

Response to reviewer comments for manuscript: “Contributions of Fine and Coarse Particles to Light Absorption at an Urban Traffic Site during Street Dust Season”

Barreira et al.

We thank the reviewers for the constructive comments regarding the paper. Below we address the specific issues point by point. The reviewers' comments are in black and our answers are in blue. Changes to the Manuscript or Supplement Information are highlighted in red (note that the lines referred in reviewer's comments correspond to the ones in the previous version of the manuscript, while the lines in our answers and in the modified text refer to the updated version).

Reviewer 1:

Major Comments

1) The manuscript reports substantial increases in absorption for $PM_{7.2}$ relative to PM_1 (20–39% depending on wavelength). This is a key result. However, the interpretation remains incomplete that is it is unclear how much of this enhancement is due to:

- increased mass in the coarse mode,
- metal-rich particles (Si, Fe, Al, Cu, Zn),
- black carbon from tyre wear or graphite from brake pads,
- filter-related artefacts (scattering, loading, multiple scattering).

For example, the abstract states:

“coarse particles enhanced absorption during dust-resuspension events” but does not quantify the relative contribution of mineral dust vs non-exhaust traffic emissions.

It could be useful to include a multivariate analysis linking absorption coefficients to XRF elemental concentrations, and normalize absorption by mass to evaluate size-dependent absorption efficiency.

We thank the reviewer for highlighting the need to better constrain the origin of the enhanced absorption observed for the $PM_{2.5-7.2}$ fraction relative to PM_1 . We agree that the increase in absorption cannot be attributed to coarse-particle mass alone without considering the chemical composition and

potential contributions from specific absorbing components and filter-related effects. As a first step, we normalized the absorption coefficients by particle mass to calculate the mass absorption efficiency (MAE). Although PM_{7.2} showed the highest absolute absorption, its MAE decreased progressively with increasing particle size across all investigated wavelengths. This indicates that the enhanced absorption of PM_{2.5} and PM_{7.2} is substantially related to a greater particle mass rather than to a higher absorption efficiency per unit mass. Nevertheless, the appreciable MAE of the 1–2.5 µm fraction and the measurable absorption associated with the coarse fraction indicate that particles larger than 1 µm may also contribute to total light absorption.

We then compared the absorption measurements for the PM_{2.5-7.2} fraction with the PMF-derived source factors reported by Aurela et al. (2026), based on measurements from the same site and sampling period. Dust showed the strongest and most consistent association with absorption ($r = 0.39\text{--}0.47$), whereas correlations with the other source factors were generally weak, suggesting a greater role of dust than non-exhaust traffic emissions in the observed coarse-particle absorption.

To investigate the chemical components associated with absorption, source-informed multiple linear regression models were applied to the PM_{2.5-7.2} fraction at 370, 520, and 660 nm, using Al, Cu, S, Na, and Cl as tracers of Dust, Non-exhaust Emissions, Combustion/Secondary Inorganic Aerosol, Aged Salt, and Fresh Salt, respectively. The models explained 75–88% of the absorption variability ($R^2 = 0.751\text{--}0.878$). Al was the only tracer showing a consistent positive association with absorption across all wavelengths ($p < 0.001$), whereas Cu was significant only at 370 nm ($p = 0.031$) and the remaining tracers were not significant. The strong Al–absorption relationship, together with the PMF results, provided further support for a contribution from mineral dust. However, correlations among the elemental tracers, particularly Al and Cu ($r = 0.82$), limited the separation of individual source contributions.

We further examined the relationship between eBC and the PMF-derived factors to assess whether carbonaceous material associated with non-exhaust emissions could contribute to the observed enhancement. eBC was strongly correlated with both Dust ($r = 0.94$) and Non-exhaust Emissions ($r = 0.88$). However, Dust and Non-exhaust Emissions were themselves strongly correlated ($r = 0.89$), preventing their individual contributions to eBC from being distinguished. Thus, the correlation between eBC and Non-exhaust Emissions does not provide direct evidence for tyre- or brake-wear-derived BC, and the available data do not allow this contribution to be quantified.

We also acknowledge the potential influence of filter-related artefacts, including multiple scattering and scattering-related apparent absorption. The AE33 dual-spot configuration accounts for filter-loading effects; however, no independent scattering measurements were available to quantify multiple-scattering effects. These artefacts therefore represent an additional uncertainty in the

absolute magnitude of the measured absorption enhancement, although their contribution cannot be quantified with the available data.

The text was modified to:

P5 L124: A source apportionment of PM_{2.5-7.2} light absorption was performed using multiple linear regression analysis to assess the relationships between light absorption and elemental tracers associated with the PMF-derived source factors identified by Aurela et al. (2026). The predictors were selected based on their contribution and specificity to the PMF-derived source factors, together with their concentrations. Al, Cu, S, Na, and Cl were used as representative tracers of dust, non-exhaust emissions, combustion/secondary inorganic aerosol, aged salt, and fresh salt, respectively. Separate multiple linear regression models were fitted for light absorption at 370, 520, and 660 nm, with absorption at each wavelength used as the dependent variable and the five elemental tracers entered simultaneously as predictors. Model performance was assessed using the coefficient of determination (R^2), while the statistical significance of individual predictors was evaluated using regression coefficients, standard errors, t-statistics, and p-values. Multicollinearity among predictors was assessed using variance inflation factors (VIF) and tolerance values. Statistical analyses were performed using jamovi (Version 2.7.38; The jamovi project, 2026), an open-source statistical software platform. The relative contribution of each elemental tracer to light absorption was estimated by multiplying the mean concentration of each tracer over the measurement period by its corresponding regression-derived mass absorption efficiency (MAE) at each wavelength. The resulting tracer-specific absorption estimates were then normalized to the sum of the positive absorption contributions from all tracers to obtain their relative contributions to the regression-derived absorption. Negative regression-derived contributions were set to zero.

Filter-based absorption measurements are subject to several uncertainties, including multiple light scattering within the filter (e.g. Weingartner et al., 2003; Yus-Díez et al., 2021), loading effects as the filter accumulates particles (e.g. Drinovec et al., 2017; Virkkula et al., 2007; Weingartner et al., 2003), and apparent absorption from scattering aerosols (e.g. Arnott et al., 2005; Bond et al., 1999). The AE33 mitigates these artefacts through a dual-spot configuration, which corrects for loading effects and tape-advancement errors, improving the reliability of absorption measurements (Drinovec et al., 2015). However, no independent scattering measurements were available in this study to quantify potential multiple-scattering or scattering-related artefacts. These effects therefore cannot be separated from the measured absorption enhancement and represent an additional source of uncertainty when interpreting its magnitude.

P9 L258: To assess whether the enhanced absorption of PM_{2.5} and PM_{7.2} fractions was primarily driven by a higher particle mass, the absorption coefficients were additionally normalized to PM mass to obtain the MAE (Fig. 2). The MAE decreased systematically with increasing wavelength for all size fractions, from 2.70, 1.83, and 0.97 m² g⁻¹ at 370 nm to 0.88, 0.65, and 0.35 m² g⁻¹ at 950 nm for PM₁, PM_{2.5}, and PM_{7.2}, respectively. Thus, although PM_{7.2} exhibited the highest absolute absorption, its absorption per unit mass was substantially lower than that of the finer fractions. This indicates that the 20–39% enhancement in PM_{7.2} absorption relative to PM₁ is largely associated with the greater mass of particles present in the coarse fraction rather than a higher intrinsic absorption efficiency.

P18 L495: Interestingly, for the PM_{2.5-7.2} fraction, a comparison of light absorption with the PMF-derived factors reported by Aurela et al. (2026), based on measurements from the same urban site and sampling period, further highlighted the role of dust in coarse-particle light absorption. Their five-factor PMF solution comprised Dust, Non-Exhaust Emissions, Aged Salt, Fresh Salt, and a mixed Combustion/Secondary Inorganic Aerosol factor. Of these, Dust showed the strongest and most consistent positive association with absorption across all wavelengths, with Pearson correlation coefficients (*r*) ranging from 0.39 to 0.47, whereas correlations with the other factors were generally weak (Table S1). Although these moderate correlations indicate that dust alone does not explain the variability in absorption, the consistent positive association suggests that dust-related coarse particles can contribute appreciably to light absorption during dust-influenced periods.

P22 L604: To further investigate the chemical components potentially associated with the observed absorption, source-informed multiple linear regression models were applied to the PM_{2.5-7.2} absorption coefficients at 370, 520, and 660 nm (Tables S2-S4). It should be noted that the absorption measurements represent the PM_{2.5-7.2} fraction, whereas the elemental measurements represent PM_{2.5-10}. This size-fraction mismatch should be considered as a limitation when interpreting the relationship between elemental composition and absorption and when extending these results to the full PM₁₀ fraction. The models explained a substantial fraction of the observed absorption variability, with R² values of 0.751, 0.844, and 0.878 at 370, 520, and 660 nm, respectively (Table S2). Among the selected elemental tracers, Al showed a strong and consistent positive association with absorption at all three wavelengths (*p* < 0.001) (Table S3). The corresponding regression coefficients decreased from 2.07 at 370 nm to 1.31 at 520 nm and 1.06 at 660 nm (Table S3). In contrast, Cu was significant only at 370 nm (*p* = 0.031), while S, Na, and Cl were not significant predictors at any of the investigated wavelengths (Table S3). However, collinearity diagnostics indicated moderate to high collinearity among some predictors, particularly Na (VIF = 10.19; tolerance = 0.098), which may reduce the stability of individual coefficient estimates (Table S4). Based on the positive regression-

derived absorption components, the dust-related contribution represented approximately 67%, 70%, and 80% of the estimated elemental contribution at 370, 520, and 660 nm, respectively. The aged-salt tracer Na accounted for approximately 25%, 23%, and 10% at the same wavelengths, while the contribution associated with S, representing combustion/secondary inorganic aerosol, remained relatively small (approximately 8%, 7%, and 8%).

The strong and consistent association of Al with absorption across all three wavelengths provides additional support for a contribution from mineral dust to the enhanced absorption of the PM_{2.5-7.2} fraction. This is consistent with the PMF analysis, in which the Dust factor showed the strongest and most consistent positive correlation with absorption, while the other source-specific elemental tracers were generally not significant in the regression. However, the regression does not provide a quantitative source apportionment of absorption, as several elemental tracers were themselves correlated, particularly Al with Cu ($r = 0.82$) and Na ($r = 0.74$) (Table S5). These covariations reflect the co-occurrence of different aerosol sources and limit the separation of their individual contributions. Nevertheless, the agreement between the PMF and elemental regression results strengthens the evidence for a dust-related contribution to coarse-particle absorption, while also indicating that the observed enhancement likely reflects a mixture of co-occurring mineral and anthropogenic components that cannot be individually resolved with the available measurements.

The relationship between eBC in PM_{2.5-7.2} and the PMF-derived factors was also examined to assess a potential contribution from carbonaceous material associated with a specific source. eBC was strongly correlated with both Dust ($r = 0.94$) and Non-exhaust Emissions ($r = 0.88$), while correlations with Combustion/Secondary Inorganic Aerosol, Fresh Salt, and Aged Salt were weaker ($r = 0.33$, 0.42 , and 0.25 , respectively) (Table S6). However, the strong correlation between Dust and Non-exhaust Emissions ($r = 0.89$) indicates a substantial co-variation between these sources (Table S6), preventing their individual contributions to eBC from being distinguished. Thus, results rather indicate that elevated eBC occurred concurrently with periods characterized by both dust and non-exhaust emissions, without demonstrating a distinct contribution from either source.

P23 L664: The consistent association of Al with coarse-particle absorption, together with the independent PMF results showing the strongest association between the Dust factor and absorption, provides additional evidence for a dust-related contribution to coarse-mode absorption. However, the covariation among elemental tracers limits the quantitative separation of dust from co-occurring anthropogenic sources, and the regression results should therefore not be interpreted as a quantitative source apportionment.

P24 L648: However, mass-normalized absorption was substantially lower in the coarse fraction than in the finer fractions, indicating that the higher absolute absorption observed for $PM_{2.5-7.2}$ was largely associated with its greater particle mass. Nevertheless, the MAE of the coarse fraction demonstrates that coarse particles can contribute intrinsically to light absorption and should not be considered optically negligible.

2) The manuscript shows that $AAE_{370/470}$ decreases from 1.12 (PM_1) to 0.77 ($PM_{2.5}$) and 0.86 ($PM_{7.2}$). This is an interesting finding, but the interpretation is too closely dependent on the classical BC/BrC paradigm, which does not hold when coarse particles contain iron oxides, tyre-derived soot, graphite from brakes, mineral dust with wavelength-dependent absorption.

The authors correctly note:

“The inclusion of coarse particles complicated the interpretation of $AAE_{470/950}$ and related biomass-burning contribution estimates.” This point should be expanded substantially. AAE-based BB% is not valid when coarse particles dominate absorption.

Therefore, it is recommended to dedicate a subsection discussing the limitations of AAE and BB% when sampling $PM_{2.5}$ – PM_{10} , supported by literature on Fe-driven absorption and non-exhaust carbonaceous materials.

Thank you for this important comment. We have substantially expanded the discussion of the limitations of AAE-based interpretation and source apportionment for larger particle size fractions. In particular, we now emphasize that the classical BC/BrC framework may not fully describe coarse-mode absorption, as coarse particles can contain other light-absorbing components, including iron oxides associated with mineral dust and carbonaceous material from non-exhaust sources such as tyre and brake wear. We also clarify that AAE should be considered primarily as an optical descriptor rather than a direct source tracer when the sampled size range extends into the coarse mode. Furthermore, we have expanded the discussion of the implications for AAE-based biomass-burning contribution (BB%) and BrC estimates, noting that the presence of non-BC absorbing components in the coarse fraction may introduce uncertainty or bias into such approaches. These considerations have also been mentioned in conclusions, where we highlight the need for further assessment of AAE-based source-apportionment methods across different PM size fractions. The text was modified to:

P11 L300: The interpretation of AAE becomes particularly challenging for coarse-mode aerosols because the classical framework linking spectrally neutral absorption primarily to BC and enhanced short-wavelength absorption to BrC does not necessarily account for the full range of absorbing

components present in larger particles. As mentioned previously, coarse particles may contain light-absorbing materials such as iron oxides associated with mineral dust as well as carbonaceous components originating from non-exhaust traffic emissions, including tyre and brake wear (e.g. Apicella et al., 2026; Di Biagio et al., 2019; Khardi, 2024). These components may exhibit their own wavelength dependence, meaning that changes in AAE across PM size fractions cannot be uniquely attributed to changes in the relative contributions of BC and BrC. Consequently, AAE should be interpreted primarily as an optical descriptor rather than a direct source tracer, particularly when the sampled size range extends into the coarse mode. This limitation is also relevant for AAE-based biomass-burning source apportionment. The estimation of BB% from AE33 measurements commonly relies on the assumption that enhanced short-wavelength absorption relative to BC is primarily associated with BrC from biomass-burning aerosol. When coarse particles contribute substantially to absorption, however, non-BC absorbing components such as mineral dust and non-exhaust carbonaceous materials may invalidate this assumption, resulting in uncertain or potentially biased BB% estimates. Thus, the observed differences in AAE between PM fractions should be considered when applying AAE-based source-apportionment approaches, particularly for size fractions extending beyond PM_{2.5}.

P24 L656: These results demonstrate that the wavelength dependence of coarse-mode absorption reflects contributions from mineral dust and non-exhaust carbonaceous components, highlighting the complexity of using AAE alone to infer aerosol sources. This variability in AAE can further influence derived aerosol parameters and source apportionment results, including BB% and BrC-related estimates. In particular, the results related to AAE_{470/950} demonstrated that the inclusion of coarse particles can complicate, and potentially compromise, BB% estimation approaches commonly implemented in the AE33-based source apportionment.

P25 L686: Further work is needed to assess the implications of coarse-mode contributions for AAE-based source apportionment, including BB% and BrC estimates particularly given the potential contribution of non-BC absorbing components in the coarse fraction, as well as the applicability of the AE33 multiple-scattering correction to this size fraction.

3) The intercomparison is well executed, showing a 14% difference between AE33 and MAAP for PM₁. However, the manuscript does not discuss whether this discrepancy increases for PM_{7.2}, where scattering artefacts are more severe.

Please, discuss whether AE33 corrections remain valid for coarse-mode aerosols and whether MAAP comparisons could be extended to PM_{2.5}/PM_{7.2}.

In addition, given that AE33 uses a fixed multiple-scattering correction ($C_{ref} = 1.39$), the applicability of this factor to coarse particles is questionable. This parameter is calibrated for submicron, BC-dominated aerosols and assumes optical behaviour typical of fine combustion particles. When applied to $PM_{7.2}$, however, it may introduce systematic biases due to the much stronger coarse-mode scattering and the presence of non-BC absorbers (e.g., Fe-oxides, mineral dust, tyre-derived carbon black). As a result, absorption in the coarse fraction may be partially overestimated. This limitation should be highlighted both in the data interpretation and in the conclusions, and future datasets including additional high-PM dust-resuspension episodes would allow a more robust evaluation of these artefacts and further reinforce the study's findings.

Thank you for this helpful comment. We have addressed the points raised regarding the applicability of the AE33 corrections and the interpretation of coarse-mode absorption. In particular, we now discuss the limitations associated with applying the fixed multiple-scattering correction ($C_{ref}=1.39$) to coarse-mode aerosol, given its different scattering properties and the potential contribution of non-BC absorbing components. We have also highlighted the resulting uncertainty in coarse-fraction absorption and derived eBC in both the discussion and conclusions. Regarding the suggested MAAP comparison, a direct comparison for $PM_{2.5}/PM_{7.2}$ could not be performed because the MAAP measurements in this study were conducted only with a PM_1 inlet. The text was modified to:

P14 L366: In addition, the fixed multiple-scattering correction applied by the AE33 ($C_{ref}=1.39$) is calibrated for submicron BC-rich particles, assuming an optical behavior typical of fine combustion particles. Therefore, this correction factor may not fully account for the optical properties of the coarse fraction, which is characterized by stronger scattering and a greater contribution from non-BC absorbing components. Consequently, increased eBC in larger size fractions does not necessarily indicate an increase in actual black carbon mass. A further limitation concerns the use of the FIDAS optical spectrometer for coarse-mode characterization since its optical response is sensitive to scattering by large particles and to assumptions regarding particle refractive index and shape, which may distort or overestimate dust-related PM peaks (e.g. Meyer et al., 2026; Yus-Díez et al., 2025). Taken together, these considerations highlight the importance of selecting the appropriate PM size fraction when quantifying light absorption and light-absorption components such as eBC, as well as when interpreting potential source contributions.

P14 L377: To better understand the contributions of different particle-size fractions to light absorption and their relative importance across the aerosol size spectrum, the absorption coefficients measured by the AE33 at seven wavelengths (370–950 nm) were partitioned into the PM_1 , $PM_{1-2.5}$, and $PM_{2.5-7.2}$ fractions (Fig. 5). The complete time series of these size-resolved absorption coefficients, along

with the corresponding PM₁, PM_{2.5}, and PM₁₀ mass concentrations for comparison, is shown in Fig. S7. This size-differentiated approach allows the contributions of submicron, 1–2.5 μm , and 2.5–7.2 μm particles to wavelength-dependent aerosol light absorption to be evaluated separately.

Across all wavelengths, PM₁ clearly dominated total absorption (values reported in Sect. 3.2). The monotonic decrease in absorption with wavelength reflects the expected spectral dependence of BC-rich aerosols, although additional enhancement from light-absorbing organics may occur at shorter wavelengths (e.g. Barreira et al., 2024). The PM_{1-2.5} fraction contributed substantially less to total absorption, with campaign-averaged coefficients ranging from 0.48 Mm⁻¹ at 880 nm to 1.13 Mm⁻¹ at 470 nm. Furthermore, it showed a relatively weak wavelength dependence compared with PM₁, indicating a lower abundance of strongly absorbing carbonaceous material. Nevertheless, the enhancement in light absorption from 370 to 470 nm in the PM_{1-2.5} fraction suggests the presence of additional light-absorbing components beyond those typically associated with PM₁ particles. The PM_{2.5-7.2} fraction also showed modest absorption, ranging from 0.85 Mm⁻¹ at 950 nm to 1.66 Mm⁻¹ at 370 nm. This does not necessarily imply that PM_{2.5-7.2} was intrinsically more absorbing than PM_{1-2.5}, because the absorption coefficient depends on both particle mass and mass-specific absorption efficiency. The observed coarse-mode absorption may therefore reflect its larger mass loading together with contributions from light-absorbing components potentially associated with mineral dust and mechanically generated particles, including brake and rail wear (e.g. Ansari and Ramachandran, 2025; Chan et al., 2002; Ito et al., 2018; Liu et al., 2022b). Without size-resolved compositional information, however, these source contributions cannot be assigned unambiguously.

The size-resolved MAE results further clarified this distinction between absorption contribution and absorption efficiency (Fig. 5). PM₁ exhibited by far the highest MAE across the investigated wavelength range, demonstrating that submicron particles were the most efficient light absorbers per unit mass. The 1–2.5 μm fraction nevertheless exhibited appreciable MAEs, particularly from 470 to 950 nm where values decreased from 0.86 to 0.41 m² g⁻¹, indicating the presence of light-absorbing material within the larger fine-particle fraction. In contrast, the MAEs of the 2.5–7.2 μm fraction were substantially lower (0.03 to 0.09 m² g⁻¹). Because PM₁₀ rather than PM_{7.2} mass concentrations were available, these values were normalized by the PM_{2.5-10} mass concentration and therefore represent an approximation of the absorption efficiency of the coarse fraction. Their low values nevertheless indicate that larger particles were much less efficient light absorbers on a mass basis than PM₁. Their measurable contribution to the absorption coefficient therefore appears to arise, at least partly, from their larger particle mass rather than from high mass-specific absorption efficiency.

Overall, PM₁ accounted for the dominant fraction of light absorption across all wavelengths, whereas the PM_{1-2.5} and PM_{2.5-7.2} fractions made smaller but measurable contributions. The PM_{2.5-7.2} fraction

contributed more to the absorption coefficient than $PM_{1-2.5}$ at several wavelengths, despite having a much lower MAE, highlighting the importance of distinguishing between a size fraction's absolute contribution to light absorption and its absorption efficiency per unit mass. These results demonstrate that light absorption was primarily associated with submicron aerosol, while particles larger than 1 μm provided additional, although substantially smaller, contributions to the overall absorption budget.

P24 L677: At the same time, the interpretation of coarse-mode absorption and derived eBC is subject to additional uncertainty because the AE33 multiple-scattering correction is calibrated primarily for fine, BC-rich particles and may not fully represent the optical properties of coarse, strongly scattering particles.

Looking forward, non-exhaust emissions are expected to become an increasingly important source of urban particulate matter. In particular, the electrification of vehicles, while reducing tailpipe emissions, may alter non-exhaust PM_{10} contributions: tyre and road wear emissions are likely to persist or potentially increase, for example due to higher vehicle mass, whereas brake wear emissions may decrease due to regenerative braking. Resuspension of road-deposited material, including mineral dust, may also remain an important contributor to coarse-mode PM, particularly under dry conditions. This underscores the importance of studying light absorption in size-resolved particle fractions to better understand the evolving contributions of both fine and coarse aerosols to urban radiative forcing and air quality. Further work is needed to assess the implications of coarse-mode contributions for AAE-based source apportionment, including BB% and BrC estimates particularly given the potential contribution of non-BC absorbing components in the coarse fraction, as well as the applicability of the AE33 multiple-scattering correction to this size fraction. Measurements across a broader range of dust-resuspension conditions would help establish the generality and robustness of the observed coarse-mode contribution to absorption and derived eBC. In parallel, the harmonization of sampling inlets and analytical methods across global measurement networks, while accounting for differences in monitoring objectives, would facilitate more meaningful comparisons across studies and regions. In particular, the use of appropriate size fractions (e.g., PM_1 or $PM_{2.5}$ for urban air quality and BC monitoring, versus PM_{10} for capturing non-exhaust contributions and broader aerosol assessments) would provide more robust and policy-relevant information for mitigating aerosol impacts on climate and health.

4) The manuscript provides a detailed description of meteorology and PM dynamics, but does not analyze how coarse-mode absorption varies with road wetness, wind speed, dust-resuspension episodes. Given the strong PM_{10} peaks during dry periods, this is essential.

Please, include an analysis of PM_{7.2} absorption vs road wetness and wind speed, and discuss changes in AAE during dust events.

We thank the reviewer for this important suggestion. However, we consider a direct correlation between PM_{7.2} absorption and road wetness or wind speed to be of limited interpretability, because these parameters primarily influence the abundance and composition of coarse particles, whereas absorption depends predominantly on their specific chemical and optical properties. Therefore, changes in PM_{7.2} mass induced by wind resuspension or road dryness do not necessarily translate proportionally into changes in PM absorption. We instead evaluated the influence of these conditions on coarse-particle concentrations and further assessed absorption through mass-normalized absorption, as described in response to Comment 1. Results showed that the highest coarse-particle concentrations occurred preferentially under dry conditions and, unexpectedly, at relatively low wind speeds (Fig. S5). The association with dry conditions is consistent with the fact that surface moisture suppresses particle entrainment. The lack of a positive relationship with wind speed suggests that resuspension at the study site is not controlled by wind speed alone, but by a combination of factors including e.g. deposited material availability, traffic-induced mechanical disturbance, road-surface conditions, and precipitation/drying history. The changes in AAE during dust events are discussed in Sect. 3.4. The text was modified to:

P9 L231: This seasonal process is consistent with the observed episodic PM₁₀ peaks during dry road periods (Figs. 1 and S5). In contrast, wet road conditions, with a maximum water accumulation of 1.2 mm during the campaign, likely suppressed PM₁₀ resuspension. Street surfaces were also occasionally covered by snow, ice, or slush (e.g. Aurela et al., 2026), further inhibiting particle resuspension. Accordingly, no elevated PM₁₀ concentrations were observed under wet surface conditions (Fig. 1 and S5). Wind direction varied substantially throughout the campaign, although prevailing winds were from the west and east, with maximum wind speeds of 4.64 m s⁻¹ and a mean of 1.24 m s⁻¹. These wind patterns likely influenced pollutant dispersion and the episodic dust events observed. Interestingly, PM₁₀ concentrations were higher during periods of relatively low wind speed (Fig. S5). This apparent inverse relationship suggests that wind speed alone did not control the observed PM₁₀ variability. Instead, elevated concentrations may reflect the combined influence of dry surface conditions, the availability of deposited material for resuspension, traffic-induced mechanical disturbance, and atmospheric stability or reduced dispersion during low-wind conditions. No significant long-range transport events were identified during the campaign.

5) The introduction mentions ACTRIS and national networks, but the discussion does not elaborate on the implications of size-cut-off variability for harmonization of eBC datasets, comparability of PM_1 vs $PM_{2.5}$ vs PM_{10} absorption, emission factor estimation, model parameterization.

This is a missed opportunity.

It could be useful to briefly discuss how coarse-mode absorption affects eBC comparability and model inputs.

Thank you for the comment. The implications of size-cut-off variability for harmonization of eBC datasets is discussed in P13L328. However, more emphasis was placed on those implications, including on the comparability between PM absorption measurements. The implications on emission factor estimation and model parameterization are discussed in P16 L422 but the text was also improved for clarity purposes. The manuscript was modified to:

P13 L341: In contrast, ACTRIS prescribes that the PM_{10} size fraction must be used, except in situations where a whole-air inlet is required, leading to higher measured eBC (ACTRIS, 2024). Consequently, measurements from different networks are not directly comparable unless the size fraction is consistent.

P13 L347: In particular, for larger particle fractions, eBC may be overestimated when non-BC absorbing components, such as mineral dust or brown carbon, contribute substantially to the measured absorption, because the absorption is converted to eBC using a BC-specific mass absorption cross-section.

P16 L420: The additional light absorption associated with the $PM_{1-2.5}$ and $PM_{2.5-7.2}$ fractions and the corresponding increase in derived eBC, though smaller than the contribution from PM_1 , can influence the total PM absorption and eBC measurements, particularly during dust events. Including these fractions leads to episodic increases in PM absorption and eBC concentrations, which may affect source-specific emission factors or models that assume eBC is confined to the fine mode. Consequently, PM absorption and derived eBC concentrations obtained using different size fractions are not necessarily directly comparable, as larger size fractions may include additional coarse-mode absorbing particles. Accounting for these contributions is therefore important for accurately interpreting urban PM absorption and eBC measurements, evaluating the relative importance of dust versus combustion sources, and incorporating measured absorption parameters into atmospheric models.

6) The analysis in Section 3.4 relies on a limited number of high-PM episodes; future campaigns capturing additional dust-resuspension events would allow a more robust statistical evaluation and further strengthen the study's conclusions.

The text in Section 3.4 was modified to:

P20L562: These findings should, however, be interpreted in light of the limitations of the available dataset. The statistical analysis of high-PM episodes was limited by the relatively small number of suitable events observed during the measurement period. In particular, additional observations during dust-resuspension events would be valuable for assessing the variability and statistical significance of the coarse-mode contribution to PM absorption and derived eBC. Future measurement campaigns covering a larger number of such events and a wider range of meteorological and source conditions would therefore provide a more robust basis for evaluating the contribution of coarse-mode aerosol and further strengthen the conclusions of this study.

P25 L689: Measurements across a broader range of dust-resuspension conditions would help establish the generality and robustness of the observed coarse-mode contribution to absorption and derived eBC.

7) The use of the FIDAS optical spectrometer for coarse-mode characterization presents methodological limitations: its optical response is strongly affected by large-particle scattering and by assumptions on refractive index and particle shape, which may distort or overrepresent dust-related PM peaks. This constraint should be acknowledged in the data interpretation and conclusions.

The text was modified to:

P14 L371: A further limitation concerns the use of the FIDAS optical spectrometer for coarse-mode characterization since its optical response is sensitive to scattering by large particles and to assumptions regarding particle refractive index and shape, which may distort or overestimate dust-related PM peaks (e.g. Meyer et al., 2026; Yus-Díez et al., 2025). Taken together, these considerations highlight the importance of selecting the appropriate PM size fraction when quantifying light absorption and light-absorption components such as eBC, as well as when interpreting potential source contributions.

Minor Comments

- **Introduction:** add a sentence noting that coarse-mode absorption is poorly constrained in climate and air-quality models.

The text was modified to:

P1 L11: However, coarse particles (2.5–10 μm ; $\text{PM}_{2.5-10}$) can also contribute during spring street dust events in northern countries, while coarse-mode absorption remains poorly constrained in climate and air-quality models.

- **Figure 2:** Absorption normalized by mass would improve interpretation.

The absorption normalized by mass was added to Figures 2 and 5. The text was modified to:

P9 L258: To assess whether the enhanced absorption of $\text{PM}_{2.5}$ and $\text{PM}_{7.2}$ fractions was primarily driven by a higher particle mass, the absorption coefficients were additionally normalized to PM mass to obtain the MAE (Fig. 2). The MAE decreased systematically with increasing wavelength for all size fractions, from 2.70, 1.83, and 0.97 $\text{m}^2 \text{g}^{-1}$ at 370 nm to 0.88, 0.65, and 0.35 $\text{m}^2 \text{g}^{-1}$ at 950 nm for PM_1 , $\text{PM}_{2.5}$, and $\text{PM}_{7.2}$, respectively. Thus, although $\text{PM}_{7.2}$ exhibited the highest absolute absorption, its absorption per unit mass was substantially lower than that of the finer fractions. This indicates that the 20–39% enhancement in $\text{PM}_{7.2}$ absorption relative to PM_1 is largely associated with the greater mass of particles present in the coarse fraction rather than a higher intrinsic absorption efficiency.

P15 L401: The size-resolved MAE results further clarified this distinction between absorption contribution and absorption efficiency (Fig. 5). PM_1 exhibited by far the highest MAE across the investigated wavelength range, demonstrating that submicron particles were the most efficient light absorbers per unit mass. The 1–2.5 μm fraction nevertheless exhibited appreciable MAEs, particularly from 470 to 950 nm where values decreased from 0.86 to 0.41 $\text{m}^2 \text{g}^{-1}$, indicating the presence of light-absorbing material within the larger fine-particle fraction. In contrast, the MAEs of the 2.5–7.2 μm fraction were substantially lower (0.03 to 0.09 $\text{m}^2 \text{g}^{-1}$). Because PM_{10} rather than $\text{PM}_{7.2}$ mass concentrations were available, these values were normalized by the $\text{PM}_{2.5-10}$ mass concentration and therefore represent an approximation of the absorption efficiency of the coarse fraction. Their low values nevertheless indicate that larger particles were much less efficient light absorbers on a mass basis than PM_1 . Their measurable contribution to the absorption coefficient therefore appears to arise, at least partly, from their larger particle mass rather than from high mass-specific absorption efficiency.

Overall, PM_1 accounted for the dominant fraction of light absorption across all wavelengths, whereas the $\text{PM}_{1-2.5}$ and $\text{PM}_{2.5-7.2}$ fractions made smaller but measurable contributions. The $\text{PM}_{2.5-7.2}$ fraction contributed more to the absorption coefficient than $\text{PM}_{1-2.5}$ at several wavelengths, despite having a much lower MAE, highlighting the importance of

distinguishing between a size fraction's absolute contribution to light absorption and its absorption efficiency per unit mass. These results demonstrate that light absorption was primarily associated with submicron aerosol, while particles larger than 1 μm provided additional, although substantially smaller, contributions to the overall absorption budget.

- **XRF:** Provide uncertainties for elemental concentrations.

The text was modified to:

P7 L187: The methodology used to quantify the absolute and relative measurement uncertainties of the ED-XRF elemental concentrations is described in Sect. S3.

S3 P20 L153:

S3 – Assessment of Energy-dispersive X-ray Fluorescence Measurement Uncertainty for Elemental Concentrations in $\text{PM}_{2.5}$ and PM_{10}

To assess the uncertainty relative to the measured concentrations, the absolute uncertainty of each measured element $u(C_{\text{element}})$ was calculated by Eq. (S5):

$$u(C_{\text{element}}) = \sqrt{\frac{s_{\text{element}}^2}{n_{\text{samples}}} + \frac{s_{\text{blank}}^2}{n_{\text{blanks}}}} \quad (\text{S5})$$

, where C_{element} is the blank-subtracted concentration of each element, s_{element} is the standard deviation of the measured concentrations of the element across the samples, n_{samples} is the number of sample replicates (3), s_{blank} is the standard deviation of the blank measurements, and n_{blanks} is the number of blank measurements (66 in total). The relative standard uncertainty $u_r(C_{\text{element}})$ was subsequently calculated by Eq. (S6):

$$u_r(C_{\text{element}}) = \frac{u(C_{\text{element}})}{C_{\text{element}}} \times 100. \quad (\text{S6})$$

The uncertainty of the “Others” category was determined by combining the individual uncertainties of the elements included in this category using the root-sum-of-squares approach.

This provides a measure of the uncertainty in relation to the magnitude of the measured concentration and allows comparison of measurement uncertainty between elements and samples with different concentration levels. For the individual elements included in $\text{PM}_{2.5}$ analysis, relative uncertainties were generally low. The median was $\sim 0.6\%$, compared with a higher mean of $\sim 1.4\%$ and standard deviation (SD) of $\sim 3.9\%$, indicating that the extreme uncertainties were relatively rare. Mean element-specific relative uncertainties were generally lowest for S ($\sim 0.1\%$), Si ($\sim 0.4\%$), and Al ($\sim 0.5\%$), and K ($\sim 0.5\%$), while Zn ($\sim 0.7\%$), Na ($\sim 0.8\%$), Fe ($\sim 1.1\%$), and Ca ($\sim 1.4\%$) showed slightly higher but still generally low

variability. Greater variability was observed for Mg (~2.4%), Ti (~2.9%), and Cl (~5.3%). The largest individual uncertainties reached 64.7% for Cl, 47.6% for Ti, and 19.9% for Mg. The “others” category had also high uncertainties, reaching 246.6% (mean of 26.1%). However, these extreme values were uncommon and mostly related to very low concentrations.

For PM₁₀, relative uncertainties were also generally low for the major elements. Overall, the mean was approximately 0.5% (median ~0.2%) and SD of approximately 2.6%, indicating that the highest uncertainties were relatively uncommon. Element-specific mean uncertainties were generally lowest for S (~0.1%) and Al (~0.2%), as well as for Na, K, Ca, and Fe (~0.3%). On the other hand, Mg (~0.5%), Si (~0.5%), Ti (~1.0%), Cl (~1.0%), and Zn (~1.2%) showed slightly higher but still generally low variability. The largest individual uncertainties were observed for Zn (~79.3%), Si (~21.0%), and “others” (~85.4%), while all remaining elements showed uncertainties below 10 %. As observed for PM_{2.5}, these extreme values were uncommon and associated with very low concentrations.

- **References:** add literature on Fe-driven absorption and non-exhaust carbonaceous particulates. Moreover, in line 94 please insert reference for traffic statistics.

Examples of relevant literature on Fe-driven light absorption and non-exhaust carbonaceous particles have been added to the discussion. The traffic statistics presented in Line 94 were provided by the City of Helsinki based on collected but unpublished data. The text was modified to:

P4 L94: On a typical weekday, around 17,000 vehicles travel along the street, with heavy-duty vehicles accounting for roughly 10 % according to municipal traffic statistics (City of Helsinki, unpublished data, 2025).

P11 L302: As mentioned previously, coarse particles may contain light-absorbing materials such as iron oxides associated with mineral dust as well as carbonaceous components originating from non-exhaust traffic emissions, including tyre and brake wear (e.g. Apicella et al., 2026; Di Biagio et al., 2019; Khardi, 2024).

- Line 224: correct “PM_{2.5}” with “PM_{7.2}”

Thank you for the comment. However, “PM_{2.5}” is correct in this sentence, as the intended comparison is between PM_{2.5} and PM₁. PM_{7.2} is already identified as having the highest absorption across all wavelengths in the same sentence.

- Line 520: delete “in the”.

The text was corrected accordingly.

Reviewer 2:

Major Comments

My first major concern is the interpretation of elemental composition in relation to the observed coarse-particle absorption. The elevated concentrations of Si, Fe and Al clearly support an important contribution from crustal or resuspended road-dust material. However, these measurements do not allow a clear distinction between mineral/resuspended dust and specific direct non-exhaust sources such as brake, tyre or rail wear. In particular, Fe can originate from several different sources, while XRF does not provide information on its chemical form. I therefore suggest that the authors moderate statements attributing the enhanced absorption specifically to non-exhaust wear emissions and distinguish more clearly between evidence for resuspended road/mineral dust and more tentative attribution to individual traffic-wear sources.

A related issue is the difference between the size fractions used for optical and elemental measurements. The coarse absorption contribution is evaluated for $PM_{2.5-7.2}$, whereas the elemental composition used to interpret it represents $PM_{2.5-10}$. Particles between 7.2 and 10 μm therefore contribute to the chemical measurements but not to the corresponding absorption measurements. This mismatch should be explicitly discussed and should be considered when interpreting relationships between composition and absorption or when extending the conclusions to the full PM_{10} fraction.

We thank the reviewer for this important comment. We agree that the elemental measurements do not allow a clear distinction between mineral/resuspended road dust and specific non-exhaust traffic sources such as brake or tyre wear. In particular, Fe can originate from multiple sources, and the XRF measurements do not provide information on its chemical form.

To further investigate the potential contribution of distinct sources to light absorption, we applied a source-informed multiple linear regression using elemental tracers associated with the PMF factors identified by Aurela et al. (2026). Al, Cu, S, Na, and Cl were used as tracers of dust, non-exhaust emissions, combustion/secondary inorganic aerosol, aged salt, and fresh salt, respectively. Regression models were fitted for absorption at 370, 520, and 660 nm, and multicollinearity was assessed using VIF and tolerance values. The relative tracer contributions were estimated from the regression-derived MAEs.

The strong association of Al with absorption, together with the Dust PMF factor, supports a contribution from mineral dust. However, correlations among elemental tracers and between the Dust

and Non-exhaust Emissions factors limited the separation of individual sources. We have therefore emphasized that the observed absorption likely reflects co-occurring mineral and anthropogenic components that cannot be individually resolved. Similarly, the strong covariance between Dust and Non-exhaust Emissions prevents their individual contributions to eBC from being distinguished.

We have moderated the interpretation of the elemental composition and its relationship to coarse-particle absorption. We now explicitly state that elemental measurements alone cannot distinguish non-exhaust traffic emissions from mineral or resuspended road dust. We have also added a statement to the revised manuscript explicitly acknowledging the mismatch between the PM_{2.5-7.2} absorption and PM_{2.5-10} elemental measurements and its implication for interpreting the relationship between elemental composition and absorption and for extending the results to the full PM₁₀ fraction.

The text was modified to:

P21 L581: Overall, the high concentrations of these crustal elements in PM₁₀ suggest that coarse-mode mineral particles may be contributing to the observed light absorption, either directly through e.g. Fe-containing mineral phases or indirectly by coating effects. While the individual correlation with absorption is weak, these elements represent a significant potential source of enhanced light absorption in the coarse fraction, providing a basis for further analysis of mineral dust and metal contributions in urban aerosols. In addition to crustal sources, non-exhaust traffic emissions, such as tyre and brake wear, may also contribute to the observed presence of elements in coarse particles (e.g. Lopez et al., 2023). However, the element measurements alone do not allow these sources to be distinguished from mineral or resuspended road dust, particularly for Fe, which can originate from multiple sources and whose chemical form cannot be resolved by XRF. These anthropogenic contributions may therefore represent additional components of the complex mixture of natural and anthropogenic particles influencing the optical properties of coarse particles in urban environments.

P22 L604: To further investigate the chemical components potentially associated with the observed absorption, source-informed multiple linear regression models were applied to the PM_{2.5-7.2} absorption coefficients at 370, 520, and 660 nm (Tables S2-S4). It should be noted that the absorption measurements represent the PM_{2.5-7.2} fraction, whereas the elemental measurements represent PM_{2.5-10}. This size-fraction mismatch should be considered as a limitation when interpreting the relationship between elemental composition and absorption and when extending these results to the full PM₁₀ fraction. The models explained a substantial fraction of the observed absorption variability, with R² values of 0.751, 0.844, and 0.878 at 370, 520, and 660 nm, respectively (Table S2). Among the selected elemental tracers, Al showed a strong and consistent positive association with absorption at all three wavelengths ($p < 0.001$) (Table S3). The corresponding regression coefficients decreased

from 2.07 at 370 nm to 1.31 at 520 nm and 1.06 at 660 nm (Table S3). In contrast, Cu was significant only at 370 nm ($p = 0.031$), while S, Na, and Cl were not significant predictors at any of the investigated wavelengths (Table S3). However, collinearity diagnostics indicated moderate to high collinearity among some predictors, particularly Na ($VIF = 10.19$; tolerance = 0.098), which may reduce the stability of individual coefficient estimates (Table S4). Based on the positive regression-derived absorption components, the dust-related contribution represented approximately 67%, 70%, and 80% of the estimated elemental contribution at 370, 520, and 660 nm, respectively. The aged-salt tracer Na accounted for approximately 25%, 23%, and 10% at the same wavelengths, while the contribution associated with S, representing combustion/secondary inorganic aerosol, remained relatively small (approximately 8%, 7%, and 8%).

The strong and consistent association of Al with absorption across all three wavelengths provides additional support for a contribution from mineral dust to the enhanced absorption of the $PM_{2.5-7.2}$ fraction. This is consistent with the PMF analysis, in which the Dust factor showed the strongest and most consistent positive correlation with absorption, while the other source-specific elemental tracers were generally not significant in the regression. However, the regression does not provide a quantitative source apportionment of absorption, as several elemental tracers were themselves correlated, particularly Al with Cu ($r = 0.82$) and Na ($r = 0.74$) (Table S5). These covariations reflect the co-occurrence of different aerosol sources and limit the separation of their individual contributions. Nevertheless, the agreement between the PMF and elemental regression results strengthens the evidence for a dust-related contribution to coarse-particle absorption, while also indicating that the observed enhancement likely reflects a mixture of co-occurring mineral and anthropogenic components that cannot be individually resolved with the available measurements.

The relationship between eBC in $PM_{2.5-7.2}$ and the PMF-derived factors was also examined to assess a potential contribution from carbonaceous material associated with a specific source. eBC was strongly correlated with both Dust ($r = 0.94$) and Non-exhaust Emissions ($r = 0.88$), while correlations with Combustion/Secondary Inorganic Aerosol, Fresh Salt, and Aged Salt were weaker ($r = 0.33$, 0.42, and 0.25, respectively) (Table S6). However, the strong correlation between Dust and Non-exhaust Emissions ($r = 0.89$) indicates a substantial co-variation between these sources (Table S6), preventing their individual contributions to eBC from being distinguished. Thus, results rather indicate that elevated eBC occurred concurrently with periods characterized by both dust and non-exhaust emissions, without demonstrating a distinct contribution from either source.

My second main concern concerns the relatively low eBC/absorption levels and the derivation of the incremental size-fraction absorption by subtraction between separate AE33 instruments. Since the

PM_{1-2.5} and PM_{2.5-7.2} absorption signals are relatively small, even modest instrument-to-instrument offsets may become important relative to the calculated coarse contribution. The occurrence of slightly negative derived absorption values under some conditions further suggests that measurement uncertainty can be comparable to the incremental signal. Although the intercomparison demonstrates good correlations between the instruments, it would be useful to quantify the absolute differences between instruments and propagate this uncertainty into the derived PM_{1-2.5} and PM_{2.5-7.2} absorption values. This would strengthen confidence in the reported coarse-mode contribution.

We thank the reviewer for this important comment. We have now quantified the instrument-to-instrument variability using the AE33 intercomparison and propagated the associated uncertainty to the incremental PM_{1-2.5} and PM_{2.5-7.2} absorption values. The uncertainty calculation and propagation procedure have been added to the Supplementary Information (Eqs. S1–S4). We have also added an assessment of the negative incremental absorption values by comparing them with the corresponding propagated measurement uncertainties. The text was modified to:

P6 L156: The intercomparison data were also used to quantify the instrument-to-instrument uncertainty and to propagate this uncertainty to the incremental PM_{1-2.5} and PM_{2.5-7.2} absorption values, as described in the Supplementary Information (Sect. S2).

S2 P19 L121:

S2 – AE33 measurement uncertainty and incremental absorption

The uncertainty associated with the AE33 absorption measurements was assessed using the intercomparison of the three co-located AE33 instruments. As described in Sect. 2.2.1, the three AE33 instruments sampled PM₁ through the same cyclone under identical sampling conditions during the intercomparison. To estimate the relative uncertainty of each instrument, the coefficient of variation (CV) for each instrument pair was first calculated as Eq. (S1):

$$CV = \frac{SD_{ij}}{(b_i + b_j)/2} \times 100 \quad (S1)$$

, where SD_{ij} is the standard deviation of the pairwise differences between instruments i and j , and b_i and b_j are the corresponding mean absorption values. The pairwise CV between the AE33 instruments ranged from 26 to 45%, depending on wavelength.

The relative uncertainty of each individual instrument was then calculated as Eq. (S2):

$$\mu_i = \sqrt{\frac{(CV_{ij}/100)^2 + (CV_{ik}/100)^2 - (CV_{jk}/100)^2}{2}}, i, j, k \in \{1, 2, 3\} \quad (S2)$$

, where μ_i represents the relative uncertainty of instrument i , and CV_{ij} , CV_{ik} and CV_{jk} are the coefficients of variation calculated for the corresponding instrument pairs, with i , j , and k denoting

the three AE33 instruments. The relative uncertainty of each instrument was estimated assuming independent relative errors resulting in values of 18–32% depending on instrument and wavelength. These relative uncertainties were subsequently converted to absolute uncertainties by multiplying each relative uncertainty by the corresponding mean absorption measurement and were propagated to the incremental size-fraction absorption using Eqs. (S3) and (S4):

$$U_{PM_{1-2.5}} = \sqrt{U_{PM_1}^2 + U_{PM_{2.5}}^2} \quad (S3)$$

$$U_{PM_{2.5-7.2}} = \sqrt{U_{PM_{2.5}}^2 + U_{PM_{7.2}}^2} \quad (S4).$$

The resulting propagated uncertainties for $PM_{1-2.5}$ absorption ranged from 1.35 to 3.43 Mm^{-1} , while those for $PM_{2.5-7.2}$ ranged from 1.40 to 2.96 Mm^{-1} , from 370 to 950 nm, respectively. These values were comparable to the standard deviations of the corresponding pairwise differences observed during the instrument intercomparison, which ranged from 1.36–3.43 Mm^{-1} (AE33_2–AE33_1), 1.67–3.45 Mm^{-1} (AE33_3–AE33_1), and 1.39–2.96 Mm^{-1} (AE33_3–AE33_2).

Because the incremental absorption was obtained by subtracting two relatively small absorption signals, negative values can occur when the difference is comparable to the measurement uncertainty. Negative incremental values were observed for 21.6–35.2% of the $PM_{1-2.5}$ data and 27.4–30.9% of the $PM_{2.5-7.2}$ data, depending on wavelength. Importantly, 94.4–97.3% of the negative $PM_{1-2.5}$ values and 86.2–88.7% of the negative $PM_{2.5-7.2}$ values were within the corresponding propagated measurement uncertainty. Thus, the large majority of the negative incremental values are consistent with the estimated measurement variability and do not necessarily indicate a physically meaningful negative absorption contribution. This uncertainty assessment was therefore taken into account when interpreting the incremental coarse-mode absorption results.

I also suggest greater caution in the interpretation of eBC in the coarse fraction. In the presence of mineral dust and other non-BC absorbers, the AE33 converts measured attenuation into eBC using a BC-specific mass absorption coefficient. Therefore, the additional eBC measured with the larger inlet should not necessarily be interpreted as an increase in actual black-carbon mass. This distinction is particularly important here because one of the main findings of the study is that coarse non-BC particles can influence the optical measurement. The terminology throughout the manuscript should make this clear.

We thank the reviewer for this important comment. We agree that eBC derived from AE33 measurements in the coarse fraction should not necessarily be interpreted as an increase in actual black carbon mass, particularly in the presence of non-BC absorbing particles. We have revised the terminology and interpretation throughout the manuscript to clarify that eBC is derived from

measured light attenuation and that increased eBC in larger size fractions may partly reflect absorption by non-BC components. We also added a discussion of the additional uncertainty associated with the AE33 multiple-scattering correction for coarse particles and clarified that AE33-derived eBC in larger size fractions should be interpreted with caution. The text was modified to:

P13 L328: Due to the differences in light absorption across size fractions, the corresponding AE33-derived eBC concentrations were evaluated across the PM₁, PM_{2.5}, and PM_{7.2} fractions (Fig. S8).

P13 L331: These results highlight the influence of the inlet cut-off size on the measured eBC. However, the observed increase should not necessarily be interpreted as an equivalent increase in actual black carbon mass, as coarse non-BC absorbing particles can contribute to the measured attenuation.

P14 L364: Nonetheless, selecting only larger particle size fractions may introduce additional uncertainty in AE33-derived eBC, because absorption in larger particles can originate from both true light-absorbing material (e.g., internally mixed black carbon) and non-BC absorbing components such as mineral dust and other light-absorbing material. In addition, the fixed multiple-scattering correction applied by the AE33 ($C_{\text{ref}}=1.39$) is calibrated for submicron BC-rich particles, assuming an optical behavior typical of fine combustion particles. Therefore, this correction factor may not fully account for the optical properties of the coarse fraction, which is characterized by stronger scattering and a greater contribution from non-BC absorbing components. Consequently, increased eBC in larger size fractions does not necessarily indicate an increase in actual black carbon mass.

Finally, the conclusions drawn from the high-dust and high-traffic subsets would benefit from some additional indication of statistical robustness, particularly because these conditions represent a relatively limited fraction of the campaign. Reporting the number of observations or independent episodes contributing to each category, together with some measure of variability or uncertainty, would help demonstrate how representative these results are.

We thank the reviewer for this suggestion. We have revised Sect. 3.4 to provide the number of observations contributing to the high-PM₁₀ dust and high-traffic exhaust categories. We have also included measures of variability for the reported eBC concentrations and PM_{2.5}/PM₁₀ ratios to better characterize the spread within these subsets. These additions provide further context for the representativeness and robustness of the differences observed between the two pollution regimes. The text was modified to:

P17 L460: This corresponded to $N = 1563$ observations for the high PM_{10} dust category and $N = 3679$ observations for the high-traffic exhaust category. The variability in studied parameters is presented as mean $\pm$ SD to account for the variability within each category.

P17 L464: The distinct aerosol regimes represented by the high PM_{10} dust and high-traffic exhaust categories were further validated using eBC concentrations (Fig. S15) as well as $PM_{2.5}/PM_{10}$ ratios (Fig. S16). Average eBC levels were considerably higher during high-traffic exhaust conditions ($0.97 \pm 1.38 \mu\text{g m}^{-3}$) compared with high PM_{10} dust episodes ($0.43 \pm 0.50 \mu\text{g m}^{-3}$) and the campaign average ($0.48 \pm 0.71 \mu\text{g m}^{-3}$), indicating the stronger influence of fresh combustion emissions in traffic-dominated periods. The $PM_{2.5}/PM_{10}$ ratio also supported this distinction, with high PM_{10} dust conditions showing a coarse-dominated aerosol ($19.6 \pm 5.2 \%$), while high-traffic exhaust periods exhibited a fine-particle-dominated regime ($62.7 \pm 16.3 \%$); the campaign-average ratio was $46.1 \pm 25.6 \%$. Together, these indicators demonstrate that the classification criteria reliably capture two contrasting aerosol environments, coarse-rich resuspension episodes versus fine-rich traffic emissions, providing a robust basis for interpreting the observed differences in light-absorption across the particle size fractions of this study (PM_1 , $PM_{2.5}$, and $PM_{7.2}$).

For the PM_1 fraction, campaign-averaged absorption was the highest under high traffic exhaust conditions, reaching up to $19.2 \pm 24.2 \text{ Mm}^{-1}$ at the shortest measured wavelength, compared to $10.8 \pm 8.1 \text{ Mm}^{-1}$ under high PM_{10} dust conditions and $11.0 \pm 14.3 \text{ Mm}^{-1}$ for the full dataset. This highlights the dominant role of traffic-related ultrafine particles in light absorption. The $PM_{1-2.5}$ fraction contributed less overall but still showed notable variation across conditions: under high PM_{10} dust episodes, campaign-averaged absorption ranged from 0.70 ± 1.15 (880 nm) to 1.59 ± 1.87 (470 nm) Mm^{-1} across the wavelengths, whereas under high traffic exhaust conditions it was near zero or slightly negative at some wavelengths (-0.27 ± 4.2 to $0.95 \pm 3.2 \text{ Mm}^{-1}$ at 370 and 470 nm, respectively), reflecting the minor role of this fraction in fresh traffic plumes. The campaign-average values for $PM_{1-2.5}$ ranged from 0.48 ± 1.26 (880 nm) to 1.13 ± 2.36 (470 nm) Mm^{-1} , indicating a modest contribution to total absorption.

For the $PM_{2.5-7.2}$ fraction, absorption increased significantly under high PM_{10} dust conditions, with mean values ranging from 1.56 ± 1.69 (950 nm) to 3.23 ± 3.74 (370 nm) Mm^{-1} . Despite the substantial variability within this fraction, the results indicate that coarse particles can make a measurable contribution to light absorption during dust-influenced conditions. In contrast, under high traffic exhaust conditions, absorption by this fraction remained close to zero, ranging from -0.04 ± 9.11 at 470 nm to $0.32 \pm 4.70 \text{ Mm}^{-1}$, at 880 nm. The large standard deviations relative to the mean values

indicate considerable variability, but the near-zero mean absorption suggests that the $\text{PM}_{2.5-7.2}$ fraction contributes little, on average, to light absorption under fresh traffic-influenced conditions.

P19 L510: To further investigate how main aerosol sources influence the wavelength dependence of absorption, the AAE was analysed for PM_1 , $\text{PM}_{2.5}$, and $\text{PM}_{7.2}$ under the previously mentioned pollution regimes (Fig. S17). For comparison purposes, the AAE values for the full campaign dataset are also shown in Fig. S17, although they were previously presented in Sect. 3.2. Under high PM_{10} dust conditions, the AAE exhibited consistently elevated values across all wavelength pairs and size fractions, indicating a pronounced wavelength dependence of absorption. For $\text{AAE}_{370/470}$, values were 1.23 ± 0.23 , 1.07 ± 0.30 , and 1.03 ± 0.25 for PM_1 , $\text{PM}_{2.5}$, and $\text{PM}_{7.2}$, respectively. Similarly, $\text{AAE}_{370/950}$ (1.30 ± 0.12 , 1.22 ± 0.14 , and 1.12 ± 0.13) and $\text{AAE}_{470/950}$ (1.32 ± 0.09 , 1.28 ± 0.10 , and 1.16 ± 0.10) decreased slightly with increasing particle size, suggesting a modest reduction in spectral dependence in the coarse fraction when near-infrared wavelengths are included. $\text{AAE}_{470/520}$ displayed the highest values overall (1.60 ± 0.18 , 1.71 ± 0.18 , and 1.59 ± 0.16), indicating strong spectral curvature in the mid-visible range during dust-dominated episodes.

In contrast, high traffic exhaust conditions were characterized by generally lower AAE values, particularly for the $\text{PM}_{7.2}$ fraction, indicating a weaker wavelength dependence of light absorption. For $\text{AAE}_{370/470}$, values were 1.04 ± 0.17 , 0.73 ± 0.13 , and 0.78 ± 0.22 for PM_1 , $\text{PM}_{2.5}$, and $\text{PM}_{7.2}$, respectively. $\text{AAE}_{370/950}$ remained close to unity across all size fractions (1.13 ± 0.08 , 1.04 ± 0.06 , and 1.03 ± 0.10), consistent with a black carbon-dominated absorption regime typical of fresh combustion emissions. The $\text{AAE}_{470/950}$ (1.17 ± 0.06 , 1.15 ± 0.07 , and 1.12 ± 0.07) was slightly higher and more invariant across particle sizes, likely due to the increased contribution of near-infrared absorption, where black carbon exhibits relatively spectrally flat behavior and reduced sensitivity to e.g. particle coating or mixing state effects. Although $\text{AAE}_{470/520}$ was somewhat higher (1.40 ± 0.09 , 1.53 ± 0.11 , and 1.44 ± 0.11), this enhancement is restricted to the mid-visible range and does not indicate strong overall spectral dependence of absorption.

P20 L543: Regarding the distinct aerosol regimes defined in this section, a shift in $\text{AAE}_{470/950}$ from the averaged values during traffic-dominated conditions to those during high-dust periods would lead to a substantial increase in the inferred BB% contribution across the studied size fractions (Fig. S18), rising by 233 % for PM_1 (from 12 ± 11 % to 40 ± 18 %), by 256 % for $\text{PM}_{2.5}$ (from 9 ± 12 % to 32 ± 18 %), and by 150 % for $\text{PM}_{7.2}$ (from 4 ± 13 % to 10 ± 17 %). However, this increase was likely not directly driven by dust itself, as BB% for $\text{PM}_{7.2}$ was higher during low-dust and traffic conditions than during high-dust periods (Fig. S18). Overall, the observed results suggest that the variability in BB% was primarily associated with particle-size effects on AAE, whereby lower $\text{AAE}_{470/950}$ values

for larger size fractions lead to reduced BB% estimates. This interpretation is further supported by the diurnal patterns: dust-related absorption peaks during daytime (Fig. S10) whereas BB% values are consistently higher in the evening (Fig. S19), indicating a decoupling between dust-driven absorption and the inferred biomass burning percentage.