

Response to anonymous referee #1: *Characterization of Brown Carbon absorption in different European environments through source contribution analysis*

November 18, 2024

Major comments

Comment 1: This is a very long paper. I would recommend that you go through to confirm that everything is needed in the main text or in the paper in general to make your points. You have an SI, but it's quite short, especially relative to the long main text.

Response 1: We appreciate your feedback regarding the length of the manuscript and the balance between the main text and the Supplementary Information (SI). We acknowledge that some sections could be streamlined without compromising the scientific message. The revised manuscript has been significantly shortened and specific material moved to the SI, now named Supplement which has been significantly extended.

Specifically, Section 3.1 has been shortened by moving the detailed discussion of the statistical evaluation to the Supplement and leaving the main message in the manuscript. Similarly, Section 3.2 has also been significantly shortened by moving the discussion of the optimization analysis of Cases 1 to 4 to the Supplement and keeping in the manuscript the analysis of Case 4 and Case 5, which are the relevant cases of the study. Finally, we have merged Sections 3.3.1 and 3.3.2 in a single and shorter section highlighting the main findings and implications of the derived refractive indexes. Some figures are now in the Supplement and we have introduced a simple and more descriptive Figure 5 that easily synthesizes the impact of the constrained refractive index in calculating absorption in the model.

We believe that the revised manuscript is more easy to read and the main findings better highlighted.

Comment 2: Figure 3 could probably be better communicated in words or the figure could go in the SI.

Response 2: We acknowledge the suggestion of the reviewer. The purpose of Figure 3 was to schematize the flow diagram of the conversion process from organic aerosol (OA) mass to OA absorption, which involves multiple steps. We believe that presenting this process visually helps clarify the methodology and enhances understanding for the readers. However, we agree with the reviewer that it is not a fundamental figure in the main manuscript and it has been moved to the Supplement. The figure is cited in the main text as *Figure S1*.

Comment 3: Similarly Table 3 should be in the SI and then referenced.

Response 3: We thank the reviewer for their suggestion. After thorough consideration, we prefer to maintain Table 3 in the main manuscript as it provides the definition of the cases discussed throughout the manuscript. The table is also used in several sections of the document as a reference for the definition of the different absorption categories. We consider that this information is easily communicated through the Table and represents the core of part of the work.

Comment 4: Related to Table and Figure 4, can you help the reader better understand any trends you are attempting to make across sites? You are showing a lot of data, so I wonder if it might be more compelling to focus your main figures on large takeaways and then put these thumbnail plots in the SI. What are the big overarching takeaways that you want the reader to know about modeling emissions/absorption across locations? Across seasons? Etc.?

Response 4: We appreciate the feedback of the reviewer. Following your suggestion, we have moved Table 4 to the Supplement and revised the main text to emphasize the broader trends and key takeaways across sites and seasons, rather than overwhelming the reader with detailed site-specific plots. Now, a detailed analysis can be found in Section S3 in the Supplement leaving the main takeaways in the main manuscript.

In particular, we focus on the most significant findings related to OA mass concentrations and source contributions, specifically highlighting the following:

- Seasonal trends: The dominance of residential emissions during winter months across all stations, driven by heating demand, and the importance of Secondary Organic Aerosols (SOA) during the warmer months, which is particularly evident in summer with peaks at sites like HYY and MSY.
- Spatial variability: Different contributions from emission sources across locations, such as the significant role of shipping emissions at coastal stations like BCN and MAR, versus agricultural emissions at inland stations like IPR and MSY.
- Model performance: The model demonstrates strong capability in capturing seasonal patterns, showing good alignment between modeled and observed concentrations. But faces difficulties with specific cases such as underestimating SOA contributions in summer or overestimating residential emissions in certain Mediterranean regions during winter.

Thank you for encouraging us to clarify and simplify our presentation. We believe this revision makes the key findings more accessible while still providing comprehensive data.

Comment 5: Perhaps instead of so many figures with separate thumbnails for each site, think through some interesting summary figures that help people pull out the main takeaways.

Response 5: Thank you for your valuable suggestion. In response, we have revised the figures to provide more insightful summary visuals that emphasize the main takeaways. Specifically, we have made the following changes:

- We moved the original scatter plot figures, Figure 6 (now Figure S4), Figure 9 (now Figure S5), and Figure 10 (now Figure S6), to the supplementary material.
- Additionally, we created a new Figure 5, which consolidates the data from Cases 4 and 5 across all stations and throughout the year. This figure now contains four scatter plots showing the monthly mean results that summarize the key trends, providing a clearer representation of the results across different stations, environments, and methodological approaches used.

We believe that these changes will make it easier for readers to extract the insights from the study, as the new summary figure reduces redundancy and offers a comprehensive view of the data.

Thank you again for your helpful feedback. We believe that this revision addresses your concerns.

Comment 6: If you are going to keep Figure 8, then it should be better saturated to show differences. It's all blues and greens and not touching the top of your color bar.

Response 6: Thank you for your feedback on Figure 8. In response to your suggestion, we have modified the color scale of the figure to emphasize the differences. The updated figure is now Figure in the revised manuscript.

Comment 7: If Figure 9 is meant to be compared to Figure 6, then perhaps a summary figure that helps the reader see the comparison would be more useful. I am not fully convinced that the current figure is needed. Is your main point that the derived k performs better than an average one? Make that point in one sentence and then cite to a statistic showing that, and if you really desire, put this plot in the SI.

Response 7: Thank you for your comment. We agree that the comparison between Figure 6 and Figure 9 could be more effectively summarized. In response, we have moved the original scatter plots, Figure 6 (now Figure S4) and Figure 9 (now Figure S5), to the supplementary material.

To address your suggestion for a more concise and comparative visualization, we have replaced these figures with a new Figure 5, which provides a panel plot. This figure presents the monthly mean for Case 4 and Case 5 across all stations, differentiating between the two approaches (derived k at each station vs. derive k combining all data). This new summary plot clearly highlights the performance differences between these two approaches. This is now explicitly stated in the results section, supported by relevant statistics.

We believe that this approach better communicates the comparison while streamlining the presentation.

Comment 8: For Figure 10, similar points to above about Figure 9.

Response 8: Thank you for your comment regarding Figure 10. In line with your suggestions for Figure 9, we have moved Figure 10 to the supplementary material, where it is now labeled as Figure S6. This decision was made to streamline the main text and focus on more essential summary figures.

We believe this change enhances the clarity and flow of the manuscript while keeping the detailed data available for readers in the supplementary section.

Comment 9: In the summary and conclusions, you don't need to go over your major methods in a ton of detail again. Make your main points clearly and efficiently.

Response 9: We thank the reviewer for their suggestion to shorten the main text and presenting the main findings clearly and efficiently. Following your suggestion, we have shorten the summary and conclusions section removing the details from the first three paragraphs presented now in a single initial paragraph and highlighting the main findings and relevant conclusions in a synthesized and clearer way in this section.

Minor comments

Comment 1: Wording could also be more concise. For example line 27 (“BrC is originating”) should be BrC originates.

Response 1: Thank you for pointing out this language improvement. We have now corrected this in the manuscript and used a more concise style.

Comment 2: For Figure 1, I would define BrC and BC with the colors that you’re using in the plot instead of labeling all of them in black to the side of each pie chart.

Response 2: In Figure 1, we now define BrC and BC using the colors that correspond to each component in the plot, brown for BrC and black for BC, and remove the labeling in each pie chart. This change improves the clarity and visual consistency of the figure.

Comment 3: In Table 5 and Figure 5, please give the cases descriptive names so that the reader can better follow along.

Response 3: Thank you for your suggestion to provide descriptive names for each case in Table 5 (no Table S4) and Figure 5 (now Figure 4 in the revised manuscript). We understand that more descriptive titles could help readers follow more easily. However, each of the cases in our study represents a unique combination of multiple categories from different sources, making it challenging to assign concise yet informative names to each case.

After considering your feedback, we attempted various naming options, but found that any descriptive names we tried to assign either became overly complex or failed to capture the unique attributes of each case. Consequently, we decided to retain the labels “Case 1” through “Case 5” for simplicity and clarity and refer the reader to Table 3 for their exact definition.

References

Response to anonymous referee #2: *Characterization of Brown Carbon absorption in different European environments through source contribution analysis*

November 18, 2024

Major comments

Comment 1: The manuscript is too long. Please consider avoiding such long papers in the future.

Response 1: We appreciate your feedback regarding the length of the manuscript. We acknowledge that some sections could be streamlined without compromising the scientific message. The revised manuscript has been significantly shortened and specific material, including figures and tables, moved to the Supplement. We believe this revision makes the key findings more accessible while still providing comprehensive information.

Specifically, Section 3.1 has been shortened by moving the detailed discussion of the statistical evaluation to the Supplement and leaving the main message in the manuscript. Similarly, Section 3.2 has also been significantly shortened by moving the discussion of the optimization analysis of Cases 1 to 4 to the Supplement and keeping in the manuscript the analysis of Case 4 and Case 5, which are the relevant cases of the study. Finally, we have merged Sections 3.3.1 and 3.3.2 in a single and shorter section highlighting the main findings and implications of the derived refractive indexes. Some figures are now in the Supplement and we have introduced a simple and more descriptive Figure 5 that easily synthesizes the impact of the constrained refractive index in calculating absorption in the model.

Comment 2: For OA mass concentration, how do you account for the collection efficiency?

Response 2: The OA mass concentrations measured with Aerosol Chemical Speciation Monitor (ACSM) instruments reported here were published in Chen et al. (2022) in the framework of the COLOSSAL Cost Action (CA16109 Chemical On-Line cOmpoSition and Source Apportionment of fine aerosols) (Freney et al., 2019). As reported in Freney et al. (2019), a composition dependent collection efficiency was calculated for each instrument and applied to each dataset following the recommendations by Middlebrook et al. (2012).

To clarify the reviewer's comment, the sentence at Lines 121-124 was modified as follows:

“Details about ACSM instruments used for OA determination, measurement principle, accuracy and treatment of sources of error as collection efficiency can be found in Ng et al. (2011), Middlebrook et al. (2012), Fröhlich et al. (2013), Freney et al. (2019) and Chen et al. (2022).”

Comment 3: For equations 1 and 2, it should be noted that many current studies show BrC can also absorb at NIR ("Optical Properties of Individual Tar Balls in the Free Troposphere", "Shortwave absorption by wildfire smoke dominated by dark brown carbon", and "Brown carbon absorption in the red and near-infrared spectral region"). Thus, assuming only BC absorbs light

at 880 nm will underestimate the babs of BrC. I suggest adding some relevant discussions. This might explain why your k is so low.

Response 3: In order to consider the Reviewer comment, the following sentence below equations 1 and 2 (from line 152 to line 171 in the revised manuscript) was accordingly modified as follows:

“ where AAE_{BC} is the Absorption Angstrom Exponent (AAE) of BC, which allows calculating $b_{abs,BC(\lambda)}$ (in units of Mm^{-1}) from the measurements of $b_{abs,BC}$ at 880 nm assuming that BrC does not absorb at 880 nm (e.g., Qin et al. (2018)). It should be noted, however, that recent studies have shown the existence of specific dark BrC components in biomass-burning (BB) smoke (tar balls or tar BrC) that can absorb radiation also in the near-infrared (e.g. Chakrabarty et al. (2010); Hoffer et al. (2016, 2017); Chakrabarty et al. (2023); Mathai et al. (2023)). Thus, a contribution to near-IR absorption from possible presence of dark BrC cannot be ruled and would lead to an underestimation of the BrC absorption reported here. However, the dark BrC contribution to absorption at 880 nm is expected to be smaller compared to that of BC. For example, Hoffer et al. (2017) reported that the absorption coefficient at 880 nm of dark BrC produced in a laboratory was 10% of that at 470 nm and, consequently, even lower compared to that at 370 nm. Similarly, Cuesta-Mosquera et al. (2023) estimated a contribution of BrC to absorption at 880 nm of 3% in a rural area in central Europe strongly affected by residential wood burning emissions in winter. Given the complexity of these specific BrC components, the imaginary refractive index (k) of tar balls generated in laboratory experiments vary over a wide range of values depending on the specific type of fuel (wood) burned and the different analytical methods employed. For example, Mathai et al. (2023) reported k values at 550 nm of tar balls measured in ambient BB plumes 10 times lower than the values reported by Hoffer et al. (2016), Chakrabarty et al. (2010) or Saleh et al. (2018); Saleh (2020) for laboratory generated particles. Also, Mathai et al. (2023) highlighted that even though Hoffer et al. (2016) and Chakrabarty et al. (2010) used similar methods, their k was at least 10 times different. Thus, due to poorly characterized optical properties, the impact of tar BrC on IR absorption at ambient conditions is still uncertain. Consequently, we follow here the common practice of considering ambient BC as the dominant absorber at 880 nm (e.g. Kirchstetter et al. (2004); Massabò et al. (2015); Liakakou et al. (2020); Zhang et al. (2020); Yus-Díez et al. (2022)). One important source of uncertainty in equations 1 and 2 is the AAE assumed for BC.”

Comment 4: It is not clear to me why you chose 370 nm instead of 550 nm for the discussion of light absorption, which is widely used for discussing aerosol optical properties. Could you justify that?

Response 4: We agree with the reviewer that the wavelength of 550 nm is commonly used to discuss the optical properties of atmospheric aerosol particles, especially for the scattering part given the high efficiency of submicrometric PM to scatter visible radiation. The same 550 nm wavelength has been also used to represent the absorption (e.g. Samset et al. (2018); Saleh et al. (2018); Saleh (2020)). Our decision to report here the OA absorption at 370 nm was due to the main fact that the OA absorption efficiency is the highest at 370 nm and for this reason the 370 nm wavelength is widely used in many publications reporting OA absorption from Aethalometer measurements. Consequently, an advantage of representing the OA absorption at 370 nm is that it can be retrieved from observations with a lower uncertainty compared to the visible (550 nm) range.

Moreover, for the k optimization we started from the k values proposed by Saleh (2020) and we used the Angstrom exponents (AAE) proposed by Saleh (2020) to report the k at 370 nm. Again, this choice was made in order to compare the modelled OA absorption with the observed

OA absorption at 370 nm with lower uncertainty. The same AAE provided by Saleh (2020) could be used to derive the optimized k reported here from 370 nm to 550 nm.

To consider the Reviewer comment, the sentence at lines 173-175 was modified as follow (now lines 200-204 in the revised manuscript):

“Figure 1 shows the average annual contributions of BC and BrC to the total absorption measured at 370 nm at twelve European stations, identified by color-coded markers indicating their background settings: yellow for urban, blue for suburban, and red for regional areas. Here we report the BrC absorption at 370 nm given that the BrC absorption efficiency is the highest in the UV spectral range and, consequently, the observed BrC absorption is less uncertain compared to the visible range.”

Comment 5: Your k at and babs at 370 nm seems too low to me. some babs even lower than some literature values reported from Arctic (see "On Aethalometer measurement uncertainties and an instrument correction factor for the Arctic" and "On Aethalometer measurement uncertainties and an instrument correction factor for the Arctic").

Response 5: The absorption reported in Figure 6 of the revised manuscript represents the contribution by OA particles to the total absorption at 370 nm. Thus, it represents a fraction of the total absorption measured at each site. Indeed, in summer, the reported OA absorption reached values lower than $1 Mm^{-1}$ especially at more remote sites. However, the OA contribution to absorption in summer is expected to be low due to the lack of important primary BrC sources as domestic biomass burning.

Moreover, here we used the values of k provided by Saleh (2020) at 550 nm. These k at 550 nm were calculated at 370 nm using the w Angstrom exponents provided by Saleh (2020). If, after the k optimization at 370 nm (Table S3 in the revised Supplement), the original w Angstrom exponents from Saleh (2020) were used to report the optimized k from 550 nm at the 370 nm, these latter will lie somewhere within the same original ranges provided by Saleh (2020).

To consider this Reviewer comment, the sentence at lines 590-591 (lines 535-540 in the revised manuscript) was modified as follow:

“Additionally, Figure 6 shows the time series of the absorption of OA at 370 nm simulated in Case 4 (all) and Case 5 (all) and the observational data for each monitoring site (by ‘stn’ in Fig. S6 in the Supplement). Although the absorption values could seem low, the annual mean OA absorption at 370 nm calculated from observations represented around 2-20% of the total measured absorption at some regional/remote sites (MSY, OPE and HYY) reaching contributions around 20-40% at the remaining sites. The OA absorption is the lowest in summer due to the lack of important primary BrC sources as domestic biomass burning. ”

Specific comments

Comment 1: Line 175-183, The discussion of BC and BrC sources lacks the evidence. Do you make these conclusions only based on the contribution? Or can any references support you? Or other data was involved.

Response 1: To consider the Reviewer comment, the sentence at Lines 175-183 (lines 200-219 in the revised manuscript) was modified as follow:

Figure 1 shows the average annual contributions of BC and BrC to the total absorption measured at 370 nm at twelve European stations, identified by color-coded markers indicating their background settings: yellow for urban, blue for suburban and red for regional areas. Here we report the BrC absorption at 370 nm given that the BrC absorption efficiency is the highest in the UV spectral range and, consequently, the observed BrC absorption is less uncertain compared

to the visible range. A low BrC contribution, around 14%, was observed at the urban sites of BCN and HEL, both affected by direct traffic emissions, making BC the dominant absorber at these sites (Okuljar et al., 2023; Via et al., 2021). MAR urban site registered higher BrC contribution (30%) likely reflecting the accumulation of biomass burning emissions in winter and the presence of BrC sources as shipping emissions (Corbin et al., 2018; Chazeau et al., 2022). Suburban stations, including SIR, KRA, and DEM, exhibit BrC contributions from 22% to 30%, reflecting a blend of local urban emissions and regional influences such as biomass burning and coal combustion. KRA is considered a pollution hotspot in Europe (e.g. Casotto et al. (2023)) with high consumption of coal and wood (both important sources of BrC) for energy production and residential heating, making the OA concentrations measured in KRA the highest among the European measuring sites included in Chen et al. (2022). DEM and SIR are affected by biomass burning emissions especially in winter, which causes a considerable accumulation of BrC during the cold season (see, e.g., Liakakou et al. (2020); ?); Savadkoobi et al. (2023)). Regional stations, represented by HYY, OPE, RIG, PAY, IPR, and MSY, display BrC contributions from 21% to 41%. These percentages indicate a mixture of biogenic sources, local emissions, agricultural activities, and transboundary pollution that affects the regional atmosphere. IPR stands out with the highest contribution (around 40%), suggesting a significant contribution of low-temperature combustion processes as residential sources (e.g. Putaud et al. (2018)). Overall, although BC typically represents the most absorbing aerosols component at these stations (usually > 70%), it is noteworthy that BrC could contribute comparably to absorption in some instances.

Comment 2: Line 214-215. It is not obvious to me how you make these assumptions. Why do you assume 50% hydrophobic species with OA/OC = 1.4 and all hydrophilic oxygenated components have OA/OC=2.1? How about other 50% hydrophobic species?

Response 2: There might be a misunderstanding in the description of the model's parameterization. Various approaches exist to represent organic aerosols (OA) in atmospheric models. In this study, we use the parameterization proposed by Pai et al. (2020). Typically, models treat primary organic aerosols as one hydrophobic and one hydrophilic component (Xian et al., 2019). This approach aims to simulate the near-field oxidation of the primary hydrophobic component. Emission inventories provide estimates of total organic carbon (OC) emissions, requiring assumptions to convert it to OA mass, the variable actually used in atmospheric transport models. In Pai et al. (2020)'s parameterization, OC emissions split equally between hydrophobic and hydrophilic aerosols. As MONARCH transports OA mass, it requires an assumption for OC-to-OA. Literature suggests ratios from 1.4 to 1.6 for hydrophobic OA species and over 2 for oxygenated species, depending on the combustion characteristics. Such values are highly uncertain and slightly different numbers are used among model parameterizations. As stated before, in this work we follow the parameterization proposed by Pai et al. (2020).

To clarify this point, the description of the scheme has been modified in the revised manuscript as follow:

Primary organic carbon (OC) emissions are emitted as 50% hydrophobic and 50% hydrophilic species. An OA/OC ratio of 1.4 is adopted for the hydrophobic component, while the hydrophilic one assumes an OA/OC ratio of 2.1 to convert OC to OA mass transported in the model.

Comment 3: Line 215-216: It is unclear how you get the conversion lifetime of 1.15 days.

Response 3: As explained in the previous comment, we adopted an OA parameterization proposed in the literature by Pai et al. (2020). The atmospheric aging of primary hydrophobic particles is simulated by its conversion to hydrophilic aerosol with a specific atmospheric lifetime.

This parameterization uses a lifetime of 1.15 days as previously proposed by other studies (Chin et al., 2002; Cooke et al., 1999).

Comment 4: Do you have references for these values in table 2, or did you derived them?

Response 4: The aerosol representation used in the MONARCH model has been described elsewhere (Spada, 2015; Obiso, 2018; Navarro-Barboza et al., 2024). The microphysical properties of the organic aerosol follow Chin et al. (2002), which in turn is based on the OPAC database (Hess et al., 1998). In order to clarify this point, Table 2 caption of the revised manuscript now includes references to Spada (2015); Chin et al. (2002); Hess et al. (1998).

References

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