The authors wish to thank each of the anonymous reviewers for taking the time to review our manuscript. While referee #2 found the manuscript to be publishable in its current, referee #1 has provided valuable input to the revised manuscript. We have responded (in blue) to each of the general and specific comments (in black) and hope they are to the satisfaction of the questions/concerned posed.

Anonymous Referee #1

General comments

The authors applied the NOAA GFDL-AM4 model to simulate air quality over India for 20 years. They used a coarse horizontal resolution and two different emissions estimates (CMIP5 and CMIP6) in the model. They compared model predictions with observed PM25 in India for six winter months and performed detailed analysis. Model with CMIP5 substantially underestimated PM25 compared to observed data in Northern India. While the model with CMIP6 improved the predictions, it still underestimated PM25. Most monitoring stations in India are located in urban areas. The model with a coarse horizontal resolution is not suitable for examining PM25 in urban areas in India.

We appreciate the reviewers concern with using this model to compare with urban air quality data in India. There is a long history of representation errors when testing gridded global model output against limited number of point observations of air quality – rural, urban, or otherwise. However, these errors exist even for relatively high resolution regional air quality models (e.g., 4–12 km) as sites are often clustered closely together. For example, there are 9 observing sites located in and around New Delhi and they are extremely diverse in their overall magnitude of $PM_{2.5}$ abundances (Figure 2a) despite being within ~25km of one another. In that sense, it's not clear how much more suitable a high resolution model would be. Furthermore, it would certainly prove difficult to obtain accurate emission data on the scale required, which is probably on the order of 1-5 km. Moreover, these are the only observations that are available and we attempt to make use of them as best as possible - not only to test the GFDL AM4 simulation of $PM_{2.5}$, but also compare modeled vs. observed meteorology-$PM_{2.5}$ relationships. In any case, we recognize this analysis has similar measurement-model mismatch issues that exist in previous works and we try to make this point in Section 3.1.

Specific comments

Line 199-200 Reference is needed for the heterogeneous uptake coefficients used in the model.

We have included a reference for the values in Table S1.

Line 204-206 Several acronyms have already been defined before and are defined here and later. No need to define the acronyms multiple times.

We have removed duplicate acronyms within the main text.

Line 215-216 Dust1, dust2, ssalt1, ssalt2, ssalt3 are not defined in the article?

Thank you for noting this. We have included a description of these components (the numbers correspond to different size bins; i.e., dust1-2 & ssalt1-3 are < 2.5µm)

Line 261-266 and Figure 1 It is not clear if NO or NO2 emissions are shown in Figure 1. "NO" is used in one sentence but "NO2" is used in the other sentence. Need clarification. How NOx emissions are being speciated into NO and NO2 emissions?

Thank you for catching this. NOx emissions are emitted as 100% NO and as such the references to NO_2 in the text and figure caption are incorrect, but they are merely a typo that we have corrected.

Figure 2 Title of Figure 2 indicates "CMIP5-dry". However, legend shows "AM4-CMIP5 (wet)". Need clarification.

Thank you for catching this. We have changed the caption for Figure 2 to read "AM4-CMIP5(wet)" as the legend indicates.

Figure 3 Observed data are taken from Kanpur site which is not clearly indicated in the Figure title.

We have added a description of the observational data (city name, lat, lon, elevation, time period, and reference) to the figure caption.

Line 325-340 The model over-predicts aerosol nitrate substantially which may result from many factors including the use of high heterogeneous uptake coefficient for N2O5 (Table S1). Recent studies (Davis et al., 2008; Reimer et al., 2009; Brown et al., 2009; Phillips et al., 2016; Chang et al., 2016) suggest a much lower value for the heterogeneous uptake coefficient. A discussion of the impact of high heterogeneous uptake coefficient for N2O5 on model results is relevant.

Davis et al., 2008: Parameterization of N2O5 reaction probabilities on the surface of particles containing ammonium, sulfate, and nitrate, Atmos. Chem. Phys., 8, 5295– 5311.

Riemer et al., 2009: The relative importance of organic coatings for the heterogeneous hydrolysis of N2O5, JGR, 114.

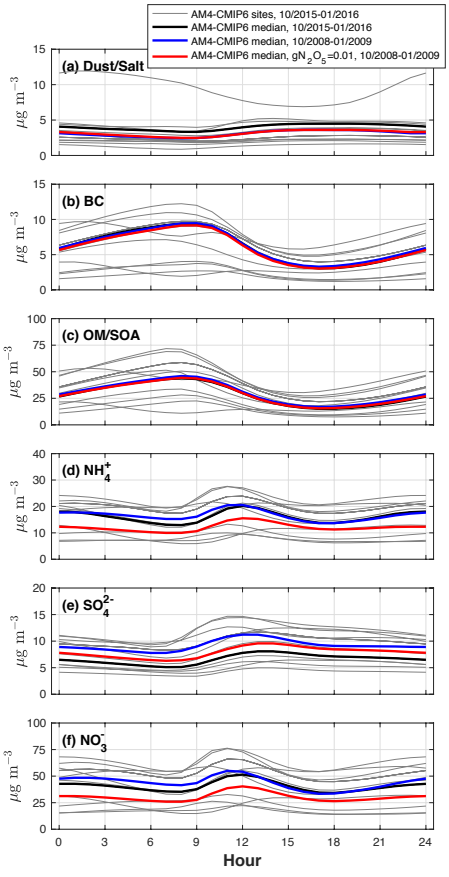
Brown et al., 2009: Reactive uptake coefficients for N2O5 determined from aircraft measurements during the Second Texas Air Quality Study: Comparison to current model parameterizations, JGR, 114.

Phillips et al., 2016: Estimating N2O5 uptake coefficients using ambient measurements of NO3, N2O5, ClNO2 and particle-phase nitrate, ACP, 16, 13231-13249.

Chang et al., 2016: Evaluating N2O5 heterogeneous hydrolysis parameterizations for CalNex 2010, JGR: Atmosphere, 121, 5051–5070.

Thank you for bringing this to our attention. The heterogeneous uptake coefficient used here for N_2O_5 is indeed significantly higher than those from recent studies, and has been the default value for the GFDL AM3 chemistry (Naik et al., 2013; Mao et al. 2013a). For the prototype AM4 version we used, this higher value was adopted, however, a lower value of 0.01 has been adopted in more recent configuration of AM4 based on updates by Paulot et al. (2016). To test how our value affects the nitrate bias, as well as the potentially aberrant nitrate diurnal cycle (per the comment below), we have run an additional simulation that covers the 2008-2009 period of the observations by Ram et al. (2012) using the updated uptake value of 0.01, which is an order of magnitude smaller than our original. We have plotted the results of this simulation in Figure S10 (diurnal cycle of $PM_{2.5}$ components), which is shown below. Nitrate is shown in panel (f). Compare the blue line (base simulation of the Ram et al. period) with the red lines (sensitivity over the same period). The effect of the updated gamma value is to reduce nitrate abundances by ~15 ug m^3 and ammonium abundances by about 5 ug m^{-3} with the largest changes at night. However, the diurnal cycle of nitrate (as with ammonium and sulfate) is qualitatively unchanged from the base simulation, with a relative maximum still occurring at midday. So, while we have reduced nitrate abundances, the seemingly aberrant diurnal cycle of nitrate is still evident even with the updated gamma and will require additional experimentation beyond the scope of this paper. We hope that this satisfies the reviewers comments/concerns. We have added the following to the paper to reflect the results of the sensitivity experiment:

We test if the seemingly aberrant NO_3^- diurnal cycle is a result of our choice of the value for the N_2O_5 heterogenous uptake coefficient (0.1), which is significantly higher than those reported by previous studies (e.g., Davis et al., 2008; Chang et al., 2016), by performing an additional simulation with an N_2O_5 uptake coefficient of 0.01 over period of the Ram et al. (2012) observations. The effect of the updated value is to reduce NO_3^- by ~15 $\mu g\ m^3$, NH_4^+ by ~5 $\mu g\ m^{-3}$, and SO_4^{2-} by ~1 $\mu g\ m^{-3}$, all with the largest changes at night (Fig S10d-f). However, the diurnal cycle of NO_3^- (as with NH_4^+ and SO_4^{2-}) is qualitatively unchanged from the base simulation, with a relative maximum still occurring at midday. So, while we have reduced nitrate abundances, the midday NO_3^- peak is still evident even with the updated gamma. One possible explanation is that the model prescribes monthly average deposition rates for NH_4NO_3 (i.e., no diurnal cycle), however, determining the cause of this midday peak will require additional experimentation beyond the scope of this paper.



Line 443-444 The sentence suggests that annual cycle is shown in the Figure. However, it shows data for 6 months, not for the year.

Thank you, we have fixed the wording to denote that it is a monthly average for six months.

Line 449-450 The sentence suggests model is biased high in RH. However, Figure 6b shows under-prediction of RH compared to observed data. Need clarification.

Thank you for bringing this to our attention. We have fixed the sentence to reflect the low-bias.

Line 492-497 Ram and Sarin (2011) analyzed measurement data and reported that nighttime aerosol nitrate level is five times greater than the day-time nitrate level. In contrast, this modeling study finds that aerosol nitrate peaks during mid-day. Despite the use of high uptake coefficient for N2O5, it finds that aerosol nitrate peaks during the day which reveals that HNO3 produced from the reaction of NO2 + OH likely dominates the production of aerosol nitrate. What caused the results to be completely opposite to that of Ram and Sarin (2011)?

See discussion above.

Figure S7 Title is not clear. Figure d-f are missing.

We have fixed the figure caption. Only figures a-c are shown.

Figure S8 Title refers to Figure 4 which should probably be Figure 5.

Thank you for catching this, we have fixed the caption.