

## **Author response to Reviewer #1 comments**

We sincerely thank the reviewer for the valuable comments. Based on the comments we received, careful modifications have been made to the manuscript. Our point-by-point response to the review comments are given below. The comments are marked in bold blue font and our responses are marked in normal black font below each comment.

### **Reviewer #1**

**The manuscript titled ‘3D assimilation and radiative impact assessment of aerosol black carbon over the Indian region using aircraft, balloon, ground-based, and multi-satellite observations’ by Kala et al. presents the three-dimensional gridded data set of black carbon (BC) aerosols over the Indian mainland utilizing data from the ground-based network, aircraft- and balloon-based measurements from multiple campaigns, and multi-satellite observations, followed by statistical assimilation techniques. It is a well-written work with excellent scientific merit, and I do not hesitate to recommend this manuscript for publication. I have a few minor suggestions below; those can be incorporated during the revision.**

We appreciate with thanks, the summary evaluation, and the positive recommendation.

#### **Specific comments:**

**Line 27-111: The introduction looks a bit wordy. Therefore, I recommend that the authors be concise in the introduction without losing the theme of this work.**

Complied with.

**Line 104-111: I would suggest removing these explanations here in the introduction since each section are well defined subsequently.**

Complied with.

**Line 16-18: Consider rewriting this sentence.**

Complied with. The sentence has been modified as: “The regional maps of BC-induced atmospheric heating derived using this dataset capture the elevated aerosol heating layers over the Indian region along with the spatial high over the Indo-Gangetic Plains.” in Page no.1 [line no.16-18]).

**Line 20-23: Rephrase this sentence.**

Complied with. The sentence has been modified as: “This could strongly influence the upper tropospheric and lower stratospheric processes, including the vertical transport of BC to higher altitudes and thus have larger implications for atmospheric stability than what would be predicted using satellite observations alone.” (Page no.1 [line no.20-23]).

**Line 32: I would suggest the authors add a few more references for the transport of BC aerosols to the high-altitude Himalayas in this context. E.g., Kompalli et al., 2016; Negi et al., 2019; Arun et al., 2019; Roseline et al., 2021; Gogoi et al., 2021; Arun et al., 2021;, etc. are the recent ones and authors can find more in the literature.**

Complied with, by adding a few more references. (Page no. 2 [line no.35-36])

**Line 41: faraway?**

Yes.

**Line 116-118: How are these seasons classified? Explain the criteria behind this.**

Based on the south-west monsoon which defines the climate of the region, there are generally four seasons considered over the Indian region. They are the cold and dry winter (December to February; DJF), pre-monsoon summer (March to May; MAM), monsoon (June

to September; JJAS) and post-monsoon (October to November; ON). In the present study, monsoon season has not been considered owing to the non-availability of sufficient aerosol measurements and the assimilated BC AAOD data, which form the base for processing the background BC absorption coefficient ( $k_{bg}$ ).

**Line 124-127: Are the Aethalometer datasets pressure corrected for the high-altitude ground-based network stations? Explain these details also in the revised version.**

Yes, the high-altitude Aethalometer measurements have been pressure corrected. A sentence has been added in the revised manuscript as: "The Aethalometer operation and corrections carried out during different campaigns are explained in detail in the references listed in Table 1". (Page no.4 [ line no.124-125])

**Line 143: Explain the uncertainties about the Absorption AOD from OMI used in this study. Also, there are a number of caveats that concern the validity of the results in this study. I would recommend that the authors also add details about this in the discussions.**

Thank you for pointing it out. The assimilated absorption AOD, OMI absorption AOD, and their uncertainties are discussed in detail in Pathak et al. (2019). The main sources of uncertainty in the OMI AAOD are the sub-pixel cloud contamination caused by the considerably larger pixel size (1325 km<sup>2</sup>), and the OMI retrievals overestimating AOD and underestimating SSA (Torres et al., 1998; 2005; 2007). Moreover, the assumptions on the vertical distribution of different aerosol species and the height of the aerosol layers contribute to the uncertainty in OMI retrievals. Torres et al. (2002) have estimated the total error in OMI AOD to be around 30% and that of SSA to be 0.05–0.1. Taking this into account, the present study has used the assimilated BC AAOD dataset generated by Pathak et al. (2019), having an uncertainty between 11% to 20% with a mean value of 15%, which is better than that of OMI AAOD.

- Pathak, H. S., Satheesh, S. K., Nanjundiah, R. S., Moorthy, K. K., Lakshmivarahan, S., and Babu, S. S.: 740 Assessment of regional aerosol radiative effects under the SWAAMI campaign–Part 1: Quality-enhanced estimation of columnar aerosol extinction and

absorption over the Indian subcontinent, *Atmospheric Chemistry and Physics*, 19, 11865–11886, <https://doi.org/10.5194/acp-19-11865-2019>, 2019.

- Torres, O., Bhartia, P. K., Herman, J. R., Ahmad, Z., and Gleason, J.: Derivation of aerosol properties from satellite measurements of backscattered ultraviolet radiation: Theoretical basis, *Journal of Geophysical Research: Atmospheres*, 103, 17099–17110, <https://doi.org/10.1029/98JD00900>.
- Torres, O., Decaeu, R., Veefkind, P., and de Leeuw, G.: OMI Aerosol Retrieval Algorithm, OMI Algorithm Theoretical Basis Document Volume III, Clouds, Aerosols, and Surface UV Irradiance, 4, pp. 47–71, 2002.
- Torres, O., Bhartia, P. K., Sinyuk, A., Welton, E. J., and Holben, B.: Total Ozone Mapping Spectrometer measurements of aerosol absorption from space: Comparison to SAFARI 2000 ground-based observations, *Journal of Geophysical Research: Atmospheres*, 110 (D10), <https://doi.org/10.1029/2004JD004611>, 2005.
- Torres, O., Tanskanen, A., Veihelmann, B., Ahn, C., Braak, R., Bhartia, P. K., Veefkind, P., and Levelt, P.: Aerosols and surface UV products from Ozone Monitoring Instrument observations: An overview, *Journal of Geophysical Research: Atmospheres*, 112 (D24), <https://doi.org/10.1029/2007JD008809>, 2007.

**Line155: How the BC AOD is estimated using OPAC model? Please elaborate more on this. Explain the constrained variables and uncertainties in doing so.**

The ground-based BC mass concentration measurements along with planetary boundary layer height data from Modern-Era Retrospective Analysis for Research and Applications-2 (MERRA2) are used to constrain the Mie scattering model Optical Properties of Aerosols and Clouds (OPAC), to obtain a near-realistic vertical variation of aerosols and thus the BC AOD. This is a good assumption for the vertical distribution of aerosols over the Indian region and is supported by several studies in the past across different seasons (Satheesh et al., 2008; Babu et al., 2010, 2011). These are already mentioned in Page 5 (lines 154 – 161). The uncertainty associated with the BC AAOD is within 11% to 20% with a mean value of 15% (Pathak et al., 2019).

- Babu, S. S., Moorthy, K. K., and Satheesh, S. K.: Vertical and Horizontal Gradients in Aerosol Black Carbon and Its Mass Fraction to Composite Aerosols over the East Coast of

Peninsular India from Aircraft Measurements, *Advances in Meteorology*, 2010, <https://doi.org/10.1155/2010/812075>, 2010.

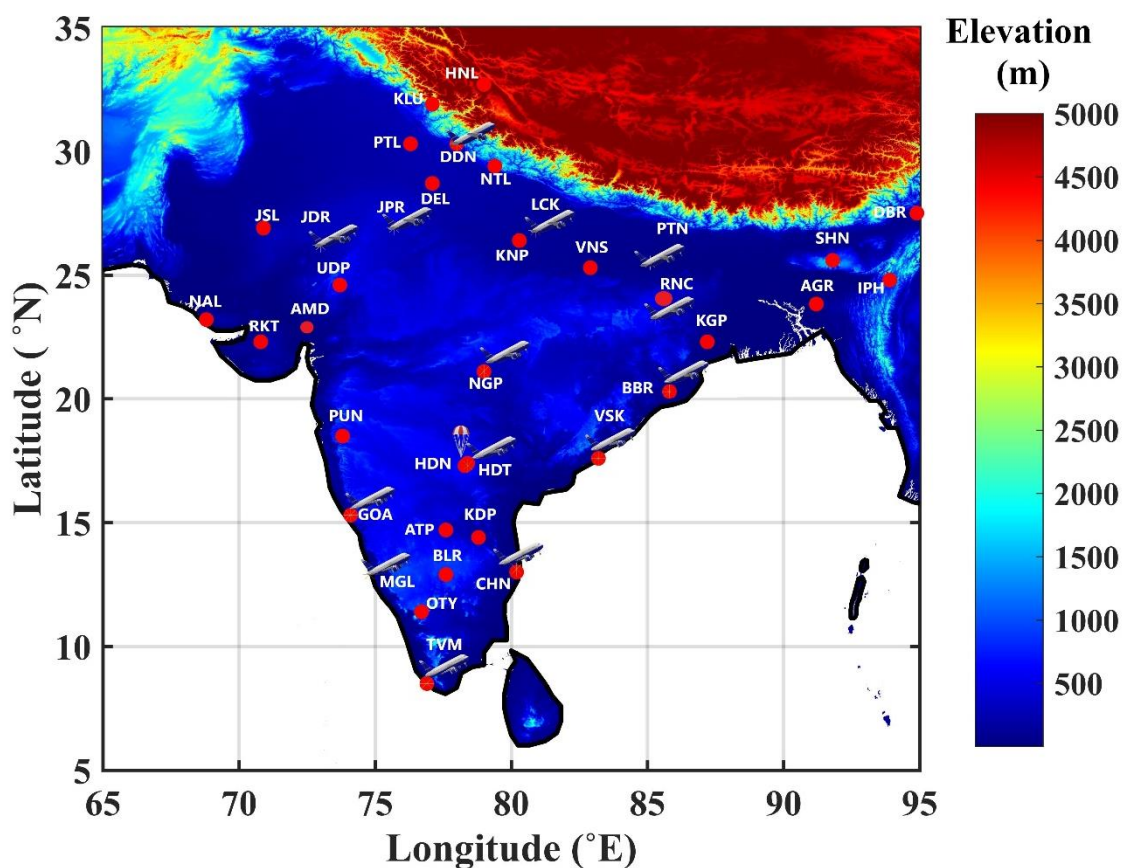
- Babu, S. S., Moorthy, K. K., Manchanda, R. K., Sinha, P. R., Satheesh, S. K., Vajja, D. P., Srinivasan, S., and Kumar, V.: Free tropospheric black carbon aerosol measurements using high altitude balloon: do BC layers build “their own homes” up in the atmosphere?, *Geophysical research letters*, 38, <https://doi.org/10.1029/2011GL046654>, 2011.
- Pathak, H. S., Satheesh, S. K., Nanjundiah, R. S., Moorthy, K. K., Lakshmivarahan, S., and Babu, S. S.: 740 Assessment of regional aerosol radiative effects under the SWAAMI campaign–Part 1: Quality-enhanced estimation of columnar aerosol extinction and absorption over the Indian subcontinent, *Atmospheric Chemistry and Physics*, 19, 11865-11886, <https://doi.org/10.5194/acp-19-11865-2019>, 2019.
- Satheesh, S. K., Moorthy, K. K., Babu, S. S., Vinoj, V., and Dutt, C. B. S.: Climate implications of large warming by elevated aerosol over India, *Geophysical Research Letters*, 35, <https://doi.org/10.1029/2008GL034944>, 2008.

**Line 174: Consider rewriting this sentence.**

Complied with. The sentence has been modified as: “As part of various field campaigns, in-situ measurements of the vertical profiles of BC aerosols have been carried out at different locations and seasons over the Indian mainland and the adjoining oceans.” (Page no.6 [line no.173-174]).

**Line 176, Figure-1: I recommend the authors to also show variabilities in the elevations of the study domain in the same map. There are different resources that provide digital elevation data.**

Thank you for the suggestion. Complied with and the revised Figure 1 is shown below. The elevation data is obtained from the United States Geological Survey Earth Resources Observation and Science (USGS EROS) archive.(Page no.4 [ line no. 119-121])



**Figure 1. Map showing the locations of surface (red markers) and aerial (aircraft and balloon) measurements. The colour scheme in the background shows the elevation data. The aircraft and balloon symbols respectively mark the aircraft and high-altitude balloon measurement locations. The station codes for the ARFI measurement sites are given in Table 1 and S1. (Page no. 25 [Line no. 897-900]).**

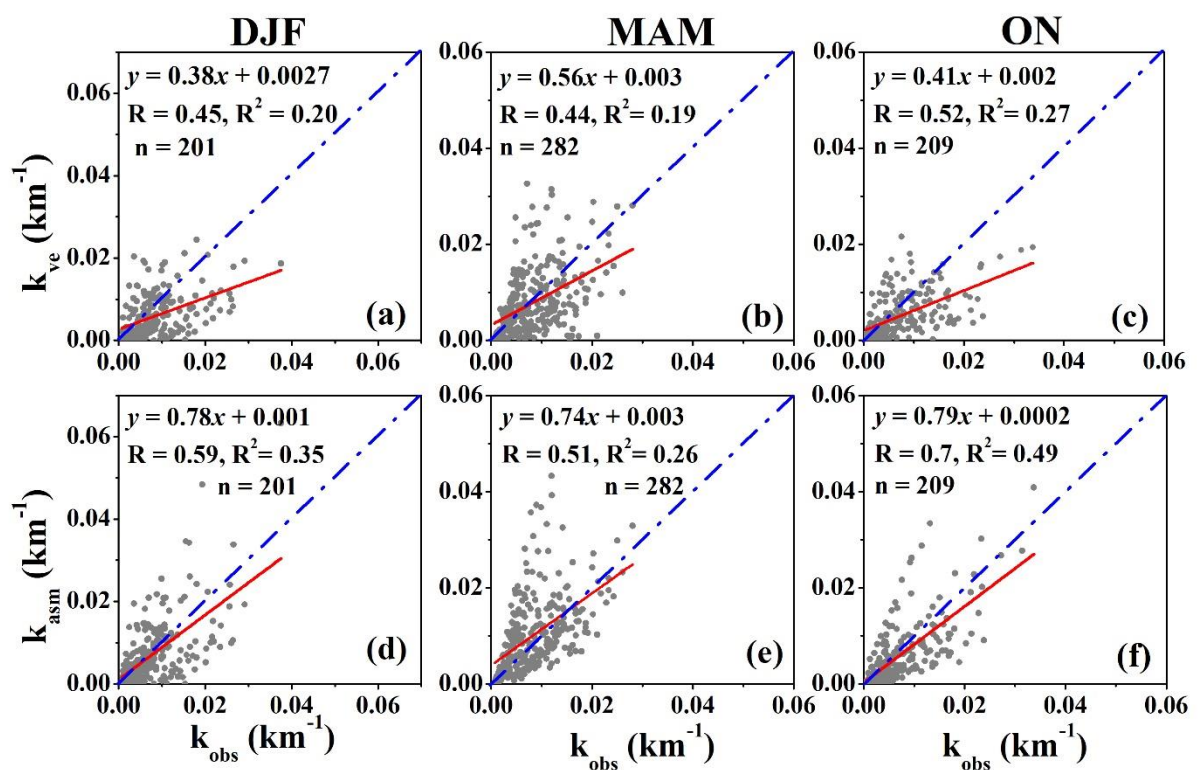
**Line 262: I suggest the authors to add some information regarding the role of crop residue burning during October-November in northern India.**

Complied with. A sentence on the role of crop residue burning during October-November in northern India has been added as: “During MAM, the IGP region is influenced by BC produced from agricultural residue burning and forest fires and the continental inflow of polluted aerosols to this region (Singh et al., 2019) whereas the crop residue burning has also been cited as a contributing cause during ON (Badarinath et al., 2006; Jain et al., 2014; Kaskaoutis et al., 2014).” (Page no. 8 [Line no. 262-265])

- Badarinath, K. V. S., Kiran Chand, T. R., and Krishna Prasad, V.: Agriculture crop residue burning in the Indo-Gangetic Plains—A study using IRS-P6 AWiFS satellite data. *Current Science*, 1085–1089, 2006.
- Jain, N., Bhatia, A., and Pathak, H.: Emission of air pollutants from crop residue burning in India. *Aerosol and Air Quality Research*, 14(1), 422–430, 2014, <https://doi.org/10.4209/aaqr.2013.01.0031>.
- Kaskaoutis, D. G., Kumar, S., Sharma, D., Singh, R. P., Kharol, S. K., Sharma, M.: Effects of crop residue burning on aerosol properties, plume characteristics, and long-range transport over northern India. *Journal of Geophysical Research: Atmospheres*, 119, 5424–5444, 2014, <https://doi.org/10.1002/2013JD021357>.

**Line 389, Figure-8: I would recommend the authors to show the R<sup>2</sup> values in these plots.**

Complied with: The modified plot with the R<sup>2</sup> values is shown below and in Page no. 30, line no. 923-927 in the revised manuscript)



**Figure 8. Scatter plots between  $k_{obs}$  and  $k_{bg}$  (top panels) and between  $k_{obs}$  and  $k_{asm}$  (bottom panels) for DJF (left panels), MAM (middle panels), and ON (right panels) seasons. The**

red line denotes the linear fit, the dashed blue line denotes the 1:1 line, and the scatter points are shown in gray. The equation of fit, correlation coefficient (R), R<sup>2</sup>, and the number of scatter points (n) are shown in each sub plots.

**Line 418: Explain the statistical significance of the derived Heating rate values and their uncertainties.**

The uncertainty of the heating rate has been evaluated by first estimating its error covariance matrix, as shown below:

$$M = (I/(k-1)) (H^*H^T) \quad (3)$$

Here, ‘I’ is an identity matrix of size n×n, ‘H’ is the heating rate matrix transformed into a matrix of size n-1, and ‘k’ is the number of time steps involved for each season. The background error covariance matrix is evaluated separately for DJF, MAM, and ON seasons. The final matrix ‘M’ consists of the variance of the heating rate for each grid, and the off-diagonal elements represent the co-variances. The uncertainty is estimated using the variance values (diagonal elements of matrix ‘M’; Niu et al., 2008; Zhang et al., 2008; Singh et al., 2017; Pathak et al., 2019). The root mean square value of the uncertainties for the derived heating rate for all the seasons is found to be ~17%. The uncertainty associated with the estimated heating rate is mentioned in Page 12, line 415 – 416.

- Niu, T., Gong, S., Zhu, G., Liu, H., Hu, X., Zhou, C., and Wang, Y.: Data assimilation of dust aerosol observations for the CUACE/dust forecasting system, *Atmospheric Chemistry and Physics*, 8, 3473-3482, <https://doi.org/10.5194/acp-8-3473-2008>, 2008.
- Pathak, H. S., Satheesh, S. K., Nanjundiah, R. S., Moorthy, K. K., Lakshmivarahan, S., and Babu, S. S.: Assessment of regional aerosol radiative effects under the SWAAMI campaign– Part 1: Quality-enhanced estimation of columnar aerosol extinction and absorption over the Indian subcontinent, *Atmospheric Chemistry and Physics*, 19, 11865-11886, <https://doi.org/10.5194/acp-19-11865-2019>, 2019.
- Singh, R., Singh, C., Ojha, S. P., Kumar, A. S., and Kumar, A. K.: Development of an improved aerosol product over the Indian subcontinent: Blending model, satellite, and



ground-based estimates, *Journal of Geophysical Research: Atmospheres*, 122, 367-390, <https://doi.org/10.1002/2016JD025335>, 2017.

- Zhang, J., Reid, J. S., Westphal, D. L., Baker, N. L., and Hyer, E. J.: A system for operational aerosol optical depth data assimilation over global oceans, *Journal of Geophysical Research: Atmospheres*, 113, <https://doi.org/10.1029/2009JD013364>, 2008.

**Line 432: I recommend the authors add some literature related to aerosol-cloud interaction and the role of BC in it.**

Thank you for the suggestion. Complied with and the sentence has been modified in the revised manuscript with additional references on aerosol-cloud interactions as: “It could also lead to increased cloud activities in the higher atmosphere as BC (aged) can act as cloud condensation nuclei and influence the aerosol-cloud interactions (for e.g., Ackerman et al., 2000; Rosenfeld, 2000; Chuang et al., 2002; Penner et al., 2004; Kaufman et al., 2005; Lin et al., 2018; Zanatta et al., 2023)”. (Page no. 12; line no. 433 - 436)

- Ackerman, A. S., Toon, O., Stevens, D., Heymsfield, A., Ramanathan, V., & Welton, E. (2000), Reduction of tropical cloudiness by soot, *Science*, 288, 1042-1047, <https://doi.org/10.1126/science.288.5468.1042>.
- Rosenfeld, D. (2000). Suppression of rain and snow by urban and industrial air pollution, *Science*, 287(5459), 1793–1796, <https://doi.org/10.1126/science.287.5459.1793>.
- Chuang, C. C., Penner, J. E., Prospero, J. M., Grant, K. E., Rau, G. H., & Kawamoto, K. (2002). Cloud susceptibility and the first aerosol indirect forcing: Sensitivity to black carbon and aerosol concentrations. *Journal of Geophysical Research: Atmospheres*, 107(D21), AAC-10, <https://doi.org/10.1029/2000JD000215>.
- Penner, J. E., Dong, X. Q., & Chen, Y. (2004). Observational evidence of a change in radiative forcing due to the indirect aerosol effect, *Nature*, 427(6971), 231–234, <https://doi.org/10.1038/nature02234>.
- Kaufman, Y. J., Koren, I., Remer, L. A., Rosenfeld, D., & Rudich, Y. (2005). The effect of smoke, dust, and pollution aerosol on shallow cloud development over the Atlantic Ocean, *Proc. Natl. Acad. Sci.*, 102(32), 11207–11212, <https://doi.org/10.1073/pnas.0505191102>.
- Lin, L., Xu, Y., Wang, Z., Diao, C., Dong, W., & Xie, S. P. (2018). Changes in extreme rainfall over India and China attributed to regional aerosol-cloud interaction during the late

20th century rapid industrialization. *Geophysical Research Letters*, 45(15), 7857-7865, <https://doi.org/10.1029/2018GL078308>.

- Zanatta, M., Mertes, S., Jourdan, O., Dupuy, R., Järvinen, E., Schnaiter, M., Eppers, O., Schneider, J., Jurányi, Z., & Herber, A. (2023). Airborne investigation of black carbon interaction with low-level, persistent, mixed-phase clouds in the Arctic summer. *Atmospheric Chemistry and Physics*, 23, 7955–7973, <https://doi.org/10.5194/acp-23-7955-2023>.

**Line 475: Why authors offer BC absorption coefficient profiles at a horizontal resolution of  $1^\circ \times 1^\circ$  and a vertical resolution of 0.5 km. What is the reason behind choosing this particular resolution? Is it possible to have a much higher-resolution dataset in this study?**

The data resolution is set to match the background data horizontal resolution of  $1^\circ \times 1^\circ$ . The background data are chosen to have a vertical resolution of 0.5 km, which matches the vertical resolution of the observational data. It will not be beneficial to attempt for a higher-resolution (than that of the individual datasets used for the assimilation) dataset.