RESPONSE LETTER#2 (egusphere-2024-3539)

Dear Anonymous Referee #2:

We would like to thank the referee for the comments. In the following paragraphs, we report the referee's comment in black and the author's answer in blue. The modification in the manuscript is marked as blue.

This manuscript presents a comprehensive analysis of the microphysical properties of refractory black carbon (rBC) aerosols, focusing on their seasonal variability and correlations with air mass trajectories at a central European background site. The data collected, along with the advanced instrumentation employed (e.g., SP2 and thermodenuder systems), provide valuable insights into the physical characteristics and atmospheric interactions of rBC. Overall, the manuscript is well-structured. However, there are a few areas where additional clarification or detail could enhance the clarity of the work.

Our response: We thank the reviewer for the thoughtful and valuable comments and suggestions, which were very helpful in improving our manuscript. We revised the manuscript carefully, as described in our point-to-point responses to the comments.

L75: Why were August and December specifically chosen to represent summer and winter? Are there other reasons for this selection, beyond the observed highest and lowest rBC concentrations?

Our response: Thanks for the comments. The reason we selected these two months is based on the rBC properties. As shown in Fig.1, the CT diurnal variation exhibited two different modes: one with a peak during the daytime (September to October) and another one with a peak at night (November to January). The most evident diurnal variations of these two modes, with relatively higher CT, were found in August and December, respectively. In addition, the highest and lowest concentrations and the most significant shift of size distribution were observed in these two months as well, as shown in Fig.2 below. These two months exhibited the most contrasting rBC properties and coincidentally fall within summer and winter. Furthermore, practice constrains our selection as well, including the limited measurement in summer, delays in ACSM data processing, and instrument maintenance.

To clarify these in the manuscript, we have revised the text to :

"In this study, we focused on August and December, the two months with the most contrasting rBC properties, which coincidentally correspond to summer and winter. As shown in Fig. S1 and S2, these months exhibited the highest and lowest BC mass concentrations, the smallest and largest rBC core sizes, and distinct diurnal variations in coating thickness."

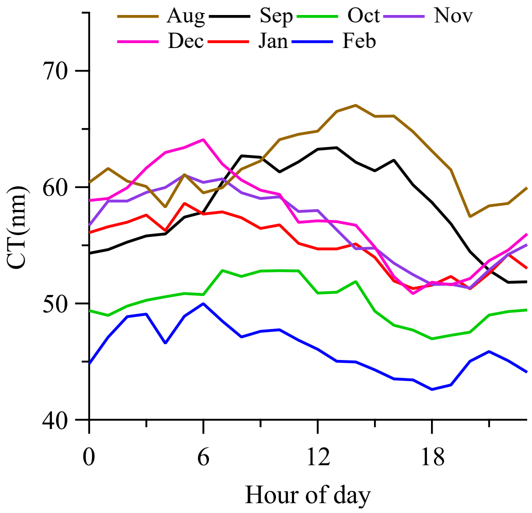

Figure.1 Diurnal variation of rBC mass concentration and coating thickness of different months.

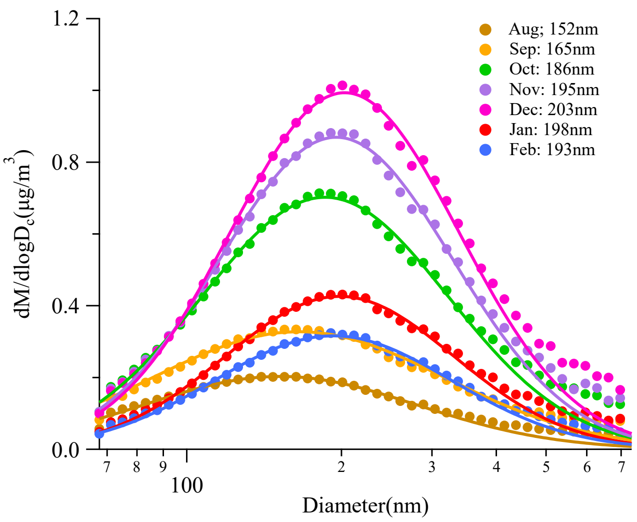

Figure.2 rBC mass size distribution of different months.

Lines 9-10: I suggest revising to: "Uncertainties persist in estimating the radiative forcing of black carbon (BC) due to an incomplete understanding of its microphysical properties."

Our response: Thanks for your comment. We have revised the sentence to "Uncertainties persist in estimating the radiative forcing of black carbon (BC) due to an incomplete understanding of its microphysical properties. "

Lines 82-83: I recommend revising to: "Figure 1 presents measurements taken at the Melpitz research site (12°56'E, 51°32'N, 86 m a.s.l.) of the Leibniz Institute for Tropospheric Research (TROPOS), located 50 km northeast of Leipzig, Germany."

Our response: Thanks for your comment. We have revised the sentence in accordance with the Referee#1's comment, which aligns closely with your recommendation.

L120: Consider adding a brief explanation of the 'LEO-fit' method for readers who may be unfamiliar with this technique.

Our response: We have added the explanation of 'LEO-fit' as follows:

"The leading edge-only (LEO) fit method, which uses a Gaussian fit of the scattering profile before coating evaporation to reconstruct the scattering signal, was applied in this study. Technical details about the LEO-fit approach can be found in Gao et al. (2007). "

Lines 165-168: I suggest revising to: "An aerosol chemical speciation monitor (ACSM, Aerodyne Research, MA, USA; Ng et al., 2011) and an Aerodyne high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS, hereafter referred to as AMS, DeCarlo et al., 2006) were used to measure the bulk chemical composition of non-refractory PM1 aerosol species, including organic aerosols (OA), nitrate, sulfate, ammonium, and chloride."

Our response: We have revised the sentence to "An aerosol chemical speciation monitor (ACSM, Aerodyne Research, MA, USA; Ng et al., 2011) and an Aerodyne high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS, hereafter referred to as AMS, DeCarlo et al., 2006) were used to measure the bulk chemical composition of non-refractory PM1 aerosol species, including organic aerosols (OA), nitrate, sulfate, ammonium, and chloride."

Lines 175-178: I recommend revising to: "Aerosol components exhibited clear seasonal variations. In summer, organic aerosols (OA) dominated, with a mean mass fraction of 55 ± 13%. In contrast, during winter, the OA fraction decreased to 29 ± 14%, while the nitrate fraction significantly increased to 29 ± 15%, compared to 8 ± 5% in summer." It is important to ensure consistency in the number of decimal places throughout the manuscript.

Additionally, while organic aerosols are the predominant component in summer, nitrate plays a key role in winter. However, the manuscript does not discuss the reasons behind these seasonal variations. I would suggest expanding the discussion on the possible drivers behind these variations. For instance, could meteorological factors like temperature and humidity be influencing the observed seasonal trends, or are they more likely related to anthropogenic activities? This would provide a more comprehensive context for understanding the data."

Our response: Thanks for your comment. We have received the sentence to "Aerosol components exhibited clear seasonal variations. In summer, organic aerosols (OA) dominated, with a mean mass fraction of 55 ± 13%. In contrast, during winter, the OA fraction decreased to 29 ± 14%, while the nitrate fraction significantly increased to 29 ± 15%, compared to 8 ± 5% in summer."

We appreciate your suggestion to expand the discussion on the drivers of these seasonal variations. Our manuscript already includes some discussions on these topics. For example, regarding the influence of anthropogenic activities, we have the discussion on the heating system in Melpitz village and residential heating emissions. Additionally, meteorological factors are considered in the discussion of the diurnal variation of the mixing state. While a more detailed and comprehensive investigation would be valuable, it goes beyond the scope of this manuscript and will be explored in future research.

L200: Why were 100-meter back trajectories chosen for the analysis? A brief explanation of this choice and the methodology used for back-trajectory and wind analysis would be beneficial in the methods section.

Our response: Thanks for your comment. We selected the 100-meter back trajectories for analysis because there was no significant difference between the back trajectories at 100 m, 500 m, and 1000 m. As shown in Figure 1, the clusters of back trajectories at different heights followed similar paths and originated from similar directions. Using the lowest altitude ensures the most direct connection to ground-based measurements and aligns well with our wind analysis without losing significant information. Given that higher-altitude trajectories exhibited similar patterns, we found 100 m to be the most relevant choice for assessing local and regional influences on rBC. In addition, we have added a new chapter about the HYSLPIT mode and NWR at the "Method" part:

2.3 Non-parametric Wind Regression and Backward Trajectory Analysis

To investigate both the local and predominant wind sector associated with transported emission sources and rBC properties, we performed a Non-parametric Wind Regression (NWR) analysis using ZeFir, an Igor-based tool developed by Petit et al. (2017). NWR smooths data over a fine grid, allowing the estimation of weighted concentrations for any wind direction (φ) and wind speed (v) pair, with weighting coefficients determined via Gaussian-like functions (Henry et al., 2009). Additionally, we employed the NOAA HYbrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT-4) model to generate 72-hour backward trajectories at 1-hour resolution, at 100 m above the sampling site's ground level. These trajectories trace air parcel origins and transport pathways, providing insight into potential pollutant source regions (Cohen et al., 2015). The resulting backward trajectories are presented in Fig. S3. Furthermore, to identify periods with similar geographical source regions and rBC physical properties, the cluster analysis was subsequently applied to the backward trajectories by using ZeFir. The optimal number of clusters was determined based on the total spatial variance (TSV) (Syakur et al., 2018) and three different clusters were identified in each season.

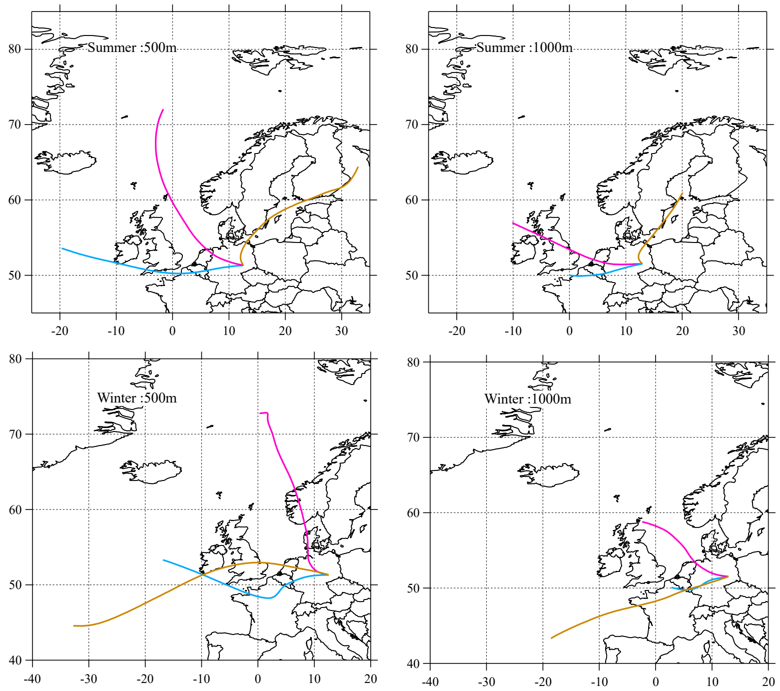

Fig.1 Back trajectory clusters of different height during summer and winter.

Lines 267-268: I recommend providing some evidence regarding local residential wood heating or burning activities in this context. Additionally, consider reorganizing this section for better clarity.

Our response: We have added the evidence regarding the residential heating and revised the sentence as follows:

"In addition, van Pinxteren et al. (2024) observed that the fraction of biomass and coal combustion emissions at Melpitz was highest during winter. Similarly, Atabakhsh et al. (2023) also found 85% of BC at Melpitz emitted from biomass and coal combustion during winter.

Therefore, residential heating could be an important emission source correlated to the large rBC core at Melpitz during this season."

L245-260: The role of biomass burning in influencing rBC size is well addressed. However, do you have any hypotheses regarding why rBC size is smaller in summer, aside from biomass burning? Could other emission sources or atmospheric processes also contribute to this seasonal variation?

Our response: Thanks for your comment. This is an important question; however, a definitive answer is beyond the scope of our manuscript. The rBC core is primarily influenced by different emission sources (Bond et al., 2013; Liu et al., 2020). The smaller size of rBC may be attributed to the lowest emission of biomass burning and coal combustion (Atabakhsh et al., 2023) in summer. In addition, the lower fraction of local emission (18%) of local emission compared to winter (34%) could also be one reason for the small size of rBC core; the large rBC could be removed during the long transportation. Furthermore, wet deposition due to precipitation and the activation of rBC as cloud condensation nuclei (CCN) could also contribute to the variation of rBC size. However, further measurements and analyses are needed to fully investigate these mechanisms..

Figure 7: Could you provide the rationale for using particle volume in this plot? A brief explanation would be helpful for clarity.

Our response: Thanks for your comment. Due to the small MMD of rBC at Melpitz, there are significantly more small rBC cores than large ones. A substantial fraction of coated rBC falls below the detection limit, preventing us from obtaining coating information. As shown in Fig.5 below, when using particle numbers, there are no significant differences in the size-reolved CT patterns across different air masses. Since rBC optical properties are correlated with particle volume, where rBC with thicker coatings or larger cores absorb more light, and inspired by Liu et al. (2014), who used the scattering enhancement of coated rBC (which is volume-dependent) to quantify the size-resolved mixing state and perform source apportionment, we decided to use particle volume for our analysis. This approach allows for a more distinct characterization of the size-resolved mixing state across different air masses.

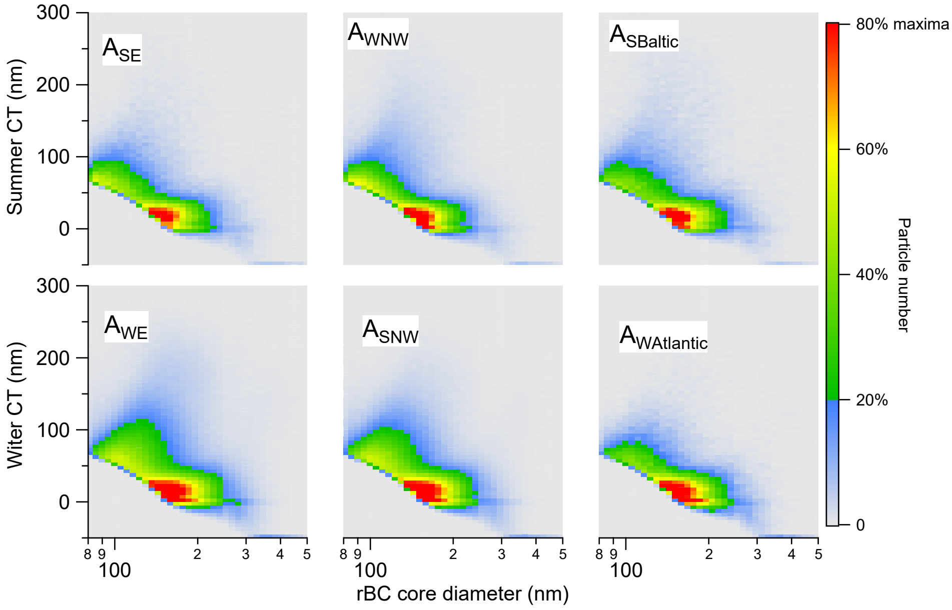

Fig.5 Size resolved CT quantified by rBC numbers of di

L325: Could you provide further evidence or examples to support the connection between "thinly coated rBC" and liquid fuel combustion?

Our response: Thanks for your question. As shown in the figure below from Liu et al. (2019), rBC from traffic emission exhibits a smaller core size and low scattering enhancement compared to rBC from the traffic and wood burning emission, which has a large core size and higher absorption enhancement. The rBC_{thin} in our study exhibited similar properties to those of rBC from traffic emissions. In addition, the mass fraction of rBC_{thin} increased during rush hours when the rBC mass concentration increased, as discussed in Section 3.3.2 of the manuscript. However, more direct evidence would require a source apportionment analysis, such as examining hydrocarbon-like organic aerosols (HOA), which we are unable to conduct currently. This could be a valuable direction in our later research, to combine the source apportionment of rBC with the size-resolved CT.

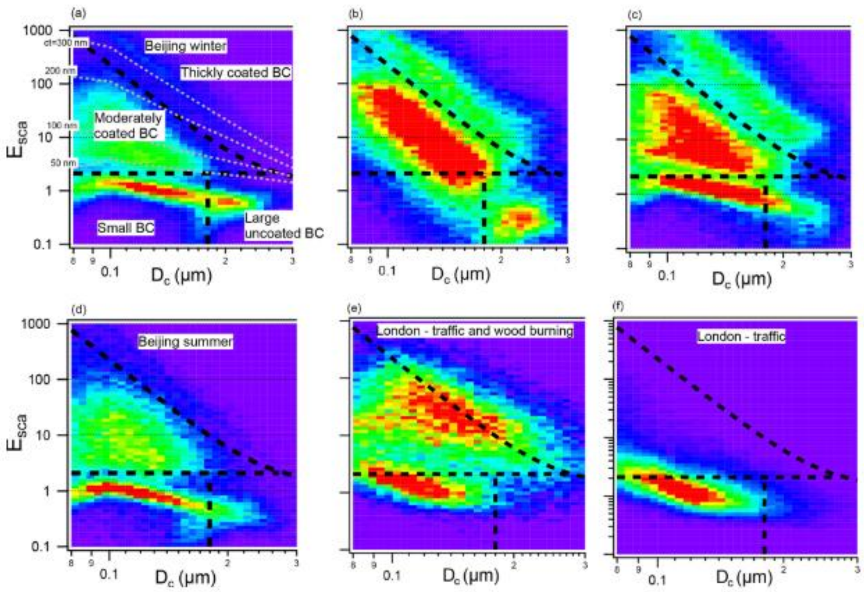

Figure 9. Scattering enhancement (E_{sca}) as a function of BC core size (D_c) for the three periods (as period I–III indicated in Fig. 3) in Beijing winter **(a–c)**, Beijing summer **(d)**, London with mixed sources **(e)**, and London with traffic source **(f)**. Each plot is coloured by particle number density (the colour scale is set to be red when the number density is above 70 % of the maxima in each panel). The particles are separated as four groups using the borders (from top to bottom) at $y = 3.38 + 0.000436 * \times \hat{-}5.7$, $y = 2.1$, $x = 0.18$, as shown by dashed lines on each plot. The dashed grey lines on **(a)** denote coating thicknesses mapped on the $E_{sca} - D_c$ plot.

L410-415: The statement "This non-simultaneous variation between CT and mfcoated may relate to local emissions and atmospheric processes" could benefit from further clarification. Could you elaborate on how these factors might contribute to this variation?

Our response: Thanks for your comment. According to the comment of Referee#1, we have removed the content related to mf_{coated} in the manuscript.

An additional suggestion would be the manuscript could benefit from a more detailed explanation of the observed relationship between rBC size and/or coatings with atmospheric chemical processes. Could the authors elaborate on how factors like photochemistry (or indicator), relative humidity, or long-range transport of pollutants might contribute to the observed changes in rBC physical properties?

Our response: We appreciate your suggestion to combine the relationship between rBC properties with atmospheric chemical processes. In our manuscript, we have discussed some of these factors, such as photochemical processes influencing coating growth and volatility during summer, and long-range transport contributing to smaller rBC core sizes, thicker coatings, and a higher fraction of low-volatility coatings. We agree that a more in-depth discussion of chemical processes would enhance the manuscript and provide further insight into rBC properties. A comprehensive analysis of these factors requires additional measurements and data. For instance, investigating photochemical influences in detail would need a thorough analysis of ACSM data, which was not yet available when this manuscript was prepared. Similarly, a systematic assessment of meteorological factors and long-range transport effects, combined with rBC properties, would extend beyond the content of a single study. This manuscript aims to provide a fundamental characterization of rBC at Melpitz. Follow-up studies will address these additional factors in more detail, including the role of chemical processes and optical properties.

References:

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