

We thank the two reviewers for their careful reading of the manuscript and their detailed comments. Our responses are shown below in blue, with new text in bold.

Anonymous Referee #1

Actually the only criticism that I have is the statement, that RO_x chemistry has only a minor role for the NO/NO_2 ratio at high altitudes. I agree, that NO_2 photolysis and the $\text{NO} + \text{O}_3$ reaction might be dominant, but due to the low temperatures the later reaction is slower in the UT. Observations indicate that HO_x (and most likely RO_2) are often enhanced in the UT due to convective injection of precursors. Thus it would be interesting to quantify the role of HO_2 and RO_2 for the NO/NO_2 ratio in the UT.

We have revised Section 4 to include a discussion of HO_2 and RO_2 . We have added the following paragraph, with the revisions to Figure 6 below.

“Zhu et al. (2016) found that GEOS-Chem underestimates the observed HCHO concentrations in the upper troposphere during SEAC4RS by a factor of 3, implying that the model underestimates the HO_x source from convective injection of HCHO and peroxides (Prather and Jacob, 1997; Müller and Brasseur, 1999). HO_2 observations over the central US in summer during the SUCCESS aircraft campaign suggest that this convective injection increases HO_x concentrations in the upper troposphere by a factor of 2 (Jaeglé et al., 1998). The bottom right panel of Figure 6 shows median modeled and observed vertical profiles of the HO_x reservoir hydrogen peroxide (H_2O_2) during SEAC⁴RS over the Southeast US. GEOS-Chem underestimates observed H_2O_2 by a mean factor of 1.7 above 8km. The middle right panel of Figure 6 shows the predicted $[\text{NO}]/[\text{NO}_2]$ ratio if modeled convective injection of HO_2 and RO_2 precursors is underestimated by a factor of 2. While such an underestimate is insufficient to reconcile simulated and observed $[\text{NO}]/[\text{NO}_2]$ concentration ratios, the contribution to the $[\text{NO}]/[\text{NO}_2]$ ratio from Reaction 6 would be much more significant than previously estimated.”

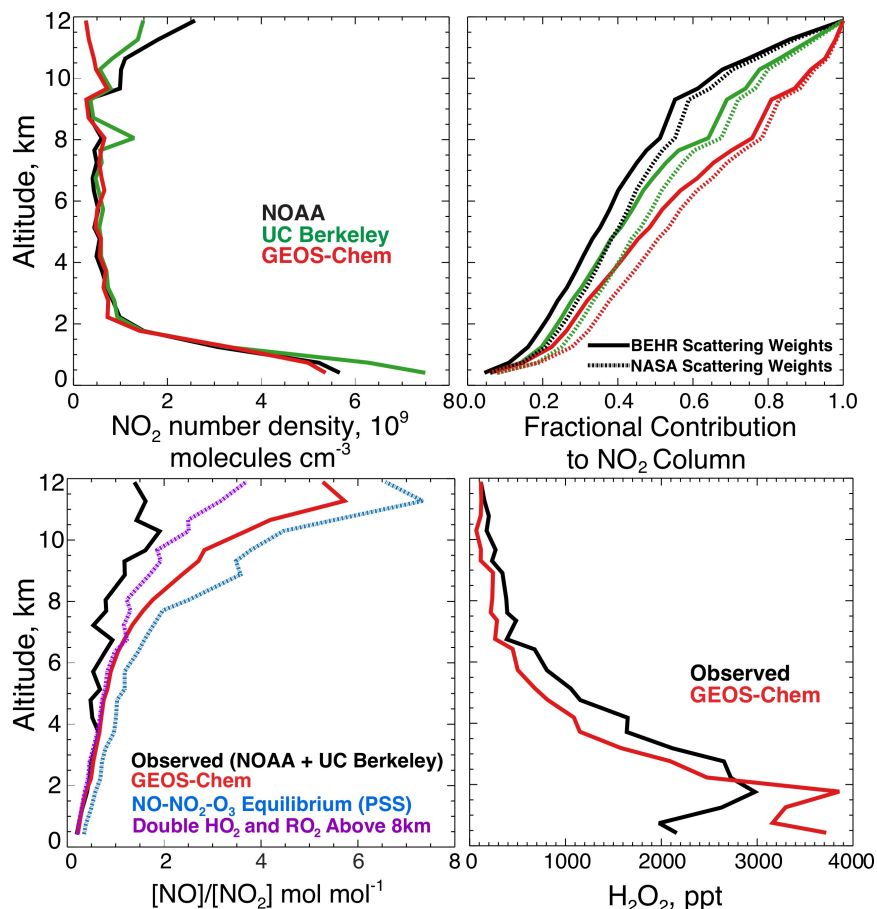


Figure 1: Vertical distribution of NO_2 over the Southeast US during SEAC⁴RS (August-September 2013) and contributions to tropospheric NO_2 columns measured from space by OMI. The top left panel shows median vertical profiles of NO_2 number density measured from the SEAC⁴RS aircraft by the NOAA and UC Berkeley instruments and simulated by GEOS-Chem. The top right panel shows the fractional contribution of NO_2 below a given altitude to the total tropospheric NO_2 slant column measured by OMI, accounting for increasing sensitivity with altitude as determined from the retrieval scattering weights. The bottom left panel shows the median vertical profiles of the daytime $[\text{NO}]/[\text{NO}_2]$ molar concentration ratio in the aircraft observations (NOAA for NO and UC Berkeley for NO_2) and in GEOS-Chem. Also shown is the ratio computed from NO- NO_2 - O_3 photochemical steady state (PSS) as given by reactions (5)+(7) (blue) and including reaction (6) with doubled HO_2 and RO_2 concentrations above 8km (purple). The bottom right panel shows the median H_2O_2 profile from the model and from the SEAC⁴RS flights over the Southeast US. H_2O_2 was measured by the Caltech CIMS (see Figure 2).

A minor point is that the titles of chapter 6 and 7 are identical.

This typo has been fixed. Section 7 now reads “7. Implications for ozone: surface air”

Anonymous Referee #3

One main flaw is the changing of the NEI for NO_x is not well addressed and justified. The necessity of the large reduction of the NEI for NO_x in the Southeast and nationally is the most important conclusion in the manuscript. However, no supporting information is utilized to verify why a reduction of 60% was suggested, besides finding it a close match to observations. The feasibility of implementing the same reduction percentages on all the other sources besides power plant emissions also needs to be justified. In addition, it is unclear how the NEI11 is

scaled to the 2013 emission, which is a fundamental piece of information to know before further modifications on the NO_x emissions.

We focus in this paper on the Southeast U.S., where emissions of non-anthropogenic NO_x are small compared to NEI11v1 emissions. We state on line 26 on page 5 the following: **“Errors in NO_x sources from soils, wildfire, or lightning cannot account for the overestimate because their magnitudes are small relative to fuel combustion, as shown below.”**

We have added a clarifying sentence for the 2013 emission factor on Page 5 line 17: **“The scaling factor for 2013 for NO_x emissions is 0.89. Further information on the use of the NEI11v1 in GEOS-Chem can be found here: http://wiki.seas.harvard.edu/geos-chem/index.php/EPA/NEI11_North_American_emissions/.”**

One other crucial problem is about the vertical mixing. The authors include this as part of the title, and provide this as a main explanation for the model-measurement bias. Yet it is barely discussed in the manuscript, for example, it is only mentioned briefly in two paragraphs and no discussion what the assimilated vertical mixing from GEOS actually looks like. If the authors think that this is an important factor, then they should discuss what the modeled values are and why they think they are overestimated. Additionally, there is no discussion on how the driving meteorology influences the near-surface turbulent mixing, which is likely important in the reanalysis data they are using. Since a large amount of ozone is produced near the surface, this section will be improved with addressing the effects of both turbulent mixing and surface ozone chemistry to understand the vertical profiles in Figure 12. In addition, the manuscript notes that daytime mixing depths are reduced by 40% in the meteorological setup of the model. It would be helpful to explain how this change influence the dynamics below the boundary layer, which further impact the vertical mixing of ozone.

We agree with the comment about the title and have changed the paper title to the following: **Why do Models Overestimate Surface Ozone in the Southeastern United States?**

We have revised Page 3 lines 18-21 to include a more detailed description of model vertical mixing and the reduction in boundary layer height.

“Turbulent boundary layer mixing follows a non-local parameterization based on K-theory (Holtslag and Boville, 1993) implemented in GEOS-Chem by Lin and McElroy (2010). Daytime mixing depths are reduced by 40% as described by Kim et al. (2015) and Zhu et al. (2016) to match lidar observations of boundary layer height from SEAC⁴RS.”

We have changed to discussion of vertical mixing throughout the paper to include a discussion of both excessive vertical mixing and chemical production of ozone likely due to excessive dryness in the model. We replace line 7 on page 2 with the following: **“This may be caused by excessively dry conditions in the model, representing another factor important in the simulation of surface ozone.”**

We replaced the paragraph on Page 12 line 19 with the following:

“It appears instead that there is a model bias in boundary layer vertical mixing and chemical production. Figure 12 shows the median ozonesonde profile at a higher vertical resolution over the Southeast US (Huntsville, Alabama and St. Louis, Missouri sites)

during SEAC⁴RS as compared to GEOS-Chem below 1.5 km. The ozonesondes indicate a decrease of 7 ppb from 1.5 km to the surface, whereas GEOS-Chem features a reverse gradient of increasing ozone from 1.5 to 1 km with flat concentrations below. Preliminary inspection suggests that this may reflect excessively dry conditions in the GEOS-5.11.0 meteorological fields, promoting boundary layer production and vertical mixing of ozone. Such a bias might not be detected in the aircraft data, collected mainly under fair weather conditions.”

We replace the concluding sentences on Page 13 line 33 with the following:

“This may be due to excessively dry conditions in the GEOS meteorological data used to drive GEOS-Chem, resulting in excessive boundary layer ozone production and mixing. Such a bias may not be detected in the aircraft data, generally collected under fair-weather conditions. Investigating this source of bias and its prevalence across models will be the topic of a follow-up paper.”

The study calculates that 54% of the ISOPO₂ radical reacts through the high-NO_x pathway compared with 62% before the NEI11 adjustment, and states the influence of changing NO_x emission on the high-NO_x pathway is weak. Since the paper attributes this weak dependence to the spatial segregation between isoprene and NO_x emissions, it would be helpful to compare the distribution of segregation with the high-NO_x pathway results to confirm this conclusion.

This work is done in Yu et al, 2016. We have clarified this sentence on page 10 line 28: **It now reads, “The lack of dominance of the high-NO_x pathway is due in part to the spatial segregation of isoprene and NO_x emissions (Yu et al., 2016).”**

The bias between simulations and observations is still large in some regions, as shown in both Figure 3 and Figure 4. In Figure 4, the manuscript uses an uneven color bar for NO_x, making it hard to distinguish the differences between observed and simulated NO_x. It would be clear to identify those differences using constant color bar scale for NO_x or provide more color contours, or to make a contour plot for the differences of NO_x and O₃ between simulations and observations. With the biases in Figure 3 and 4, the changes on NO_x emissions could have a regional dependence.

We have changed the scale on Figure 4 for NO_x to a linear rather than a log scale.

We provide the spatial correlation coefficient for the Southeast US and the continental US inset in Figure 3 to show the good agreement between model and observation.

We state on Page 7 line 21 that for NO_x, “The spatial correlation coefficient is 0.71.” I have added the following to the caption of Figure 4 to clarify the good spatial agreement between model and observation for both NO_x and O₃: **“The spatial correlation coefficient is 0.71 for both NO_x and O₃. The normalized mean bias is -11.5% for NO_x and 4.5% for O₃.”**

We add the following sentence to Page 6 line 1: **“There is no information in the spatial pattern of bias that would warrant a more location-specific or source-specific reduction.”**

The sentence “no indication of regional patterns of model bias that would point to the need for a more selective adjustment of NO_x emissions” is not clear to me. It would be better to draw a conclusion about regional patterns after analyzing the model biases in Figure 3 and 4.

See reply to #4 above.

We added the following clarification on page 7 line 13: **“We see from Figure 3 that the model with decreased NO_x emissions reproduces the spatial variability in the observations with minimal bias over the Southeast US domain shown in Figure 1 and across the rest of the country.”**

The domains of the maps are not consistent in Figures 1, 3, 4, 5 and 7, which is confusing as they come from the same simulations. Also, it would be helpful to add lat/lon labels for the contour maps.

The maps are intended to either show the CONUS, Southeast US, and/or or the Gulf of Mexico. We have added lat/lon labels to the contour maps for Figures 1, 4, 5, and 7 to clarify their domains.

Technical corrections:

1. Same title for Section 6 and Section 7. Please clarify the differences between these two sections.
 - a. **“7. Implications for ozone: surface air”**