Responses to comments from Anonymous Referee #2

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 #2 are as follows. The referee comments are shown in red color and our responses are shown in black color.

1 Answers to major comments from Anonymous Referee #2

Comment 1: Page 8: Line 3. It was mentioned that PBL height is used from MERRA-2 and it has been validated previously by Sathyanadh et al. (2017). Though Sathyanadh et al. (2017) mentioned that good correlation between 0.74-0.83 is seen when MERRA-2 PBLH when compared with radiosonde and radio occultation (done for very few stations that too for one year 2011 only), our experience is that it underestimates heavily the PBLH. Since this is the one of important parameter while calculating AAOD, it is suggested to show detailed comparison of MERRA-2 with existing IMD radiosonde derived PBL heights or GPS RO measurements for the said period 2008-2016. A figure showing the PBL altitudes for the respective stations will be highly useful while interpreting the results particularly the AAODs.

We are thankful towards the reviewer for correctly pointing out that the validation of planetary boundary layer height (PBLH) by MERRA-2 dataset, provided in Sathyanadh et al. (2017) is limited in terms of duration (May to September 2011). Therefore, in the view of importance of accurate PBLH for our studies, it is pertinent to validate PBLH provided by MERRA-2 over the Indian region for the entire duration. Accordingly, we have validated the MERRA-2 PBLH for the duration of 11 years (2008 to 2018) with those estimated using radiosonde measurements (downloaded from *http://weather.uwyo.edu/upperair/sounding.html*) over the Indian region. However, due to unavailability of continuous radiosonde measurements over many of the locations of ground-based ARFINET and/or AERONET stations, we have considered radiosonde measurements from 8, subregional representative locations (Figure 1), the Aerosol Optical Depth (AOD) and Black Carbon (BC) measurements from which are employed for constructing assimilated products. The details regarding lat-lon coordinates and broad geographical features for these stations are provided in the Table S1 and S3 from the supplementary material, along with other ARFINET and AERONET stations, data from which is used for the assimilation study.

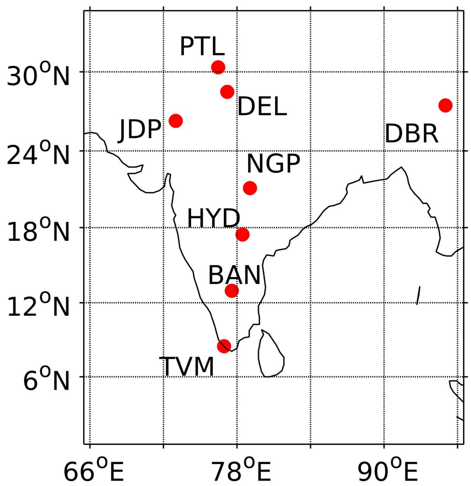

Figure 1: Locations of the ground stations, radiosonde measurements from which are used for the purpose of validating PBLH derived by MERRA-2. These subregional representative stations form a subset of ground-based observatories, AOD and BC mass concentration measurements from which are employed for construction of assimilated AOD and Absorption AOD (AAOD) products.

The radiosonde measurements at these stations (figure 1) are usually performed twice a day, at 00 GMT and 12 GMT and provide vertical distribution temperature, pressure, relative humidity. Further, these fundamental thermodynamic fields are used to derive the vertical profiles for virtual potential temperature (θ_v), which are also provided in the respective data files.

In order to estimate PBLH from the radiosonde data, we have computed the gradient in the virtual potential temperature ($\triangle\theta_v$) at each given altitude. The height (above surface) at which the $\triangle\theta_v$ exceeds 3 $^{\circ}$k km^{-1} is considered as PBLH (Kompalli et al., 2014; Nair et al., 2011) at that location. The planetary boundary layer is likely to be deeper during daytime vis-a-vis nighttime, due to stronger solar heating during the day. Due to this, shallower PBL occurring in the early morning (00 GMT) may not be always captured with the provided radiosonde profiles. In the view of this, we have employed PBLH estimated using radiosonde measurements during daytime (12 GMT) only, for the present validation purposes.

The hourly averaged PBLH (12 GMT) given by MEERA-2 for that particular day, are bi-linearly interpolated to the locations of stations shown in figure 1, in order to get spatio-temporally collocated estimate of MERRA-2 PBLH. The scatter plots between the collocated PBLH and those estimated from radiosonde measurements for 8 locations, during year 2008 to 2018, are presented in figure 2.

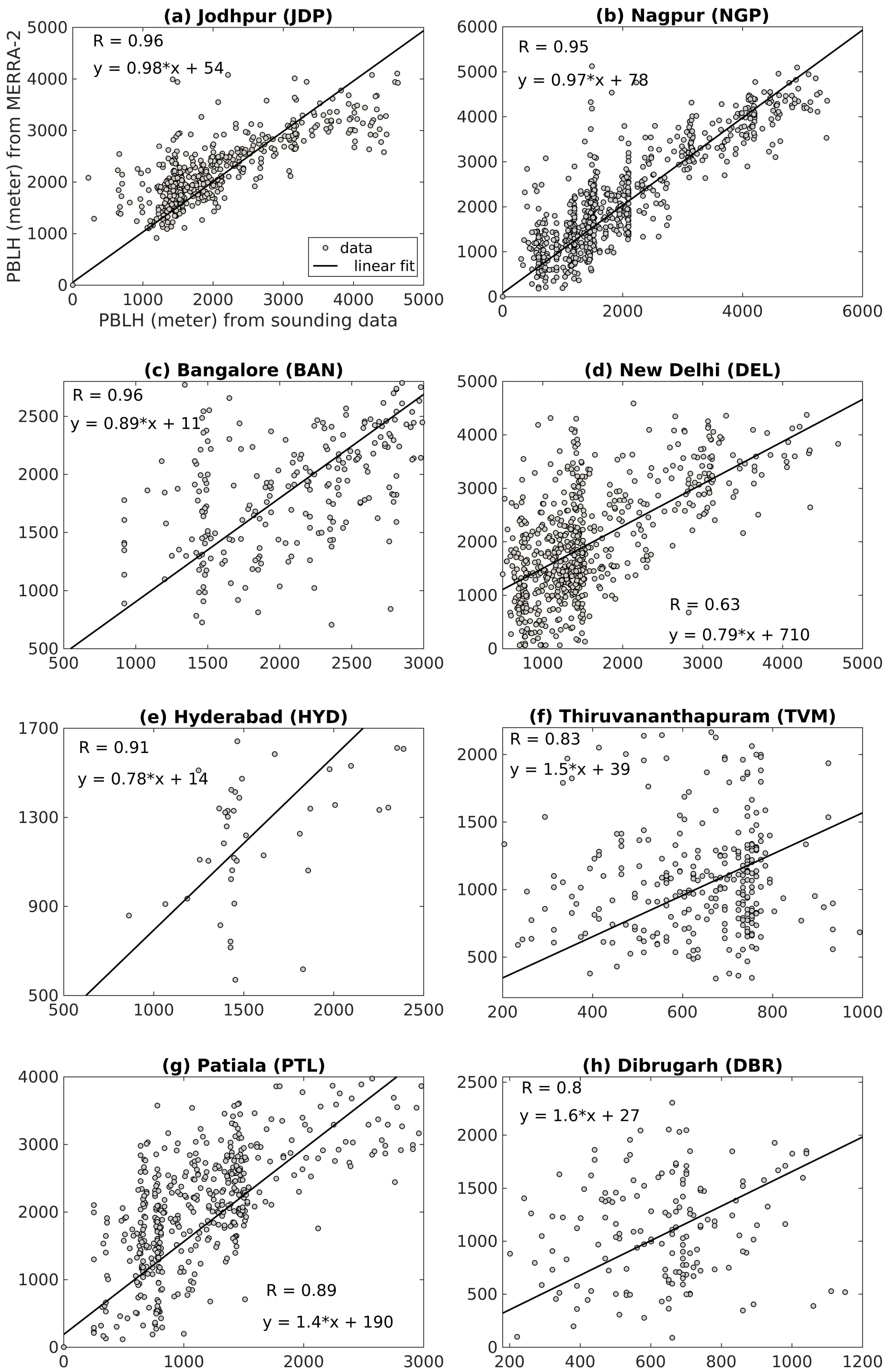

Figure 2: Comparison of spatio-temporally collocated MERRA-2 PBLH with those derived from radiosonde measurements performed at 8 representative locations during year 2008 to 2018. The correlation coefficient (R) (significant at 95% confidence limit) and the equation of linear regression between the two PBLH estimates are provided in each of the figures.

It can be seen from figure 2 that, PBLH provided by MERRA-2 dataset are well-correlated with those estimated using radiosonde data, although the correlation coefficient is varying from 0.63 to 0.96, w.r.t the location. The equations for linear regression between the two PBLH estimates suggest that, PBLH given by MERRA-2 are underestimated over majority of the stations (Figure 2a to 2e), which is in line with the general observation made by reviewer. Nonetheless, substantially overestimated PBLH values by MERRA-2 are apparent for some of the stations (Figure 2f to 2h).

We agree with the reviewer that, in order to enhance the accuracy of AAODs estimated from ground-based BC mass concentration measurements, one would use PBLH values derived from radiosonde data. However, due to limited temporal sampling (daily 2 profiles only) and unavailability of radiosonde measurements at every location of ARFINET observatory (34 in number), we had to rely on the PBLH product provided by MERRA-2 reanalysis dataset, which is well correlated with observations. As suggested by the reviewer, we are adding figure 2 and its pertinent explanation in the modified version of manuscript.

Comment 2: Validation with independent measurements of AAOD: I am surprised to very poor correlations in AAOD shown in Figure 3. Since correlations are poor, how to trust the data for further applications. Perhaps need to be re-checked while using actual PBL heights.

We agree with the observation made by referee about the correlation between merged AAODs and independent ground-based AAODs which is slightly 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 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 AAOD 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 from the previous version of manuscript), which underlines the substantial improvement brought in due to assimilation.

Comment 3: I suggest adding another panel in Figures 4-5 showing the difference between SR AOD and MG AOD along with dAOD. I do not understand why the difference is not shown throughout the Indian region similar to that shown for dAAOD in Figures 6-7.

The difference between the MG and SR AOD which is indicated by dAOD (i.e MG AOD - SR AOD) is shown in Figure 4c and 5c (page no. 18 and 19 from earlier version of manuscript). Similarly, spatial variation of dAAOD (i.e. MG AAOD - SR AAOD) is demonstrated in Figure 6c and 7c (page no. 18 and 19 from earlier version of manuscript), for the two representative cases. As can be seen from Figure 4c and 5c that non-zero dAOD values are being demonstrated over the regions represented by ground-based AODs and they (dAOD values) smoothly reduce to zero as one moves away from the locations of ground-based observatories. On the other hand, Figure 6c and 7c are showing dAAODs having wider spatial coverage The differences in the nature of regional distribution for dAOD (Figure 4c and 5c) and dAAOD (Figure 6c and 6c) are primarily due to difference between nature of assimilation methods employed for AOD and AAOD.

As detailed in section 3.1 (earlier version of manuscript), for AOD assimilation, we have employed Weighted Interpolation Method (WIM) (a variation to the Successive Correction Method) which ensures that MG AODs are always bounded by SR and GR AODs and are guaranteed to have less uncertainties than those in SR AODs (section S2 from the earlier version of supplementary material). WIM provides a localized approach towards assimilation and merges the GR AODs with SR AODs at the grid points within specified radius and height of influence. Nevertheless, for AAOD assimilation, WIM could not smoothly merge GR AAODs with the gridded background data (i.e. SR AAODs), possibly due to the relatively weaker correlation between the two (R = 0.36, Figure 3a from the earlier version of manuscript) vis-a-vis GR AOD and SR AOD (R = 0.77, Figure 2a from the earlier version of manuscript). In the view of this, we have employed, a widely used data assimilation technique, 3D-VAR, which merges the scattered observations with the gridded data in the patterns dictated by the background error covariance matrix (B). In the present work, B is constructed employing long-term time series (year 2005 to 2016) for the monthly OMI AAODs over the Indian region (section S3 from the earlier version of supplementary material). Due to this, the differences between MG and SR AAODs are being seen over entire Indian region (Figure 6c and 7c), unlike in case of AOD where the differences between the merged and satellite product are prominent over the regions represented by ground-based AODs.

Comment 4: In the abstract it is listed as 44 stations for AOD and 32 stations for AAOD. However, I am unable to see them in the list of stations provided in the supplementary information.

Table S1 and S2 from the supplementary material (previous version) provide the lists of 27 ARFINET and 20 AERONET stations, AOD measurements from which are employed for assimilation purpose. As three locations are common among ARFINET and AERONET, the total tally of stations reduces to 44. The list of 34 ARFINET stations providing BC mass concentration measurements, is given in Table S3 from earlier version of the supplementary material.

2 Answers to minor comments from Anonymous Referee #2

Comment 1 Page 2: Line 21: remove repeated word have

This rectification has been incorporated in the modified manuscript.

Comment 2 Figure 1: dot size used in this figure is too small to recognize different colors. Size should increase up to 3-4 times similar to that shown in Figures 4-7

We have modified the Figure 1 demonstrating locations of all ground-based stations, data from which is used in the current work. The modified form of Figure 1 is incorporated in the updated version of manuscript.

Comment 3 I suggest moving Figure 9 to supplementary information as the regional coordinates are already mentioned in Table 1. This figure is not adding much.

The suggested change has been implemented in the modified form of manuscript.

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