical about the value of publishing this work in the form of a scientific article. The most limiting points remain, even in this revised version and as mentioned by reviewer R3, in the choices made by the authors in the experimental methodology (which does not allow the effects of the generation conditions on the properties of the soot to be dissociated) but also in certain assumptions considered in the analysis and comparison of the results.

I therefore still do not recommend this article for publication in Aerosol Research, but strongly encourage the authors to submit it to a journal that publishes technical papers.

We thank the reviewer for the comment. This study aims to provide tools for field studies. Conducted under laboratory conditions, it allowed us to improve our understanding of mass metrology of carbonaceous particles using different measurement techniques. It thus opens perspectives for studies in which mass concentrations vary over time and where the reference gravimetric method would not be applicable. In terms of advancement, we have proposed an extension of Olfert's model to account for the dependence of the effective density at 100 nanometers on the OC/TC ratio. In the revised version, we will highlight this new aspect in conclusion. In summary, we thank the reviewer for the thorough work which helped to improve the manuscript and the analysis of the data.

Answer to Referee 2

Reviewer: Verify that the figure references in the text are using the numbering from the revised version. At least lines 259, 273, 308 and 310 seem to be referring to the wrong figure. Figure 8 caption refers to comparison against gravimetric mass concentration, but I believe this should refer to TEOM in the revised manuscript.

Answer: Indeed, a number of numbering errors remained. We hope that we have improved the manuscript as a whole, including the referencing of equations and figures. Thank you for your careful review.

Answer to Referee 3

Reviewer: I am satisfied with the revisions made so far; however, I believe the article still requires additional work and significant text improvement. I recommend that the manuscript be reconsidered after major revisions.

Answer : In the submitted version, to avoid any confusion between TC and PM, which was indeed a source of misunderstanding and possible error for the reader, we removed TC information from Figure 5, and considered he OC/EC analyzer only for the determination of OC/TC ratio.

Furthermore, the text has been revised, particularly regarding the MA300 aethalometer, where we focused on updating the principles for evaluating mass concentration based on the actual features of the MA300 used in this study. We hope that these improvements will be considered satisfactory. We thank the reviewer for the thorough work, which helped to improve the quality of the manuscript and the analysis of the data.

Reviewer: the manuscript remains unclear regarding whether TC measurements are treated as actual determinations of particle mass concentration or merely as a parameter describing particle composition (OC/TC content, as suggested in the title). In line 137, the statement "The second method for determining the mass concentration of generated soot is based on the Sunset Lab OC/TC" is incorrect. This method can only measure the mass of carbon atoms in PM, not the total particle mass as achieved by gravimetric methods. While it is possible to approximate PM mass from the carbonaceous aerosol (CA) mass, as I mentioned in my previous review, this requires the OC–OA ratio.

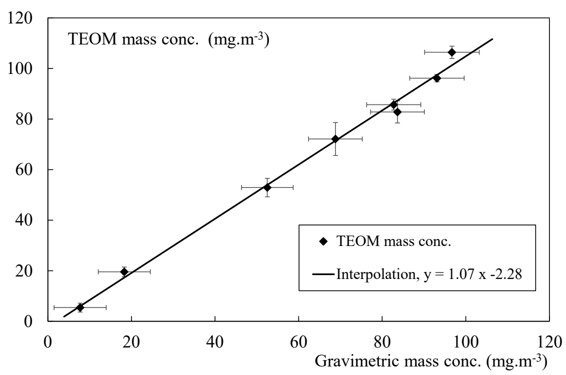
Answer:
In the case of particles that are predominantly carbonaceous, such as those produced by the CAST, we had indeed considered that TC could serve as an estimate of PM. This approach is, however, ambiguous and methodologically approximate. We therefore prefer to remove any reference to TC as an estimate of PM and use the OC/EC analyzer solely for evaluating the OC/TC ratio.

The original sentence "The second method for determining the mass concentration of generated soot is based on the Sunset Lab OC/EC field instrument (Sunset Laboratory), which has been used for both the measurement of the OC/TC ratio"
Has been modified to:

"The Sunset Lab OC/EC field instrument (Sunset Laboratory) has been used for the measurement of the OC/TC ratio"

Reviewer: the mass concentration of TC cannot be equal to the PM mass concentration because TC accounts only for the carbon fraction and excludes other components. Therefore, I do not understand how Figure 5 presents these values as equivalent. It may serve as a reasonable estimate in the case of a CAST mini-CAST burner under controlled conditions, but it will always be lower than the actual PM mass.

Indeed, we are well aware that TC and PM can only be considered equivalent in the theoretical case of particles composed solely of carbon. Figure 5 may create confusion for the reader, which should be avoided. The purpose of the thermo-optical method was to provide information on the OC/TC ratio, which is an important factor for optical methods. We therefore simply propose removing the measured TC information from Figure 5, in order to compare gravimetric PM with the TEOM by reversing the axes as suggested.



Reviewer: the mass concentration of TC cannot be equal to the PM mass concentration because TC accounts only for the carbon fraction and excludes other components. Therefore, I do not understand how Figure 5 presents these values as equivalent. It may serve as a reasonable estimate in the case of a CAST mini-CAST burner under controlled conditions, but it will always be lower than the actual PM mass.

We believe that we have responded to this remark.

Reviewer: Additionally, the statement in line 143, "The IMPROVE A protocol (Chow et al., 2001) has been used to reduce these pyrolytic conversions," is misleading and should be clarified. A more accurate statement would be: "Filters were analyzed using the OC/EC method based on the IMPROVE_A thermal protocol."

Indeed, the sentence has changed.

Reviewer: the IMPROVE_A thermal protocol was developed as part of the U.S. Interagency Monitoring of Protected Visual Environments (IMPROVE) program and was not designed to minimize the formation of pyrolytic carbon during analysis. In fact, reducing pyrolytic carbon was the main driving force behind the development of the EUSAAR_2 thermal protocol. It is also concerning that the response to my previous review included the sentence: "We estimated only the OC/TC using the Sunset Semi-Continuous Organic Carbon/Elemental Carbon (Sunset OC/EC) aerosol analyzer that utilizes the modified National Institute for Occupational Safety and Health thermo-optical method to determine total carbon (TC), organic carbon (OC), and elemental carbon (EC) at near real-time."

This reference to the NIOSH thermal protocol is inconsistent with what is written in the manuscript, and with the analysis shown in the figure provided in your response, which clearly does not follow NIOSH but IMPROVE_A. This raises a legitimate question as to whether the response text was critically reviewed or if automated tools (e.g., AI) were used without proper verification.

Indeed, it was the IMPROVE_A protocol that was used, and not the NIOSH protocol. The mentioned sentence comes from the article "Characterization of the Sunset Semi-Continuous Carbon Aerosol Analyzer" (full ref. below), which we referred to during the review response phase. It was present in full in the intermediate versions and should have been removed; we apologize that it may have given the impression of using AI.

Jace J. Bauer, Xiao-Ying Yu, Robert Cary, Nels Laulainen & Carl Berkowitz (2009) Characterization of the Sunset Semi-Continuous Carbon Aerosol Analyzer, Journal of the Air & Waste Management Association, 59:7, 826-833,

Reviewer: I agree with the explanation that heating the second branch of the system (filter sampling line) does not significantly affect the results and that condensation would likely introduce a larger error due to particle losses. However, it is essential that this consideration is explicitly mentioned in the manuscript. Furthermore, the authors should clearly state that the sampling system was designed and operated in accordance with ISO 8178 requirements.

Indeed, we have specified: « The sampling system was designed and operated in accordance with ISO 8178 requirements, which specify standardized procedures for measuring exhaust emissions from internal combustion engines"

Reviewer: I agree with the additional description of the microAeth MA350 operating principle; however, this explanation is based on Drinovec et al. (2015), which was developed for the Aethalometer AE33. Although the general principle is similar, the filter tape material differs between the two instruments. Therefore, it is important to focus only on the values calculated specifically for the MA350. Parameters reported for the Aethalometer AE33 - such as the multiple scattering coefficient and leakage - are not relevant in this manuscript and are confusing the reader. Please include and discuss only the values of C, MAC, and leakage that were actually applied in your analysis. Please discuss their uncertainties (also in regards to OC/TC content) as well.

Indeed, the theoretical explanation provided was based on Drinovec's article and does not correspond to the MA300 used in this study. We have therefore revised this section to reference only the values used in the MA300. Additionally, we have adjusted the notations to align more closely with recent publications on the subject.

Furthermore, we have expanded the discussion on the uncertainty assessment of this type of instrument, particularly regarding the potential variability of MAC values depending on the composition or morphology of the collected aerosols, by referring to recent publications.

Chakraborty, M., Giang, A., Zimmerman, N., 2023. Performance evaluation of portable dual-spot micro-aethalometers for source identification of black carbon aerosols: Application to wildfire smoke and traffic emissions in the Pacific Northwest. Atmos Meas Tech 16, 2333–2352. https://doi.org/10.5194/amt-16-2333-2023

Sipkens, T.A., Corbin, J.C., Chen, K., Rivellini, L.-H., Abbatt, J., Olfert, J.S., 2025. Quantitative uncertainty and post-processing for micro-aethalometers measuring black carbon. https://doi.org/10.5194/egusphere-2025-4209

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Reviewer: According to EN16450, the reference method for PM mass concentration can only be offline filter-based gravimetric measurement (filter weighting). Candidate automatic measurement systems (AMS) for equivalency assessment include TEOM, beta-ray attenuation, light scattering, etc. To strengthen your analysis, I recommend plotting the offline gravimetric method on the x-axis in Figure 5 and comparing it to TEOM on the y-axis. This will demonstrate two critical points: (1) That the two sampling branches in your experiment are equivalent. (2) That TEOM (as a candidate AMS) is equivalent to the reference offline gravimetric method. Only after establishing this equivalency should TEOM be compared with all other online mass concentration measurement methods as you did in Fig. 8 and 9.

Indeed, we followed this advice, which improves both the correctness and the readability of the text.

Reviewer: Finally, I still do not understand how TC values can be equal to PM (TEOM) values—please refer to my comment in the first section of this review for details.

We have updated Fig. 5, so the reader will have no ambiguity: the OC/EC analyzer is used to obtain the OC/TC ratio.