

Author Responses and Planned Revisions to Reviewer (#1) Comments

Budhavant et al. reports a work depicting the extent of the long-range transport of pollutants from the Indo-Gangetic Plain to Maldives via Bangladesh, and the effects of atmospheric processes happened during the transport on the properties of light absorbing species (BC and BrC). This is an interesting study with the aerosol sampling done over strategically located sites. However, there are several other studies on the similar topic using the sampling at the same locations (Bosch et al., 2014; Dasari et al., 2019; Nair et al., 2023, etc.).

We appreciate the overall positive feedback. In contrast to the cited earlier studies, this is the first study to use all three of the widely distributed receptor observatories. This enabled for the first time to constrain the optical properties of both BC and BrC, water-soluble ions, and additional parameters. Their strategic positioning allows for a comprehensive investigation of the far-reaching South Asian outflow, with the key novel insight being the doubling of the MAC of BC aerosols.

Author shall explicitly mention the novelty of the current study over the reported studies. There are some interesting results but their explanations are not satisfactory, and sometimes there are conflicting arguments while explaining the observed data. Multiple parameters (MAC₆₇₈, MAC₃₆₅, OC/BC, Cl/Na, [NH₄], [SO₄], AAE) indicate that a better data churning is required. Attribution of all observations to long-range transport and processes is not convincing. The data shall be segregated with respect to wind back trajectories and different categories shall be discussed differently with more converging inferences. There is a sufficient room for modifications before the manuscript can be considered for publication in ACP.

We will follow the reviewer's recommendations and further develop the interpretations. The three major findings of this study are:

- A near doubling of BC mass absorption cross-section (BC-MAC) from the IGP exit (at BCOB) to the Indian Ocean receptor sites, with the offered explanation backed up by ancillary data being a coating enhancement process.
- A decrease in BrC-MAC over the same over-ocean transport, with the hypothesized mechanism being the photochemical degradation of light-absorbing organic moieties.
- An enhancement of Cl/Na signals the significant presence of anthropogenic Cl at the exit of IGP, possibly affecting atmospheric oxidation.

We agree to explore segregating the data into wind sector categories for further evaluation of the results.

Major Comments:

L74: Why the water-insoluble BrC is ignored? There are several studies showing water-insoluble BrC is a considerable part of BrC. A comparison of water-soluble BrC alone with BC gives an incomplete and biased picture of BrC effects.

WS-BrC enhances the light absorption of BC and is evaluated also using its MAC to estimate the radiative forcing of both EC and WS-BrC. The absorption Angstrom exponent (AAE) is another optical property of BrC, indicating the wavelength dependence of light absorption, which helps extrapolate the optical properties of WS-BrC. Therefore, it is crucial to study the light absorption characteristics of both EC and WS-BrC together to quantify the relative light absorption contribution of WS-BrC to EC, which is currently lacking in the literature, especially in rural and receptor sites in South Asia. We appreciate the reviewer's suggestion and will both consider these aspects in future studies and will also here revise the current manuscript to include recognition of water-insoluble BrC.

L189-193: If this logic is true, why didn't OC come from that region? What processes can remove only OC but not SO₄²⁻ and NH₄⁺ coming from same region?

The high SO₄²⁻ and NH₄⁺ concentration at MCOH comes mainly from central and eastern India, where thermal power plants, construction industries, and petroleum refineries emit pollutants like SO₂ and NO_x that are further oxidized in the atmosphere OC to BC decreases significantly during long-range transportation due to selective processing and/or washout of OC. We have earlier shown this system that OC has a shorter lifetime and undergoes more transformation than both BC, SO₄, and NH₄, making it a complex mixture (Budhavant et al., 2020). The differential removal rates during transportation processes have a greater impact on OC than on BC, leading to a relatively higher fraction of BC at the MCOH and MCOG sites, constituting a larger relative share of the total carbonaceous aerosols than BC (Budhavant et al., 2018, 2020).

L204-208: Discussion on Cl/Na ratio is weak and too hypothetical. The seawater ratio of Cl/Na is 1.8 (on mass basis). A dotted line with ratio 1.8 can be added to the Figure 4. At BCOB site, Cl/Na ratio is varying from 0 to 11. It shall be explained that what sources or processes are adding or removing particulate Cl from the samples. There shall be convincing supporting evidences.

We are grateful for the suggestion and will make several revisions to Figure 4, including adding a dotted line with the seawater ratio of 1.8. An enhancement of Cl may be very important. While this is not the central focus of the paper, it is of such potential significance for regional atmospheric chemistry that we feel it behoves the manuscript to bring this out. We will include more detailed descriptions to support the arguments in the revised manuscript.

L217-221: The inherent assumption in this statement is that all BC at both the receptor sites is transported from South Asia, which is not concurred by wind trajectories.

Although receptor sites MCOG and MCOG have slightly different trajectory paths, by far most of the trajectories originate from South Asia, especially during synoptic measurements. Please refer to Figure 1 and Figures S1-S5, which depict the AMBTs, fire spots, and AOD across the sampling region. While there may be other small sources on the route, such as ships and small islands, their contribution to total geographical and BC emissions is negligible. We can thus conclude that most BC emissions in the region come from South Asia. We will re-analyze this aspect during revisions and assert more clearly what the data shows.

L225-227: This is a strong but hypothetical statement. It has to be proven with other supportive measurements. Why the difference in MAC678 can't be due to different sources of BC over these sites? In fact, this inference may be more logical because OC/EC ratios are also different at those sites.

There are very few local and insignificant sources of BC over these small ocean islands compared to the heavily polluted air masses originating in the subcontinent. The decreasing OC/EC ratios are explained by higher washout of OC than EC and higher oxidation of OC than of EC, as discussed and referenced in the manuscript text. Nevertheless, we will consider the reviewer's concerns and will look to revise the text on this subject for increased clarity.

L248-250: As per the reasons given for MAC678, shouldn't MCOG have a lower MAC365 compared to MCOH?

We would like to clarify that BC and BrC have very different wavelength-dependent absorption properties and drastically differ in their involvement with other components during long-range transport. It appears that the bleaching process is becoming less significant, possibly because the most labile components have already been consumed around MCOH.

L282-284: This is quite illogical and not convincing. There are several inferences which contradicts each other.

Our description may not fully support our argument about the radiative forcing at BCOB, so we will revise the text in the revised manuscript.

Table 1: How come SO₄ and NH₄ conc are similar or even higher than BCOB. It is counter intuitive and warrants satisfactory justification. Concentrations of other species like OC, WSOC, NO₃, K, etc. look as per expectations.

As explained earlier, the high SO₄²⁻ and NH₄⁺ concentration at MCOH comes mainly from central and eastern India, where thermal power plants, construction industries, and petroleum refineries emit pollutants like SO₂ and NO_x that are further oxidized in the atmosphere OC to BC decreases significantly during long-range transportation due to selective processing and/or washout of OC. Nevertheless, we will consider the reviewer's concerns and will look to revise the text on this subject for increased clarity.

The increase in BC MAC₆₇₈ is attributed to coating during transport from IGP to MCOH and MCOG. However, back trajectories analysis shows that winds were not from the IGP for a considerable time period. How can the higher MAC₆₇₈ be justified in those samples?

In certain situations, the winds in the IGP region do not originate from within the area. Instead, they sometimes come from southern India or the Bay of Bengal. However, during winter, polluted winds from the IGP can reach the Bay of Bengal, leading to similar signals being detected over the MCOH and MCOG regions. This is especially noticeable during synoptic observations. We will elaborate on these aspects in the revised ms.

Fig. 1: 10-days air mass back trajectories are used in this paper, which is in contrast to most of the studies which are using 5 or 7 day back trajectories. The reason of using 10 day back trajectory shall be explained.

The lifespan of BC is influenced by wet and dry deposition, with fine-mode aerosols tending to persist for longer periods. Factors such as humidity, wind speed, temperature, and mixing state also affect its lifespan, which typically ranges from one to two weeks. Based on our earlier studies on this Indian Ocean receptor, it has become clear that anthropogenic aerosols at this time may have quite a long residence time, even surpassing two weeks (e.g., Budhavant et al., 2020, 2023). This is the key motivation for having longer BT times than 5-7 days. This study examines air mass back trajectories to identify potential sources of BC and other aerosol components at the MCOG station, which is even further from MCOH. Given this long-distance travel, the focus on the dry season (increasing longevity), the fact that BC has longer lifetimes than other aerosols, and the experiences from earlier studies, a BT time horizon of ten days is deemed appropriate. We will in the revised manuscript elaborate on this motivation.

Fig.2: This data shall also be plotted in different ways. Samples with similar BC fractions are showing quite different MAC₆₇₈. It shall be explained.

We will revise the manuscript to acknowledge and explain that similar BC fractions may display different MAC₆₇₈ due to differences in, e.g., coating and internal mixing.

Fig. 3: As per this plot, WIOC is the dominant fraction of OC. A significant part of this WIOC could be BrC, which is not measured, reflecting limitation of this work. Further, in many samples with low WSOC fraction, MAC₃₆₅ is quite high. Possible reasons shall be discussed.

This point is already made above. While this and earlier studies are showing that WSOC is the dominant fraction of OC and an analytically tangible fraction that contains light-absorbing moieties, the manuscript will be revised on this aspect to more explicitly acknowledge that there is also WIOC.

Fig. 4: In many of the samples collected over MCOH and MCOG, OC/BC looks close to 1, which is not normal. How is it inferred?

It is correct that the ratios of OC/BC are relatively low compared to other studies conducted in mainland South Asia, except in certain urban cities such as Dhaka and Chennai. Nevertheless, the OC/EC ratios reported for MCOH in many other earlier studies align with the current study's findings on this ratio. This suggests decreasing OC/BC between IGP exit and after transportation over the ocean, indicating selective washout and bleaching reactions of organic carbon, as discussed in detail in the m (page 6, line 183-185).

It would be better to plot WSOC/OC rather than WSOC/TC because WSOC/OC ratio can be better interpreted.

We will implement this change suggested by the reviewer in Figure 4 during revisions.

Fig.5: As the major focus of this paper is on BC and BrC, it would be appropriate to calculate RF for BC and BrC, and their contribution to total aerosols RF.

Thank you for your suggestion, but it is currently beyond the scope of our study. However, we will consider it for future studies.

Fig.S2-S4: Why 10 days and not 5 or 7 days? Why at 50 m only? It would be better to add a few higher altitudes relevant for long-range transport.

Please refer to our response to the back trajectories of Figure 1 a few comments earlier. The sampling towers of the three atmospheric observatories have an altitude below 20 meters on their islands. We hence used a height of 50 meters to compute the air mass back trajectories for the sampled boundary layer.

Fig, S5: AOD data appears to follow expected trends, unlike chemical data.

We believe both AOD and chemical data can be understood, as discussed in the manuscript.

Table S1: SO₄, NH₄, nss-Ca, nss-Mg are not correlated with any species, why? Where is Na and Cl? Major ions data (absolute concentrations) shall also be given here.

Nss-SO₄ comes mainly from industries, coal power plants, and ships; NH₄ comes mainly from agriculture, including animal husbandry and NH₃-based fertilizer application, whereas nss-Ca and nss-Mg are primarily sourced from soil. Thus, it is not expected that all these are correlated with each other. While certain correlations do exist between some measured ions, such as nss-SO₄ and NO₃, nss-Ca, and nss-Mg, they were not explicitly detailed in the manuscript since the paper primarily focuses on optical features. Additionally, we will include Na and Cl in Tables S1-S3 and provide their concentrations in Table 2.

Table S2: Why do NO₃, Ca, and Mg are not correlating with any other species?

Please refer to our previous response to the comment above.

Table S3: Why does K not correlating with any other species?

Please refer to the response to the earlier comment.

Minor Comments:

L53-56: add appropriate references from this region.

We will add more references.

L120: Eq 3: How was the babs measured? Why at 365 nm only?

The absorption spectrum of water extracts was measured using a Hitachi absorption spectrophotometer-2010, in the range of 190 to 1200 nm (as described in the Methods section; lines 118-126). BrC is an organic aerosol moiety that absorbs light of shorter wavelengths. It is mainly absorbed in the ultraviolet and near-visible wavelengths, which gives it a brownish or yellowish appearance. To accurately measure the levels of BrC present in particles, the absorption coefficient between 360 and 370 nm (average of 365 nm) is commonly used and allows for inter-comparison of BrC between studies

L255-257: There are numerous recent studies and MAC365 shall be compared with the recent studies.

We will compare our results with even more earlier reports of this property in the revised version of the manuscript.

L262-264: mention the wavelength range used for AAE calculation for the better clarity for readers.

To prevent any potential interference from light-absorbing solutes such as ammonium nitrate, sodium nitrate, and nitrate ions, which have absorption peaks near 308, 298, and 302 nm, respectively, the AAE was based on the 330-400 nm range, as is common praxis. This is already mentioned in the manuscript in the earlier section, lines 125-126.