Response to comments by Referee #2

The revised version is greatly improved and responds to most of my concerns. There is however still one major issue. Concerning the impact of pNO3- photolysis, there is something that does not add up. The authors claim that "The photolysis of pNO3- does not change the pNO3- concentration much (~1%, see Figure S1) because deposition dominates the sink." However, their Fig. R1 (in the response to Reviewer#1) shows that the pNO3- photolysis rate is of the order of 1E-5 s-1 in the lower troposphere (except in January). How could such a fast sink rate be negligible in comparison with deposition? Unless I'm wrong, the deposition lifetime should be at least one day. There is something going on that requires further investigation. A possible explanation could be that the pNO3- sink is nearly exactly compensated by enhanced HNO3 conversion to the particulate phase. In which case, the pNO3- photolysis would have a big impact on gas-phase HNO3 concentrations. Is this the case, and if so, can you show the impact in the paper and evaluate whether this is reasonable and in line with observations?

Thanks for pointing this out. The pNO_3^- photolysis rate over CONUS is indeed comparable to its deposition loss rate, which we have now corrected in our revised manuscript.

The reason for the limited impact of pNO_3^- photolysis on pNO_3^- concentration is that the loss of pNO_3^- is compensated by fast oxidation of NO_x back to HNO_3 because pNO_3^- photolysis releases both NO_x and OH (from HONO photolysis). We have now included this discussion in our revised manuscript.

Response to comments by Referee #3

The authors provided an analysis of the decreasing trends in tropospheric NO2 columns over the US. They found that the consideration of aerosol nitrate photolysis can lead to an increase in model NO2 by 13% and a decrease in the retrieved NO2 by 7%, and the combined effects can lead to a 45% reduction in the difference between modeled and retrieved changes in the year of 2009 and 2017. The topic is interesting, and the analysis is helpful for a better understanding of free tropospheric NO2. I recommend the paper for publication after consideration of the points below.

We thank the reviewer for their thoughtful comments. Our response to the comments is as follows:

Comments:

1. **There are large interannual variabilities in tropospheric NO2 columns. The conclusion derived in this analysis represents the impacts of aerosol nitrate photolysis in the years 2009 and 2017. It cannot represent the impact on the trends in the period of 2009-2017.**
   Thanks for pointing this out. To clarify, we have now made some edits in Section 3.

2. **Have the authors considered the location and time consistency in simulations and OMI observations when calculating the mean NO2 columns over the CONUS? The OMI instrument crosses the equator at a local time of 13:45, and lots of data are filtered by the quality filters. To provide an accurate comparison, we should only consider modeled NO2 matching the temporal and spatial locations of OMI observations.**
   Yes, we have. We now clarify this in the revised manuscript.

3. **Lines 176: "Shah et al. (2023) found that incorporating aerosol nitrate photolysis in GEOS-Chem largely corrected the model's underestimation of NOx over the oceans during the ATom aircraft campaign".**
   I checked Shah et al. (2023) and found "GEOS-Chem reproduces the shape of the PSS-inferred NO2 profiles throughout the troposphere for SEAC4RS and DC3 but overestimates NO2 concentrations by about a factor of 2" in the Abstract. I assume I perhaps have made some misunderstanding, but it seems that the discussions are not consistent.
   Shah et al. (2023) did a model comparison against several aircraft campaigns, including ATom over the oceans and SEAC^{4}RS and DC3 over the US. For clarification, we now add a discussion of Shah et al. (2023) to the revised manuscript.

4. **The calculation of the enhancement factor is coarse. Considering the low SSA, it suggests an enhancement by a factor of 10 everywhere in the middle and upper troposphere. Can we apply the same factor to simulate free tropospheric NO2 globally?**
   In the upper troposphere, our calculated enhancement factor (EF) is about 10-20 over land at northern mid-latitudes, and up to 100 over the oceans in the southern hemisphere (see Figure 5 of Shah et al., (2023)). These values are within the range of EF that we currently know from field and laboratory studies (Ye et al., 2017; Romer et al., 2018; Andersen et al., 2023) and can be applied globally to test their effects. However, as we stated in the

conclusion section, further studies are needed in the future as these rates are still very uncertain. To clarify, we have made some edits in Section 2.

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