

Author Responses and Planned Revisions to Reviewer (#2) Comments

The Budhavant manuscript describes the results of SAPOEX campaign at three sites in South Asia. There is potential with this dataset, but the authors need to do more to connect across the results in each section (e.g. back trajectory analysis, aerosol composition, optical properties, radiative forcing). For example, the absolute concentrations of aerosol composition were only presented as campaign averages, which made it difficult to compare to the aerosol radiative forcing which was presented and discussed as an average and by month. There was often an over-simplification of the results, for example, the assertion that the MCOH site received transported air masses from the BCOB site: this did not seem to be always the case so it added confusion when discussing composition and aging. Overall the manuscript needs more refining of focus and connection among the different sections. Even in the introduction, the discussion of source and processing impacts on BC and BrC was over-simplified and lacking in precision.

We appreciate this constructive feedback and concrete suggestions to enhance our manuscript. We are committed to making these necessary revisions and providing supplementary information to support our findings. We will scrutinize for opportunities to relate the parts closer to each other and synthesize the overall findings.

Detailed comments:

Ln 25: can this be better linked in the abstract to the aerosol optical properties?

Yes. The revised manuscript will link the similar pattern between Br-MAC and total radiative forcing to aerosol optical properties.

Ln 46: this is an awkward phrase here. please edit

We will edit this text bit for clarity.

Ln 47: is this referring specifically to this region? this is undoubtedly true for BC, but WSOC may have other sources? e.g. biogenic and SOA?

Thank you for bringing this additional dimension up. We acknowledge that WSOC may have additional sources, such as biogenic SOA, and we will make the necessary changes to the manuscript.

Ln 47 – 56: this paragraph is difficult to follow as written. the authors need to clarify their purpose here. There is some confusion as they are trying to simultaneously discuss BC and WS-BrC. It doesn't really work and needs editing for clarity.

We recognize that our initial is blurred. We will edit thoroughly, likely breaking the paragraph up into two separate ones for the two aspects.

Ln 49: This sentence is awkward and the logical transition here is unclear

We will revisit to address the issue.

Ln 65: edit for clarity

We will edit for clarity.

Ln 88: What is the particle size here? TSP? PM_{2.5}

We have used PM_{2.5} samples. This will be explicitly mentioned here.

Ln 111: are there any concerns about the high loading on these filters? typically filter-based photometers limit the filter loading to that which corresponds to a 50% transmission. the filters collected for this offline analysis would not have their loading limited by light transmission. i understand that this correction is intended to address the filter loading, but these correction schemes were originally designed for online instruments which have a filter advancement/change at a set transmission threshold. can this concern be addressed?

Yes, the filter loading is a concern in this highly polluted air (especially for BCOB). We have invested substantial consideration into any effects of this and have thus previously reported and discussed filter-loading corrections and these are cited in this manuscript (e.g., Budhavant et al., 2020).

Ln 180-182: its not clear how these facts are relevant here. please remove or expand the discussion to make this more clear.

We will revisit this part and remove any irrelevant parts, and add more supporting explanations as needed.

Ln 191-195: what about biogenic SO₄? do you have a constraint on the possible marine contribution that goes beyond sea salt? Additionally, this rationale of sources from central and east India is a bit confusing as the BTs indicate that air masses predominantly leave the Indian subcontinent near BCOB or from west India before traveling to MCOH. If the aerosol composition from the west side of India is markedly different (e.g. higher SO₄ fraction) than the IGP and BCOB, than the aging discussion of MCOH representing aged BCOB aerosol needs to be more refined.

A previous study at MCOH found that DMS contributes only up to 3% to nss-SO₄ in polluted air (Granat et al., 2010). We will evaluate the SO₄/BC load for samples coming from west India vs from the Bay of Bengal. The “synoptic” comparison will be focused on winds coming from the IGP and southern India. The revised manuscript will be updated to this effect.

Ln 208: Is the EC supposed to be BC?

We confirm that this is BC.

Ln 217-223: I'm still stuck on the SO₄ discussion. the provenance of the SO₄ seems very relevant in determining if these 3 sites do represent different ages of the same air mass. as

briefly mentioned in this section, increased SO₄ would also seem to be very relevant for the coatings question. however some of the previous discussion of the loss of water soluble fraction during aging and transport (WSOC) seems to conflict with this. certainly the increase in SO₄ at MCOH, absolute concentration as well as an extreme increase in relative contribution, is very relevant for coating of BC and internal mixture of BC and SO₄ aerosol. i would like to see more discussion of this potentially conflicting observations between wsoc and so₄, and using the three sites as steps in the aging process of one air mass. If the rationale for higher SO₄ at MCOH is a shift in geographic source region, than the discussion of aging of two different aerosol systems needs to be included.

We agree with this thinking. We will return to these aspects and seek to both clarify and provide additional information by incorporating two distinct aerosol systems.

Ln 265-267: are these differences in AAE significant? the std deviation is relatively high.

We have noted the differences mentioned, but they do not seem significant. Hence, we will proceed with making the necessary changes.

Ln 282-284: can the authors discuss why the atmospheric forcing was higher at MCOH while the surface concentrations for summed species was lower? it is also a bit difficult to interpret the relationship between the aerosol radiative forcing and the rest of the surface aerosol discussion as the time scales are not well aligned for aerosol concentrations and the ARF. overall, i'd like to see better connections among the sections of the discussion.

In general, MCOH has lower radiative forcing than BCOB. However, during January, MCOH had slightly higher atmospheric forcing due to outflow from IGP, as shown in Figures 5, 2, and 3. We will provide more clarity in the revised version. We will also seek to connect/synthesize the findings in the different sections further.

Table 1: this AAE is calculated off a very narrow range in wavelength. is 400 nm the longest wavelength measured here? What are the potential shortcomings of reporting AAE for such a narrow range in wavelength? Also, it is difficult to assess the trends in the ambient concentrations when only the averages for the entire period are reported. It is fine to present the ratios in the figures, but useful to also be able to see changes in the absolute concentrations as well.

We have measured the wavelength range from 190 to 1200 nm, yet, as elarated above in response to reviewer 1, the AAE is customarily reported for the range of 330 to 400 nm. One reason is 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. Further AAE is more dependent on linear ratios for shorter wavelengths, while for longer wavelengths, the correlation is weaker. We plan to revise to include these motivations in the additional information for Table 2.