Author response to Reviewer #2 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 #2

This paper attempts to generate 3D composite grid data of BC from data obtained from various observation platforms and estimate the radiative effects of BC considering the vertical profiles. This study potentially provides materials with some implications to improve our understanding of BC over India, which can lead to a solid contribution to ACP. I am afraid, however, that there is critical issues need to be addressed before this paper can be considered for publication.

We appreciate the summary evaluation and the positive comments.

Major comments:

#1

In the earth sciences, assimilation generally refers to the process of integrating different types of observational data into a numerical model in order to improve the accuracy and reliability of model predictions and simulations. Even though the authors are using the mathematical methods used for assimilation, in this study they just combined the various observations together to create composite data. Using the word assimilation in the title and texts is misleading to the reader (and I misunderstood it too). I suggest using a different word.

We appreciate your valuable feedback. However, we would like to retain the title as such because the mathematical framework employed in the present study is similar to the statistical assimilation studies involving numerical models. As such, we believe that the term 'assimilation' could be used in this context as well, which is further in line with the data and methodology followed in Pathak et al. (2020) as well. However, we emphasize that this is different from the dynamical assimilation methods used in weather predication and climate models.

- Pathak, H. S., Satheesh, S. K., Moorthy, K. K., & Nanjundiah, R. S. (2020). Assessment of regional aerosol radiative effects under the SWAAMI campaign–Part 2: Clear-sky direct shortwave radiative forcing using multi-year assimilated data over the Indian subcontinent. Atmospheric Chemistry and Physics, 20(22), 14237-14252, https://doi.org/10.5194/acp-20-14237-2020.

#2

It is difficult to understand the process of creating composite data from the description in the text and Figure 3. First, each of the data used should be described, and then the process shown in Fig.3 should be described carefully and in sequence.

Thank you for the comment. The flow chart has been modified and the revised figure 3 is shown below and in Page no. 26 (line no. 904-906) in the revised manuscript. Each data set and the methodology are explained in Sect. 2, along with the references discussing them in detail.

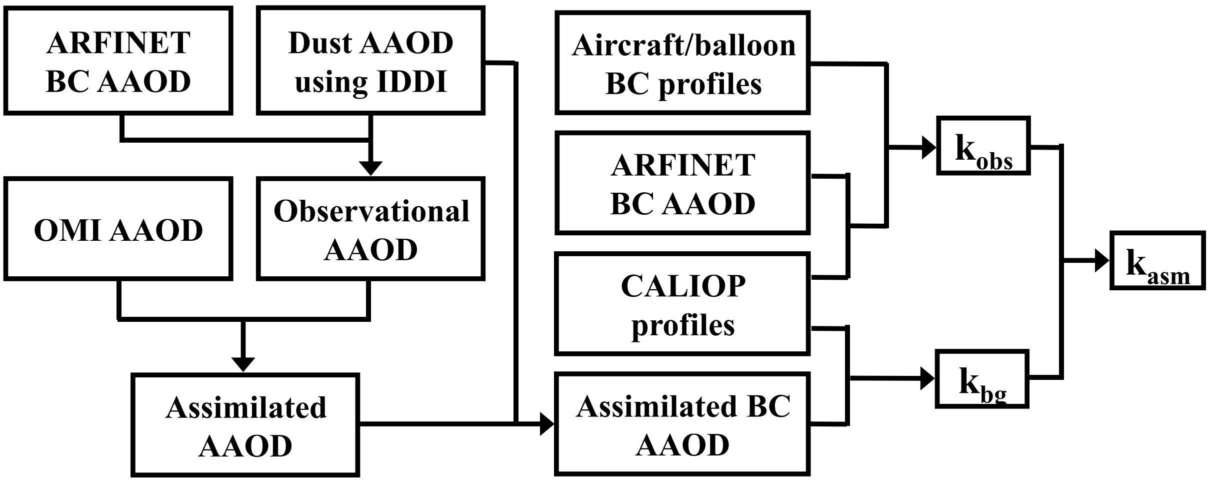

Figure 3. Flowchart describing the various data sets and steps involved in the data assimilation. k_{obs}, k_{bg}, and k_{asm} respectively represent the observational, background and assimilated aerosol absorption coefficients.

#3

The authors are using the assimilation method in the wrong way. Figure 3 shows that common data (ARFINET BC AAOD, CALIPSO profiles) is used to generate both k_obs and k_bg. In other words, k_obs and k_bg are not independent. This does not satisfy the preconditions for maximum likelihood estimation, which is the basis of the variational method. In this case, the covariance between k_obs and k_bg must be taken into account.

Thank you for the comment, which along with the previous comment have been considered to revise the flow chart (Figure 3) to be more comprehendible. It can be seen from the modified flowchart (shown in the previous comment) that k_{obs} is generated using aircraft and balloon profiles measured as part of various campaigns and AFRINET BC AAOD (weighted with CALIOP profiles). On the other hand, k_{bg} is generated merely using the assimilated BC AAOD developed in Pathak et al. (2019) weighted with CALIOP profiles and do not include in-situ measured profiles. Pathak et al. (2019) have used near-surface BC measurements, MERRA-2 planetary boundary layer height, dust AAOD estimated using Infrared Difference Dust Index, and OMI AAOD to generate this assimilated BC AAOD product but the airborne measurement data have not been used . These two different data sets have been used in the construction of background and observational data in the present study. Hence, we believe that the background and observational data can be considered to be independent.

Specific comments:

#1

Line 141: It is very difficult to determine the absorption of dust alone from satellite observations. Please discuss the uncertainty of that and the uncertainty in the BC AAOD obtained.

The uncertainty associated with BC AAOD has been estimated to be varying from 11% to 20% with a mean value of 15% (Pathak et al., 2019). Estimation of dust AAOD using multiple data sets is explained in the second paragraph of Sect. 2.2. and extended in the penultimate paragraph of Sect. 2.4 in the revised manuscript.

#2

Line 223: I am concerned about the very simple method of calculating background error covariance. Because the matrix A is only a deviation from the climatic value not including information of uncertainty of k_bs. Did you not try the method of calculating from the uncertainty of the data sets used to generate K_bg? I also concerned that the simple multiplication of the deviations (i.e., equation (3)) can properly estimated the off-diagonal components (i.e., the covariance in the spatial direction) of matrix B. Have you examined the structure of the covariance closely? This also relates to the advantage of the 3D-Var as pointed out in line 367-369.

Thank you for these comments. The background error covariance can be derived either from climatological data or forecasts (Lewis et al., 2006). We have estimated the covariance matrix from climatological data (Eq. 3) as it effectively captures the spatial covariance structure (Pathak et al., 2019). Due to the relatively short data availability duration, it can be safely assumed that there are no discernible increasing or decreasing trends in the absorbing aerosol loading during the assimilation period. Thus, the deviations in monthly mean BC absorption coefficient across different years is considered as anomalies which are further employed for co-variance estimation. The square root of diagonal elements of co-variance matrix provides the estimates for the uncertainties in the background data at respective locations, while off-diagonal elements signify the cross-covariance values, which provide valuable insights on how the aerosol emissions from a grid influence the neighboring grids, and vice versa. It should be noted here that the present methodology does not employ any external estimates of the uncertainties in the background data.

Following up with reviewer R#2 suggestion, we have carefully examined the spatial covariance (off-diagonal components) within the error covariance matrix. For explanation, we have selected two representative locations, one each from North India and Peninsular India, at an altitude of 2 km amsl, for MAM season. The results, representatives of which are shown in Figure S4 in the supplementary section (Page no. 2 – 3; line no. 12 - 41), indicate that the covariance matrix adequately captures the spatial covariance for nearby locations. In the top panel of Figure S4, around the source region from peninsular India (marked by black diamond symbols), the covariance is high at an altitude of 2 km, indicating that the nearby grids are getting strongly influenced by the source region. Similarly, in the bottom panel of Figure S4, where a source from north India is considered, high covariance is observed over the Indo-Gangetic Plain at an altitude of 2 km. A similar pattern of high covariance is observed for other source locations as well (plots are not shown).

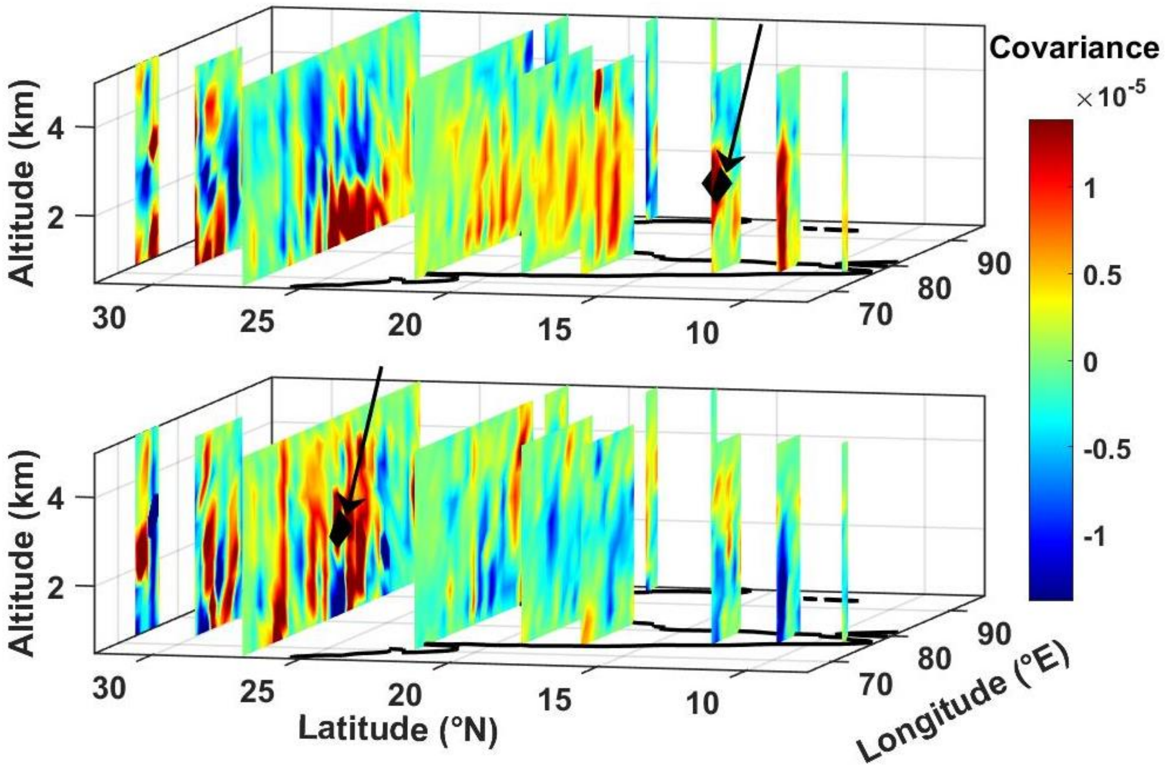

Figure S4. Spatial variation of the covariance between a single grid (marked with black diamond and arrow) and rest of the grids for peninsular (top panel) and northern (bottom panel) India.

- Lewis, J. M., Lakshmivarahan, S., and Dhall, S.: Dynamic data assimilation: a least squares approach, Vol. 104, Encyclopedia of Mathematics and its Applications, Cambridge University Press, Cambridge, 2006.
- 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.

#3

Line 360: The values of Delta_k in Figure 7 show very fine-scale variation, particularly

in MAM. What is the cause of this? As a result, we also see fine-scale spatial variation in k_asm compared with Assimilated BC AAOD (Figure 4). This fine-scale variation is a realistic result?

The assimilation of k_{obs} with k_{bg} has brought the fine-scale variation in the vertical profiles of k_{asm} (because of incorporating realistic/measured profiles in the assimilation process) and this is reflected in δk. Such fine scale variations have been reported over the Indian region in several earlier studies (cited in the manuscript) and their signature in the assimilated BC AAOD data set is a realistic result. In fact, it is an important outcome of the study and fills a long-standing research gap, which would be helpful in improving the climate model simulations over the Indian region.

#4

Line 367-369: In the method of obtaining from deviations as in equation (3), apparent correlations may appear, especially between grids separated by a distance. Have you examined about this?

Thank you for pointing it out. We have examined this and observed that a noticeable apparent correlation exists between the grids over western India and the Indo-Gangetic plain during the MAM season. This correlation could be attributed to a composite effect resulting from long-range dust transport as well as strong BC emissions occurring over the entire Indian region. However, after considering the absence of such a prevalent correlation in other regions and seasons, we have determined that its impact on the assimilation process is not expected to be detrimental.

#5

Line 387-394: It is obvious that K_asm is more consistent with K_obs than K_bg; the discussion using Fig.8 makes no sense. If authors want to verify Kasm, authors should do so with independent data.

We acknowledge the significance of validating k_{asm} (assimilated data) using independent sources. However, it is important to note that in our study, the availability of k_{obs} (observed data) for validation purposes is limited. The scarcity and sparsity of k_{obs} pose serious challenges in reserving a specific subset solely for validation, as it would largely reduce the already limited data available for assimilation. Considering these constraints, our intention in Figure 8 was to investigate the extent to which the assimilation process improved the correlation between k_{bg} (background data) and k_{obs}. Nevertheless, we appreciate the reviewer suggestion to focus on validating k_{asm} using independent k_{obs}. As such, we have performed the validation of k_{asm} against independent k_{obs}. Among the 35 stations, we utilized k_{obs} data from 25 stations for assimilation and reserved k_{obs} data from 10 stations for validation. To ensure statistical significance and an adequate number of data points in the analysis, the validation datasets from all three seasons were consolidated. The results from this analysis are shown in Figure RC2. It can be observed that after assimilation, using the new data subsets, k_{obs} still aligns more closely with k_{asm} as compared to k_{bg}, highlighting the robustness of our methodology.

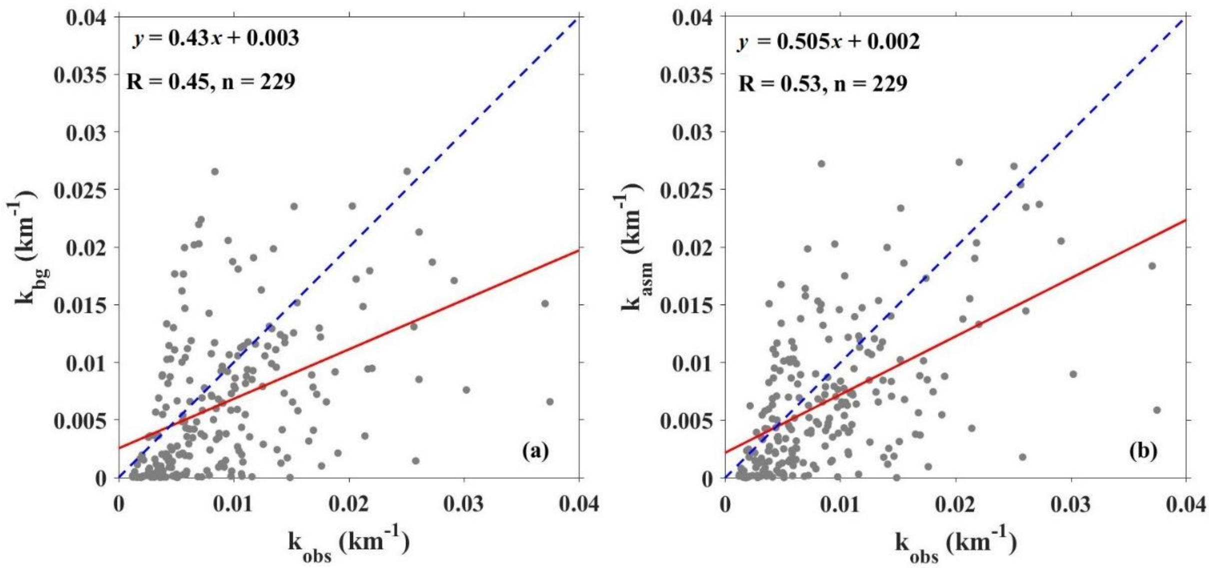

Figure RC2. Scatter plots between profiles of (a) k_{obs} and k_{bg} and (b) k_{obs} and k_{asm}. The red line denotes the linear fit, the dashed blue line denotes the 1:1 line, and the scatter points are shown in gray. The equation of fit, correlation coefficient (R), and the number of scatter points (n) are shown in each sub plots.

#6

Line 413-425: Very interesting point. If there are any previous studies that make similar points, please include them.

Thank you. The vertical profiles of aerosol single scattering albedo and the possibility of obtaining more realistic aerosol radiative forcing profiles were shown in Vaishya et al. (2018) and Manoj et al. (2020) using aircraft measurements (limited temporally and spatially to smaller domains as compared to the present study) and are already cited in the manuscript.

- Manoj, M.R., Satheesh, S.K., Moorthy, K.K. and Coe, H.: Vertical profiles of submicron aerosol single scattering albedo over the Indian region immediately before monsoon onset and during its development: research from the SWAAMI field campaign. Atmospheric Chemistry and Physics, 20(6), pp.4031-4046, https://doi.org/10.5194/acp-20-4031-2020, 2020.

- Vaishya, A., Babu, S. S., Jayachandran, V., Gogoi, M. M., Lakshmi, N. B., Moorthy, K. K., and Satheesh, S. K.: Large contrast in the vertical distribution of aerosol optical properties and radiative effects across the IndoGangetic Plain during the SWAAMI–RAWEX campaign, Atmospheric Chemistry and Physics, 18, 17669-17685, https://doi.org/10.5194/acp-18-17669-2018, 2018.

#7

LIne437-450: Were you able to find any traces of self-lofting in this data set?

No, the self-lofting of aerosols occurs due to processes associated with relatively lower time scales as compared to the time intervals used in this study to generate the assimilated data set.

#8

Line426-470: Although related to the need for an accurate vertical profile of BC, it is mostly redundant as it is mostly a description of previous studies. The description should be more concise in conjunction with the results of this study.

Complied with.