Answer to referees

Higher absorption enhancement of black carbon in summer shown by two year measurements at the high-altitude mountain site of Pic du Midi Observatory in the French Pyrenees

We thank the three reviewers for evaluating the manuscript and providing us constructive and useful comments. Referees have common concerns which we addressed to the best of our possibilities:

- In order to better highlight the objectives and the main results of the paper (1) we modified the title, (2) clearly listed the scientific questions addressed in the introduction section, (3) modified the conclusion into a section "Summary and implications for climate models", (4) moved Figure 2 from the meteorological section to the Supplement, (5) moved Figure S7 and Figure S9 from the Supplement to the main text in Sections 3.4.1 and 3.4.2, respectively.
- We clarified and completed the section on aerosol optical properties by adding a more complete description of the observed parameters and by reworking the design of Figures 3 and 5.
- We added two elements in the Supplement describing the processing of the SP2 measurements: a Section S1 comparing BC mass concentrations obtained from our software on PYTHON and the PSI SP2 Toolkit running on IGOR and a Section S2 to explain the processing of the rBC core size distribution. A Section S5 about the discrimination of Free Tropospheric/ Planetary Boundary Layer conditions has been added in the Supplement.
- We changed the "BC" nomenclature to "rBC" to follow the recommendations by Petzold et al. (2013) for measurements performed by a SP2.

Please find below reviewer comments in black and our responses in blue. The line numbers in the responses refer to the new version of the paper.

Anonymous Referee #3

Major comments

Tinorua et al. provides a description of an important black carbon data set collected at a high-altitude mountain site in France – these sorts of data sets and measurements are indeed essential to improving climate models. However, the impact of this data set is not compellingly described in the manuscript – there are some sections bogged down in numbers and lists, some paragraphs that making sweeping general statements without the rigorous analysis / discussion that I would expect of an ACP paper, and perhaps most importantly, the implications of the findings on climate models (as described in the article introduction) are limited to a brief 6-line paragraph at the end of the manuscript.

These problems of narrative, hypothesis generation and testing, and rigor can even be seen in the title, which is overly generic – I would like to see the authors more precisely focus and describe their key findings and make sure these findings are well supported in the paper, clear in the title and that the methods are described in more detail. In summary, I think the manuscript has a lot of

potential after major revisions to sharpen the focus. I look forward to this paper being published in ACP after making these adjustments.

There are also some grammatical / style issues throughout that would benefit from an additional proofread, and the time series figures spanning the full 2-year campaign are generally overwhelming. I would recommend that some of these figures be moved to the SI and that focused vignettes of the data be presented in the main manuscript. Additionally, the Supporting Information lacks a lot of detail and description; it is simply a collection of figures, which limits its utility.

Specific Comments

COMMENT #1 : Title: I recommend making the title more descriptive of the findings. It's too generic as written.

REPLY : The title has been changed as follows:

"Higher absorption enhancement of black carbon in summer shown by two year measurements at the high-altitude mountain site of Pic du Midi Observatory in the French Pyrenees"

COMMENT #2 : Line 54: Can you define what is meant by "short periods" – currently subjective

REPLY : The sentence in lines 56-57 has been completed as follows :

"However, most of these measurements were performed in the Planetary Boundary Layer (PBL) and over short periods from a few hours to as long as a season."

COMMENT #3 : Figure 1: Map resolution is poor – can you make this clearer?

REPLY : Figure 1 has been edited. It now has a better resolution. Maps at the regional and local scales have also been added.

COMMENT #4 : Lines 89-91: I would like to see a bit more description of the inlet and its design – what is the total flow rate and diameter? What are the particle losses? If there is a citation that describes this inlet in more detail perhaps it could be provided.

REPLY :

A Whole Air Inlet (WAI) is installed at the PDM as recommended by ACTRIS for measurement sites frequently in clouds and/or freezing conditions. This inlet samples clouds droplets and interstitial aerosol particles up to a diameter of around 10 µm.

Section 2.2.1 in lines 96-102 has been modified as follows:

"All particle-measuring instruments sampled air taken in parallel from a whole air inlet, located 2 m above the building roof. This inlet is used for the long-term observations in mountainous sites and designed to maintain an isokinetic and laminar flow. The main flow rate was fixed at about 460 l min^{-1}. The splitter was fixed at the end of the stainless tube. The hat of the whole air inlet and the stainless tube were both thermo-regulated in order to avoid frost and gradually regulate the temperature of the samples air to the measurement room. The air was heated to around 20°C in order to perform aerosol in-situ measurements at a relative humidity lower than 30 %. The

instrumental room temperature was regulated at around 20°C. The annual cycle of the dew point temperature varied between about -10° and +5°C."

COMMENT #5 : Line 101-102: The process of developing the size distributions shown in Figure S1 in the Supplement are not described – I don't see any descriptive text in the SI, either. Can you provide some more detail in the SI? Feel like there is a big jump here. How are you measuring size distribution? How do you know that all of the ultrafine particles are indeed BC? I think I am just missing a few steps here because I don't have the relevant expertise / background.

REPLY : We agree that we did not provide sufficient elements to understand the issue. We have reformulated the sentence and added a Section S2 in the Supplement to describe the processing of rBC size distribution measured by the SP2 and the estimated missing mass fraction.

The sentence in lines 118-120 was revised as :

"However, the observed size distributions showed that an important fraction (around 12%) of M_{BC} at diameters below 90 nm is not measured by the SP2 (Figure S1 in the Supplement)"

The following text has been added to the Supplement :

"Text S2 : Information about the rBC size distribution processing

The SP2 measures rBC cores from mass equivalent diameters of 90 to 580 nm. Fig. S2 shows the two-year average of the daily rBC cores size distributions. It can be noticed in Fig. S2 that the number size distribution measured by the SP2 did not cover the full size range of rBC at PDM. This is particularly true for the rBC particles below 90 nm, where the major fraction of the M_{rBC} was missed by the SP2. In order to estimate the missing rBC mass fraction undetected by the SP2 (e.g. the mass size distribution under 90 nm and over 580 nm), the daily rBC size distributions were fitted with a sum of three lognormal functions as :

$$\frac{dM}{dln(D_p)} = \sum_{i=0}^{3} \left(\frac{M_i}{\sqrt{2\pi}\ln(\sigma_{g,i})} \exp\left[\frac{-\ln^2(D_p/D_{g,i})}{2\ln^2(\sigma_{g,i})} \right] \right)$$

with M_i, $D_{g,i}$ and $\sigma_{g,i}$ representing the rBC mass concentration, the geometric mean diameter and the geometric standard deviation of the mode i, respectively. The same function with two modes has been used to fit the number size distribution.

The fitting parameters were constrained in the following ranges : Mode 1 : $50 < D_{g,1} < 100$ nm and $1.2 < \sigma_{g,1} < 3$; Mode 2 : $150 < D_{g,2} < 250$ nm and $1.3 < \sigma_{g,2,} < 2.9$; Mode 3 : $350 < D_{g,3} < 500$ nm and $1 < \sigma_{g,3} < 3$. "

COMMENT #6 : Section 2.2.3: I see now that you are describing the SMPS in the 10-1000 nm size range – would recommend re-ordering the manuscript so you aren't introducing results before you have mentioned the methods. My question on how you know the size distribution below 90 nm is BC still stands.

REPLY : Section 2.2.3 focuses on the measurement of aerosol properties. The SMPS was used to measure aerosol size distribution between 12.6 nm and 532.6 nm size range. We used a SP2 to measure the rBC size distribution and it is described in section 2.2.2.

COMMENT #7 : Lines 124-126: I do not believe these in preparation materials were provided for review – can you provide more details on the calculation of the C value in the response to reviewers?

REPLY : The following sentence has been added in Section 2.2.3, lines 144-146:

"The multiple scattering parameter used to correct the measured attenuation was set to 3.22, according to the value obtained at λ=880 nm by Yus-Díez et al. (2021) at the mountainous site of Montsec d'Ares located less than 200 km from the PDM. "

COMMENT #8 : Section 2.3: I find the description of the equations (1-3) to be confusing to follow. You mention on line 143 the calculation of the aerosol absorption coefficient at 635 nm using another AAE calculation but the justification for this approach seems to be missing. I believe this is linked to the nephelometer wavelength ranges but its difficult to follow the narrative.

REPLY : We agree that there was missing elements about the method used to calculate AAE between 450 and 635 nm. Therefore, we have added the measurement wavelengths of the aethalometer in Section 2.2.3 and we have completed the text in Section 2.3, lines 162-166, as follows:

"The spectral dependence of σ_{ap} was characterized by the Absorption Angstrom Exponent ($AAE_{aer,450-635}$) calculated between 450 and 635 nm as follows :

$$AAE_{aer,450-635} = \frac{-\log\left(\frac{\sigma_{ap,450}}{\sigma_{ap,635}}\right)}{\log\left(\frac{450}{635}\right)}$$

For this calculation, $\sigma_{ap,\,470}$ and $\sigma_{ap,\,660}$ from the aethalometer were adjusted at the wavelengths of 450 and 635 nm measured by the nephelometer using the AAE calculated from the aethalometer between 370-470 nm and 590-660 nm. "

COMMENT #9 : Line 161: You are citing material that to my knowledge is not available for review – please provide details even if it is confidentially to reviewers only. I would like to see some of these details from the in preparation manuscript if possible.

REPLY : Some details has been added in Section 2.3, lines 197-199:

"In addition, we filtered out periods when dust were sampled at PDM for the calculation of MAC_{rBC} since Yuz-Diez et al. (2021) observed significant biases in the multiple scattering correction of the aethalometer AE33 during such events."

COMMENT #10 : Lines 167-171: I find the justification of choice for MAC_bare to be lacking – what are the implications of your choice? Is this still standard practice given that you are citing a paper from 2006? Given the interest in the subject matter I require more convincing that this is the appropriate method.

REPLY : A text in Section 2.3, lines 206-215, has been added to justify the use of Mie theory to calculate $MAC_{bare,rBC}$:

"The calculation of $MAC_{bare,rBC}$ using Mie's theory assume a simplified spherical assumption of rBC morphology. However rBC may exhibit complex morphologies whose optical behavior is

imperfectly predicted by Mie's theory, introducing a bias in the retrieved $MAC_{bare,rBC}$ (Saleh et al., 2016). It might be considered that Mie's theory is suitable for estimating the absorption of highly aged rBC, which exhibit an internally mixed core-shell structure. China et al. (2015) used this method to calculate the E_{abs} of rBC in a high-altitude site of the Azores Islands because the large majority (70%) of these long-range transported particles were found highly compacted. Several studies found that Mie's scattering model captures basic optical properties of BC in biomass burning plumes (Liu et al., (2017), Denjean et al., (2020), Zanatta et al., (2018)) calculated MAC_{rBC} of heavily coated rBC particles from the Arctic region using Mie's theory and found consistent results with direct measurements. "

COMMENT #11 : Figure 2: I'm not sure this figure has a lot of value in the main manuscript given that you have already described the findings in the text in Section 3.1. I would rather see this figure in the SI and instead have you provide more details on the methods section, which I found to be lacking.

REPLY : We agree with this comment. Figure 2 has been moved to the SI.

COMMENT #12 : Figure 4: The time series are difficult to follow because you are plotting the full two years. I am not sure this is the most effective way to communicate this information. Perhaps you can re-arrange to highlight the seasonality you describe in the text and move the full time series to the SI.

REPLY: We thank the reviewer for the suggestion . We decreased the time resolution of Figure 4 and Figure 6 (now Figure 3 and 5) to one month.

COMMENT #13 : Lines 253-254: If this is a key conclusion of the work then this should be better reflected in abstract / title of the paper. Also given that it was unexpected, can you probe more deeply why you think this was observed and provide more detail that you are confident that it isn't an artifact of your sampling methods or calculations? Would just like to see this better and more specifically justified.

REPLY: We are confident in the sampling method that follows the ACTRIS and WMO-GAW guidelines. The uncertainties on the calculation of optical parameters and rBC concentration have been quantified. We clarified and completed the section on aerosol optical properties by adding a more complete description of the observed parameters and by reworking the design of Figures 3 and 5.

COMMENT #14 : Lines 297-298: I feel like you could be more precise describing how different techniques and correction factors influence the differences. This feels overly generalized as written. Additionally, if you have confidence in your very high MAC value, then what are the implications of this?

REPLY: The paragraph comparing M_{rBC} and MAC_{rBC} has been modified and completed in Section 3.3, l. 359-368:

"The ambient MAC_{rBC} was around 9.2 ± 3.7 m^2 g^{-1} at λ=880 nm (Fig. 5d). Several studies previously reported MAC_{rBC} values between 8.9 to 13.1 for measurements at λ=637 nm in European mountain stations (Pandolfi et al., 2014; Yus-Díez et al., 2022; Zanatta et al., 2016). By using a AAE of unity, these values can be converted to MAC_{rBC} between 6.4 and 9.5 m^2 g^{-1}at λ=880 nm. These studies used different measurement techniques, analysis method and correction factors from ours for estimating MAC_{BC} that makes difficult the comparison of MAC_{rBC} derived from different instruments. Pandolfi et al. (2014) performed a linear regression between $\sigma_{ap,637}$ values measured by a Multi-Angle Absorption Photometer (MAAP) and daily M_{EC} values from off-line filter-based measurements by a SUNSET OCEC Analyzer. Yus-Díez et al. (2022) and Zanatta et al. (2016) retrieved MAC_{BC} with these instruments by calculating the ratio between the two parameters instead of a linear regression. Because of the absence of a standard method for quantifying M_{BC}, the absolute uncertainties on the MAC_{BC} obtained in the literature are very high ranging from ±30 to 70% (Zanatta et al., 2016)."

The implication of our results on BC representation in the climate models has been detailed in the last Section "Summary and implications for climate models" .

COMMENT #15 : Figure 7: You should have a consistent scale for panels (a) and (b).

REPLY : The scales of Figure 7 (now Fig. 6) have been changed.

COMMENT #16 : Line 320: Please avoid sensational language like "remarkable". Do you mean a notable diurnal profile?

REPLY : Yes, it was a linguistic confusion. This was changed in lines 399-400, as follows:

"There was a notable opposite diurnal profile between seasons in E_{abs} with midday showing a minimum around 1.7 in winter, and a maximum around 2.9 in summer."

COMMENT #17 : Section 4.1 and wet scavenging: Conclusions about the significance of precipitation I think would benefit from a more rigorous statistical test vs descriptive observations that you have provided. Additionally, this whole section reads as quite speculative. This is important given that this is one of your main conclusions of note.

REPLY : The Figure (now Fig. 9) showing the influence of precipitation to the rBC core size distribution has been added to the main text for clarity. We also modified the text in section 4.1 to include values and statistics, lines 415-429:

"Figure 8a shows median $\Delta M_{rBC}/\Delta CO$ of 2.1 ng m^{-3} $ppbv^{-1}$ for air masses affected by precipitation, against 0.7 ng m^{-3} $ppbv^{-1}$ without precipitation during the transport of the air masses. The reduction of $\Delta M_{rBC}/\Delta CO$ by a factor of three suggests that a significant removal process of rBC from the precipitation occurred long the transport pathway, apart from vertical transport from the PBL. This result is confirmed by the dependence of $\Delta M_{rBC}/\Delta CO$ to RH in Fig. 8b, where a sudden decline of $\Delta M_{rBC}/\Delta CO$ appeared for highest RH>80%, going from median $\Delta M_{rBC}/\Delta CO$ between 2.0 and 2.4 ng m^{-3} $ppbv^{-1}$ for RH<80% to a median $\Delta M_{rBC}/\Delta CO$ of ~ 0.4 ng m^{-3} $ppbv^{-1}$ above 80% of RH.

Figures 8c-d show in contrast little influence of precipitation and RH on the rBC absorption enhancement, with a constant median E_{abs} value of around ~ 2.1.

To better understand the negligible impact of rBC wet scavenging on MAC_{rBC}, we compared the measured rBC core size distribution of air masses affected or not by precipitation during their transport and under high RH conditions or not (Figure 9). A two-fold lower M_{rBC} in precipitation conditions compared to that without precipitation provides additional evidence for the dominant role of wet scavenging for rBC. The same result appeared by comparing rBC core size distribution under wet or dry conditions. However wet scavenging did not significantly altered the modal diameter of rBC core size distribution. ”

COMMENT #18 : Lines 348-350: Can you clarify why precipitation events were removed? Because of the influence on the ΔBC/ΔCO ratio? Was slightly unclear to me.

REPLY : The objectives of the analysis of $\Delta M_{rBC}/\Delta CO$ ratio in Section 3.4.2 is to investigate the sources of rBC transported in the free troposphere and coming from the boundary layer. In order to avoid potential variation of $\Delta M_{rBC}/\Delta CO$ ratio due to wet deposition rather than a difference in rBC source, precipitation events (Air masses for which precipitation occurred along 72-h back trajectories computed by the HYSPLIT model) were removed in the section 3.4.2.

A text in Section 3.4.2, lines 443-448 has been added to precise the reasons why precipitation events were removed:

“As explained in Section 3.3, $\Delta M_{rBC}/\Delta CO$ ratio depends on the condition of combustion (fuel type, efficiency) and wet deposition by precipitation (Baumgardner et al., 2002; Taylor et al., 2014). We observed in section 3.4.1 a large decrease of $\Delta M_{rBC}/\Delta CO$ when precipitation occurred during the transport of the air masses. In order to investigate the influence of rBC sources on rBC properties, precipitation events (air masses for which precipitation occurred along 72-h back trajectories) were removed in this section.”

COMMENT #19 : Lines 378-379: Can you describe what these additional measurements would be? And the more precise source apportionment?

REPLY : We modified the sentence in lines 477-478 to precise which additional measurements could allow to validate our hypothesis :

“Additional measurements of the aerosol chemical composition and in particular of a tracer of biomass burning in the atmosphere such as levoglucosan should be performed at PDM to confirm this. “

COMMENT #20 : Lines 420-427: To me these are the most important lines in the manuscript – the implications of your findings. I am a little disappointed that this is relegated to one brief paragraph and that your major conclusion re: wet scavenging is not as thoroughly assessed in the paper as is likely warranted given the conclusion. I would like to see the implications section for climate models more rigorously discussed.

REPLY: We added the implications of our results for climate models application in a last section “Summary and implications for climate models” as follows:

“4. Summary and implications for climate models

Continuous two-year measurements of refractive BC (rBC) properties and additional aerosol characteristics have been performed at the high-altitude mountain site Pic du Midi in the French Pyrenees. The classification of the dominant aerosol type using the spectral aerosol optical properties indicates that rBC is the predominant absorption component of aerosols at PDM and controls the variation of SSA throughout the two years. The lower SSA in summer (∼0.93) than in winter (∼0.97) is correlated with a higher rBC number fraction, whereas the influence of BrC and dust was found negligible.

One key parameter to constrain BC absorption and associated radiative forcing in climate models is the refractive index of BC, and in particular the resulting MAC_{BC}. It was not clear if BC at high-altitude mountain sites should have a thicker or thinner coating than in urban or plain sites or even should be coated at all. On the one hand, the longer BC lifetime and the low temperature in the free troposphere (FT) favor thicker coating due to enhanced condensation of low-volatility compounds in colder environment. On the other hand, the low concentrations of particles and gaseous precursors in the FT may limit the coating processes. Our two-year long observations show that the overall net effect is a strong absorption enhancement with a mean E_{abs} value of 2.2 ± 0.9.

The value of 7.5 $m^2\ g^{-1}$ at λ = 550 nm of T. Bond & Bergstrom (2006) is the most common MAC_{BC} used in climate models. The recommendation was based on a compilation of experimental results for freshly generated BC at and near sources obtained earlier than the early-2000s. Nevertheless this value is largely under the MAC_{rBC} found in this study (9.2 $m^2\ g^{-1}$ at λ = 550 nm, which can be converted to 14.7 $m^2\ g^{-1}$ at λ = 550 nm assuming AAE=1). The review by Moteki (2023) has also come to a similar conclusion. The reasons behind this bias should be better understood, in the light of observations such as those provided in the present study.

This study has notably shown the high variability of rBC properties measured in a remote site, where they have undergone long-range transport and aging. Certain causes of the large variability in MAC_{rBC} have been eliminated and highlighted:

- Wet deposition is regarded as the main sink of BC, constraining its lifetime and size distribution, and thus atmospheric its concentration and optical properties. Our direct $\Delta M_{rBC}/\Delta CO$ measurements show the important role of wet deposition as a sink of rBC with around 67 % removed in the atmosphere by precipitation. However, we found a negligible impact of rBC wet removal process on both rBC size distribution and E_{abs}. This result may be due to the combination of large rBC particles reaching PDM ($D_{rBC,core}$ around 180 nm) and high critical supersaturation in precipitating clouds. The BC wet removal process was found to be one of the most misrepresented process in the representation of BC in models (Textor et al., 2006; Yu et al., 2019), leading to overestimated BC tropospheric concentrations and lifetime and in fine, a higher simulated radiative forcing (Samset et al., 2014; Schwarz et al., 2013). Substantial controversial and ambiguous issues in the wet scavenging processes of BC are apparent in current studies (Yang et al., 2019) . Our results suggest that a bulk wet deposition parameterization (which does not account for particle size dependent scavenging) could realistically represent the actual BC wet scavenging at this site.
- rBC core was found to have a mean $D_{rBC,core}$ of 179 nm ± 28 nm, being reasonably independent of the season and day. There was no clear relationship between MAC_{rBC} and $D_{rBC,core}$, which indicates that the variation in rBC core size was not responsible for the MAC_{rBC} variability. Similar observations of rBC core size distribution in the atmosphere provided observational evidence of the stable distribution with a mode centered of around 200 nm approximately one day after emission (Liu et al., 2010; Schwarz et al., 2010; Shiraiwa et al., 2008). This self-similarity could greatly simplify the representation of MAC_{BC} in model simulations since a description of BC mixing state becomes the determinant factor of model performance when estimating BC optical properties and radiative forcing.
- Different time scales of air movements and atmospheric processes affect MAC_{rBC} throughout the year. MAC_{rBC} values were found higher in summer (geometric mean of 10.3 $m^2\ g^{-1}$),

when the influence of regional-scale motions dominates the rBC load, than in winter (geometric mean of 8.3 m^2 g^{-1}), when the influence of local-scale motions outweighs the rBC load. There are three possible explanations for this. (i) The plumes traveling in the FT tend to have a longer lifetime providing sufficient time for rBC aging during transport. In winter this results in a strong diurnal variability of M_{rBC} (E_{abs}) with higher (lower) values in the middle of the day linked to the injection of rBC originating from the planetary boundary layer (PBL). However the aging timescale can not be the only explanatory factor since thermally driven PBL injection did not significantly impact M_{rBC} and E_{abs} in summer and higher values have been observed in summer than in winter for similar FT conditions. (ii) The source of rBC emission was different between the winter and summer seasons. Combining $\Delta M_{rBC}/\Delta CO$ with air mass transport analysis, we observed additional sources from biomass burning in summertime leading to higher M_{rBC} and E_{abs}. (iii) Different aging processes occur between seasons, such as photochemical activity that could explain the observed amplification of light absorption by rBC around noon.

The complexity and diversity of BC mixing states in the real atmosphere cannot be represented in climate models, and therefore these models generally use simplified schemes. A fixed e-folding timescale (1–3 days) is commonly used as the turn-over time for converting fresh BC particles into aged ones (Myhre et al., 2013). In addition, atmospheric models necessarily approximate the full complexity and diversity of BC composition, which can lead to mismatches with observed E_{abs} (Fierce et al., 2020).. The findings presented here suggest that different dynamic processes governing BC light absorption occur during the day and night, and between summer and winter. A parameterization of BC aging explicitly based on aerosol microphysical processes, in which the conversion rate is considered to vary depending on the environmental conditions (e.g., temperature, photochemical activity,...) and some key species (e.g., aerosol, coating precursors,...) may be required to adequately represent the true variability of MAC_{BC}."

References:

Baumgardner, D., Raga, G., Peralta, O., Rosas, I., Castro, T., Kuhlbusch, T., John, A., & Petzold, A. (2002). Diagnosing black carbon trends in large urban areas using carbon monoxide measurements. *Journal of Geophysical Research: Atmospheres*, *107*(D21), ICC 4-1-ICC 4-9. https://doi.org/10.1029/2001JD000626

Bond, T. & Bergstrom. (2006). Light Absorption by Carbonaceous Particles : An Investigative Review. *Aerosol Sci. Tecnol.*, *40*, 27-67. https://doi.org/10.1080/02786820500421521

China, S., Scarnato, B., Owen, R. C., Zhang, B., Ampadu, M. T., Kumar, S., Dzepina, K., Dziobak, M. P., Fialho, P., Perlinger, J. A., Hueber, J., Helmig, D., Mazzoleni, L. R., & Mazzoleni, C. (2015). Morphology and mixing state of aged soot particles at a remote marine free troposphere site : Implications for optical properties. *Geophysical Research Letters*, *42*(4), 1243-1250. https://doi.org/10.1002/2014GL062404

Fierce, L., Onasch, T. B., Cappa, C. D., Mazzoleni, C., China, S., Bhandari, J., Davidovits, P., Fischer, D. A., Helgestad, T., Lambe, A. T., Sedlacek, A. J., Smith, G. D., & Wolff, L.

(2020). Radiative absorption enhancements by black carbon controlled by particle-to-particle heterogeneity in composition. *Proceedings of the National Academy of Sciences*, *117*(10), 5196-5203. https://doi.org/10.1073/pnas.1919723117

Liu, D., Flynn, M., Gysel, M., Targino, A., Crawford, I., Bower, K., Choularton, T., Jurányi, Z., Steinbacher, M., Hüglin, C., Curtius, J., Kampus, M., Petzold, A., Weingartner, E., Baltensperger, U., & Coe, H. (2010). Single particle characterization of black carbon aerosols at a tropospheric alpine site in Switzerland. *Atmospheric Chemistry and Physics*, *10*(15), 7389-7407. https://doi.org/10.5194/acp-10-7389-2010

Liu, D., Whitehead, J., Alfarra, M. R., Reyes-Villegas, E., Spracklen, D. V., Reddington, C. L., Kong, S., Williams, P. I., Ting, Y.-C., Haslett, S., Taylor, J. W., Flynn, M. J., Morgan, W. T., McFiggans, G., Coe, H., & Allan, J. D. (2017). Black-carbon absorption enhancement in the atmosphere determined by particle mixing state. *Nature Geoscience*, *10*(3), Article 3. https://doi.org/10.1038/ngeo2901

Moteki, N. (2023). Climate-relevant properties of black carbon aerosols revealed by in situ measurements : A review. *Progress in Earth and Planetary Science*, *10*(1), 12. https://doi.org/10.1186/s40645-023-00544-4

Myhre, G., Samset, B. H., Schulz, M., Balkanski, Y., Bauer, S., Berntsen, T. K., Bian, H., Bellouin, N., Chin, M., Diehl, T., Easter, R. C., Feichter, J., Ghan, S. J., Hauglustaine, D., Iversen, T., Kinne, S., Kirkevåg, A., Lamarque, J.-F., Lin, G., … Zhou, C. (2013). Radiative forcing of the direct aerosol effect from AeroCom Phase II simulations. *Atmospheric Chemistry and Physics*, *13*(4), 1853-1877. https://doi.org/10.5194/acp-13-1853-2013

Pandolfi, M., Ripoll, A., Querol, X., & Alastuey, A. (2014). Climatology of aerosol optical properties and black carbon mass absorption cross section at a remote high-altitude site in the western Mediterranean Basin. *Atmospheric Chemistry and Physics*, *14*(12), 6443-6460. https://doi.org/10.5194/acp-14-6443-2014

Saleh, R., Adams, P. J., Donahue, N. M., & Robinson, A. L. (2016). The interplay between assumed morphology and the direct radiative effect of light-absorbing organic aerosol. *Geophysical Research Letters*, *43*(16), 8735-8743. https://doi.org/10.1002/2016GL069786

Samset, B. H., Myhre, G., Herber, A., Kondo, Y., Li, S.-M., Moteki, N., Koike, M., Oshima, N., Schwarz, J. P., Balkanski, Y., Bauer, S. E., Bellouin, N., Berntsen, T. K., Bian, H., Chin, M.,

Diehl, T., Easter, R. C., Ghan, S. J., Iversen, T., … Zhang, K. (2014). Modelled black carbon radiative forcing and atmospheric lifetime in AeroCom Phase II constrained by aircraft observations. *Atmospheric Chemistry and Physics*, *14*(22), 12465-12477. https://doi.org/10.5194/acp-14-12465-2014

Schwarz, J. P., Samset, B. H., Perring, A. E., Spackman, J. R., Gao, R. S., Stier, P., Schulz, M., Moore, F. L., Ray, E. A., & Fahey, D. W. (2013). Global-scale seasonally resolved black carbon vertical profiles over the Pacific. *Geophysical Research Letters*, *40*(20), 5542-5547. https://doi.org/10.1002/2013GL057775

Schwarz, J. P., Spackman, J. R., Gao, R. S., Perring, A. E., Cross, E., Onasch, T. B., Ahern, A., Wrobel, W., Davidovits, P., Olfert, J., Dubey, M. K., Mazzoleni, C., & Fahey, D. W. (2010). The Detection Efficiency of the Single Particle Soot Photometer. *Aerosol Science and Technology*, *44*(8), 612-628. https://doi.org/10.1080/02786826.2010.481298

Shiraiwa, M., Kondo, Y., Moteki, N., Takegawa, N., Sahu, L. K., Takami, A., Hatakeyama, S., Yonemura, S., & Blake, D. R. (2008). Radiative impact of mixing state of black carbon aerosol in Asian outflow. *Journal of Geophysical Research: Atmospheres*, *113*(D24). https://doi.org/10.1029/2008JD010546

Taylor, J. W., Allan, J. D., Allen, G., Coe, H., Williams, P. I., Flynn, M. J., Le Breton, M., Muller, J. B. A., Percival, C. J., Oram, D., Forster, G., Lee, J. D., Rickard, A. R., Parrington, M., & Palmer, P. I. (2014). Size-dependent wet removal of black carbon in Canadian biomass burning plumes. *Atmospheric Chemistry and Physics*, *14*(24), 13755-13771. https://doi.org/10.5194/acp-14-13755-2014

Textor, C., Schulz, M., Guibert, S., Kinne, S., Balkanski, Y., Bauer, S., Berntsen, T., Berglen, T., Boucher, O., Chin, M., Dentener, F., Diehl, T., Easter, R., Feichter, H., Fillmore, D., Ghan, S., Ginoux, P., Gong, S., Grini, A., … Tie, X. (2006). Analysis and quantification of the diversities of aerosol life cycles within AeroCom. *Atmospheric Chemistry and Physics*, *6*(7), 1777-1813. https://doi.org/10.5194/acp-6-1777-2006

Yang, Y., Fu, Y., Lin, Q., Jiang, F., Lian, X., Li, L., Wang, Z., Zhang, G., Bi, X., Wang, X., & Sheng, G. (2019). Recent Advances in Quantifying Wet Scavenging Efficiency of Black Carbon Aerosol. *Atmosphere*, *10*(4), Article 4. https://doi.org/10.3390/atmos10040175

Yu, P., Froyd, K. D., Portmann, R. W., Toon, O. B., Freitas, S. R., Bardeen, C. G., Brock, C., Fan, T., Gao, R.-S., Katich, J. M., Kupc, A., Liu, S., Maloney, C., Murphy, D. M., Rosenlof, K. H., Schill, G., Schwarz, J. P., & Williamson, C. (2019). Efficient In-Cloud Removal of Aerosols by Deep Convection. *Geophysical Research Letters*, *46*(2), 1061-1069. https://doi.org/10.1029/2018GL080544

Yus-Díez, J., Bernardoni, V., Močnik, G., Alastuey, A., Ciniglia, D., Ivančič, M., Querol, X., Perez, N., Reche, C., Rigler, M., Vecchi, R., Valentini, S., & Pandolfi, M. (2021). Determination of the multiple-scattering correction factor and its cross-sensitivity to scattering and wavelength dependence for different AE33 Aethalometer filter tapes : A multi-instrumental approach. *Atmospheric Measurement Techniques*, *14*(10), 6335-6355. https://doi.org/10.5194/amt-14-6335-2021

Yus-Díez, J., Via, M., Alastuey, A., Karanasiou, A., Minguillón, M. C., Perez, N., Querol, X., Reche, C., Ivančič, M., Rigler, M., & Pandolfi, M. (2022). Absorption enhancement of black carbon particles in a Mediterranean city and countryside : Effect of particulate matter chemistry, ageing and trend analysis. *Atmospheric Chemistry and Physics*, *22*(13), 8439-8456. https://doi.org/10.5194/acp-22-8439-2022

Zanatta, M., Gysel, M., Bukowiecki, N., Müller, T., Weingartner, E., Areskoug, H., Fiebig, M., Yttri, K. E., Mihalopoulos, N., Kouvarakis, G., Beddows, D., Harrison, R. M., Cavalli, F., Putaud, J. P., Spindler, G., Wiedensohler, A., Alastuey, A., Pandolfi, M., Sellegri, K., … Laj, P. (2016). A European aerosol phenomenology-5 : Climatology of black carbon optical properties at 9 regional background sites across Europe. *Atmospheric Environment*, *145*, 346-364. https://doi.org/10.1016/j.atmosenv.2016.09.035

Zanatta, M., Laj, P., Gysel, M., Baltensperger, U., Vratolis, S., Eleftheriadis, K., Kondo, Y., Dubuisson, P., Winiarek, V., Kazadzis, S., Tunved, P., & Jacobi, H.-W. (2018). Effects of mixing state on optical and radiative properties of black carbon in the European Arctic. *Atmospheric Chemistry and Physics*, *18*(19), 14037-14057. https://doi.org/10.5194/acp-18-14037-2018