## Reply to Reviewer #2

We are thankful to Reviewer #2 for his/her comments and suggestions.

The comments from Reviewer #2 are in italics followed by our responses:

## Specific comments:

1. In section 3.1, authors evaluated model simulated AOD using MODIS derived AOD and found that model underestimate the AOD by a factor of 2. Model is not able to capture the high AOD belt over Indo-Gangetic Plain (IGP) and most of northern parts of India (Figure 1). Dust transport from West Asia (MODIS AOD) is also not captured in the modelled AOD (Figure 1). Authors need to check dust emission flux to figure out how well model able to simulate dust source regions. A brief discussion about species-wise AOD information could be useful for explaining the underestimation of model simulated AOD.

We agree with the reviewer that emissions may contribute to the underestimation of AOD. For the dust emissions used in this study, it is described in the Section 2.1, Lines 18-19, on Page 16907,

"Dimethyl sulfide, dust, and sea salt emissions are calculated online as for the GOCART model (Ginoux et al., 2001; Chin et al., 2002)".

And also, this dust emission scheme, which is implemented in the WRF-Chem (MOZCART) that we use, has been evaluated extensively over India by Kumar et al. (2014). Therefore, we cited that paper in the Section 2.1, Lines 3-5, on Page 16907,

"This version of the WRF-Chem aerosol and chemistry modules has been used and evaluated in studying effects of dust aerosols on tropospheric chemistry during the pre-monsoon season in northern India (Kumar et al., 2014)"

Other factors such as meteorological conditions (such as relative humidity, boundary layer dynamics and soil moisture in the case of dust) could also contribute to the AOD underestimation in addition to emissions. However, a comprehensive analysis of model schemes and boundary conditions is required to provide such information. We think that it is beyond the scope of this paper. This limitation and the main goal of the current study are given on Lines 10-13, Page 16922,

"Resolving the mismatch between simulated and observed aerosol extinction profiles requires possible upgrades of multiple model physics schemes and quantification of key parameters that could affect vertical distribution of aerosols, for instance, biomass burning injection heights (Grell et al., 2011), boundary layer height and near-surface winds (Nair et al., 2012). Additionally, high-quality measurements at different locations are also needed for model evaluation over longer time periods, and it is recommended for future studies over this region"

To address the contribution to the AOD by individual aerosol species raised by the reviewer, we plot the species-specific aerosol burdens as a proxy for understanding the contribution to the AOD by individual aerosol species, since in our model AOD is calculated assuming the internal aerosol mixtures, and the species-specific AOD is not available. Fig. S1 below shows the calculated aerosol burdens for March 2012. This new figure is now included in the Supplementary Material as well as the following discussions:

"In our model, aerosol optical depth (AOD) is calculated with the internal mixing assumption. In order to attribute the AOD underestimation to major aerosol types, we plot the species-specific aerosol burdens as a proxy for understanding the contribution to the AOD by

individual aerosol species. Fig. S1 shows the aerosol burdens calculated for March 2012. Since dust is the dominating species over northwestern India semi-arid regions and the adjacent Arabian Sea, it is the main contributor to the underestimation of AOD over these regions. In contrast, anthropogenic sulfate, oc, and bc are the main components of aerosol loadings (thus AOD) in polluted northern and northeastern India, as well as in the long-distance transported aerosols over the downwind of southwestern Indian sub-continent."



Figure S1. Calculated aerosol burdens  $(mg/m^2)$  of (a) Sulfate, (b) BC, (c) OC, and (d) Dust for March 2012

We also added the following additional discussions in the text, Section 3.1, Lines 1-3 on Page 16911 as follows,

"These discrepancies could be attributable to episodic dust activities not reproduced by WRF-Chem (as shown in Fig. S1 that dust aerosols are dominating species) or to overestimation associated with the MODIS satellite retrievals over highly reflective surfaces such as deserts and clouds over the ocean. In other aerosol-concentrated areas, anthropogenic pollutants such as sulfate, BC, and OC are the main contributors to the AOD underestimation (Fig. S1)"

2. Authors discussed the evaluation of modelled AOD for March 2012, even though the simulations are available for eight months (From August 2011 to March 2012). It would useful if authors use the entire simulation period for the model evaluation.

The reason that we only presented the AOD comparison for March 2012 is because the groundbased lidar observations of aerosol vertical profiles are available only for that month. Since the objective of this paper is to "identify altitude-related bias" and examine the subsequent responses due to the mis-represented vertical profiles of aerosol extinctions, we decided not to include the model results of AOD from other months for which we do not have vertical profile measurements to evaluate. Justifications of limiting our analysis to one month (March 2012) were given in Section 2.1, Lines 12-17, on Page 16908,

"The model-data analysis and discussions here center on simulations in March 2012, for two reasons. First, during this pre-monsoon month, ground-based lidar measurements are available at Nainital and Kanpur (in northern India) and used with satellite observations to characterize bias in the calculated aerosol extinctions. As discussed later, it is important to have independently calibrated ground-based measurements because of the uncertainty associated with satellite data."

For the reviewer's reference, the AOD comparison for other months is shown below in Fig. R2.1. It indicates underestimation in the model-calculated AOD similar to that for March 2012. Resolving these mismatches between simulated and observed AOD requires development of a verification database extending from field campaigns, ground-based and aircraft measurements for evaluation of model simulated boundary layer dynamics and aerosol concentrations and chemical composition. This could then lead to possible upgrades of model physics schemes and quantification of key parameters including emissions, biomass burning injection heights, boundary layer height and near-surface winds etc. We consider that it is beyond the scope of this paper and certainly deserves further investigation.



Figure R2.1 Time averaged AOD between Aug 2011 and March 2012 from (left) MODIS/Terra, and (right) WRF-Chem simulations

3. Authors found that "83% of the model low-bias is due to aerosol extinctions below 2 km". A brief discussion about the vertical distribution of anthropogenic and wildfire emissions treatment in

## the model would be useful to the reader here. How this treatment could influence the uncertainty in the vertical distribution of extinction?

Following the suggestion by the reviewer, we now include the following sentence in the "Methodology" (Section 2.1) describing the vertical distribution of aerosol emission treatment, Line 20, on Page 16907:

"Primary aerosol emissions including all the anthropogenic, biomass burning and natural sources are injected into the lowest level of the model and transported by advection and updrafts. Calculations of optical properties of aerosol assume..."

We also add more and revise the following discussions in the "Summary and Discussion" (Section 4), Lines 10-13, on Page 16922:

"Resolving the mismatch between simulated and observed aerosol extinction profiles requires possible upgrades of multiple model physics schemes and quantification of key parameters that could affect vertical distribution of aerosols, for instance, biomass burning injection heights (Grell et al., 2011), boundary layer height and near-surface winds (Nair et al., 2012) as well as a. Additionally, high-quality measurements at different locations are also needed for model evaluation over longer time periods, and it is recommended for future studies over this region"

It should also be noted that the biomass-burning source over this region is primarily due to seasonal burning of agriculture waste concentrated over the agricultural region to the west and during December and early January (first crop also known as kharif). A second crop is also harvested during spring and is associated with biomass burning peaks in April/May (Rabi crops).

Two references added:

Grell, G., Freitas, S. R., Stuefer, M., and Fast, J.: Inclusion of biomass burning in WRF-Chem: impact of wildfires on weather forecasts, Atmos. Chem. Phys., 11, 5289-5303, 2011.

Nair, V. S., Solmon, F., Giorgi, F., Mariotti, L., Babu, S. S., and Moorthy, K. K.: Simulation of South Asian aerosols for regional climate studies, J. Geophys. Res., 117, D04209, doi:10.1029/2011JD016711, 2012.

4. Authors separated the effect of absorption and scattering properties using two simulations (Case 1 and Case 2). But between Case 1 and Case 2, there can be considerable changes to the aerosol distributions. How does this contribute to the uncertainty in simulated aerosol extinction profiles?

As described in the Section 3.2, Lines 12-17, on Page 16913, sensitivity studies including both Case I and Case II are conducted by "optimizing matching of the observed aerosol vertical profiles. The calculated aerosol extinctions in the lowest eight model layers (below ~850 hPa, at 1.5-3 km above sea level in the simulated model domain) are increased by a factor of 2 at each time step..." That means that in both sensitivity cases, the simulated aerosol concentrations are kept the same as in the control run, and aerosol extinction profiles (sum of scattering and

absorption) were adjusted to the same level (i.e., a factor of 2 increases), although their scattering or absorbing fractions are different.

This is clearly shown in the model results as in Fig. 1(c) for AOD and Fig. 2 for extinction profiles. Only one curve is needed on each panel to represent the results from sensitivity studies (both Case I and Case II) labeled as "Double extinction below 850 hPa" in Fig. 1(c) and "Model (increased ext)" in Fig. 2". We have also clarified this point in the discussions of results, Section 3.3, Lines 15-17, on Page 16915,

"Because aerosol extinctions (thus AODs) in Cases I and II are increased to the same level, the TOA radiative effects of aerosols are similar for the two cases..."

And in the Section 3.4, Line 7, on Page 16917,

"Forced by the same aerosol extinction profiles with the bias correction..."

Therefore, the calculated AOD distributions and extinction profiles are identical for Case I and Case II. There are no "considerable changes to the aerosol distributions" nor "contribute to the uncertainty in simulated aerosol extinction profiles" between Case I and Case II.

Technical comments:

Page 16903, Line 26: Wrong citation year (Pan et al., 2015).

Corrected.

Page 16905, Lines 10-14: Recent multi-model evaluation paper (Quennehen et al., 2015) is missing from the manuscript.

Reference Quennehen et al. (2015) is now added as follows on Line 14, Page 16905:

"A recent study by Quennehen et al. (2015) examined six global and one regional models with CALIPSO-derived backscatter profiles at 532nm during August and September 2008, and the multimodel mean backscatter is also underestimated between 0 and 2 km over northern India and eastern China."

Page 16907, Lines 15: Compiled SO2 emissions is confusing. Rewrite the sentence.

It is now revised as follows:

"The total SO2 emissions in South Asia with updated emissions over India are 9.36 Gg yr<sup>-1</sup>, slightly less than the default GOCART emissions (10 Gg yr<sup>-1</sup>)"

Page 16910, Lines 17: The geographic pattern of AOD distributions is not reasonably well captured. Rewrite the sentence.

It is now revised as:

"...the overall geographic pattern of AOD distributions is simulated reasonably well except for over the Arabian Sea"

Figure 2: Why MPL data extinction profiles peak is different than other data sets?

The MPL profile at Nainital in Figure 2(a) is similar to other data sets. So we assume that the reviewer refers to the monthly mean MPL extinction profile depicted in the Figure 2(b) for Kanpur, which has a peak around 600m above the surface different from the CALIPSO data and model results. This MPL extinction profile is derived by monthly averaging the quality-assured MPLNET level 2 daytime products (Welton et at., 2001). As shown below (Fig. R2.2) for the 23 days in March 2012 with data available, these MPL daily profiles downloaded from their data archive also have a peak around 600m above the surface, and the reason is not clear from the MPLNET website. Nevertheless, this does not affect our method and main conclusions, since we did not try to match the MPL profiles but applied a systematical bias correction of a factor of 2 to the model predictions below 850hPa.



Figure R2.2 MPL daily profiles available from the MPLNET for 23 days in March 2012 at Kanpur