Responses to comments from Anonymous Referee #1

June 10, 2019

We are thankful towards both the reviewers for their constructive comments and suggestions which not only provided the deeper insights into the present work but also helped to improve the overall quality of the manuscript. The responses to the specific comments/suggestions from the Anonymous Referee #1 are as follows. The referee comments are shown in red color and our responses are shown in black color.

1 Answers to comments from Anonymous Referee #1

Comment 1: For such comparative estimation a higher resolution data from satellites such as level 2 data available almost daily be ideal, while level 3 data at a lower resolution on a monthly basis has been reported to be systematically underestimated.

We agree with reviewer that level-2, daily data would be more appropriate for construction of assimilated product. However, level-2, daily satellite data often consists of large data gaps, primarily due to clouds. This issue is even more prominent in case of retrievals using space-borne measurements by Ozone Monitoring Instrument (OMI) which has coarser resolution (13 km $\times$ 25 km) than that of MODIS. In addition, the ground-based AOD measurements from ARFINET can be performed only in clear-sky conditions, which limits the availability of ground-based AODs at daily time scale.

In order to improve the regional climate impact assessment for aerosols, one needs to construct quality improved, spatially homogeneous and temporally continuous datasets for aerosol properties, which is also envisioned in South West Asian Aerosol Monsoon Interactions (SWAAMI), a joint Indo-UK field experiment. In accordance with this, we have employed level-3, monthly satellite products, as background datasets from which wide and continuous spatial coverage is inherited by assimilated products.

Comment 2: Comparison between AODs and AAODs are expressed in terms of R, R2 has used to explain such variation. R values lower

We agree with reviewers observation about the correlation between merged AAODs and independent ground-based AAODs (R) being low yet significant (at 95% confidence level). However, there are genuine reasons behind the same which are as explained below.

The merged AAOD (MG AAOD) product is developed by systematically assimilating Ozone Monitoring Instrument (OMI) retrieved AAODs and those estimated from ground-based BC measurements as well as satellite-based infrared measurements, employing 3D-VAR, a widely used assimilation technique based on weighted least square error minimization. The OMI AAODs which form the background data for the AAOD assimilation are demonstrating relatively weaker yet significant correlation (R=0.36) with independent ground-based AAODs (GR AAOD) (Page no.16, figure 3a). Such weak correlation between OMI and GR AAODs could be primarily because of differences in the estimation procedures. OMI-near UV (OMAERUV) algorithm which is employed for retrieval of AOD and AAOD, makes use of the measurements of the upwelling radiation at 354 and 388 nm at the top of atmosphere (TOA). This algorithm exploits the prominent interaction between molecular scattering and the aerosol absorption as well as lower surface reflectance in UV wavelength range. The AOD and AAODs are further retrieved using the look up tables (LUT) consisting of pre-computed reflectance values (at the TOA) derived by a set of aerosol models which consider specific vertical distribution for each of the aerosol types (Torres et al., 2005, 2007). For carbonaceous aerosols, those models consider exponential profile with maximum concentration occurring at 3 km above ground level. On the other hand, during estimation of AAODs corresponding to surface level BC mass concentration, we have considered uniform distribution of BC within the PBL and exponential decay above it. Being based on the common inferences drawn from the extensive aircraft and balloon measurements of BC over different regions of India (Suresh Babu et al., 2010; Babu et al., 2011), the vertical distribution considered in our work is better representative for the Indian region. This difference between vertical distribution of aerosols considered during estimation of OMI and GR AAOD could have lead to weak correlation between the two. In addition, the uncertainties in OMI AAODs emanating from assumptions about height of an aerosol layer and sub-pixel cloud contamination could have further assisted in reducing the correlation between OMI AAOD and their ground-based counterpart. This weak correlation between OMI and GR AAODs could be one of the primary reasons behind the observed correlation between merged and GR AAODs (R = 0.47, figure 3b, page no 16 from the earlier version of manuscript). As suggested by reviewer, R^2 values are also now included in figure 2 and 3 from the earlier version of manuscript,

As described by 3D-VAR, the weights given to each parent dataset are inversely proportional to the uncertainties in the respective datasets (Kalnay, 2003; Lewis et al., 2006). Accordingly, GR AAODs are inversely weighted with respective uncertainties (specified in equation 14, page no. 15 of earlier version of manuscript), which certainly limits the signature of ground-based measurements in assimilated AAODs. This factor also could have further restricted the correlation between merged AAODs with its ground-based counterpart. In spite of this, the point to be highlighted here is that the correlation shown by merged AAOD with independent GR AAOD (R = 0.47, figure 3b, page no 16 of earlier form of manuscript) is about 30% higher than that shown by OMI AAOD (R = 0.36, figure 3a, page no 16 of the previous version of manuscript), which underlines the substantial improvement brought in due to assimilation.

Comment 3 dAOD and dAAOD values are large, sometimes as equal to AOD or AAOD

We agree with this observation made by reviewer that dAOD (i.e. MG AOD - SR AOD) and dAAOD (i.e. MG AAOD - SR AAOD) values are sometimes as large as AOD and AAODs shown by respective satellite products.

The merged AOD and AAODs are constructed by systematically assimilating GR AOD and AAODs with corresponding satellite products employing data assimilation schemes as explained in section 3.1. Therefore the dAOD and dAAOD values are primarily associated with differences between corresponding satellite retrievals and ground-based measurements. The MODIS retrieved AODs tend to be underestimated as compared to AERONET AODs over highly polluted, smoke covered regions (Zhang and Reid, 2006). However, cloud contamination leads to systematic overestimation of MODIS AODs irrespective of AOD ranges (Zhang and Reid, 2006). This issue is even more prominent in case of OMI AAODs which tend to be overestimated due to sub-pixel cloud contamination (Torres et al., 2005, 2007). In addition, the assumptions regarding vertical distribution of absorbing aerosols made by the aerosol models used in OMI-near UV (OMAERUV) algorithm, can also lead to OMI AAODs being substantially different from their ground-based counterpart. The difference between satellite retrieved and ground-measured values, is the main reason behind the kind of dAOD and dAAOD values shown in our work.

However, the validation exercise (section 4.1) has demonstrated that the assimilated products are better confirming with independent ground-based measurements than their satellite counterparts, which indicates the improved accuracy of assimilated products vis-a-vis respective satellite datasets.

Comment 4 The uncertainties are reported to be lower than satellite product, how and why this happens is not clear.

We agree that the explanation about the uncertainty reduction for assimilated products is not directly given in the earlier version of the manuscript.

We would like draw referee's kind attention towards that fact that uncertainties in assimilated AOD and AAODs are guaranteed to be lower than those in respective satellite products, as guaranteed by the assimilation techniques employed, Weighted Interpolation Method (WIM, used for AOD assimilation) and 3D-VAR (used for AAOD assimilation). Theoretical proof for property of variance minimization for WIM is given in section S2 in the supplementary material (earlier version). Although we did not provide the similar proof for 3D-VAR, the appropriate references (Kalnay, 2003; Lewis et al., 2006) for the same were given in the earlier form of manuscript (line no 13-15, page no. 15). Nonetheless, in the modified version of the manuscript, we are including the the proofs for variance minimization property demonstrated by WIM and 3D-VAR in section 3.

Comment 5 Using BC mass and dust to construct SSA can result in uncertainty, for example, the aethalometer measurements are reported to have uncertainties in BC mass measured at different environmental conditions using the same attenuation coefficients valid for urban regions.

We agree with this important point raised by reviewer about sources of uncertainties in SSA emanating from those in measurements of BC mass and dust. The uncertainties in BC mass concentration measurements made by Aethalometer, are reported to be 2 to 5 % (Hansen and Novakov, 1990; Babu et al., 2004; Dumka et al., 2010). In the present work, we have considered 5% uncertainties in BC mass concentration measurements while estimating the uncertainties in BC AAODs, as mentioned in line 30, page no. 14 of the earlier version of manuscript. The details regarding estimation of uncertainties in BC AAODs are provided on line no. 26-30 on page no. 14 and line no. 1-3 on page no.15 of the earlier form of manuscript. Similarly, the uncertainties in dust AAODs emanating from vertical heterogeneities in dust and its optical properties are estimated to be 25%, as mentioned on line no.4, page no. 15. The composite variance in GR AAODs (i.e. BC AAOD + Dust AAOD) estimated by equation 14 (as given on page no. 15 of earlier version of manuscript) was used to form diagonal elements of observation error covariance matrix (R) used in 3D-VAR (equation 13 in earlier form of manuscript).

Thus, the uncertainties in BC mass measurements and dust, were taken into account while constructing assimilated AAODs which are further used along with assimilated AODs to estimate SSA (equation 17 on page no. 22 in earlier form of manuscript). The uncertainties in SSA are then estimated from those in MG AODs and AAODs, as described in line no. 13-15, page no. 23 from the earlier version of manuscript. Thus, the uncertainties contributed by BC and dust AAODs are being taken into account to estimate those in SSA.

Comment 6 As far as the methodology is concerned several approaches are mentioned with variations in planetary boundary layer height etc., sensitivity of these assumptions not explained.

We understand the concern raised by the reviewer regarding robustness of assumptions which is usually examined through sensitivity analysis. However, we would like note that, variance in planetary boundary layer height (PBLH) data is not assumed, rather estimated using long-term time series (year 2000-2013) of PBLH data provided by MERRA-2 reanalysis product. In order to do so, we have constructed error covariance matrix using monthly mean, MERRA-2 PBLH after removing long-term trend (if existing) and seasonal variation. This covariance matrix is constructed employing the methodology explained in section S3.1, page no. S6 from the earlier version of supplementary material. The diagonal terms of the covariance matrix provide estimate for the variance in PBLH for the corresponding grid points, which we have considered in the subsequent calculations. Therefore, we would like to kindly note that the prescribed sensitivity analysis is not warranted.

We understand that this was not explained in the earlier version of manuscript due to which it was seeming that the variance in PBLH is assumed, which is not the case. In the view of this, we are including these details in the modified version of supplementary material.

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