Revisions of the paper
The synergy of morphology and mixing state on the absorption of coated black carbon soot

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First of all, we would like to thank the two anonymous reviewers and Dr Schnaiter for their careful reading of the paper and their helpful comments. The following provides a point-by-point response to the reviewer's comments (comments in black, answers in blue).
To note that in the revised version of the manuscript, in addition to changes made to reply to the specific comments received, we have also performed some small adjustments in the text and figures to correct typos or to improve readability.

Point-by-point response to reviewers

Comments by Dr. M. Schnaiter

This is a very interesting manuscript on the absorption enhancement of black carbon (BC) through coating with sulphuric acid and SOA mass. However, the manuscript lacks a discussion of these results in the context of earlier work, in particular the study by Schnaiter et al. (2005), who performed similar experiments in the AIDA chamber. The experimental setup and conditions in the AIDA chamber were very similar to those used in the present study in the CESAM chamber. Notably, Schnaiter et al. (2005) reported absorption enhancements of 1.8 to 2 even for BC aggregates that fully collapsed during the experiments, which is somewhat in contradiction to the low absorption enhancements presented in the present study.
Good scientific practice requires that new results be discussed in the context of existing literature. In this case, a comparison with Schnaiter et al. (2005) is essential, particularly given that the experiments were conducted under very similar conditions, namely in aerosol chambers with comparable instrumentation and experimental procedures. Without such a comparison, it remains unclear how this work advances our understanding of the BC absorption enhancement relative to studies conducted more than two decades ago.

We thank Dr. Schnaiter for his comment. In order to follow his suggestion, and in line with comments from another reviewer, we added a new Section in the Discussion part entitled "4.3 Comparison to previous chamber studies investigating soot ageing". This section places our findings better in the context of the existing literature and provides a more detailed comparison with simulation chamber studies employing similar experimental approaches. To this end, we identified two particularly relevant chamber studies that investigated the effects of organic coatings on soot over extended ageing periods: Schnaiter et al. (2005) and Metcalf et al. (2013). However, to the best of our knowledge, no comparable chamber studies examining the formation and ageing of sulfuric acid coatings on soot have been reported. We therefore added the following text to compare our results with these previous studies and to better contextualize our findings within the existing literature.

"Only a limited number of chamber studies have examined both the formation and subsequent ageing of coatings on soot particles. Although their soot generation methods, coating protocols, and the extent of soot and coating characterization differ from those employed in the present study, two studies, the ones by Schnaiter et al. (2005) and by Metcalf et al. (2013), investigated the formation and ageing of organic coatings on polydisperse soot and are discussed below. To the best of our knowledge, no comparable chamber studies examining the formation and ageing of sulfuric acid coatings on soot have been reported.

Schnaiter et al. (2005) investigated the impact of α-pinene SOA coatings on diesel soot using experiments conducted in the 84 m³ AIDA chamber. In their study, the soot underwent five successive coating steps through the in situ ozonolysis of α-pinene before being aged for an additional 10 h in the chamber, resulting in total experiment durations of up to 25 h. They reported absorption enhancement factors of 1.8-2.1 at wavelengths of 450, 550, and 700 nm for collapsed soot aggregates. These values are at the upper end of the range observed in our SOA experiments, particularly for the SOA-S_{low} experiment, which most closely resembles the conditions in terms of the soot-to-α-pinene precursor ratio as investigated at the AIDA chamber. In contrast to our observations, Schnaiter et al. (2005) found that E_{abs} remained nearly constant during the ageing period. This discrepancy is likely explained by fundamental differences in the experimental protocols, particularly the ageing conditions. In our experiments, the aerosol was aged under slight continuous dilution and in the absence of additional α-pinene precursor. Under these conditions, the more volatile SOA components gradually evaporated, leading to a reduction in the coating mass. In the AIDA experiments, however, the soot experienced continuous SOA growth throughout the ageing period. This was achieved by first introducing ozone into the chamber and subsequently supplying a continuous flow of air saturated with α-pinene vapor, thereby sustaining ongoing SOA condensation onto the soot particles. These conditions maintained the particle coating and likely explain why E_{abs} remained nearly constant during the 10 h ageing period. Schnaiter et al. (2005) further showed that the Mie core–shell model cannot accurately reproduce the optical behaviour of thinly coated soot particles, although its agreement with observations improves with increasing coating-to-soot ratios.

Metcalf et al. (2013) performed chamber experiments with fullerene soot, a surrogate for fractal carbonaceous particles, in the 28 m³ Caltech Teflon chamber. The soot was exposed to several types of organic coatings, including SOA formed from the high-NOx photooxidation of naphthalene and from the high- and low-NOx photooxidation of α-pinene. The experiments consisted of an initial irradiation period lasting several hours, during which SOA formed and the chamber temperature increased, followed by several hours of dilution accompanied by a gradual decrease in temperature. Consistent with our observations, Metcalf et al. (2013) reported absorption enhancement factors of 1.25–1.35 at 405 nm and 1.15–1.30 at 781 nm for the α-pinene SOA experiments. They also observed that the SOA coating partially evaporated during the course of the experiments, particularly during the dilution phase. The extent of evaporation depended on the chamber temperature, both during the irradiation period, which influenced the formation of more or less semi-volatile SOA components, and during dilution, where the magnitude of the temperature decrease controlled the degree of evaporation. In our experiments, temperature variations during dilution were limited to 1–2 K, suggesting that changes in coating mass were primarily driven by dilution rather than by temperature-induced

volatility changes. Unlike our observations,, Metcalf et al. (2013) demonstrated that E_{abs} can be accurately predicted when the coating mass or thickness is known and the particle can be represented by a simple core–shell geometry. However, this approach relies on fullerene spherical particles instead of fractal soot, thereby limiting the representativeness of the results for atmospheric conditions.

Overall, these previous studies have provided evidence that coating-induced absorption enhancement occurs and provide key complementary observations to our study. However, they have important limitations because they provide only limited information on the simultaneous evolution of particle coating and morphology throughout the different stages of the coating process and, in the case of Metcalf et al. (2013), rely on simplified particle morphologies. Consequently, it remains difficult to directly compare these studies or derive predictive relationships applicable to atmospheric soot. The present work addresses this gap by systematically characterizing both coating evolution and soot restructuring throughout the ageing process, so to move towards a mechanistic understanding of E_{abs}. Although such measurements are experimentally challenging, this approach provides new insights into the underlying processes and demonstrates that this combined characterization is essential for developing a predictive understanding of coating-induced absorption enhancement. "

Reviewer 1

This manuscript by Heuser et al. "The synergy of morphology and mixing state on the absorption of coated black carbon soot" presents interesting and relevant new results on how coating and particle morphology affect black carbon absorption and further climatic implications. The data are highly valuable and original, the study is well written and the topic is important. I find the manuscript suitable for publication after the authors address a few points that would help clarify the interpretation of the results. My main comments concern the interpretation of the mobility-diameter changes and low Eabs values, the role of particle-to-particle heterogeneity, and the uncertainty characterization of the extinction-minus-scattering absorption method.

Major comment 1: Interpretation of restructuring, heterogeneity, and low Eabs

The main conclusion of the manuscript is that the BC absorption response cannot be explained by morphology alone but it follows from synergy between morphology and particle-to-particle mixing-state heterogeneity. I think this is a plausible and interesting conclusion, but in the current version the underlying evidence could be tied together more explicitly. At times, it is difficult to identify which results the authors base their conclusions on.

In particular, the SOA experiments appear to show an initial decrease in mobility diameter, which may indicate restructuring or compaction of soot aggregates. However, it is not fully clear to me whether this behavior should be interpreted primarily as aggregate collapse / void filling, or connected to changes in mass loading or evolving coating distribution. It would therefore help if the authors could discuss more explicitly the physical interpretation of the mobility-diameter decrease, including its timing relative to SOA formation and whether the data suggest a step-by-step process (e.g. initial restructuring followed by continued coating growth or aging).

The main conclusion from the manuscript is that the BC response to coating cannot be explained by coating amount alone, but is affected by morphological organization of both the soot and the coating-soot system. The controlled laboratory approach we deployed provides evidence that coating formation dynamics, coating type (non-volatile/volatile), and further ageing can affect the composition, morphology, and size of the soot and the coating-soot system, ultimately ruling the E_{abs} magnitude and its modifications.

For the SOA experiments, the mobility diameter decrease occurred quite rapidly after coating formation for experiments SOA-S_{ref} and SOA_{high}-S_{high}, whereas it increased for the SOA-S_{low} experiment. In combination with the mobility exponent D_{fm} and the TEM images, the measurements suggested that the coating formation induced a transition towards more spherical and compacted particle morphologies. For SOA-S_{low}, the CMD evolution combined with D_{fm} and TEM, additionally support the interpretation that the reduction in particle size associated with restructuring was masked by the larger amount of condensed coating material, resulting in a net increase in CMD. During subsequent ageing the CMD then remained nearly constant for SOA-S_{ref} and SOA_{high}-S_{high}, indicating limited additional restructuring, while decreased for the SOA-S_{low} experiment, consistent with continued restructuring during the first hours after coating formation for the more heavily coated soot.

In order to better clarify the interpretation of the CMD decrease, part of Sect. 3.3 was revised as follows:

"The D_{fm} rises to 2.06 ± 0.04 (SOA_{high}- S_{high}), 2.24 ± 0.05 (SOA-S_{ref}), and 2.61 ± 0.04 (SOA-S_{low}), indicating that the coating formation induced a transition towards more spherical and compacted particle morphologies. This restructuring is also reflected by the marked and simultaneous decrease in the CMD for SOA-S_{ref} and SOA_{high}-S_{high}. In contrast, the CMD of SOA-S_{low} increases rapidly after coating formation. The TEM images additionally confirm the coating-induced soot restructuring in all experiments. They show that the majority of soot collapsed to form small and compact aggregates (Fig. 2f-g; more illustrative images in Fig. S5), and, when visible from microscopy, the coating is identified to encapsulate soot particles. The net increase in CMD observed for SOA-S_{low} is then attributed to the larger amount of condensed coating material for this experiment, which masks the reduction in particle size associated with soot restructuring (Fig. 2b, Fig. S3). After this initial evolution, the soot CMD for SOA-S_{ref} and SOA_{high}- S_{high} stabilizes as a consequence of minimal further SOA deposition (α-pinene precursor consumed) and negligible coagulation (as discussed in Text S2, Fig. S10). In contrast, for SOA-S_{low} the CMD of the soot peak is found to continuously decrease, interpreted as ongoing restructuration."

Related to this, the manuscript emphasizes the importance of particle-to-particle heterogeneity in mixing state, but this aspect remains somewhat qualitative. If possible, I encourage the authors to add at least one simple quantitative characterization of heterogeneity from the available single-particle measurements (for example, fraction of heavily coated particles, or evolution of the rBC mass distribution) to help the reader to connect the discussed heterogeneity to the observed absorption response.

We agree with the reviewer on the need for a more quantitative evaluation of particle-to-particle heterogeneity. However, two technical limitations prevented such an analysis.

First, the availability of TEM images varied substantially across experiments. Depending on the particle concentration, the number of images acquired ranged from only a few tens to more than 300. This uneven distribution of images among the different experimental conditions and phases precluded a statistically robust clustering analysis and meaningful comparison across the various experimental stages. Instead, we performed a qualitative visual assessment of the TEM images. This analysis consistently indicated that the coating was heterogeneous both at the individual particle level (i.e., the spatial distribution of the coating on the surface of each particle) and at the particle population level (i.e., the particle-to-particle mixing state). Examples images are provided in the manuscript and Supporting Information, to provide illustration of detected soot from TEM measurements.
The paragraph describing TEM measurements and analyses in Sect. 2.2 was revised to clarify this aspect:
"*Morphological information of the soot particles were also retrieved using transmission electron microscopy (TEM). Soot particles samples were collected over different moments of the experiments using three TEM grids (200-mesh copper grid with a Formvar/ carbon film, Agar Scientific) glued onto the Teflon-pore filters (47mm Nuclepore Whatman, 0.8 µm nominal pore size). Sampling was performed with a custom-made stainless-steel filter holder operated at a flow rate of 2-4 L min-1. The total sampling time was adjusted to ensure similar area loadings between different samples. Nevertheless, the total number of TEM images acquired varied considerably among experiments and particularly for the aged conditions, ranging from a few tens to more than 300 images per sampled grid set. The TEM images were acquired with a JEOL® 100CXII and a JEOL® JEM2010 microscopes and processed using the same protocol as described in Heuser et al. (2025). A qualitative visual assessment of the TEM images was performed to characterize the aged sampled soot. However, the variation in the number of images across experiments precluded a statistically robust clustering analysis and, by consequence, a more quantitative comparison across the various experimental stages."*

Second aspect concerns the SP2 measurements. The SP2 alone is capable of providing quantitative (coating thickness) and semi-quantitative (thick vs thin coating) information on the degree of internal mixing. However, it is fundamental to retrieve a resolvable scattering signal to infer at least the temporal occurrence of the peak or the amplitude. During the CESAM experiments, the scattering and position-sensitive detector did not fully resolve the scattering signal, preventing detailed analysis on coating thickness and thin/thick coating fractions. Similar issue was reported in recent AIDA experiments with the same SP2 model (Zanatta et al., 2025).
These limits are reported in Section 2.2.
"Due to instrumental limits of the SP2 scattering detector, a detailed quantification of the coating thickness (leading-edge-only, LEO-fit) or frequency of thinly and thickly coated rBC cores (delay-time) was not possible, as reported for the same SP2 instrument in a recent experiments in the AIDA (Atmospheric Interactions and Dynamics in the Atmosphere) chamber (Zanatta et al., 2025)".

This point becomes especially important in the SOA experiments where E_{abs} appears to decrease strongly and in some cases even fall below 1. Since the coatings are non-absorbing, it is difficult to interpret the result physically. I would therefore ask the authors to discuss more explicitly whether such low values are connected to true population-level microphysical effects, or whether they may partly reflect normalization choices, dilution, wall losses, evolving size distributions, or other measurement uncertainties.

The decrease of E_{abs} below the value of 1 for aged soot in the SOA coating experiment is due to a combination of i/ soot collapse occurring from the early stages of the experiment and 2/ subsequent coating evaporation during ageing time. No correction for dilution or wall losses were applied because our analysis relies on intrinsic parameters or ratios for which these corrections are not required, allowing us to avoid the introduction of a potential bias due to the correction or the propagation of uncertainties.

In order to better clarify the E_{abs} values for ageing soot state during SOA experiment, the text in Sect. 3.3 was revised as follows:

"After 15 hours of ageing in the dark, a significant decrease of $M_{R,bulk,}$ and E_{abs} (0.83-1.04) at rather constant CMD is observed for the SOA-S_{ref} and SOA$_{high}$- S_{high} experiments. The reduction of $M_{R,bulk}$, concurrently with the absence of further SOA production, negligible coagulation, invariance of CMD, and a slight reduction in the soot density, indicates a loss of coating mass on the particles. Such a loss can occur during the continuous dilution of the chamber volume, despite minimal, which can lead to the evaporation of some of the more volatile compounds of the SOA (Grieshop et al., 2007), while low-volatile organics remain on the soot and continue to affect the absorption (Zhang et al., 2023). Such a loss of coating may cause further compaction of the soot (Corbin et al., 2023), as TEM pictures show dominantly collapsed particles (Fig. 2g). Therefore, the pronounced decrease in E_{abs} (down to values lower than 1) after 15 h of ageing is attributed to the combined effects of coating mass loss through evaporation and continued soot restructuring, which further reduced the particle size relative to the pre-coated soot."

Major comment 2: Measurement description and uncertainty of EMS-derived absorption

A second point that would benefit from clarification is the absorption derived from the extinction-minus-scattering (EMS) approach. Because EMS uncertainty can increase substantially at high SSA I think the manuscript should describe more explicitly the instrumental setup, the applicable measurement range, and the uncertainty limits relevant for the experiments discussed here.

The manuscript mentions multiple CAPS and nephelometer instruments, but it is not fully clear which instrument combinations were used to derive absorption in different experiments. I would therefore ask the authors to specify this more clearly and to provide, if possible, a concise uncertainty characterization of the EMS method in the conditions relevant to this study (e.g. table2 in supplement). In particular, it would be useful to indicate over what ranges of absorption coefficient and SSA the EMS method was considered reliable.

I would also appreciate a short clarification of the statement that EMS absorption was "validated against reference filter-based measurements". Why was the filter-based method treated as the

reference and how well did the two methods agree? Over what concentration and SSA ranges was this comparison performed?

More generally, a few additional experimental details could help the interpretation: for example, whether the aerosol was dried (since both sulphuric acid and organic aerosol can take up water over a broad RH range) and what the relevant soot number concentrations / absorption coefficients / SSA values were during the key experimental stages. Some of these details are in the supplementary material (SSA), but they could be linked more explicitly to the discussion of uncertainty.

We thank the reviewer for this comment. The Sect. 2.2 was modified to:

- clarify the experimental setup, in particular for optical measurements and experimental details;
- add uncertainty for each measurement, including a specific discussion for the extinction-minus-scattering absorption coefficient and clarification on the measurement range and its reliability with respect to uncertainties;
- better description was provided on the comparison between EMS and filter-based techniques. Moreover, the concept of "reference" techniques was removed, which mostly refer to the fact that filter-based techniques are widely used, but not reference in the sense of artefact-free.

The revised text for the optical measurements reads as follows:

"*Spectral optical properties were monitored with a nephelometer (TSI Model 3563 Integrating Nephelometer, 1–sec resolution, sampling flow rate 2 L min-1, Anderson et al. (1996)), providing the scattering coefficient (bscat) at 450, 550, and 700 nm, and two Cavity Attenuated Phase Shift monitors (CAPS PMex,, Aerodyne Research, both operated at 1–sec resolution, sampling flow rate 0.85 L min-1, Kebabian et al. (2007); Massoli et al. (2010)) measuring the extinction coefficient (bext) at 450 and 630 nm. Two additional CAPS single scattering albedo monitors (CAPS PMSSA, 1–sec resolution, sampling flow rate 0.85 L min-1, Onasch et al. (2015), Yu et al. (2024)) were additionally deployed in some experiments to provide bext at 450 and 630 nm to compare against CAPS PMex data (the scattering signal from the CAPS PMSSA was not considered in this analysis). The bext by the CAPS PMex and PMSSA agreed within their uncertainty (<6%). The aerosol scattering coefficient measured at angles between 7° and 170° from the nepheleometer were corrected based on Massoli et al. (2009) to obtain the fully integrated 0-180° bscat by using a real refractive index of 1.95 (Bond and Bergstrom, 2006). Total uncertainty on bsca was estimated at about 8%. The scattering Ångström exponent was calculated via the power-law fit of the spectral bscat and used to interpolate the scattering coefficient at 630 nm.*

The aerosol absorption coefficient (b_{abs}) at 450 and 630 nm was calculated from online measurements throughout each experiment as b_{ext} from the CAPS PMex minus b_{scat} from the nephelometer. The uncertainty on the derived b_{abs} was estimated to range within 10-30%. As fully described in Heuser et al. (2025), the b_{abs} derived with extinction-minus-scattering was compared against three filter-based measurements operating between 375 and 850 nm: the Multi Angle Absorption Photometer (MAAP, Petzold et al. (2005)), the Multi-Wavelength Absorbance Analyzer (MWAA, Massabò et al. (2013)), and the polar photometer of the University of Milano (PP_UniMI, Vecchi et al. (2014)). The comparison of the b_{abs} at 450 and 630 nm shows an average difference of 7% and a strong correlation of R^2=0.94 between

extinction-minus-scattering and filter-based techniques, which is within the combined measurement uncertainty. Greater data dispersion was observed for absorption coefficients below 10 Mm^{-1}, a value that can be considered the lower detection limit for b_{abs}. During the experiments, the measured absorption coefficients ranged from 10 to 10^3 Mm^{-1}, indicating that the entire range of b_{abs} values in the present analysis falls within the reliable measurement range."

Major comment 3: More quantitative comparison to previous chamber studies

The manuscript would also benefit from a clearer comparison with previous chamber studies that have also reported larger absorption enhancement, including studies where restructuring of soot aggregates was also observed. Although these are qualitatively discussed in the introduction, the reasons for the differences are not further discussed.

I suggest that the authors add a brief but more quantitative comparison discussing which differences in experimental design, soot generation, coating protocol, definition of Eabs, or measurement approach could possibly explain the discrepancies. This would help readers place the current findings in context and better understand whether the results reflect very different soot/coating regime or methodological differences.

We thank the reviewer for this comment. In order to follow this suggestion, and in line with comments from Dr. Schnaiter, we added a new Section in the Discussion part entitled "4.3 Comparison to previous chamber studies investigating soot ageing". This section places our findings better in the context of the existing literature and provides a more detailed comparison with simulation chamber studies employing similar experimental approaches. To this end, we identified two particularly relevant chamber studies that investigated the effects of organic coatings on soot over extended ageing periods: Schnaiter et al. (2005) and Metcalf et al. (2013). However, to the best of our knowledge, no comparable chamber studies examining the formation and ageing of sulfuric acid coatings on soot have been reported. We therefore added the following text to compare our results with these previous studies and to better contextualize our findings within the existing literature.

"Only a limited number of chamber studies have examined both the formation and subsequent ageing of coatings on soot particles. Although their soot generation methods, coating protocols, and the extent of soot and coating characterization differ from those employed in the present study, two studies, the ones by Schnaiter et al. (2005) and by Metcalf et al. (2013), investigated the formation and ageing of organic coatings on polydisperse soot and are discussed below. To the best of our knowledge, no comparable chamber studies examining the formation and ageing of sulfuric acid coatings on soot have been reported.

Schnaiter et al. (2005) investigated the impact of α-pinene SOA coatings on diesel soot using experiments conducted in the 84 m^3 AIDA chamber. In their study, the soot underwent five successive coating steps through the in situ ozonolysis of α-pinene before being aged for an additional 10 h in the chamber, resulting in total experiment durations of up to 25 h. They reported absorption enhancement factors of 1.8-2.1 at wavelengths of 450, 550, and 700 nm for collapsed soot aggregates. These values are at the upper end of the range observed in our SOA experiments, particularly for the SOA-S_{low} experiment, which most closely resembles the conditions in terms of the soot-to-α-pinene precursor ratio as investigated at the AIDA

chamber. In contrast to our observations, Schnaiter et al. (2005) found that E_{abs} remained nearly constant during the ageing period. This discrepancy is likely explained by fundamental differences in the experimental protocols, particularly the ageing conditions. In our experiments, the aerosol was aged under slight continuous dilution and in the absence of additional α-pinene precursor. Under these conditions, the more volatile SOA components gradually evaporated, leading to a reduction in the coating mass. In the AIDA experiments, however, the soot experienced continuous SOA growth throughout the ageing period. This was achieved by first introducing ozone into the chamber and subsequently supplying a continuous flow of air saturated with α-pinene vapor, thereby sustaining ongoing SOA condensation onto the soot particles. These conditions maintained the particle coating and likely explain why E_{abs} remained nearly constant during the 10 h ageing period. Schnaiter et al. (2005) further showed that the Mie core–shell model cannot accurately reproduce the optical behaviour of thinly coated soot particles, although its agreement with observations improves with increasing coating-to-soot ratios.

Metcalf et al. (2013) performed chamber experiments with fullerene soot, a surrogate for fractal carbonaceous particles, in the 28 m³ Caltech Teflon chamber. The soot was exposed to several types of organic coatings, including SOA formed from the high-NOx photooxidation of naphthalene and from the high- and low-NOx photooxidation of α-pinene. The experiments consisted of an initial irradiation period lasting several hours, during which SOA formed and the chamber temperature increased, followed by several hours of dilution accompanied by a gradual decrease in temperature. Consistent with our observations, Metcalf et al. (2013) reported absorption enhancement factors of 1.25–1.35 at 405 nm and 1.15–1.30 at 781 nm for the α-pinene SOA experiments. They also observed that the SOA coating partially evaporated during the course of the experiments, particularly during the dilution phase. The extent of evaporation depended on the chamber temperature, both during the irradiation period, which influenced the formation of more or less semi-volatile SOA components, and during dilution, where the magnitude of the temperature decrease controlled the degree of evaporation. In our experiments, temperature variations during dilution were limited to 1–2 K, suggesting that changes in coating mass were primarily driven by dilution rather than by temperature-induced volatility changes. Unlike our observations,, Metcalf et al. (2013) demonstrated that E_{abs} can be accurately predicted when the coating mass or thickness is known and the particle can be represented by a simple core–shell geometry. However, this approach relies on fullerene spherical particles instead of fractal soot, thereby limiting the representativeness of the results for atmospheric conditions.

Overall, these previous studies have provided evidence that coating-induced absorption enhancement occurs and provide key complementary observations to our study. However, they have important limitations because they provide only limited information on the simultaneous evolution of particle coating and morphology throughout the different stages of the coating process and, in the case of Metcalf et al. (2013), rely on simplified particle morphologies. Consequently, it remains difficult to directly compare these studies or derive predictive relationships applicable to atmospheric soot. The present work addresses this gap by systematically characterizing both coating evolution and soot restructuring throughout the ageing process, so to move towards a mechanistic understanding of E_{abs}. Although such measurements are experimentally challenging, this approach provides new insights into the

underlying processes and demonstrates that this combined characterization is essential for developing a predictive understanding of coating-induced absorption enhancement. "

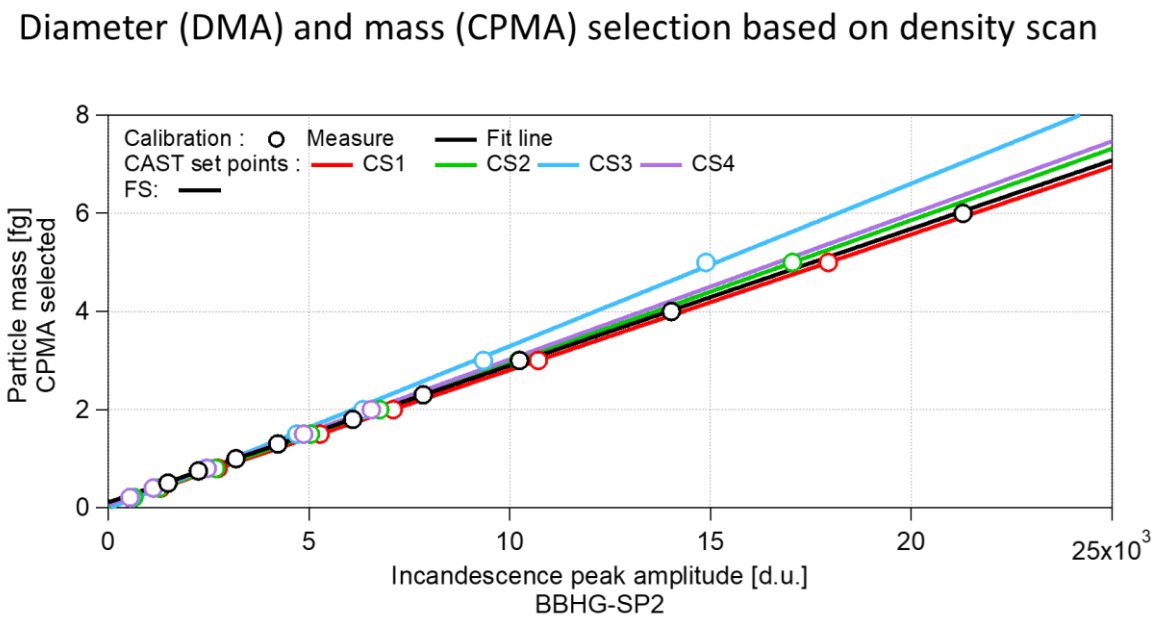
Minor comments

- Please clarify the aims of the control experiments in lines 95–100 more explicitly: what specific effects were expected, and in which measured quantities?

The text in Sect. 2 was revised to more clearly specify the objectives of the control experiments. The new text reads as follows: "*Ten control experiments were performed to evaluate the impact on the soot properties of each compound, process, and protocol used in the coating experiments. These aimed to quantify the effects of soot lifetime and coagulation under dry and humid conditions, of irradiation under dry and humid conditions, and of the presence of O3 and OH-radicals under humid conditions on particles' physico-chemical and optical properties, with particular emphasis on particle density, size, and light absorption. Further control experiments were performed with non-absorbing submicron ammonium sulphate and sulphuric acid aerosols with well-characterized properties to validate the performance and consistency of the optical and physico-chemical measurements, as presented and discussed by Heuser et al. (2025).*"

- Please clarify how the SP2 was calibrated (e.g. calibration material and consistency across calibration approaches). Was size-selected mini-CAST soot or fullerine used? Were the results similar?

The response of the SP2 incandescence detectors was tested with fullerene and with CAST-soot (CS) produced at the different miniCAST setpoints discussed in Heuser et al. (2025) for soot of varying elemental-to-organic carbon fractions (the most elemental CS1 is considered in the present analysis). The figure below shows that a linear relationship between the amplitude of the incandescence peak and the particle mass selected with a CPMA was maintained with all soot materials. It is interesting to note that CS1, the most elemental CAST-soot, showed calibration coefficient very similar to fullerene soot, indicating a similar presence of refractory black carbon material, dominating the overall composition. The sampling of other soot types (CS2, CS3, CS4) resulted in a steeper slope, indicating a weaker incandescence response at parity of total mass, as confirmed by the increasing OC content as shown in Heuser et al. (2025).



Calibration details were added in Section 2.2:
"*The SP2 was calibrated with fullerene-soot material, mass-selected with the CPMA. The calibration curve of fullerene showed comparable coefficients to the most elemental CS1 particles produced at fuel-lean conditions (Heuser et al., 2025).*"

- Lines 193–194: please elaborate briefly on the comparison between EMS and the filter-based reference method.

This paragraph was expanded to include more details on the comparison between EMS and filter-based techniques (see one of the previous comments).

- Was the aerosol dried before optical measurements?

The aerosol was not dried before measurements, including the optical ones. This point was specified in Sect. 2.2 "*The aerosol was measured without drying to preserve realistic RH conditions.*".

- Please comment briefly on the atmospheric relevance of the selected miniCAST operating point.

The miniCAST is chosen as it is a reference instrument providing repeatable and controllable soot generation, allowing for both fuel-lean to fuel-rich conditions, as discussed in (Moore et al., 2014). The miniCAST fuel-lean soot point selected in this study is often considered to be comparable to diesel and aircraft emissions (i.e., kerosene-type) (Ess et al., 2021; Moore et al., 2014; Saffaripour et al., 2017). A sentence was added in Sect. 2.1 in relation to this aspect:
"*For all experiments, the propane, mixing N_2, quench N_2, oxidation, and dilution air flows were kept at 0.03, 0, 2, 0.75, and 5 L min^{-1}, respectively, as corresponding to the first operation point of the miniCAST, producing a fuel-lean combustion with a global equivalence ratio of 0.91. Fuel-lean miniCAST soot is widely regarded as representative of diesel and aircraft (kerosene-type) emissions (Ess et al., 2021; Moore et al., 2014; Saffaripour et al., 2017).* "

- Line 30: the sentence “The range of measured Eabs … cannot be predicted using a fixed Eabs “ is not logical and should be rephrased.

The sentence was modified as “*The range of measured E_{abs} resulting from soot processing cannot be reliably represented using a fixed E_{abs} value or by a core-shell optical model, two approximations commonly used in climate models.* »

- Line 73: “the” → “a”.

As the “the” refers to “CESAM chamber” we kept the sentences unchanged.

- Line 285: typo (“butv”?).

The typo was corrected.

- In Fig. S11 and related figures, comparison between Ntot and rBC soot distributions would be easier if the color scales were harmonized, or if an additional panel were added for soot-mode total number.

The colour scales were set to optimize the data visualization in each panel. Harmonizing the colour scale across all panels would reduce the clarity of the particle size distributions; therefore, we prefer to keep the colour scales different as they are in the original version. Concerning the soot total number concentration, the third panel already includes a comparative representation of the soot-mode total mass, the mass of secondary aerosols, and the total soot concentration. Adding an additional panel showing the total number concentration would further increase the complexity of an already quite information-rich figure, compromising its readability.

- Could you please comment on the apparent drop in enhancement factor when the lamp is switched on in the supplementary (control experiment) figures?

We thank the reviewer for this comment that helped to improve the description of the control experiments. First, it should be noted that, for the control experiments, the definitions of the mass absorption cross-section (MAC) and enhancement factor (EF) are based on the total particle mass (m_{total}) rather than the soot mass (m_{soot}). In experiments where particle mass changes due to physicochemical processing, this definition can lead to variations in the calculated MAC and EF values. However, these variations should be interpreted differently from the EF values reported for the coating experiments, where the changes are primarily driven by an increase in coating mass and quantified by m_{soot}. To clarify this distinction, the following sentence was added to the captions of Figs. S17–S23: *"Note that, unlike Figs. S11–S16, the MAC and EF values are calculated here using only (m_{total})."*

For several control experiments, no significant changes in effective density were observed, resulting in nearly constant MAC and EF values. This is the case for the dry experiments (Figs. S19–S20). In contrast, the irradiation humid experiment (Fig. S22) and the irradiation humid experiment in the presence of ozone (Fig. S23) exhibited more pronounced changes in effective density, leading to a measurable decrease in EF. These changes in effective density indicate that physicochemical transformations occurred during the experiments. Specifically, illumination under humid conditions (Fig. S22) may promote the photooxidation of surface organic carbon, as previously reported by (Han et al., 2012). In the humid experiment with

ozone (Fig. S23), the increase in effective density is likely associated with ozone uptake by the particles, consistent with observations from previous chamber studies (Kamm et al., 1999; Zelenay et al., 2011).

Some more discussion are provided in Sect. 3.1 to clarify the observations for these control experiments. The revised text reads as follows:

"*No significant changes in the soot physico-chemical and optical properties are likewise observed for the control experiments for the various conditions tested. The effective density measurements (Fig. 3a) and the mobility exponents determined from them are found to be rather stable compared to the fresh soot and the pre-coating steps. The only exception was a moderate increase in ρeff(Dm) observed during the irradiated humid experiments, both in absence and in presence of ozone. These changes in effective density indicate that some, despite minor, physicochemical transformations occurred during the experiments. Specifically, illumination under humid conditions may promote the photooxidation of surface organic carbon, as previously reported by Han et al. (2012). In the humid experiment with ozone, the increase in effective density is likely associated with ozone uptake by the particles, consistent with previous chamber studies (Kamm et al., 1999; Zelenay et al., 2011). The long-term ageing control experiments, Soot coagulation dry 1 and 2, also showed some deviations for Dfm, with values around 1.96±0.01. These deviations are however attributed to measurement bias due to the very low aerosol concentrations after 24 hours of coagulation and dilution. The MAC values remain for all conditions inside one standard deviation of the values of fresh soot as reported in Heuser et al. (2025) (Fig. S1). Timelines and results of the ageing control experiments are shown in the supplementary information (Fig. S17-S23).*"

Reviewer 2

This manuscript presents a systematic chamber study of absorption enhancement by coated soot, with particular emphasis on the combined effects of coating amount, morphological restructuring, and particle-to-particle mixing-state heterogeneity. The experimental design is comprehensive, including H2SO4 and α-pinene SOA coating experiments, multiple control experiments, and extended ageing periods. The combined use of SMPS, CPMA, SP2, TEM, nephelometer, and CAPS measurements provides a valuable multi-constraint dataset linking soot size, effective density, refractory BC mass, morphology, and optical properties. Overall, the manuscript is scientifically valuable and suitable for publication after minor revision. The following comments mainly request clearer process attribution, more quantitative use of the existing measurements, and a fuller description of uncertainty. They do not require additional experiments, and most can be addressed through clarification, reanalysis of existing data, or more cautious wording.

1. The timing of soot collapse should be clarified. Figure 2b suggests that SOA-Sref and SOAhigh-Shigh already undergo collapse during the initial coating stage, while Eabs increases, which appears inconsistent with the later interpretation that collapse reduces absorption enhancement. The authors could explain whether different mechanisms dominate the optical response during the initial coating stage and subsequent ageing. In addition, the continuous decrease in MR_bulk during processing should be clarified; is SOA evaporation responsible?

As correctly noted by the reviewer and illustrated in Fig. 2, the soot collapse and restructuration for SOA-S_{ref} and SOA_{high}-S_{high} experiments occurred rapidly during the initial coating stage. The E_{abs} increase observed in this phase was due to the presence of coating material on the soot particles. The CMD remained then constant during subsequent ageing, while the $M_{R,bulk}$ showed a significant reduction after 15 hours of ageing in the chamber. The reduction of $M_{R,bulk}$, concurrently with the absence of further SOA production, negligible coagulation, invariance of CMD, and a slight reduction in the soot density, indicated a loss of coating mass on the particles due to the evaporation of some of the more volatile compounds of the SOA. At this later stage, the E_{abs} decrease can be attributed to the combined removal of the coating by evaporation, combined to the fact that the soot size was reduced due to compacting in the initial experiment phase and continuing during the experiments.
The different mechanisms affecting E_{abs} are discussed in Sect. 4.1. Nonetheless, to clarify the processes affecting the soot absorption evolution for SOA experiment, we added the following text at the end of Sect. 3.3
"Therefore, the pronounced decrease in E_{abs} (down to values lower than 1) after 15 h of ageing is attributed to the combined effects of coating mass loss through evaporation and continued soot restructuring, which further reduced the particle size relative to the pre-coated soot."

2. Figs in the manuscript show SOA and H2SO4 may have different impacts on BC collapse. This is interesting because previous studies typically using one type of coating materials, e.g., Peng et al. (2016, PNAS) and Zhang et al. (2020, PNAS) ??. Could the author confirm this and make a short discussion?

As correctly noted by the reviewer we observe a different impact on BC collapse due to the different dynamic of coating formation, as observed between H_2SO_4 and SOA experiments, and their further ageing. A more rapid collapse is identified for the SOA case, for which coating establishment is quicker, and a more gradual coating formation and particle restructuration is observed for the experiments with sulfuric acid. Nonetheless, as discussed in Sect. 4.1 (*text reported below*) this difference is not only compound-related but linked to the dynamic of coating formation:
"*The shape, i.e. restructuration of the particles, and the coating distribution are found to vary with experiments and ageing time. The slower production of H_2SO_4 impacts the shape and thus the morphology of the soot less in the initial coating formation stages than the much faster SOA formation, which rapidly reduces mobility diameter and compacts soot particles. Only after long term ageing, the majority of particles for H_2SO_4 experiments is restructured, accompanied by a reduced E_{abs} for the aged and collapsed H_2SO_4-coated soot. Based on previous flow reactor-type measurements observing soot compaction during rapid H_2SO_4 condensation (Khalizov et al., 2009b; Zhang et al., 2008), it can be assumed that the difference is not purely compound-related but likely a result of the dynamics of the coating formation.*"

3. The morphology and mixing-state evidence should be presented more quantitatively. Figures S4 and S5 are selected images, yet the main text refers to the majority of particles and to mixing-state control of Eabs. Please report the number of particles

analysed and, if available, morphology-class fractions; otherwise, soften the causal wording. Relevant studies, such as Huang et al. (2024, One Earth), could also be cited to strengthen the interpretation and improve the credibility of the morphology–optics linkage.

We agree with the reviewer on the need for a more quantitative evaluation of particle-to-particle heterogeneity. However, two technical limitations prevented such an analysis.

First, the availability of TEM images varied substantially across experiments. Depending on the particle concentration, the number of images acquired ranged from only a few tens to more than 300. This uneven distribution of images among the different experimental conditions and phases precluded a statistically robust clustering analysis and meaningful comparison across the various experimental stages. Instead, we performed a qualitative visual assessment of the TEM images. This analysis consistently indicated that the coating was heterogeneous both at the individual particle level (i.e., the spatial distribution of the coating on the surface of each particle) and at the particle population level (i.e., the particle-to-particle mixing state). Examples images are provided in the manuscript and Supporting Information, to provide illustration of detected soot from TEM measurements.

The paragraph describing TEM measurements and analyses in Sect. 2.2 was revised to clarify this aspect:

"*Morphological information of the soot particles were also retrieved using transmission electron microscopy (TEM). Soot particles samples were collected over different moments of the experiments using three TEM grids (200-mesh copper grid with a Formvar/ carbon film, Agar Scientific) glued onto the Teflon-pore filters (47mm Nuclepore Whatman, 0.8 µm nominal pore size). Sampling was performed with a custom-made stainless-steel filter holder operated at a flow rate of 2-4 L min-1. The total sampling time was adjusted to ensure similar area loadings between different samples. Nevertheless, the total number of TEM images acquired varied considerably among experiments and particularly for the aged conditions, ranging from a few tens to more than 300 images per sampled grid set. The TEM images were acquired with a JEOL® 100CXII and a JEOL® JEM2010 microscopes and processed using the same protocol as described in Heuser et al. (2025). A qualitative visual assessment of the TEM images was performed to characterize the aged sampled soot. However, the variation in the number of images across experiments precluded a statistically robust clustering analysis and, by consequence, a more quantitative comparison across the various experimental stages."*

In order to soften the causal wording, we additionally changed the following text in Sect. 3.2:

Original version

"*The decrease in E_{abs} is* ***attributed*** *to a size reduction of the absorbing soot core, as* ***evidenced in TEM images analysis*** *(Fig. 1g, Fig. S4), showing significant particles' restructuration.* »

Revised version

"*The decrease in E_{abs} is* ***likely associated*** *to a size reduction of the absorbing soot core,* ***as observed in TEM images*** *(Fig. 1g, Fig. S4), showing significant particles' restructuration.* »

We also thank the reviewer for pointing at the very interesting paper by Huang et al. (2024), which is now cited at two different points in the Introduction providing further support to the contextualisation of our study.

"*Field observations led to diverse results, with some studies reporting E_{abs} up to 3 (Liu et al., 2015, 2017; Zhang et al., 2018), while others calculate small E_{abs} despite large coating masses (Cappa et al., 2012b, 2019; Chan et al., 2010; Healy et al., 2015;* ***Huang et al., 2024****). The spread of observations suggests that the mechanisms leading to absorption enhancement do not depend solely on the amount of coating mass deposited on the BC. Key aspects evidenced to affect E_{abs} magnitude and variability include (i) the single particle properties and mixing state including the morphological organization of the soot and its coating (Chen et al., 2024;* ***Huang et al., 2024****; Liu et al., 2017; Sedlacek III et al., 2012), i.e. the way secondary species distributes over the fractal soot including void filling or encapsulating soot with consequent collapse of the aggregate;* "

4. The radiative-effect calculation should include a clearer uncertainty assessment. Table 3 reports relative RF differences without showing how uncertainty in measured SSA, extinction-minus-scattering absorption, assumed refractive indices, and retained parameters affects the range. It is suggested to propagate the dominant remaining uncertainties or provide a sensitivity interval.

Calculations have been made to evaluate the uncertainty on the ΔRF. To this aim the SSA values are considered as the primary sources of uncertainty, while the Rs and β are considered as fixed parameters. The resulting uncertainty on ΔRF is estimated at 20%, taking into account statistical uncertainties on SSA measurements and the sensitivity of the modelled SSA to the assumed complex refractive index. The following text was inserted:

"*The uncertainty on ΔRF was estimated at ~20% based on the error propagation formula and considering only the uncertainty on the measured SSA (12-25%, Heuser et al. (2025)) and the sensitivity of the modelled SSA on the assumed complex refractive (15% on average based on the comparison of the SSA values calculated with 1.95-0.79i and 1.46-0.3i).*"

Technical comments

- Around line 248, the Mie-derived Eabs is described as the ratio of uncoated to coated babs, whereas Sect. 2.3.1 defines Eabs as coated relative to prior/uncoated absorption.

The text was modified in order to clarify the definition and calculations. In Sect 2.3.1 the text was changed as: "*The absorption enhancement factor E_{abs} was calculated at 450 and 630 nm and 3–min resolution as the ratio of the MAC after the coating generation to its value prior to coating generation (E_{abs}= MAC_{coated}/MAC_{prior}).*" In Sect 2.3.2 the text was changed as: *"The Eabs($M_{R,bulk}$) was calculated as the ratio of the babs of the coated and uncoated soot for the given $M_{R,bulk}$*".

- Please give the explicit conversion equation between SP2-derived mrBC and msoot and clarify whether the reported factor of 1.2–1.8 represents mtotal/mrBC or the inverse.

The reported factor (R) represents $R=m_{total}/m_{rBC}$. Hence, the conversion equation is $m_{total}=m_{rBC}*R$ and R value indicate that the total mass of soot is slightly larger than the refractory mass, in line with EC/TC retrieval. In order to clarify this point, the text was modified as follows in Sect. 2.2:

"*The ratio of the m_{total} to the SP2 fullerene-calibrated mrBC ($R=m_{total}/m_{rBC}$) was determined for each experiment prior to coating formation and ranged from 1.2 to 1.8. This ratio was used*

to convert m_{rBC} into the total soot mass (m_{soot}) representing the total carbonaceous mass of soot, including both elemental and organic carbon fractions, according to $m_{soot}=R \cdot m_{rBC}$. This approach enabled continuous tracking of the soot mass concentration throughout the experiments."

- Around line 247, "the the size distribution" should be corrected to "the size distribution."

The typo was corrected.

- Around line 285, "due to condensation; butv also coagulation" should be corrected to "due to condensation, but also coagulation."

The typo was corrected.

- Around line 292, "TEM images depicts" should be corrected to "TEM images depict."

The typo was corrected.

- Please use either "ageing" or "aging" consistently throughout the manuscript.

The text was revised and the formulation "ageing" is now used consistently throughout the manuscript.

- The main text refers to Figs. S17–S25 for the control experiments, whereas the Supplement contains figures only through Fig. S23. Please correct the figure range.

The figure range was corrected.

References

Baldo, C., Formenti, P., Nowak, S., Chevaillier, S., Cazaunau, M., Pangui, E., Di Biagio, C., Doussin, J.-F., Ignatyev, K., Dagsson-Waldhauserova, P., Arnalds, O., MacKenzie, A. R., and Shi, Z.: Distinct chemical and mineralogical composition of Icelandic dust compared to northern African and Asian dust, Atmospheric Chemistry and Physics, 20, 13521–13539, https://doi.org/https://doi.org/10.5194/acp-20-13521-2020, 2020.

Baldo, C., Formenti, P., Di Biagio, C., Lu, G., Song, C., Cazaunau, M., Pangui, E., Doussin, J.-F., Dagsson-Waldhauserova, P., Arnalds, O., Beddows, D., MacKenzie, A. R., and Shi, Z.: Complex refractive index and single scattering albedo of Icelandic dust in the shortwave spectrum, EGUsphere, 2023, 1–42, https://doi.org/10.5194/egusphere-2023-276, 2023.

Battaglia, F., Formenti, P., Giorio, C., Cazaunau, M., Pangui, E., Bergé, A., Gratien, A., Pereira, D. L., Bertin, T., de Brito, J. F., Romanias, M. N., Michoud, V., Baldo, C., Chevaillier, S., Noyalet, G., Decorse, P., Picquet-Varrault, B., and Doussin, J.-F.: Formation and composition of organic aerosols from the uptake of glyoxal on natural mineral dust aerosols: a laboratory study, Atmospheric Chemistry and Physics, 25, 12409–12431, https://doi.org/10.5194/acp-25-12409-2025, 2025.

Caponi, L., Formenti, P., Massabó, D., Di Biagio, C., Cazaunau, M., Pangui, E., Chevaillier, S., Landrot, G., Andreae, M. O., Kandler, K., Piketh, S., Saeed, T., Seibert, D., Williams, E., Balkanski, Y., Prati, P., and Doussin, J.-F.: Spectral- and size-resolved mass absorption efficiency of mineral dust aerosols in the shortwave spectrum: a simulation chamber study, Atmospheric Chemistry and Physics, 17, 7175–7191, https://doi.org/https://doi.org/10.5194/acp-17-7175-2017, 2017.

De Haan, D. O., Hawkins, L. N., Wickremasinghe, P. D., Andretta, A. D., Dignum, J. R., De Haan, A. C., Welsh, H. G., Pennington, E. A., Cui, T., Surratt, J. D., Cazaunau, M., Pangui, E., and Doussin, J.-F.: Brown Carbon from Photo-Oxidation of Glyoxal and SO2 in Aqueous Aerosol, ACS Earth Space Chem., 7, 1131–1140, https://doi.org/10.1021/acsearthspacechem.3c00035, 2023.

Di Biagio, C., Formenti, P., Balkanski, Y., Caponi, L., Cazaunau, M., Pangui, E., Journet, E., Nowak, S., Caquineau, S., Andreae, M. O., Kandler, K., Saeed, T., Piketh, S., Seibert, D., Williams, E., and Doussin, J.-F.: Global scale variability of the mineral dust long-wave refractive index: a new dataset of in situ measurements for climate modeling and remote sensing, Atmospheric Chemistry and Physics, 17, 1901–1929, https://doi.org/https://doi.org/10.5194/acp-17-1901-2017, 2017.

Di Biagio, C., Formenti, P., Balkanski, Y., Caponi, L., Cazaunau, M., Pangui, E., Journet, E., Nowak, S., Andreae, M. O., Kandler, K., Saeed, T., Piketh, S., Seibert, D., Williams, E., and Doussin, J.-F.: Complex refractive indices and single-scattering albedo of global dust aerosols in the shortwave spectrum and relationship to size and iron content, Atmospheric Chemistry and Physics, 19, 15503–15531, https://doi.org/https://doi.org/10.5194/acp-19-15503-2019, 2019.

Di Biagio, C., Doussin, J.-F., Cazaunau, M., Pangui, E., Cuesta, J., Sellitto, P., Ródenas, M., and Formenti, P.: Infrared optical signature reveals the source–dependency and along–transport evolution of dust mineralogy as shown by laboratory study, Scientific Reports, 13, 13252, https://doi.org/10.1038/s41598-023-39336-7, 2023.

Doussin, J.-F., Fuchs, H., Kiendler-Scharr, A., Seakins, P., and Wenger, J. (Eds.): A Practical Guide to Atmospheric Simulation Chambers, Springer International Publishing, Cham, https://doi.org/10.1007/978-3-031-22277-1, 2023.

Ess, M. N., Bertò, M., Irwin, M., Modini, R. L., Gysel-Beer, M., and Vasilatou, K.: Optical and morphological properties of soot particles generated by the miniCAST 5201 BC generator, Aerosol Science and Technology, 55, 828–847, https://doi.org/10.1080/02786826.2021.1901847, 2021.

Han, C., Liu, Y., Ma, J., and He, H.: Key role of organic carbon in the sunlight-enhanced atmospheric aging of soot by O2, Proceedings of the National Academy of Sciences, 109, 21250–21255, https://doi.org/10.1073/pnas.1212690110, 2012.

Heuser, J., Di Biagio, C., Yon, J., Cazaunau, M., Bergé, A., Pangui, E., Zanatta, M., Renzi, L., Marinoni, A., Inomata, S., Yu, C., Bernardoni, V., Chevaillier, S., Ferry, D., Laj, P., Maillé, M., Massabò, D., Mazzei, F., Noyalet, G., Tanimoto, H., Temime-Roussel, B., Vecchi, R., Vernocchi, V., Formenti, P., Picquet-Varrault, B., and Doussin, J.-F.: Spectral optical properties of soot: laboratory investigation of propane flame particles and their link to composition, Atmos. Chem. Phys., 25, 6407–6428, https://doi.org/10.5194/acp-25-6407-2025, 2025.

Huang, X.-F., Peng, Y., Wei, J., Peng, J., Lin, X.-Y., Tang, M.-X., Cheng, Y., Men, Z., Fang, T., Zhang, J., He, L.-Y., Cao, L.-M., Liu, C., Zhang, C., Mao, H., Seinfeld, J. H., and Wang, Y.: Microphysical complexity of black carbon particles restricts their warming potential, One Earth, 7, 136–145, https://doi.org/10.1016/j.oneear.2023.12.004, 2024.

Kamm, S., Möhler, O., Naumann, K.-H., Saathoff, H., and Schurath, U.: The heterogeneous reaction of ozone with soot aerosol, Atmospheric Environment, 33, 4651–4661, https://doi.org/10.1016/S1352-2310(99)00235-6, 1999.

Moore, R. H., Ziemba, L. D., Dutcher, D., Beyersdorf, A. J., Chan, K., Crumeyrolle, S., Raymond, T. M., Thornhill, K. L., Winstead, E. L., and Anderson, B. E.: Mapping the Operation of the Miniature Combustion Aerosol Standard (Mini-CAST) Soot Generator, Aerosol Science and Technology, 48, 467–479, https://doi.org/10.1080/02786826.2014.890694, 2014.

Saffaripour, M., Tay, L.-L., Thomson, K. A., Smallwood, G. J., Brem, B. T., Durdina, L., and Johnson, M.: Raman spectroscopy and TEM characterization of solid particulate matter emitted from soot generators and aircraft turbine engines, Aerosol Science and Technology, 51, 518–531, https://doi.org/10.1080/02786826.2016.1274368, 2017.

Wang, J., Doussin, J. F., Perrier, S., Perraudin, E., Katrib, Y., Pangui, E., and Picquet-Varrault, B.: Design of a new multi-phase experimental simulation chamber for atmospheric photosmog, aerosol and cloud chemistry research, Atmos. Meas. Tech., 4, 2465–2494, https://doi.org/10.5194/amt-4-2465-2011, 2011.

Zanatta, M., Bogert, P., Ginot, P., Gong, Y., Hoshyaripour, G. A., Hu, Y., Jiang, F., Laj, P., Li, Y., Linke, C., Möhler, O., Saathoff, H., Schnaiter, M., Umo, N. S., Vogel, F., and Wagner, R.: AIDA Arctic transport experiment – Part 1: Simulation of northward transport and aging effect on fundamental black carbon properties, Aerosol Research, 3, 477–502, https://doi.org/10.5194/ar-3-477-2025, 2025.

Zelenay, V., Monge, M. E., D'Anna, B., George, C., Styler, S. A., Huthwelker, T., and Ammann, M.: Increased steady state uptake of ozone on soot due to UV/Vis radiation, Journal of Geophysical Research: Atmospheres, 116, https://doi.org/10.1029/2010JD015500, 2011.