

Responses to reviewer comments:

Referee report to egusphere-2025-1954

Referee comments are highlighted in red, our responses are in green, and changes made in the manuscript are shown in blue.

Comments:

Md. Al-amin Hossen et al. have carefully reviewed the manuscript and implemented the stated concerns the reviewers have referred to. In my opinion, they also sufficiently improved the text and the scientific discussion to be published as research article in ACP. I only have some last minor things, which need to be addressed before final publication.

Response: We thank the reviewer for the careful evaluation of our manuscript. We revised the manuscript according to the comments of the reviewer to ensure the quality of the manuscript so that it will publish in ACP.

- Line 26: change 'influence' to 'influences'

Response: Thank you for your suggestion. We have changed the word 'influence' to 'influences'. New line is given below.

Light-absorbing organic carbon, collectively known as brown carbon (BrC), significantly **influences** climate and air quality, particularly in urban environments like Dhaka, Bangladesh.

- Line 53: change 'shorter' to 'short'

Response: Thank you for your suggestion. We have changed the word 'shorter' to 'short' and corrected line is given below.

Brown carbon (BrC) is a type of organic carbon that absorbs light at **short** wavelengths, such as 300-400 nm (Laskin et al., 2015).

- Line 54 and 56: now you introduced the abbrev. BrC, so you should use it

Response: Thank you for your suggestion. We have used abbreviation of brown carbon (BrC) in line 54 and 56 as follow:

In contrast to black carbon, **BrC** exhibits a wavelength-dependent absorptivity that increases significantly towards the higher energy end of the spectrum (Wang et al., 2023). Similar to black carbon, **BrC** also has a positive radiative effect, which lowers the total cooling effect of atmospheric aerosols caused by light scattering (Feng et al., 2013).

- Line 60: change ‘automobile’ to ‘vehicle’

Response: Thank you for your suggestion. We have changed the word ‘automobile’ to ‘vehicle’ in the revised manuscript as follow:

The two main sources of BrC were **vehicle** emissions and biomass burning.

- Line 71: again use ‘BrC’ instead of brown carbon from now on

Response: Thank you for pointing this out. We have consistently used the abbreviation *BrC* instead of “brown carbon” from line 71 in the revised manuscript, as it was already introduced earlier.

- Line 90: good new discussion paragraph. Maybe you should use a citation for these arguments listed here

Response: Thank you for your suggestion. We have cited the articles in this paragraph as follow:

Aerosol surface morphology plays an important role in the light-absorbing properties and reactivity of BrC. Irregular or porous particle surfaces can enhance the adsorption of gaseous precursors, such as phenolic compounds, thereby influencing their heterogeneous oxidation and nitration pathways in the atmosphere (Liu et al., 2020; Wang et al., 2024). Moreover, morphological features can affect how BrC and its phenolic components interact with cloud or fog droplets, altering aqueous-phase reaction rates and the formation of light-absorbing secondary products.

Liu, D., He, C., Schwarz, J. P., and Wang, X.: Lifecycle of light-absorbing carbonaceous aerosols in the atmosphere, *NPJ Clim Atmos Sci*, 3, 40, 2020.

Wang, Y., Huang, R.-J., Zhong, H., Wang, T., Yang, L., Yuan, W., Xu, W., and An, Z.: Predictions of the optical properties of brown carbon aerosol by machine learning with typical chromophores, *Environ Sci Technol*, 58, 20588–20597, 2024.

- Line 137: point at the end of the sentence is missing

Response: Thank you for pointing this out. We have used a point in the manuscript now as follow:

Fig.1: Schematic diagram of the overall sampling and analysis procedure from July 2023 to January 2024 at the Dhaka North site and the Dhaka South site.

- Line 210: I rather meant that this is common knowledge. So, you can remove this section. However, just write in the result section 3.4.3 that you used the r -value for correlation analysis according to Hauke and Kossowski, 2011.

Response: Thank you for the suggestion. We have removed the section as advised and now mention in Section 3.4.3 that the r -value was used for correlation analysis according to Hauke and Kossowski (2011).

The new line in Section 3.4.3 is given below.

In this study, the r -value was used for correlation analysis according to Hauke and Kossowski (2011).

- Figure 2: Why do you put the Table (f) into the Figure. Rather put it as a stand-alone Table. In this case, you might also be able to enlarge panel (e).

Response: Thank you for the valuable suggestion. We have removed Table (f) from Figure 2 and presented it as a stand-alone table and also enlarge panel (e) in Figure 2 for better clarity.

Table -2: Relative abundance of surface elements in PM_{2.5} obtained from Energy Dispersive X-ray Spectroscopy

Elements	Atomic No.	Weight %
C	12	29.84
O	16	44.61
Al	13	0.62
Na	11	2.96
K	19	0.63
N	7	0.02
S	16	0.61
Ca	20	0.55
Cr	24	0.01
Pb	82	0.04
As	33	0.06
Hg	80	0.09

- Line 276: so, Fig. 2f will be Table later

Response: Thank you for pointing it out. We have removed the table from Figure 2 and represent it as a single table (Table 2) in the revised manuscript. We also removed Table (b) from Figure 5.

Table-2 shows that the quantity of carbon (C) and oxygen (O) was very high in PM_{2.5}, along with varying amounts of minerals (Ca, Fe, K, Al), sulfur, nitrogen, and trace metals (As, Cr, Cd, Pb, Hg).

- Fig3: if possible also try to enlarge it. It is your key Figure

Response: Thank you for your suggestion. We have enlarged Figure 3 in the revised manuscript according to your suggestion.

- Line 481: Please always write again the whole name in the ‘Conclusion’ section. E.g. for BrC and ATR-FTIR

Response: Thank you for the suggestion. We have written out the full forms of all abbreviations, including BrC and ATR-FTIR, in the Conclusion section as follows.

4. Conclusions

This study provides comprehensive insights into the compositions, surface elements, and formation mechanism of brown carbon in the atmosphere of Dhaka, Bangladesh, through the identification and quantification of seven key phenolic precursors and an aqueous-phase nitration experiment. We observed that PM_{2.5} predominantly exhibited spherical, irregular, and chain-like morphologies, with carbonaceous (C, O, N, S), mineral (Ca, Fe, K, Al), and trace elements (Pb, Cr, Hg, Cd, As) identified on their surfaces. Attenuated Total Reflectance - Fourier Transform Infrared Spectroscopy (ATR-FTIR) analysis revealed prominent functional groups, including carbonyls, nitro groups (–NO₂), and aromatic conjugated systems, suggesting the presence of complex organic structures. Among the phenolic precursors, 4-nitrophenol ($2.20 \pm 1.21 \mu\text{g m}^{-3}$) and 2-hydroxyphenol ($2.31 \pm 1.39 \mu\text{g m}^{-3}$) showed the highest concentrations, especially during winter. A strong seasonal and spatial variation was evident, with higher levels in Dhaka South, indicating the influence of urban emissions and combustion activities. Correlation analysis revealed strong positive relationships among most compounds ($r > 0.9$), suggesting shared emission sources such as biomass and fossil fuel combustion.

These findings suggest that phenolic compounds are not only abundant in the urban air of Dhaka but also highly reactive, forming nitrated derivatives such as 4-nitrophenol that contribute

substantially to **brown carbon**. The detection of nitrated and hydroxylated phenols further highlights their transformation via atmospheric oxidation processes. The similarity in chemical behavior and high co-occurrence among these phenolic compounds confirms their common origin and potential for secondary aerosol formation.

Compared to other urban regions like Jinan, China, and Strasbourg, France, the concentrations of phenolic and nitrated phenolic compounds in Dhaka are substantially higher, likely due to more intense and unregulated combustion activities (Li et al. 2020, Delhomme et al. 2010). Our findings align with prior studies (e.g., Wang et al., 2018; Li et al., 2020) identifying catechols and nitrophenols as dominant **brown carbon** components but extend the understanding by quantifying these compounds across seasons and locations within a South Asian megacity.

While this study presents robust data on PM_{2.5} composition and **brown carbon** precursors, limitations include the relatively short sampling period and limited spatial coverage. Additionally, although aqueous-phase nitration experiments were conducted, the ambient contribution of such processes remains to be quantified. Future studies should explore broader geographic regions, perform year-round monitoring, and couple laboratory findings with atmospheric modeling for deeper mechanistic insights.

The results underscore the significant contribution of phenolic and nitrated phenolic compounds to atmospheric **brown carbon** in Dhaka, with implications for both regional air quality and climate forcing. The demonstrated aqueous-phase nitration pathway of catechol at acidic pH suggests a potentially underappreciated route for **brown carbon** formation under humid and polluted conditions. This highlights the importance of regulating combustion-related emissions and considering aqueous-phase chemistry in climate models to accurately assess the radiative effects of organic aerosols.

- Line 483: Do not repeat 'particles' after PM_{2.5} as it is already included in 'particulate matter'

Response: Thank you for your suggestion. We have removed the particle word after PM_{2.5}.