We thank the referees for their constructive comments and suggestions made to improve and clarify our manuscript. Our responses are given below. For clarity, reviewer comments are given in black, our responses in blue, and changes to the text of the manuscript in green. All the received reviewer comments are numbered as are their respective author replies – this way, cross-references in the present rebuttal letter are easily possible. We have also numbered individual points within longer reviewer comments to have clear and structured responses, without changing the original wording.

RC1: The manuscript examines the seasonal difference between AMS-derived organic carbon (OC_{AMS}) and thermal-optical OC (OC_{TOT}) measurements from three Central European sites using AMS / ACSM and thermal-optical transmission methods. These comparisons are relevant to long-term aerosol monitoring, and the study has the potential to provide useful insights into differences between these two commonly used OC measurements approaches However, there are several important issues that should be addressed before the manuscript is suitable for publication.

We thank the reviewer for the careful evaluation of our manuscript and for the constructive comments and suggestions. We appreciate the reviewer's valuable feedback, which has helped us improve the clarity, robustness, and interpretation of our results. We have carefully addressed all comments and revised the manuscript accordingly, as detailed in our point-by-point responses below.

Major comments:

The observational finding of substantial wintertime differences between OCAMS and OCTOT appears reasonably robust, particularly because the agreement between online and offline TOT measurements suggests that the large winter discrepancies are unlikely to arise solely from filter-analysis analytical artifacts. However, the manuscript proceeds from this observational result to relatively specific mechanistic interpretations involving coal combustion, refractory organics, dust transport, BrC, and ultimately an OA threshold for identifying conditions under which OCAMS maybe biased low. These interpretations are not yet sufficiently supported because the analyses do not adequately distinguish among instrumental effects, circular reasoning arising from shared variables, sampling differences, and genuine aerosol-composition effects. In several cases, the analyses are partly circular or do not directly test the proposed mechanisms. The manuscript should be revised by first rigorously establishing the robustness and causes of the inter-method differences and then providing independent evidence for the proposed explanations. Otherwise, the corresponding interpretations and conclusions should be removed or substantially toned down.

We thank the reviewer for this important comment. We acknowledge the concerns regarding the distinction between instrumental uncertainties and aerosol-composition effects, as well as the need for independent evidence to support our proposed arguments. Therefore, we have expanded the evaluation of instrumental and sampling-related uncertainties, included additional independent analyses, and clarified the limitations of the proposed OA:eBC warning indicator. We have also revised the discussion and conclusions to avoid attributing the observed discrepancies to specific mechanisms without sufficient evidence. These revisions are described in detail in our responses to the individual comments below.

R1C1: (1) A key example is the proposal of OA:eBC as a diagnostic threshold for the discrepancy between OCAMS and OCTOT. OCAMS is calculated directly from AMS OA as OCAMS = AMS OA/(OM:OC). Thus, the two quantities are intrinsically coupled through the shared AMS OA measurement. If the AMS underestimates OA for any reason, both OCAMS and OA will decrease simultaneously. Consequently, the observation that low OA is associated with low OCAMS/OCTOT cannot, by itself, demonstrate a source- or matrix-dependent effect. **(2)** This concern is further strengthened by Figure 8, which pools observations from three sites and two distinct seasons. FRY winter appears to occupy much of the low-OA:eBC/large-discrepancy region, whereas summer observations largely occupy the high-OA:eBC/small-discrepancy region. **(3)** The apparent relationship may therefore reflect differences among site-season groups rather than a continuous relationship within a given aerosol regime. In addition, the proposed OA:eBC threshold of approximately 5 appears to be visually selected rather than statistically established. **(4)** Given these concerns, it is important that the authors redo this analysis using independent source indicators. For example, examine against eBC, independently derived source tracers, PMF factors, PAHs, potassium, levoglucosan, elemental tracers, or other available source information. At minimum, the relationship should be demonstrated separately within each site and season and tested using a multivariable or mixed-effects model. Any threshold should be determined statistically and validated rather than inferred visually from the pooled dataset.

R1A1: We thank the reviewer for this comment. We would like to address this bigger comment in four sections as follows:

1) Mathematical coupling between OA:eBC and OC_{AMS}

We thank the reviewer for this important comment. We agree that OA:eBC and OC_{AMS} are not mathematically independent because OA_{AMS} contributes to both quantities. Therefore, if OA were under-recovered by the AMS, both OC_{AMS} and the measured OA:eBC ratio would decrease. OA:eBC is therefore not used as independent evidence for the cause of the OC_{AMS}-OC_{TOT} discrepancy. Importantly, our interpretation of the relationship between the OC_{AMS}-OC_{TOT} discrepancy and aerosol/air-mass conditions is supported by independent evidence, rather than by the OA:eBC ratio. Independent evidence is provided by the air-mass cluster analysis. These clusters were determined from backward trajectories and therefore do not depend on OA_{AMS}, OC_{AMS}, OC_{TOT}, or eBC. For the C1W/C2W air-mass trajectory clusters, the OC_{AMS}-$OC_{Offline}$ slopes were 0.38, 0.12, and 0.48 at MEL, FRY, and NAOK, respectively, compared with 0.51, 0.27, and 0.76 for the remaining clusters. Thus, at all three sites, the magnitude of the AMS-TOT discrepancy changed with independently classified air-mass conditions. This provides evidence independent of OA:eBC that the AMS-TOT discrepancy is associated with differences in aerosol/transport conditions. We nevertheless agree that trajectory analysis cannot uniquely separate source, composition, and transport effects, and we have therefore avoided assigning the discrepancy to a single mechanism.

We also examined whether the particularly low measured OA:eBC ratio at FRY could be explained by OA_{AMS} under-recovery, as suggested by the reviewer. As a sensitivity test, the measured mean winter OA:eBC at FRY was 1.12. Assuming 50% OA under-recovery would increase the ratio to 2.24, while assuming even 70% under-recovery would increase it to 3.73. Thus, substantial hypothetical OA under-recovery would still leave FRY with a comparatively low OA:eBC ratio. This calculation is not intended to determine the true OA:eBC ratio, neither to show that OA_{AMS} is unbiased nor to establish the source or cause of the discrepancy. It simply shows how strongly the measured ratio would change under hypothetical OA under-recovery.

We have clarified this point in Section 3.4.2 of the revised manuscript as follows: "It should be noted that OA:eBC and OC_{AMS} are not mathematically independent because AMS-measured OA contributes to both quantities."

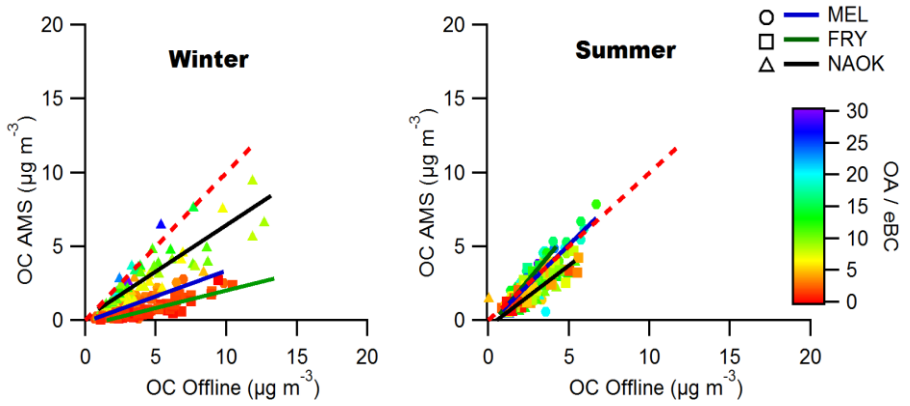
2) Interpretation of Fig. 8 and differences between sites and seasons

We also agree that the pooled pattern in Fig. 8 partly reflects differences between sites and seasons and therefore should not be interpreted as explaining a continuous relationship between OA:eBC and the OC_{AMS}-OC_{TOT} discrepancy within each individual site or season. The measured OA:eBC is rather proposed as a potential practical screening parameter rather than as an independent diagnostic of the cause of the discrepancy. Fig. 8 therefore shows the measurement conditions under which comparatively low measured OA:eBC and poor OC_{AMS}-OC_{TOT} agreement occurred together in this dataset, not to show that OA:eBC causes or independently explains the discrepancy. From a measurement perspective, a low measured OA:eBC ratio can result from genuinely low OA concentrations relative to eBC, underestimation of OA by the AMS, or a combination of both.. For the proposed screening purpose, knowing the exact cause is not required at the first step: in each case, a low measured OA:eBC identifies conditions in which AMS derived OC may warrant additional verification by an independent carbon measurement. Indeed, if AMS under-recovery itself causes the measured OA:eBC ratio to decrease, this does not invalidate its use as a warning flag; it represents exactly the type of condition for which additional verification of AMS derived OC would be warranted. The ratio also provides more context than the absolute OA_{AMS} concentration alone because it expresses measured OA relative to independently measured eBC. We have revised the manuscript Section 3.4.2 to clarify that this is a warning indicator to prompt further checking, not an independent means of identifying the mechanism responsible for the discrepancy. Its wider applicability will require validation against independent OC measurements in additional environments and datasets: "The pattern in Figure 8 partly reflects differences between sites and seasons and should not be understood as independent evidence for the cause of the AMS-TOT OC discrepancy, but rather as an indication of measurement conditions where the two methods may disagree."

We have also added the following study from Rovira et al. (2026) to Section 3.4.2: "A recent study by Rovira et al. (2026) used the eBC/OA ratio, calculated from Aethalometer-derived eBC and AMS/ACSM-measured OA at 17

European sites including NAOK and found that increasing eBC/OA ratios were associated with increasing contributions of primary relative to secondary organic aerosol, demonstrating the ratio's sensitivity to aerosol source composition."

To further address the reviewer's concern, we have also revised Figure 8 to show winter and summer observations in separate panels, with different symbols identifying the three measurement stations (MEL, FRY, and NAOK). This allows the seasonal and site-specific differences to be distinguished more clearly, while retaining the OA:eBC color scale. The figure caption has also been updated accordingly. These changes make the comparison easier to interpret and avoid confusion arising from pooling observations from different sites and seasons in a single plot:



"Figure 8: Comparison of OC_{AMS} (PM_1) with $OC_{Offline}$ ($PM_{2.5}$) as a function of OA:eBC ratio during winter and summer at all three stations combined. The symbols represent the three measurement stations (circles: MEL; squares: FRY; triangles: NAOK), while the color scale shows the organic aerosol to equivalent black carbon (OA:eBC) ratio. Solid lines represent the site-specific orthogonal distance regression fits, and the red dashed line represents the 1:1 relationship."

3) Statistical determination of the OA:eBC warning value

Regarding the OA:eBC threshold, we agree with the reviewer that the previously suggested value of approximately 5 was visual. We have therefore identified the threshold statistically, using our winter dataset. The results of this analysis have been added to the Supplement (Table S5) and as a text block (Text S2): "To examine whether measured OA:eBC could be used as a warning indicator for substantial OC disagreement, we performed receiver operating characteristic (ROC) analysis using the winter dataset. ROC analysis helps identify a cutoff value that best distinguishes between two groups of observations (Fawcett, 2006). We considered two cases, where $OC_{AMS}/OC_{Offline} < 0.7$ and < 0.5, meaning AMS-derived OC being more than 30% and 50% lower than offline thermal-optical OC, respectively. The analysis gave OA:eBC cutoffs of 6.77 and 5.09, with an area under the ROC curve (AUC) values of 0.961 and 0.966, respectively (Text S2, Table S5). AUC shows how well the two groups can be distinguished from each other, with values closer to 1 showing better separation. These cutoffs are based on our combined winter dataset and are suggested as preliminary warning values rather than fixed thresholds applicable to all measurement conditions."

Table S5: Results of ROC analysis using the combined winter observations from MEL, FRY, and NAOK (n = 203).

OC underestimation	OA:eBC cutoff	AUC	Sensitivity (%)	Specificity (%)
>30%	6.77	0.961	84.6	96.4
>50%	5.09	0.966	91.9	96.3

Section 3.4.2 of the revised manuscript reads as follows: "A receiver operating characteristic analysis of our winter dataset (n = 203) was performed to provide a statistically relevant threshold value. Results gave OA:eBC cutoffs of

6.77 and 5.09 for $OC_{AMS}/OC_{Offline}$ < 0.7 and < 0.5, respectively (Table S5). Based on the observations at these three stations, it is suggested that measured OA:eBC values below approximately 5 may be used as a warning indicator, where AMS derived OC should be interpreted with caution and, where possible, compared with an independent OC measurement. Although this value results from three different datasets, it is not implied that OC underestimation cannot occur above this value, or that it must occur below it nor is OA:eBC ≈ 5 proposed as a diagnostic criterion for AMS derived OC underestimation, but as a practical warning value that may encourage researchers to check their measurements. When possible a systematic comparison between OC_{AMS}, OC_{TOT} and OA:eBC at different environments and seasons is strongly recommended to further evaluate the potential variability of these quantities."

4) Independent evidence from source indicators

Following the reviewer's suggestion, we also examined individual independent source indicators, including eBC and levoglucosan, but no clear relationship with the OC_{AMS}-OC_{TOT} discrepancy was observed (for example as can be seen in the figure below). We do not interpret this absence of a relationship as evidence that aerosol source or composition is unimportant. The winter aerosol at these sites represents mixtures of several combustion sources, and individual tracers are not unique to one source. This is explained by an independent multi-tracer analysis of samples from the same campaign using PAHs, n-alkanes, and saccharide tracers (Arora et al., 2026b). At FRY, for example, levoglucosan and retene covaried with several high molecular-weight PAHs, showing substantial mixing of biomass-burning and coal-related signatures; MEL also showed a mixed combustion signature, whereas NAOK showed a clearer biomass-burning signature. Therefore, individual tracers cannot be expected to represent these mixed source conditions adequately. Indeed, the independent study used the combined chemical information in a PCA/k-means analysis and resolved distinct biomass-burning, coal/mixed-combustion, and background/biogenic source-related

factors. This supports our decision not to assign the AMS-TOT discrepancy to the concentration of any single tracer.

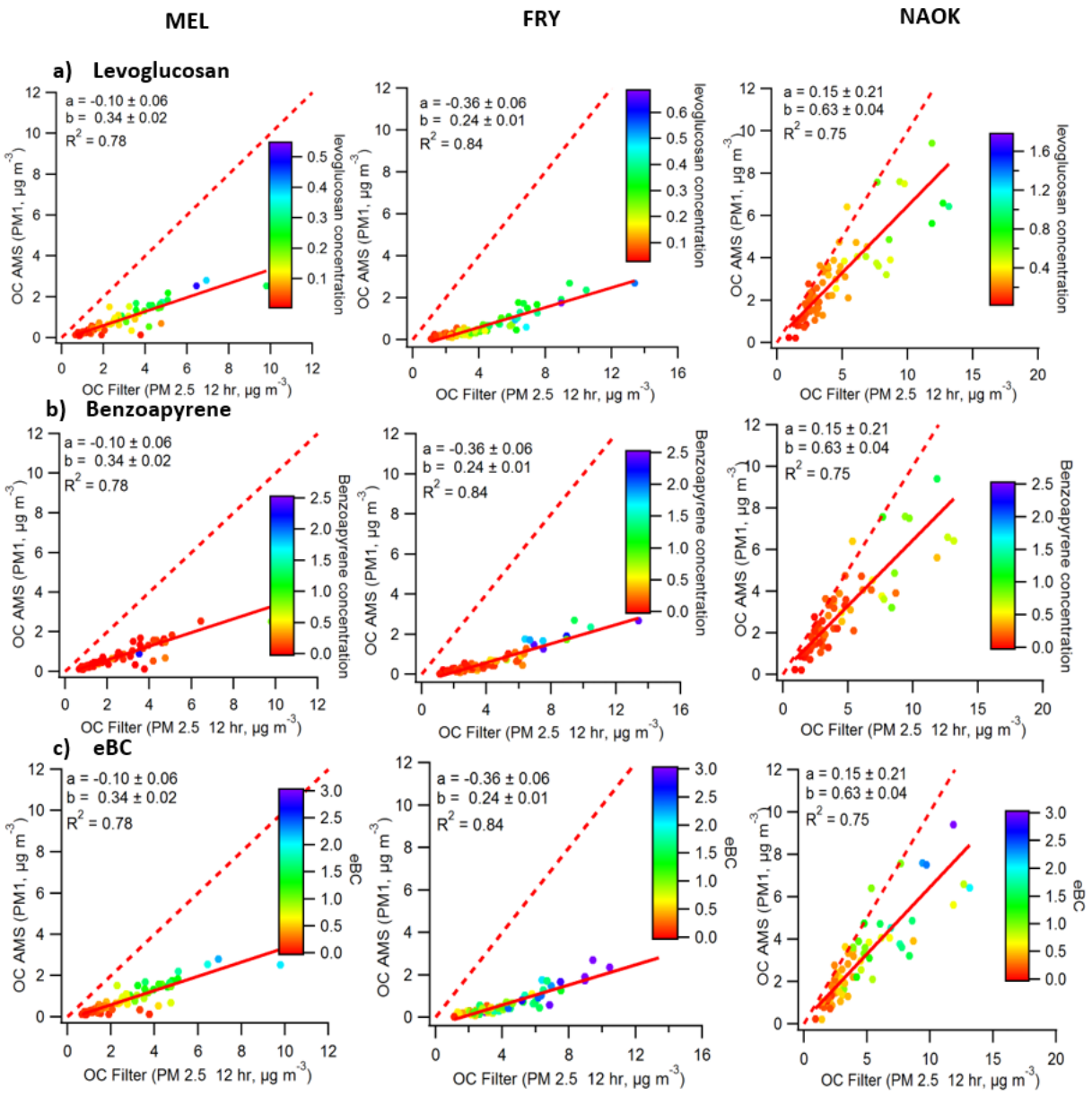


Figure: Comparison of organic carbon concentrations derived from AMS/ACSM measurements (OC_{AMS}, PM_1) and offline thermal-optical transmission measurements (OC_{TOT}, $PM_{2.5}$) at Melpitz (MEL), Frýdlant (FRY), and Košetice (NAOK) during the winter 2021 campaign. Data points are color-coded according to the concentrations of (a) levoglucosan, (b) benzopyrene, and (c) equivalent black carbon (eBC), as shown by the corresponding color scales. Solid red lines represent the orthogonal linear regression fits, with the regression coefficients (a and b) and coefficients of determination (R^2) provided in each panel. Dashed red lines show the 1:1 relationship. The comparisons were used to investigate whether individual, independently measured combustion-related tracers were associated with the observed differences between AMS/ACSM and TOT-derived OC.

We also do not use its OA:eBC ratio of 1.12 to identify FRY as coal influenced. The low measured OA:eBC at FRY is therefore described only as being consistent with the low OA:eBC ratios previously reported for coal combustion (Bond et al., 2004; Lin et al., 2023), rather than as evidence used to assign the source therefore Section 3.4.2 was

changed to: "FRY showed the lowest winter OA:eBC ratio (1.12), consistent with the low ratios previously reported for coal combustion(1.10 -1.30; Bond et al., 2004; Lin et al., 2023). However, this agreement alone cannot be used to identify the dominant source at FRY. Independent molecular-tracer analysis of samples from the same campaign suggested mixed biomass-burning and coal-related signatures at FRY (Arora et al., 2026b). MEL showed an intermediate winter OA:eBC ratio (3.06), while NAOK showed a higher ratio (10.21)."

R1C2: Instrument-related uncertainties also require more careful evaluation before the OC discrepancies are attributed primarily to aerosol composition or source characteristics. A major source of AMS measurement uncertainty is its collection efficiency for particles. While the manuscript discusses RIEs, high-molecular-weight compounds, ion transmission, organonitrates, and the Pieber effect, it does not quantitatively assess uncertainties associated with CE. This is particularly important because three different instruments were used—an HR-ToF-AMS at MEL, a C-ToF-AMS at FRY, and a ToF-ACSM at NAOK—and the largest OCAMS–OCTOT discrepancy occurs at FRY. The manuscript should clearly state how **(1)** CE was determined or assumed for each instrument, site, and season; whether composition-dependent CE was applied; what calibrations were performed; and **(2)** whether independent measurements (such as SMPS and particle mass concentration measurements) are available to inform about possible differences in inlet transmission, vaporizer performance, and overall mass quantification among the instruments.

R1A2: We thank the reviewer for this comment. We have addressed the points as follows:

1) Collection Efficiency, Calibrations and FRY discrepancy

We thank the reviewer for this important comment. To address uncertainties related to collection efficiency (CE) and instrument calibration, we have expanded Section 2.4 to clarify the CE treatment applied at each station, provide the available seasonal CE statistics, and specify the calibration procedures and relative ionization efficiencies (RIEs). The following text has been added: "For the HR-ToF-AMS at MEL, a composition-dependent collection efficiency (CDCE) was applied following Middlebrook et al. (2012), whereas fixed collection efficiencies of 0.5 were used for the C-ToF-AMS at FRY and the ToF-ACSM at NAOK, as described in Arora et al. (2026a). At MEL, the mean CE was 0.51 ± 0.08 (range: 0.45-0.99) during winter and 0.450 ± 0.001 (range: 0.45-0.85) during summer. Ionization efficiency calibrations were performed following ACTRIS recommendations using size-selected ammonium nitrate and ammonium sulfate (Freney et al., 2016). The RIE values for nitrate, sulfate, ammonium, OA, and chloride were 1.1, 1.2, 4.0, 1.4, and 1.3 at MEL; 1.1, 1.2, 3.3, 1.4, and 1.3 at FRY; and 1.1, 0.8, 3.6, 1.4, and 1.3 at NAOK, respectively (Arora et al., 2026a)."

2) We have additionally revised Section 3.5.2 to discuss the implications of these comparisons, particularly at FRY during winter, where the largest OC_{AMS}-OC_{TOT} discrepancy occurred. The following text has been added: "The independent AMS/ACSM-SMPS particle-volume comparisons (Table S6) gave regression slopes of 0.92, 1.46, and 1.41 during winter and 1.14, 1.20, and 1.56 during summer for MEL, FRY, and NAOK, respectively. At FRY during winter, where the largest OC_{AMS}-OC_{TOT} discrepancy occurred, the AMS/SMPS volume slope was 1.46 (R^2 = 0.83), suggesting that the C-ToF-AMS did not show an overall particle-volume deficit relative to the independent SMPS measurement. If anything, the deviation was in the opposite direction with the AMS-derived particle volume being higher than the SMPS-derived volume. Thus, an overall underestimation of aerosol volume alone cannot explain the observed OC discrepancy. Nevertheless, uncertainties associated with CE, particle transmission, and vaporization efficiency, particularly for individual aerosol components, cannot be excluded."Independent measurements (SMPS and particle mass concentration measurements)

To assess possible differences in overall aerosol quantification among the instruments, we have also included the independent AMS/ACSM-SMPS particle-volume comparisons previously reported in Arora et al. (2026a). These cover all three stations and both seasons. We have added the following text to Section 2.6: "The independent comparison of AMS/ACSM-derived particle volume with SMPS-derived particle volume was available for all three stations during both winter and summer campaigns, as previously reported in Arora et al. (2026a). It has also been included in the Supplement (Table S6) to assess possible instrument-related differences in overall aerosol mass quantification."

A table has been added to the Supplement with the following caption: "Table S6: Linear regression results for comparisons of AMS/ACSM-derived particle volume with SMPS-derived particle volume at Melpitz (MEL), Frýdlant (FRY), and Košetice (NAOK) during the winter and summer 2021 campaigns. The regression slopes (b), their reported uncertainties, and coefficients of determination (R^2) were obtained from Arora et al. (2026a)."

Season	Station	Slope (b ± uncertainty)	R^2
Winter	MEL	0.92 ± 0.00	0.96
Winter	FRY	1.46 ± 0.02	0.83
Winter	NAOK	1.41 ± 0.02	0.82
Summer	MEL	1.14 ± 0.01	0.91
Summer	FRY	1.20 ± 0.01	0.88
Summer	NAOK	1.56 ± 0.01	0.88

R1C3: The MEL winter volume closure shown in Figure S13 is useful, but it does not establish quantitative closure for the other instruments or seasons. In particular, no equivalent mass or volume closure appears to be presented for FRY, where the largest discrepancy is observed. Providing AMS/ACSM mass or volume closure for each site and season, especially FRY winter, would substantially strengthen the assessment of whether instrument-specific measurement biases contribute to the observed differences.

R1A3: We thank the reviewer for this suggestion. As discussed in detail in our response to R1C2, SMPS measurements were available for all three sites and both seasons, and the corresponding AMS/ACSM-SMPS particle-volume comparison was previously reported for these campaigns (Arora et al., 2026a). This comparison includes FRY winter, for which the AMS-SMPS volume slope was 1.46 ± 0.02 (R^2 = 0.83). Thus, despite FRY winter showing the largest OC_{AMS}-OC_{TOT} discrepancy, the C-ToF-AMS did not show an overall particle-volume deficit relative to the SMPS; rather, the deviation was in the opposite direction. To address the reviewer's comment directly and make this information readily available in the present study, we have now added the all-site/all-season AMS/ACSM-SMPS particle-volume comparison to the Supplement (Table S6) and revised Sections 2.6 and 3.5.2 accordingly.

R1C4: The OM:OC calculation and HR-versus-UMR data treatment need to be carefully explained as well. here appear to be internal inconsistencies between the equations provided in the manuscript and several values reported in Table S1. This directly affects the quantity being evaluated, so it is not a minor presentation issue. For example, the manuscript states: and . However, several numbers in Table S1 do not reproduce from these equations. For example, as printed in the Supplement:

MEL winter: f_44=0.16 -> OM:OC ≈ 2.16, not 2.64.

MEL summer: f_44=0.14 -> OM:OC ≈ 2.05, not 2.57.

NAOK summer: f_44=0.18 -> OM:OC ≈ 2.27, not 2.81.

Table S1 nevertheless reports 2.64, 2.57, and 2.81, respectively. These differences are large enough to substantially alter OCAMS.

R1A4: We thank the reviewer for identifying this inconsistency. The reviewer is correct that several OM:OC values previously reported in Table S2 were inconsistent with Eqs. (1) and (2) and had been incorrectly transferred to the table. The OM:OC values have now been corrected accordingly in Table S2. We would also like to clarify the HR versus UMR treatment, as the presentation in the previous version of Table S1 did not make this distinction sufficiently clear. As described in Section 2.4, MEL was measured with an HR-ToF-AMS, and the time-resolved OM:OC ratios obtained from the HR elemental analysis using PIKA and the Improved-Ambient method were used to derive OC_{AMS}. The f_{44} based OM:OC values at MEL were included only for comparison with the HR-derived values and were not used for the calculation of OC_{AMS}. At FRY and NAOK, where only UMR data were available, OM:OC was derived from f_{44} using Eqs. (2) and (3), and $OC_{AMS/ACSM}$ was then calculated according to Eq. (4), as described in Section 2.4. We have corrected the erroneous entries in Table S2 and revised the table and its caption to clearly separate the f_{44} derived OM:OC values from the HR-derived values at MEL:

Table S2: Campaign-mean f44, O:C, and OM:OC values obtained from the UMR f44-based parameterization (Eqs. 1-2), together with campaign-mean OM:OC obtained independently from HR elemental analysis (PIKA, Improved-Ambient method) at MEL. The values shown are means of the time-resolved values; the time-resolved values, rather than the rounded campaign means shown here, were used to calculate OC. At MEL, HR-derived time-resolved OM:OC was used for the OC_{AMS} concentrations reported in the main analysis, whereas the f44-derived values are provided for comparison. At FRY and NAOK, time-resolved f44-derived OM:OC was used to calculate $OC_{AMS/ACSM}$.

Season	Station	Mean f_{44}	O:C from f_{44}	OM:OC from f_{44}	HR OM:OC (PIKA)	OM:OC method used for OC_{AMS}
Winter	MEL	0.16	0.77	2.16	2.05	HR (PIKA)
Winter	FRY	0.24	1.11	2.61	–	f_{44}, Eqs. 1–2
Winter	NAOK	0.19	0.9	2.33	–	f_{44}, Eqs. 1–2
Summer	MEL	0.14	0.68	2.05	1.93	HR (PIKA)
Summer	FRY	0.15	0.73	2.11	–	f_{44}, Eqs. 1–2
Summer	NAOK	0.18	0.85	2.27	–	f_{44}, Eqs. 1–2

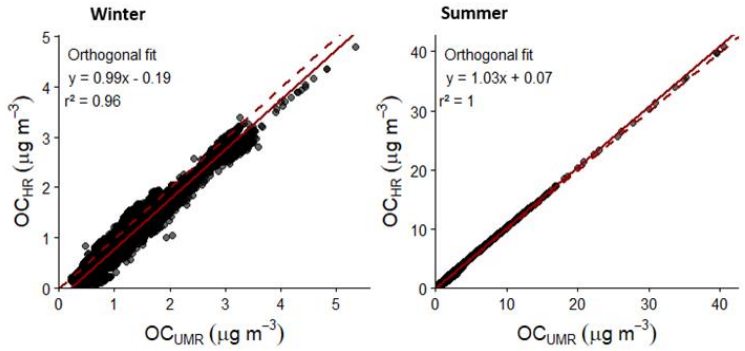
The revised table now reports the campaign-mean f_{44}, O:C, and OM:OC values and explicitly identifies the OM:OC method used for OC determination at each site. The caption further clarifies that these are means of the time-resolved values and that the time-resolved values, rather than the rounded campaign means, were used to calculate OC. The corrections concern the values reported in Table S2 and do not affect the $OC_{AMS/ACSM}$ data used in the analysis, the reported intercomparison results, or the conclusions of the manuscript.

R1C5: A related inconsistency occurs in Section 3.2, which is presented as an evaluation of the influence of mass spectral resolution on OCAMS, but Figure S3 actually plots HR OA against UMR OA, not derived OC. The figure caption says “organic carbon,” whereas the axes say OA. Agreement in OA does not establish agreement in OC because the HR and UMR OM:OC estimates can differ. It is important that the authors explicitly document the exact time-resolved equation used to calculate OCAMS for every site, identify which MEL calculation was used in Figures 5–8, reproduce Table S1, and then redo the HR-versus-UMR comparison using derived OC rather than OA.

R1A5: We thank the reviewer for pointing out this inconsistency. The reviewer is correct that the original Fig. S3 compared OA obtained from HR and UMR processing, whereas the figure caption incorrectly referred to organic carbon. The original OA comparison was intended to evaluate the consistency of the OA concentrations obtained from

HR and UMR processing before application of the respective OM:OC conversion. However, we agree that agreement in OA alone does not establish agreement in the subsequently derived OC because OM:OC is determined differently for the HR and UMR approaches. We have therefore retained the OA comparison and added a direct comparison of the resulting time-resolved HR and UMR-derived OC concentrations at MEL to Fig. S3:

Figure S3: Intercomparison of OC between UMR and HR during winter and summer at MEL using HR-ToF-AMS



To make the calculation fully explicit, Section 2.4 has been revised as follows:

At MEL, the availability of high-resolution mass spectra enabled elemental analysis using the Improved-Ambient (I-A) method (Canagaratna et al., 2015). AMS data were processed using SQUIRREL (version 1.65B) for unit mass resolution (UMR) and PIKA (version 1.25B) for high resolution (HR) within Igor Pro (version 9.02, WaveMetrics, Inc.). For the OC_{AMS} concentrations used in the subsequent intercomparisons at MEL (Figs. 5-8), the time-resolved OM:OC ratio obtained from HR elemental analysis in PIKA was used according to:

$$OC_{AMS,HR}(t) = \frac{OA_{HR}(t)}{(OM{:}OC)_{HR}(t)} \quad (1)$$

The UMR f_{44} based calculation at MEL was performed independently for comparison with the HR approach and was not used to generate the MEL OC_{AMS} concentrations. At FRY and NAOK, where HR elemental analysis was not available, OC was derived at each time point from the UMR f_{44} signal (Org44/Org) using the following empirical relationships:

$$(O{:}C)_{UMR}(t) = 0.079 + (4.31 * f_{44}(t)) \quad (2)$$

$$(OM{:}OC)_{UMR}(t) = 1.17 + (1.29 * (O{:}C)_{UMR}(t)) \quad (3)$$

$$OC_{AMS,UMR}(t) = \frac{OA_{UMR}(t)}{(OM{:}OC)_{UMR}(t)} \quad (4)$$

Thus, HR-derived time-resolved OM:OC was used for MEL, whereas time-resolved f_{44}-derived OM:OC was used for FRY and NAOK. The corresponding campaign-mean f_{44}, O:C, and OM:OC values are provided in Table S2; these means are reported for summary purposes only, whereas the time-resolved values were used to calculate OC.

Section 3.2 has also been made more explicit and expanded to state:

"Because agreement in OA alone does not account for differences in the HR and UMR-derived OM:OC ratios, the resulting OC concentrations were also compared directly (Fig. S3). The HR and UMR-derived OC showed strong agreement, with slopes of 0.99 (R^2 = 0.96) in winter and 1.03 (R^2 = 1.00) in summer."

Thus, despite the different approaches used to determine OM:OC, the resulting HR and UMR-derived OC concentrations at MEL were highly consistent. Table S2 and the Fig. S3 caption have also been revised accordingly to differentiate between the HR and UMR-derived quantities.

R1C6: More generally, important information needed to evaluate the comparability of the datasets is scattered throughout the manuscript and Supplement. A detailed summary table should therefore be provided that compiles the relevant information for all datasets analyzed in this study. At a minimum, the table should include the measurement sites, sampling period and season, instrument types and model, measured quantity, sampling size cut (PM_1 vs. $PM_{2.5}$), number of valid offline samples or paired observations used in the comparisons, and the native and/or averaged time resolution of the online measurements. For the AMS/ACSM measurements, it would also be useful to indicate the instrument configuration and any important differences among sites that may affect comparability.

R1A6: We thank the reviewer for this helpful suggestion. We agree that the information required to assess the comparability of the datasets was previously distributed between the Methods and Supplement. We have therefore added a new summary table to the Supplement (Table S1), compiling the measurement period, instrument type and model, measured quantity, aerosol size cut, time resolution, and number of valid samples/observations for each site and season:

Table S1: Overview of the measurement datasets and instrumental configurations used for method comparisons at Melpitz (MEL), Košetice (NAOK), and Frýdlant (FRY) during the winter and summer 2021 campaigns. A dash means that the corresponding comparison was not available.

Measurement	**MEL**	**NAOK**	**FRY**
Location	51.5333° N, 12.9333° E; 86 m a.s.l.	49.5734° N, 15.0805° E; 534 m a.s.l.	50.9407° N, 15.0698° E; 366 m a.s.l.
Sampling periods	Winter: 1 Feb-20 Mar 2021; Summer: 1 Jul-30 Aug 2021		
Offline OC/EC instrument / analysis	DIGITEL DHA-80; Sunset Laboratory Model 5L, EUSAAR-2		
Offline measured quantity / size cut	OC, EC / $PM_{2.5}$		
Offline sampling resolution	12 h		
Online OC/EC instrument	Sunset Laboratory Model 4		
Online measured quantity / size cut	OC, EC / $PM_{2.5}$	OC, EC / $PM_{2.5}$	OC, EC / PM_1
Native online resolution	2 h	4 h	2 h
AMS/ACSM instrument	HR-ToF-AMS	ToF-ACSM	C-ToF-AMS
AMS/ACSM measured quantity / size cut	OA (OC derived) / NR-PM_1		
Native AMS/ACSM resolution	2.5 min	10 min	5 min
AMS/ACSM configuration relevant to OC determination	High-resolution analysis; Improved-Ambient method using PIKA	Unit-mass-resolution analysis; OM:OC estimated from f_{44}	Unit-mass-resolution analysis; OM:OC estimated from f_{44}

Paired AMS/ACSM-offline observations (winter / summer)	74 / 86	66 / 84	67 / 89
Paired AMS/ACSM-online observations (winter / summer)	– / 519	159 / 266	373 / 283
Paired offline-online observations (winter / summer)	– / 84	73 / 81	74 / 57

The table also summarizes the site-specific AMS/ACSM configurations and the main instrumental differences relevant to the comparisons, including the use of different AMS/ACSM instruments and the PM_1 versus $PM_{2.5}$ size cuts. The corresponding information in the Methods has been retained and the following sentence was added at the end of section 2.4 "An overview of the measurement periods, instrument configurations, measured quantities, aerosol size cuts, native time resolutions, and numbers of paired observations used in the intercomparisons at each site and season is provided in Table S1."

R1C7: Finally, the conclusions and recommendations should be more closely aligned with what the present dataset can establish. The recommendations that TOT should serve as the primary reference for winter OC trends, that AMS-derived OC should under certain conditions be regarded only as a lower bound, and that OA:eBC < 5 identifies problematic measurement conditions are broad conclusions based on three Central European sites, two relatively short seasonal campaigns, three different AMS/ACSM instruments, and a mechanism that remains unresolved. A more defensible and still important conclusion is that, under the winter combustion-influenced conditions encountered in this study, OCAMS was substantially lower than EUSAAR-2 OCTOT, whereas agreement was considerably better in summer. The causes of this seasonal discrepancy remain uncertain and likely involve multiple instrumental, sampling, and aerosol-composition factors. The broader recommendations should therefore be moderated accordingly unless they can be supported by additional independent analyses.

R1A7: We thank the reviewer for this comment. As discussed in our responses to R1C1-R1C3, we have revised the manuscript to clarify that the OA:eBC value of approximately 5 is proposed as a preliminary warning indicator rather than a universal threshold. We have also included independent AMS/ACSM-SMPS comparisons and air-mass analyses to further evaluate the observed discrepancies. Furthermore, as clarified in our responses to R1C26 and R2C6, we have revised the manuscript to emphasize that TOT is not considered an absolute reference, but rather a standardized measurement method, supported by the reasonable agreement between online and offline TOT measurements and have removed the statement suggesting that OC_{AMS} should be treated as a lower bound for OC. The potential contributions of instrumental, sampling, and aerosol-composition effects have also been addressed in Section 3.5, as detailed in our previous responses. We have additionally made the following change in Section 4 (Conclusion) to address the reviewer's remaining concerns: "Based on the observations at the three stations investigated here, larger discrepancies between OC_{TOT} and OC_{AMS} were observed during winter whereas it was more reliable during summer; thus, OC values from the AMS should be interpreted with caution during winter, particularly when OA:eBC ratio value falls below ≈ 5 which is proposed as a preliminary warning value rather than a strict diagnostic threshold."

As well as in Abstract: "The results suggest that measured OA:eBC may serve as a preliminary warning indicator for OC_{AMS}-OC_{TOT} disagreement and provide practical recommendations to improve OC quantification under winter combustion conditions."

Minor comments:

R1C8: Abstract, lines 15–16: remove "negligible influence of particle size differences." FRY demonstrates that size-cut mismatch cannot explain the large winter discrepancy, but it does not show that size effects are negligible everywhere.

R1A8: We thank the reviewer for this clarification. We agree that the persistence of the substantial winter OC discrepancy at FRY, where both the AMS and semi-continuous TOT analyzer use nominal PM_1 size cuts, shows that

differences in sampling size cuts alone cannot explain the observed discrepancy. However, this observation does not establish that particle size effects are negligible across all sites and measurement conditions. We have therefore removed the statement suggesting a negligible influence of particle size differences from the Abstract and replaced it with: "Semi-continuous and offline TOT measurements showed generally good agreement across sites and seasons, while the substantial winter AMS-TOT discrepancy at FRY persisted despite nominally matched PM_1 size cuts."

R1C9: Methods, Sect. 2.4: provide more information about AMS/ACSM calibration, CE, RIE-OA, fragmentation-table treatment, and instrument-specific QA/QC. Referring readers primarily to another publication is insufficient for a measurement-comparison paper.

R1A9: We thank the reviewer for this comment. As detailed in our response to R1C2, we have expanded Section 2.4 to clarify the site-specific CE treatment, calibration procedures, and RIE values, and Section 2.6 to describe the independent AMS/ACSM-SMPS volume comparisons. The instrument configurations and data-processing software are already described in Section 2.4 and Table S1, as discussed in our response to R1C6.

R1C10: Provide sample sizes (data point numbers) in all the scatter comparison figures.

R1A10: We thank the reviewer for this suggestion. To avoid overcrowding the scatter plots with additional information, we have added a new Table S1 in the Supplement, which summarizes the number of paired observations for the AMS/ACSM-offline TOT, AMS/ACSM-online TOT, and offline-online TOT comparisons at each site and during each season. We also clarified in Results Section 3. that all valid paired observations listed in Table S1 were used in the corresponding regression analyses, without any further exclusion: "The numbers of valid paired observations used in the regression analyses for each method comparison, site, and season are summarized in Table S1. These observations were also used in the cluster-based comparisons, where they were separated according to their assigned clusters." We believe this provides the requested information clearly while keeping the figures easy to read.

R1C11: Table S1: verify all OM:OC, O:C, and f44 values against Eqs. 1–2 and specify whether the table contains means of time-resolved ratios or ratios calculated from mean values. Several entries appear inconsistent with the stated equations.

R1A11: We thank the reviewer for pointing this out. As explained in our response to R1C4, some OM:OC values were incorrectly transferred to the original supplementary table. These have now been corrected and checked against Eqs. 1-2 (now 2-3). The corrected values are presented in Table S2. We have also clarified in the table caption that the reported values are means of time-resolved ratios, rather than ratios calculated from campaign-mean concentrations. As explained in our responses to R1C4 and R1C5, time-resolved OM:OC ratios were used to calculate AMS/ACSM-derived OC, with HR-derived values used at MEL and f44-based values at FRY and NAOK.

R1C12: 3.1: slopes of 0.79–0.88 should not simply be described as "close to unity" without discussing the systematic 10–20% difference and its relevance to subsequent comparisons.

R1A12: We thank the reviewer for this comment. We agree that slopes of 0.79-0.88 suggest a systematic difference between the online and offline TOT measurements and should not be described as simply "close to unity." We have revised Section 3.1 to quantify this difference: "For OC, winter slopes were 0.85 ± 0.06 (R^2 = 0.71) at FRY and 0.79 ± 0.06 (R^2 = 0.66) at NAOK, while summer slopes were 0.80 ± 0.09 (R^2 = 0.51) at MEL and 0.88 ± 0.12 (R^2 = 0.62) at FRY (Fig. 3). Although these slopes suggest systematic differences of approximately 12-21% between online and offline TOT measurements, the methods remain reasonably consistent overall."

R1C13: Lines 187–207: the statement that the lower MEL R2 "could be attributed to enhanced SOA production" needs evidence. Enhanced SOA does not inherently reduce agreement between two OC methods.

R1A13: We thank the reviewer for this comment. We agree that enhanced SOA production does not necessarily explain the lower R^2 between the two TOT methods. We have therefore removed this interpretation and revised the sentence in Section 3.1 as follows: "The summer results at MEL (0.80 ± 0.09, R^2 = 0.51) and FRY (0.88 ± 0.12, R^2 = 0.62) followed a similar pattern (Fig. 3), although the agreement was weaker at MEL, as suggested by the lower R^2."

R1C14: 3.2/Fig. S3: clarify OA versus OC. The current figure evaluates OA, despite the section and caption referring to OC.

R1A14: We thank the reviewer for this comment. This point has already been addressed in our response to R1C5. As explained there, Fig. S3 now includes comparisons of both OA and OC derived from HR and UMR analysis. Section 3.2 has also been revised to differentiate between the two comparisons and to discuss the agreement in the resulting OC concentrations.

R1C15: Lines 289–291: saying the MEL comparison demonstrates "reliable and reproducible OC estimation across instruments and years" is too strong. Similar slopes in different campaigns do not establish accuracy.

R1A15: We thank the reviewer for this comment. We agree that similar results across different campaigns do not establish the accuracy of AMS-derived OC. We have therefore revised the statement in Section 3.3 to emphasize consistency with previous observations rather than implying independent validation of the method: "The MEL results obtained with the HR-ToF-AMS are consistent with those reported using an ACSM in a previous comparison with daily $PM_{2.5}$ filter measurements (Poulain et al., 2020). This similarity suggests that comparable inter-method differences were observed across the two campaigns, but does not independently establish the accuracy of AMS-derived OC."

We have also added the following discussion of Rovira et al. (2026) to Section 3.3, providing further context for the wintertime OC discrepancies: "A similar wintertime discrepancy was reported by Rovira et al. (2026) at Košetice, where C-ToF-AMS-derived OC (PM_1) showed poor correlation with offline thermal-optical OC ($PM_{2.5}$). Therefore, the authors excluded the winter observations from their analysis, whereas the summer observations showed better agreement. Although the reasons remain unclear, they suggested that differences in winter aerosol composition, particularly heating-related emissions, might contribute."

R1C16: Line 300: "size effects were excluded" should be changed to something like "nominal inlet size-cut differences cannot explain the FRY discrepancy." AMS aerodynamic-lens transmission and particle properties still matter.

R1A16: We thank the reviewer for this comment. Similar to our response to R1C8, we have also revised the Results section 3.3 to clarify this: "At FRY, both the AMS and semi-continuous TOT analyzer used nominal PM_1 size cuts, yet substantial winter discrepancies persisted. This shows that differences in nominal sampling size cuts alone cannot explain the observed disagreement. Nevertheless, differences in particle transmission characteristics, including the size-dependent transmission of the AMS aerodynamic lens, cannot be excluded."

R1C17: 3.4.1: explain whether C1W/C2W grouping was defined independently before examining the OC discrepancy. Otherwise there is a risk of post hoc classification.

R1A17: We thank the reviewer for this comment. We clarify that C1W and C2W correspond to air-mass clusters previously identified in Arora et al. (2026a), independently of the present OC intercomparison. The labels C1W and C2W are used here to identify these existing winter clusters; they do not represent a new classification based on the observed OC discrepancies. We have clarified this in Section 3.4.1: "The air-mass clusters were previously identified in Arora et al. (2026a), independently of the OC discrepancies examined in this study (Fig. S6). The labels C1W and C2W refer to two of these pre-existing winter clusters and do not represent a new classification based on the OC comparison."

R1C18: Lines 367–374: direct evidence is needed for the claim that transported mineral dust carried enough refractory organic material to influence the OC comparison.

R1A18: We thank the reviewer for this comment. We agree that the amount of refractory organic material associated with transported mineral dust was not directly measured in this study. We have therefore clarified this in Section 3.4.1 as follows: "Long-range transport of mineral dust may introduces a significant fraction of refractory organic material, which is not efficiently sampled by the AMS due to the aerodynamic lens cutoff and the standard vaporization

temperature (~600 °C) (Capes et al., 2008). This could potentially contribute to the lower AMS-derived OC concentrations observed during these transport periods. However, the amount of refractory organic material associated with the transported dust was not directly measured in this study, and its contribution to the observed OC discrepancy therefore remains uncertain."

R1C19: 3.5.1: the section title repeats the title of Sect. 3.5.

R1A19: We thank the reviewer for pointing this out. We have revised the title of Section 3.5.1 to avoid repetition: "Uncertainties associated with thermo-optical OC measurements"

R1C20: Lines 469–476/Fig. S4: remove the "PC subtraction" as a correction or justify it rigorously from thermal-optical theory.

R1A20: We thank the reviewer for this comment. As also clarified in our response to R2C6, subtracting the pyrolytic carbon (PC) fraction was not intended as a correction to the EUSAAR-2 OC measurements. Instead, it was used as an extreme sensitivity test to examine the possible influence of PC assignment on the differences between AMS/ACSM- and TOT-derived OC. We have extensively revised Section 3.5.1 and the caption of Fig. S4 (in response to R2C6) to make this distinction clear. We also clarify that PC is operationally determined from the thermal-optical analysis and that subtracting the entire PC fraction does not represent an alternative estimate of the true OC concentration.

R1C21: Lines 527–545: the RIE sensitivity analysis is performed mainly at MEL, while FRY has the most severe discrepancy. Its generalization to FRY and NAOK therefore needs qualification.

R1A21: We thank the reviewer for this comment. As also discussed in our response to R2C5, the MEL sensitivity analysis cannot be directly generalized to FRY and NAOK because of differences in aerosol composition and instrument configuration. We have therefore clarified this limitation in Section 3.5.2 by adding: "The RIE sensitivity analysis at MEL cannot be directly extrapolated to FRY and NAOK. Therefore, the contribution of RIE variability to the pronounced winter OC discrepancy at FRY remains uncertain."

R1C22: Lines 539–541: an OSc range from –1 to +1 should not simply be described as "fresh, less-oxidized POA." OSc approaching +1 is highly oxidized. Interestingly, the Supplement states that most values were –1 to 0; the main text and SI should be made consistent.

R1A22: We thank the reviewer for pointing this out. We agree that an OSc range extending to +1 cannot be described entirely as fresh, less-oxidized POA. The predominant range was actually between −1 and 0, as already reported in the Supplement (Fig. S12). We have therefore corrected the sentence in Section 3.5.1 to make the manuscript consistent with the Supplement and to avoid overinterpreting the oxidation state: "Consistent with this, the measured OSc of organics at MEL during winter was predominantly between –1 and 0 (Fig. S12), suggesting a substantial contribution from less-oxidized organic aerosol, which may be associated with uncertainties in RIE and detection efficiency."

R1C23: 3.5.2: the Pieber-effect discussion is overly qualitative. This can potentially be evaluated looking of periods of The argument that three instruments are unlikely to show the same artifact does not exclude a systematic seasonal dependence associated with nitrate-rich winter aerosol.

R1A23: We thank the reviewer for this comment. We agree that the use of different instruments does not completely exclude a systematic seasonal interference associated with nitrate-rich aerosol. We have therefore revised the discussion in Section 3.5.2 as follows: "However, the use of different instruments does not completely rule out a systematic seasonal influence of this interference, particularly under nitrate-rich winter conditions. Therefore, a possible contribution of the Pieber effect to the observed OC discrepancies cannot be excluded."

R1C24: 3.6: distinguish clearly among BrC absorption, inferred BrC mass, MAC_{OA}, and MAC_{BrC}; explain uncertainty and independence of these quantities.

R1A24: We thank the reviewer for this comment. The calculation of BrC absorption, BrC mass, and MAC_{OA} is described in Text S1 of the Supplement. We have also clarified the distinction between these quantities and their uncertainties in Section 3.6 by adding: "BrC absorption was estimated from the wavelength-dependent optical measurements, whereas BrC mass was inferred using site-specific MAC_{OA} values derived from OA concentrations (Text S1). Therefore, the estimated BrC mass is not independent of OA and is subject to uncertainties associated with the assumed optical properties and MAC_{OA} values."

We have also revised the interpretation of MAC_{BrC} in Section 3.6 to avoid overstating its significance: "...although the MAC_{BrC} values are subject to uncertainty."

As explained in our response to R2C7, we have additionally revised Text S1 to clarify the data treatment and uncertainties associated with the BrC analysis. We have also added a direct comparison between BrC absorption and the OC method discrepancy. For this comparison, we used BrC absorption rather than inferred BrC mass to avoid introducing the additional dependence on AMS/ACSM derived OA.

R1C25: Conclusion, line 575: "(range of slopes and R2)" is an unfinished placeholder and needs to be replaced.

R1A25: We thank the reviewer for pointing this out. We have removed the unfinished placeholder and revised the sentence in the Conclusion as follows: "Across all stations, a pronounced seasonal pattern was observed, with generally better agreement between AMS/ACSM and TOT-derived OC during summer, while larger discrepancies were observed during winter."

R1C26: Conclusion: replace statements implying that OCTOT is the absolute truth with language such as "AMS-derived OC was systematically lower than EUSAAR-2 TOT OC." Both are operationally defined measurements with different uncertainties.

R1A26: We thank the reviewer for this comment. We agree that neither method provides an absolute measure of OC and that both are subject to method-specific uncertainties. However, we would like to distinguish between the operational definition of OC and the degree of standardization of the measurement procedures. EUSAAR-2 TOT determines OC using a defined and standardized thermal-optical protocol, whereas AMS/ACSM-derived OC is estimated indirectly from OA concentrations using OM:OC ratios, introducing additional uncertainties associated with aerosol composition and the OM:OC determination. Therefore, our recommendation does not imply that TOT represents the true OC concentration since there are several uncertainties associated as extensively detailed in section 3.5.1 Uncertainties associated with thermo-optical OC measurements, but rather that its standardized protocol provides a more consistent basis for intercomparison. As explained in our response to R2C6, we have revised Recommendation 2 in the Conclusion to clarify this distinction while we explicitly acknowledge the uncertainties associated with TOT, including charring and OC-EC split point determination.

In addition, as described in our response to R1C25, we have revised the first paragraph of the Conclusion to describe the observed seasonal differences without implying that either method represents the absolute reference.

R1C27: The Supplement title differs from the manuscript title, and Figure S3 says "organic carbon" while plotting OA. These should be harmonized.

R1A27: We thank the reviewer for pointing out these inconsistencies. We have corrected the Supplement title to match the manuscript title: "Determination of Aerosol Particle Organic Carbon (OC) by Aerosol Mass Spectrometry (AMS) and Thermo-Optical Transmission (TOT) Analysis: Spatial and Seasonal Differences"

The terminology in Fig. S3 has also been corrected from organic carbon (OC) to organic aerosol (OA), as described in our response to R1C14.

Reviewer 2

The manuscript titled "Determination of Aerosol Particle Organic Carbon (OC) by Aerosol Mass Spectrometry (AMS) and Thermo-Optical Transmission (TOT) Analysis: Spatial and Seasonal Differences" fundamentally focuses on measurement-comparability challenges across instruments and addresses this by integrating observations from three Central European sites over two seasons. I think comparing offline and semi-continuous thermal-optical measurements is useful for the broader community, and the authors made a comprehensive effort to examine possible sources of disagreement for OC between AMS/ACSM and thermo-optical technique. Still, there are avenues where further improvements are necessary. For example, the conclusion that winter combustion conditions and more particularly, the low OA:eBC ratios, determine when AMS-based OC diverges from TOT OC is somehow undersupported with the current analysis. This, I think is my primary comment. The measurements have different particle-size cuts and time resolutions whose treatment should be more clearly described. The OA:eBC ratio is not independently validated. The paper would benefit from a stronger consideration of these limitations and more careful interpretations. Following are my complete comments:

R2C1: pp 4, lines 85–94; pp 5, lines 112–145; pp 10, lines 245–250: The compared measurements do not consistently represent the same aerosol size fraction. AMS/ACSM measures PM_1. Offline TOT uses $PM_{2.5}$ filters. The online TOT cut is $PM_{2.5}$ at MEL and NAOK but PM_1 at FRY; meanwhile, the AMS/ACSM cuts also vary among sites. The statement that size effects are excluded at FRY does not resolve the size-cut mismatch at the other comparisons. It would be good to constrain the size-cut contributions, clearly note which comparisons are size matched, and qualify conclusions that rely on PM_1–$PM_{2.5}$ comparisons.

R2A1: We thank the reviewer for this comment. As explained in our responses to R1C6, R1C8, and R1C16, we have clarified the instrument-specific size cuts in Table S1 and revised the Abstract and Section 3.3 to avoid excluding particle-size effects. At FRY, both the AMS and online TOT measurements used nominal PM_1 size cuts, yet a substantial winter OC discrepancy remained. Therefore, the discrepancy in this size-matched comparison cannot be attributed to a PM_1-$PM_{2.5}$ size-cut mismatch. However, differences in particle transmission cannot be excluded, and the PM_1-$PM_{2.5}$ mismatch remains a potential source of uncertainty in the other comparisons.

To make this limitation explicit for all sites, we have added the following sentence to Section 3.3: "Because AMS/ACSM measurements represent PM_1 aerosol whereas offline TOT measurements represent $PM_{2.5}$, differences in the sampled size fractions may contribute to the observed OC discrepancies. The magnitude of this contribution was not independently quantified and may vary between sites and seasons."

R2C2: pp 4, lines 97–110; pp 5, lines 112–120; pp 10–11, lines 245–275: The time matching and regression methods need to be described in enough detail to reproduce the analysis. Please specify how AMS/ACSM data were averaged to match the 12-hour offline filters and the 2-hour or 4-hour online TOT measurements; how incomplete or missing intervals were handled; and how many paired observations enter each fit.

R2A2: We thank the reviewer for this comment. We agree that the temporal matching procedure and treatment of incomplete measurement intervals should be described more explicitly. As also noted in our response to R1C6, we have added a new overview table (Table S1) to the Supplement. We have also added the following paragraph to Section 2.6 (Data Quality Assurance):

"For all inter-method comparisons, measurements were temporally matched according to the sampling interval of the lower time-resolution method. For comparison with offline TOT, the high time-resolution AMS/ACSM data were averaged over the corresponding 12 h filter sampling intervals (06:00-18:00 and 18:00-06:00 UTC). For comparison with online TOT, AMS/ACSM data were averaged over the corresponding online TOT measurement intervals. Intervals containing no valid AMS/ACSM measurements were excluded. Missing data were not interpolated and only temporally matched observations for which data from both methods were available were retained for the intercomparisons and orthogonal regression analyses. The resulting numbers of paired observations for each site, season, and method comparison are reported in Table S1."

R2C3: pp 14, lines 339–355; pp 15, lines 370–377; pp 22, lines 589–593: The proposed OA:eBC threshold is not yet established as a practical indicator. The conclusion recommends caution when winter OA:eBC is below approximately 5, but the evidence appears to be a visual or site-season association across a limited number of campaigns. OA:eBC

also contains AMS OA—the quantity used to calculate OCAMS—so its relationship with OCAMS–TOT differences may partly suggest mathematical coupling. If possible, the relationship should be quantitatively tested using paired observations, accounting for uncertainty and sensitivity to site and season, and assess whether it remains after considering the measurement-size mismatch. At the very least, the OA:eBC < 5 should be presented as a preliminary observation specific to these campaigns rather than a general threshold or recommendation.

R2A3: We thank the reviewer for this comment. As already discussed in detail in our response to R1C1, we have addressed the mathematical coupling between OA:eBC and OC_{AMS}, the influence of site-season differences, and the statistical basis of the proposed warning value. We agree that OA:eBC is not mathematically independent of OC_{AMS} and therefore cannot independently explain the observed AMS-TOT discrepancies. We have therefore clarified this limitation in Section 3.4.2. Furthermore, as described in R1A1, we have performed a receiver operating characteristic (ROC) analysis which gave OA:eBC cutoffs of 6.77 and 5.09 for $OC_{AMS}/OC_{Offline} < 0.7$ and < 0.5, respectively (Table S5). These results provide a quantitative basis for the proposed warning value of approximately 5.

As also discussed in R1A1, the pooled relationship partly reflects differences between sites and seasons, and the proposed value is therefore not presented as a universal threshold. The potential influence of particle-size differences has already been addressed in our responses to R1C8 and R2C1, including the comparison at FRY, where the substantial winter discrepancy persisted despite nominally matched PM_1 size cuts. To address these concerns, we have already revised Section 3.4.2 and the Conclusion (Section 4) to emphasize that OA:eBC ≈ 5 is proposed only as a preliminary warning indicator based on the conditions investigated in this study, rather than a universally applicable diagnostic threshold.

R2C4: pp 12, lines 293–315; pp 13–15, lines 321–377; pp 20–21, lines 535–565: Source attribution and proposed mechanisms should be framed as hypotheses. The authors attribute the largest discrepancy in measurements to coal influence at FRY, transported dust, refractory organics, and combustion matrix effects, but the evidence presented is largely indirect. The merged trajectory clusters include dust episodes but it is acknowledged that mean trajectories do not identify the dust source. OA:eBC is not a unique source tracer and brown carbon absorption does not directly establish the mass of refractory OC missed by AMS. I suggest to meticulously distinguish measured results from interpretations.

R2A4: We thank the reviewer for this comment. As already discussed in our responses to R1C1, R1C17, and R1C18, we have extensively revised the manuscript to distinguish the observed AMS-TOT discrepancies from their possible source and composition related explanations. In particular, the air-mass clusters were defined independently of the OC intercomparison (R1C17), and we have explicitly clarified that the amount of refractory organic material associated with transported dust was not directly measured and that its contribution to the observed discrepancy remains uncertain (R1C18). Furthermore, as discussed in R1A1, we have clarified that OA:eBC is not an independent source tracer and cannot establish the cause of the discrepancy. The coal-related influence at FRY is supported by independent molecular-tracer analysis of samples from the same campaign (Arora et al., 2026b), rather than inferred solely from OA:eBC. The interpretation of BrC absorption and its limitations has also been clarified in our previous responses, distinguishing the observed optical properties from the possible presence of refractory organic material. Therefore, we retain the observed seasonal and site-specific differences as the main findings, while presenting coal-related combustion effects, transported dust, refractory organics, and aerosol-matrix effects as possible contributing mechanisms rather than independently established causes.

R2C5: pp 5, lines 124–145; pp 9, lines 227–243; pp 17–20, lines 432–508: I think the AMS/ACSM OC estimation discussion should be further expanded. The authors apply the f_{44}-based equations to UMR data at FRY and NAOK, while MEL uses HR data and the I-A method. The UMR–HR comparison at MEL is used to suggest similar behavior elsewhere, but that somehow does not validate the UMR conversion at other sites or for their aerosol compositions. Please state precisely how OC was calculated at each site, identify the applicable OM:OC method and corrections, and discuss the limitations of extrapolating the MEL HR–UMR result to FRY and NAOK. The reported uncertainty should include the uncertainty in the f_{44}-to-O:C and OM:OC parameterizations.

R2A5: We thank the reviewer for this comment. The site-specific OC calculation procedures, the OM:OC method applied at each site, and the direct comparison of HR and UMR-derived OC at MEL have already been clarified in response to R1C4 and R1C5 through revisions to Section 2.4, Table S2, Section 3.2, and Fig. S3. In response to the

additional points raised here, we agree that the close HR-UMR agreement at MEL should not be interpreted as validation of the UMR-derived OC at FRY and NAOK, because aerosol composition and instrument configuration differed among the sites. We have therefore revised Section 3.2 to state: "The close agreement between HR and UMR-derived OC at MEL suggests that the effect of these limitations was small under the aerosol conditions encountered at MEL; however, the magnitude of this uncertainty cannot be assumed to be the same at FRY and NAOK because aerosol composition and instrument configuration differed among the sites. The MEL comparison is therefore considered a site-specific evaluation of the UMR approach rather than a validation of UMR-derived OC at FRY and NAOK."

We have also addressed the uncertainty associated with the f_{44}-based parameterization in Section 3.5.2 and have therefore added: "Canagaratna et al. (2015) reported an error of approximately 13% for the f_{44}-based O:C parameterization for secondary OA, with larger uncertainty expected for primary OA. This parameterization uncertainty is additional to the uncertainty associated with the AMS/ACSM OA mass measurement."

R2C6: pp 16–17, lines 385–430; pp 22, lines 598–602: **(1)** The interpretation of pyrolytic carbon (PC) subtraction and TOT as a reference needs clarification. Subtracting PC from winter OC changes the slopes substantially, including a NAOK slope above unity, but the manuscript does not make clear how PC is defined, measured, and assigned to OC, or how the uncertainty of that subtraction is handled. **(2)** It may be useful to show that calculation (SI-relevant) and clarify whether the reported PC quantity is a measured fraction or an operationally defined fraction of the thermogram. **(3)** A standardized protocol is not necessarily an unbiased absolute reference as the authors also discuss possible charring and split-point uncertainty. Please qualify the conclusion that TOT provides "a more stable reference for absolute OC levels" and justify what "stable" means in this context.

R2A6: We thank the reviewer for this comment. We have addressed the points as follows:

1) PC subtraction and its influence on the OC comparison

We thank the reviewer for this important comment. We have clarified that subtraction of the complete PC fraction was not intended as an alternative correction of EUSAAR-2 OC or as an estimate of "true" OC. It was used as an intentionally extreme sensitivity test to assess whether PC/split-point uncertainty could contribute to the observed discrepancy. The complete operationally determined PC contribution was subtracted from the corresponding offline OC concentration for each sample. The thermal-optical OC-EC split is known to be sensitive to the distinction between charred OC and pre-existing EC (Chow et al., 2001; Yang and Yu, 2002), and optical correction may in some cases overcompensate across the OC-EC split (Malm et al., 2020). Thus, some fraction of the observed discrepancy could potentially arise from uncertainty associated with PC/split-point assignment together with the other method and aerosol/matrix dependent factors investigated in the manuscript. We have therefore replaced the previous interpretation in Section 3.5.1 with: "To assess the influence of PC assignment on the OC_{AMS}-$OC_{Offline}$ comparison, the complete operationally determined PC fraction was subtracted from $OC_{Offline}$ for each sample (Fig. S4) as an intentionally extreme sensitivity test. This was not intended as an alternative correction of EUSAAR-2 OC or as an estimate of "true" OC nor does it assume that the entire PC fraction is erroneously assigned to OC. Complete PC subtraction changed the winter slopes from 0.34 to 0.61 at MEL, from 0.24 to 0.44 at FRY, and from 0.63 to 1.24 at NAOK. The substantial changes show that the comparison is sensitive to PC assignment, whereas the slope above unity at NAOK shows that complete removal of PC can overcorrect the comparison. Thus, uncertainty associated with the optical PC/OC-EC split may contribute to the observed discrepancy, but PC alone cannot explain it. During winter, PC fractions were higher at FRY (35%, 1.97 μg m^{-3}), MEL (34%, 1.52 μg m^{-3}), and NAOK (28%, 1.18 μg m^{-3}), compared with summer (~18-22%, ~0.5-0.6 μg m^{-3}). Despite the lower PC fractions in summer, agreement between OCAMS and OCOffline was already substantially better. Moreover, the OC_{AMS}-$OC_{Offline}$ relationship showed no systematic dependence on PC/TC (Fig. S5), consistent with the combined influence of multiple method, site, season, and aerosol-matrix dependent factors."

Regarding uncertainty, PC and OC are derived from the same thermogram and share dependence on the optically determined OC-EC split; their uncertainties are therefore not independent. We did not assign a separate analytical uncertainty to the PC-subtracted values, which are used only for this sensitivity analysis. We have added the following sentence to Section 3.5.1: "Because PC and OC are derived from the same thermogram and share dependence on the optically determined OC-EC split, their uncertainties are not independent; therefore, the PC-subtracted values are used

only as a sensitivity analysis and are not treated as an alternative OC measurement with an independently propagated analytical uncertainty."

2) Operational definition of PC and calculation procedure

We agree that the previous version did not sufficiently distinguish the operationally determined pyrolytic carbon (PC) fraction from an independently measured atmospheric carbon fraction, and that the purpose of the PC-subtraction analysis was not explained clearly enough. We have therefore added the following clarification to Section 3.5.1: "In the EUSAAR-2 TOT procedure, PC is not determined as an independent atmospheric carbon fraction but is operationally derived from the thermogram using the laser-transmittance correction (Cavalli et al., 2010). During the subsequent oxidizing phase, the OC-EC split point is determined from recovery of the transmittance signal, and carbon associated with charring is assigned to OC according to this optical correction. So the reported PC quantity depends on the thermal-optical protocol and optical split-point determination and should not be interpreted as an independently measured chemical fraction."

We have also revised the Fig. S4 caption to state explicitly how the sensitivity analysis was performed: "Figure S4: Sensitivity analysis of the influence of the operationally determined pyrolytic carbon (PC) fraction on the comparison of OC_{AMS} with $OC_{Offline}$ measurements after applying pyrolysis corrections in a) winter (top) and b) summer (bottom) at MEL, FRY and NAOK. For this analysis, the complete PC contribution was subtracted from the corresponding offline OC concentration for each sample. This represents an intentionally extreme sensitivity test and not an alternative correction or estimate of true OC; it does not imply that the complete PC fraction is erroneously assigned to OC. Solid red lines show orthogonal distance regression fits, and dashed red lines indicate the 1:1 relationship. Slopes (b), intercepts (a), and R^2 values are provided for each site. Course of time is in color for each season."

3) TOT as an operationally defined measurement

Finally, we agree with the reviewer that a standardized protocol is not necessarily an unbiased absolute reference. We have therefore removed the statement that OC_{TOT} provides "a more stable reference for absolute OC levels" and revised the conclusion to: "2. OC_{TOT} provides a more standardized and methodologically consistent basis for comparing OC concentrations, with OC determined directly using a defined thermal-optical protocol and showing reasonable agreement between independent online and offline TOT measurements. In contrast, OC_{AMS} is an indirectly derived quantity and is better suited for resolving temporal variability and composition. Wintertime OC trend analyses in monitoring networks are therefore recommended to be anchored primarily to TOT based OC, with AMS based OC used in a complementary way. However, TOT remains an operationally defined measurement and is subject to uncertainties associated with charring, OC-EC split-point determination, and other method-dependent effects."

R2C7: pp 20–21, lines 525–565: **(1)** The brown carbon analysis is not sufficiently connected to the OC comparison. Brown carbon is used to help explain AMS–TOT disagreement, but the section mainly interprets seasonal and site-level absorption differences. **(2)** The reported brown carbon absorption standard deviations are large relative to some means, particularly in summer, and MAC values also vary strongly. The SI shows brown carbon absorption and MAC calculations but further expansion is needed to describe uncertainty and data filtering, and test whether either quantity covaries with the OC residuals. If no such relationship is tested, present this section as contextual optical characterization rather than evidence explaining the method discrepancy. **(3)** Some source-specific interpretations (for example, "coal-derived" or "tar-rich") should be softened unless supported by independent source or molecular measurements.

R2A7: We thank the reviewer for this comment. We have addressed the points as follows:

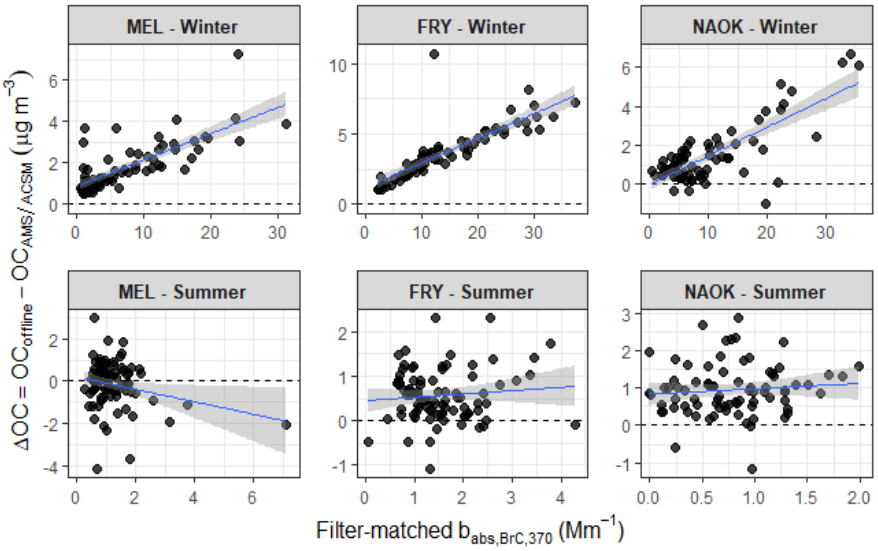
(1) Relationship between BrC absorption and the OC discrepancy

We agree that the previous version did not sufficiently connect the BrC analysis to the OC comparison. We have therefore revised this section and added a direct analysis of the relationship between BrC absorption and the difference between $OC_{Offline}$ and $OC_{AMS/ACSM}$. BrC absorption at 370 nm ($b_{abs,\ BrC,370}$) was averaged over the corresponding 12-h offline filter sampling periods and compared with the OC difference, defined as Δ OC = $OC_{Offline}$-$OC_{AMS/ACSM}$. The

analysis was performed separately for each site and season. We used BrC absorption rather than BrC mass because the BrC mass calculation depends on MAC, which was derived using OA. Since $OC_{AMS/ACSM}$ is itself derived from $OA_{AMS/ACSM}$, using the directly derived optical absorption avoids introducing this additional OA dependence. The following text has been added to Section 3.6 of the manuscript: "To test whether BrC covaried directly with the OC method discrepancy, $b_{abs,\ BrC,370}$ was averaged over each corresponding 12-h offline filter sampling period and compared with $\Delta OC = OC_{Offline} - OC_{AMS/ACSM}$ separately by site and season. In winter, $b_{abs,\ BrC,370}$ positively correlated with ΔOC at MEL (Pearson $r = 0.77$, $p < 0.001$), FRY ($r = 0.81$, $p < 0.001$), and NAOK ($r = 0.77$, $p < 0.001$); Spearman correlations were also positive ($\rho = 0.77$, 0.93, and 0.50, respectively; all $p < 0.001$). No corresponding positive relationship was observed in summer. These results show that higher winter BrC absorption coincided with a larger $OC_{Offline}$ - $OC_{AMS/ACSM}$ difference, supporting an association between the discrepancy and BrC organic-aerosol composition during this season which could be influenced by both the emission sources and atmospheric evolution (Forrister et al 2015). However, this does not necessarily mean that BrC itself causes the discrepancy or that BrC is quantitatively missed by AMS/ACSM."

The following figure was added to the supplement:

Figure S16: Relationship between BrC absorption at 370 nm ($b_{abs,\ BrC,370}$) and the OC method difference $\Delta OC = (OC_{Offline}-OC_{AMS/ACSM})$ at MEL, FRY, and NAOK during winter (top row) and summer (bottom row). BrC absorption was averaged over the corresponding 12-h offline filter sampling periods. Black points represent individual filter-matched observations, blue lines show linear regressions, and grey shading indicates the 95% confidence interval. The dashed horizontal line denotes (ΔOC =0), corresponding to agreement between the two OC estimates.



(2) Uncertainty, data filtering, and variability in BrC measurements

Text S3 has additionally been revised to describe data treatment and uncertainty: "Negative calculated $b_{abs,\ BrC,370}$ values were not included; this affected <5% of observations in each site-season dataset. For comparison with OC, the high time-resolution BrC absorption data were averaged over the corresponding 12-h offline filter sampling periods. As a coverage sensitivity test, the analysis was repeated using only filter periods containing ≥75% valid BrC data; the positive winter relationships were retained. Because BrC mass depends on the applied MAC, which was derived using OA, the comparison with the OC discrepancy was performed using $b_{abs,\ BrC,370}$ rather than BrC mass."

We further clarify the large standard deviations: "The large variability in both BrC absorption and MAC, particularly under the low BrC absorption observed in summer suggests substantial within campaign variability. Therefore, the campaign average values are used only for optical characterization, whereas the relationship with ΔOC is evaluated from the individual filter-matched observations."

(3) Limitations of source-specific interpretations

Finally, we removed/softened source-specific statements and made it explicit that our present analysis cannot assign any sources to FRY: "However, the present optical measurements do not establish the presence of these specific compounds or independently identify their source… Overall, the distinct wintertime BrC profile at FRY, characterized by a high absorption coefficient, enhanced absorption at shorter wavelengths, high MAC and a low OA:eBC ratio is consistent with strongly light-absorbing, combustion-influenced organic aerosol. However, the present optical measurements do not provide molecular identification of the BrC components or independently establish their specific sources and cannot alone explain the discrepancies between OC determined by AMS and TOT protocols, and targeted molecular-level analyses are required to confirm such a connection."

Minor comments

R2C8: pp 6, lines 167–170; pp 7, lines 185–193; pp 8, lines 207–225: The subsection numbering in Section 3 repeats "3.1.1" for TC, OC, and EC. Please correct the numbering.

R2A8: We thank the reviewer for noticing this numbering error. We have corrected the subsection numbering in Section 3.1 as follows: "3.1.1 TC comparison", "3.1.2 OC comparison", "3.1.3 EC comparison".

R2C9: pp 15, lines 349 and 362–365: Please resolve the winter NAOK OA:eBC inconsistency: section 3.4.2 gives 10.21 in one place and 10.75 later (lines 349 and 362–365). Ensure the abstract, text, tables, and figures use the same value.

R2A9: We thank the reviewer for identifying this inconsistency. We have corrected both occurrences of 10.75 to 10.21 in Section 3.4.2 to ensure consistency throughout the manuscript.

R2C10: pp 11, lines 260–275: The online OC comparison at MEL is described as showing OCOnline lower than OCAMS (section 3.3), whereas the offline comparison has a slope of 1.27 for OCAMS relative to OCOffline. Please make the regression orientation explicit and explain the difference between the two comparisons.

R2A10: We thank the reviewer for this comment. Both comparisons show higher AMS-derived OC at MEL during summer. The inconsistency comes from the wording, not the results. The difference is that Fig. 5 compares OC_{AMS} with offline TOT (slope = 1.27), whereas Fig. 6 compares OC_{AMS} with online TOT (slope = 1.39). In both cases, OC_{AMS} is plotted on the y-axis. We have clarified the regression orientation in the figure captions and revised Section 3.3 to make the consistency between the two comparisons explicit. The following sentence has been added to the Fig. 5 caption: "OC_{AMS} is plotted on the y-axis and $OC_{Offline}$ on the x-axis."

Similarly, the following sentence has been added to the Fig. 6 caption: "OC_{AMS} is plotted on the y-axis and OC_{Online} on the x-axis."

We have also replaced the sentence in Section 3.3 with: "At MEL during summer, OC_{AMS} was higher than OC_{Online} (slope = 1.39, R^2 = 0.87), consistent with the OC_{AMS}-$OC_{Offline}$ comparison (slope = 1.27, Fig. 5)."

R2C11: pp 7, lines 185–190: The statement that the lower R^2 at MEL in the online/offline OC comparison "could be attributed to enhanced SOA production" (section 3.1) is speculative. Please either support this with analysis or remove/qualify it.

R2A11: We thank the reviewer for pointing this out. As also addressed in our response to R1C13, we have removed the speculative explanation linking the lower R^2 at MEL to enhanced SOA production.

R2C12: pp 1–2, lines 12–27 and 51–73; pp 10–11, lines 245–275: Please check terminology and notation throughout: use OCAMS/OCACSM consistently, define "OCTOT" when it first appears outside the abstract, and standardize "semi-continuous," "online," and "offline."

R2A12: We thank the reviewer for pointing out these inconsistencies. We have revised the terminology and notation throughout the manuscript, defined OC_{TOT} at its first occurrence outside the abstract (Section 1), and standardized the use of "semi-continuous," "online," and "offline." We have also clarified in Section 2.4 that OC_{AMS} is used as a collective notation for OC derived from either AMS or ACSM measurements, ensuring consistency with the notation used in the figures.

Community Comments

Dear Authors,

I looked very briefly at your interesting article, and have some brief comments. I apologize for not having enough time to expand these comments into a full review.

CC1: The figures present Clusters 1,2 and Other, then later Winter/Summer. It would be easier to interpret if consistent terminology was used. If impossible, perhaps add a table explaining how I should map Clusters to Seasons. (This is from a very rapid reading.)

CCA1: We thank the reviewer for this comment. Figures 5 and 6 compare AMS/ACSM- and TOT-derived OC during winter and summer. These comparisons show that AMS/ACSM substantially underestimated OC during winter, while the two methods showed much better agreement during summer. Therefore, in Figure 7, we focused only on winter to investigate whether the disagreement between the two methods depended on where the air masses came from. We divided the winter measurements into two groups:

- C1W and C2W: Air masses transported from the North Atlantic across Western Europe (C1W) and from the Iberian Peninsula and Western Mediterranean (C2W). These clusters were also associated with three Saharan dust transport events (5-10 February, 21-26 February, and 2-4 March 2021), particularly during periods dominated by C2W.
- C3W-C6W: Air masses arriving through other pathways, including Scandinavia, Eastern Europe, and the Norwegian Sea.

The air-mass clusters and Saharan dust events were previously identified and discussed in our earlier publication (Arora et al., 2026a), independently of the present OC intercomparison. In Figure 7, we used this existing classification to examine whether the larger wintertime OC discrepancies were associated with long-range transport, including periods influenced by Saharan dust.

Thus, all clusters shown in Figure 7 belong to winter. They are not different seasons but represent different air-mass origins within the winter dataset. The complete winter and summer air-mass classifications are provided in Figure S6. To make this clearer, we have revised the caption of Figure 7: "Figure 7: Comparison of OC_{AMS} (PM_1) with $OC_{Offline}$ ($PM_{2.5}$), illustrating the impact of clusters grouped according to the previously established air-mass trajectory clusters (Arora et al., 2026a). Top panels represent Clusters 1 and 2, and bottom panels represent the rest of the clusters (3, 4, 5, and 6) for MEL, FRY, and NAOK stations during winter. The dashed red line indicates a 1:1 relationship, b = slope, and a = intercept. The corresponding air-mass trajectories are shown in Fig. S6." Since Figure 7 only examines winter measurements, no additional table mapping clusters to seasons is needed.

CC2: Was "long-range transport from the North Atlantic" perhaps associated with wildfires in North America? (We have seen that "tar brC", i.e. partially carbonized char, produces OC signals but is insoluble and refractory. Direct TOA evidence is published here https://doi.org/10.1038/s41612-019-0069-5 in Figure S6, we also have direct AMS evidence (not yet published but presented at the Canadian Aerosol Conference, July 10, 2026. https://www.caar-acra.ca/cac-2026-abstracts/chemical-composition-of-tarballs-and-%E2%80%9Ccharballs%E2%80%9D-formed-by-pyrolysis-of-wildfire-like-organics

CCA2: We thank the reviewer for this interesting suggestion and for sharing the relevant studies. The air-mass trajectories associated with the winter clusters C1W and C2W suggest transport from the North Atlantic across Western Europe and from the Iberian Peninsula and Western Mediterranean, respectively. Our previous study (Arora et al., 2026a) also identified three Saharan dust transport events during the winter campaign, particularly associated with C2W. However, we have no direct evidence linking these air masses to North American wildfire emissions.

We agree that partially carbonized biomass-burning particles, such as tarballs and charballs, could contribute to the observed differences between AMS/ACSM and TOT-derived OC. Corbin et al. (2019) demonstrated that tar brown carbon can be refractory and remain undetected by conventional AMS measurements. The additional findings

presented at the Canadian Aerosol Conference in 2026 further support this possibility by showing reduced AMS sensitivity to partially carbonized tarballs. These findings are relevant to our study because refractory organic material could contribute to the wintertime OC discrepancies. However, we did not directly measure tarballs or charballs, and therefore cannot determine their contribution to the observed differences. We have added a brief discussion of this possible mechanism to Section 3.4.1, while avoiding a specific attribution to North American wildfire emissions: "Partially carbonized combustion-derived organic material, such as tar brown carbon, may also contribute to the observed discrepancies. Corbin et al. (2019) demonstrated that such material can be refractory and remain largely undetected by conventional AMS measurements, although its contribution to the present OC discrepancies cannot be determined without direct measurements."

CC3: I congratulate the authors on immediately including their data publicly, however, only high-level data are on Zenodo. This public data set would be further improved by including the sub-categories OC1, OC2, OC3, OC4 etc as well as the absorption data.

CCA3: We thank the reviewer for the positive feedback on our data availability and for this helpful suggestion. We agree that including the individual thermal-optical carbon fractions and aerosol light-absorption measurements would make the dataset more useful for further research. We have therefore updated our Zenodo repository to include the individual carbon fractions (OC1-OC4, etc.) and the aerosol light-absorption data. The Data Availability statement in the manuscript has also been updated accordingly.

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