Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2019-731-RC2, 2019

Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Space-time variability of ambient $PM_{2.5}$ diurnal pattern over India from 18-years (2000–2017) of MERRA-2 reanalysis data" *by* Kunal Bali et al.

Anonymous Referee #2

Received and published: 20 December 2019

Air quality is an important environmental concern. It is more important for countries like India where ambient PM levels are above air quality standard limits. There is a need to understand the temporal and spatial pattern and the sources of pollution over India to take necessary measures.

Studies over India lacks long term analysis of PM2.5 at regional scale. This study claims to present the first space-time variability of ambient PM2.5 diurnal pattern in India for an 18-year (2000-2017) using the bias corrected MERRA2 data. While the objectives of the paper are interesting, the results presented in the paper can be highly uncertain.

Extensive description and evaluation of the MERRA2 Aerosol reanalysis products (1980 onwards) have been presented by Randles et al., (2017) and Buchard et al (2017). In addition to this, Buchard et al (2017) also presented some case studies. Both studies point out and conclude that caveats that must be considered when using this new reanalysis product for future studies of aerosols and their interactions with weather and climate. I am sure this applies Air quality studies as well.

After reading the present manuscript, it gives me impression that authors have not fully understood how the MERRA2 aerosol products has been created, what are the limitations and whether it can be used to address the objectives of the paper. This assessment is in line with the assessment made by referee #1. Following concerns can be addressed before it is accepted for publication

1. Emission annual trend

Firstly, emissions are an important factor to study the spatial and temporal variability and trend. It is important to understand in detail the spatial and temporal scale of the inventories used in the simulation. Authors, in the paper as well as pre-review response, has mentioned that the MERRA-2 products use anthropogenic (EDGARv4.2) and biogenic sulfate (AeroCom Phase II) and carbonaceous aerosols (scaled RETROv2). This gives the impression that most of the anthropogenic emissions are from EDGARv4.2 which is normally available until 2012. However, only anthropogenic SO2 is used from EDGARv4.2 and that is from 1980-2008. The exhaustive list is given in Table 1 of Randles et al., (2017) and also discussed in Randles et al., (2016) where most of the anthropogenic emission is from AeroCom Phase II from 1980-2006. Moreover, Buchard et al (2017) has also mentioned that MERRA-2 anthropogenic emissions vary on a yearly basis, and emissions databases do not extend to 2013 (e.g., 2006 and 2008 are terminal years for anthropogenic OC/BC and SO2 databases, respectively) and same emission is repeated after 2006/2008 until recently Randles et al., (2016).

Therefore, when the terminal years for the anthropogenic emissions are 2006/2008 and constant afterward, can it be used for trend analysis? I am sure one must be very cautious using this data to derive the trend up to 2017.

2. Emissions grid resolution

The native resolution of most of the emissions used for the MERRA2 simulation is over 1deg x 1deg resolution (other than biomass). These emissions datasets are re-gridded to the native model grid. How it could impact the analysis of the paper can be discussed.

3. Hourly analysis

The authors presented the analysis on the hourly scale. The validity of the simulated hourly scale surface PM2.5 concentration lies in the fact how well the meteorology is simulated and the diurnal/hourly profiles used to process the emission. As far as I understand, the seasonal cycle is used (Figure 2.2 of Randles et al., 2016) to speciate annual emission to monthly emissions. There is no mention of diurnal cycle, therefore I assume that no diurnal profile is used for MERRA2 simulations. Moreover, the analysis and comparison of surface PM2.5 across US with MERRA aerosol products presented by Buchard et al (2016 and 2017) were not presented at hourly scale. When the MERRA2 data has been used for hourly scale, then columnar products are used rather than surface products (section 4d of Buchard et al 2017). In a comparison of MERRA2 PM2.5 with surface PM2.5 over North China by Song et al., (2018) has shown that MERRA-2 cannot follow the diurnal variation of PM2.5 but reproduce a good daytime variation of AOD. In this case, once should be concerned about the validity of the results presented at hourly scale.

4. Calculation of PM2.5 from MERRA2.

Authors calculated the PM2.5 by adding up dust and sea-salt in size bins smaller than 2.5m, hydrophilic and hydrophobic OC, BC and sulfate (assuming the entire load is within PM2.5). However a different formula is used in Buchard et al (2016) and Song et

al., (2018). The mass of sulfate is multiplied by 1.375 and OC is multiplied by a factor between 1.2 and 2.6. Authors can comment on why they used unit factor of sulfate and OC in their calculation. Moreover, the SOA ,which dominates in IGP and missing Nitrate aerosols can also be discussed. Also, a larger overestimation of dust and sea salt in MERRA2 (Buchard et al) can be discussed.

5. CPCB PM2.5 data.

Authors have now provided the list of monitoring stations (Table 1 of suppl material). However the manuscript lacks the description of the CPCB PM2.5 data used in this study. Authors need to provide more information about the methodology/technique/instrument used for the measurement of ambient PM2.5. They need to provide the environment type of each station in table 1, whether they are urban, rural, traffic or background sites. As authors have mentioned that the PM2.5 monitoring started in India by the Central Pollution Control Board (CPCB) in 2008-2009, so all the stations will not have continuous measurements from 2009-2017. Therefore, authors also need to provide the period of valid measurement available and missing period if any. If some stations have continuous measurements from 2009-2017, then there is a chance that the monitoring instrument might have changed. They can mention whether the instrument/technique has changed and how the data has been inter-calibrated. As far as I am aware, CPCB provides the data as measured from the instrument without any quality control. One must do quality control before using the data. Authors may also provide the steps of quality control.

6. Use of CBCP data for this study

To the best of my knowledge and the locations provided by the authors in the table 2. It can be confirmed that most of the stations (it appears that more than 90%) of the stations are in Urban areas. Buchard et al (2016 and 2017) have restricted the analysis of PM2.5 across US over suburban and rural sites because PM2.5 concentrations are generally higher and less uniform in urban areas, such stations are not representative of the grid-box mean values that MERRA estimates. In this case, I doubt that CPCB urban data is suitable for comparison and bias correction.

7. MERRA2 PM2.5 evaluation and bias estimation

The detailed evaluation of MERRA2 PM2.5 has not been presented in the paper other than mean diurnal plot. Before going for bias correction, it is important to know the temporal and spatial biases in the model. A detailed statistical evaluation has to be presented. The evaluation can be presented for a limited period when most of the data is available. Please refer Song et al., (2018) https://doi.org/10.1016/j.atmosenv.2018.08.012

8. Bias correction methodology.

Although MERRA2 aerosol reanalysis products are better than non-assimilated products, it can have biases, therefore it was calibrated across India (spatially) and during 2009-2017 (temporally) using the CPCB data measured at 80 sites mentioned in supplementary material table 1. To obtain the collocated CBCP and MERRA2 PM2.5, authors have either averaged all CPCB sites within a MERRA-2 grid ($0.5^\circ \times 0.625^\circ$) OR re-grid the MERRA-2 data from 0.5 x 0.625 degree resolution to 0.05 x 0.05 degree resolution and then extracted the PM2.5 values at CPCB coordinates (as per reply to the pre review comments). Authors use 50% CPCB data for bias correction and 50% for validation. Please clarify how the 50% data was selected, was it random or continuous.

First, authors need to address the issues related to CPCB data quality, its availability during 2009-2017 and its spatial representativeness as most of them are in Urban area and are within the same grid. Second, the bias correction methodology needs further clarification as it seems as per the manuscript that authors do two types of bias correction (or calibration). One for in-situ 80 sites and another for the Indian grid. For 80 sites, authors obtain a linear relation between MERRA2 and CPCB PM2.5 and then get the calibration factor as a function of CPCB PM2.5 which is then added to MERRA2

to correct it. (Line 164-171). For Indian grids, authors calculate the calibration factor as a function of MERRA2 2.5 value. To find out the linear regression, authors have binned the data in 500 bins (0-500 ug/m3) (in this way the data becomes independent of the time and location).

For the linear relation used for 80 sites, authors get a liner line with a slope of 0.228 between CPCB and MERRA2. This shows that there is a huge underestimation of MERRA2 PM2.5 most probably because of the use of Urban PM2.5. It is even more surprising liner line between bias (CPCB-MERRA2) has a slope of 0.772. It can be interpreted that model bias has a better correlation then the model estimate. And if the model bias is more than the model estimate then one must rethink before using this data for further analysis.

Finally, the authors find a bias-corrected relation BCM=0.99*CPCB+0.005. Rounding off and further simplification, this equation reduced to BCM=CPCB. It means, all the MERRA2 values are replaced by CPCB values. In this way, authors will certainly get good correlation (0.94) between bias-corrected MERRA2 and validation CPCB PM2.5. Authors can check and report the correlation between validation and the data used for bias correction. By using this methodology, you are overfitting the MEERA2 data. I don't think this is the right way to do the bias correction. There are several papers on bias correction methodology that authors can refer to.

9. Overall comment I have no further comments on the rest of the analysis as it depends on how good is the bias-corrected MERRA2 data. As the moments it appears that 1. MERRA2 PM2.5 may not be suitable for hourly analysis. 2. MARRA2 PM2.5 can not be used for trend analysis because of constant emissions after 2008. 3. MERRA2 aerosols are not suitable for Urban PM2.5 analysis. Because of this, The MERRA2 model bias is more than the MERRA2 estimates. This suggests urban PM2.5 should not be used for bias correction. 4. A robust method of bias correction is required.

Hence, I am less confident that the results presented in the paper are valid. This is important because the authors claim that these results will help formulate better air pollution mitigation plans by evidence-based policy actions at the regional and national levels. I feel that authors need to be extra cautious and discuss the limitations before publishing these kinds of results. At the moment, it would be appropriate the perform a detailed evaluation of the MERRA2 products over India and present the biases and uncertainties across different temporal scales and geographical regions of India.

10. Some of the minor suggestions

Use either bias corrected MERRA (BCM) or calibrated uniformly. This paper has not been referred: Central Pollution Control Board (CPCB) Ambient air quality statistics for Indian metro cities, Central Pollution Control Board, Zonal Office, Bangalore, 2003. Authors can discuss India specific assimilation used for MERRA2 in detail as indicated by referee#1.

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2019-731, 2019.