

Responses to the comments by Dr. P. Kasibhatla

Thank you for providing valuable comments. Please find our responses (in black) to your comments (in blue) below.

This is an interesting paper that suggests a possible explanation for typical model over-prediction of surface ozone over CONUS (Figure 7). It is not clear however if model simulations are improved both in terms of surface O₃ predictions, as well as O₃ vertical profiles (especially in the boundary layer and just above the boundary layer). While comparisons with measured vertical profiles of JNO₂ are shown in Figure 3, no corresponding comparisons of vertical profiles are shown for O₃. It would be useful to show these comparisons (and provide histograms as is done for JNO₂) with simultaneous aircraft O₃ measurements, especially given the overprediction of JNO₂ in the boundary layer in the GOES simulation compared to the CNTR simulation for the NOMADSS flights (Figure 3).

In terms of model evaluation, it would be also useful to show comparisons of the modeled Ox vs NO_z relationship against observations (as is done in Travis et al., 2016) as a check on modeled ozone production efficiency.

1. O₃ vertical profile comparison

The influence of satellite cloud corrections on vertical profile of O₃ is shown in Figs. P1 (SEAC⁴RS) and P2 (NOMADSS). Only aircraft data over land within the southeast region (latitude: 25–40N, longitude: 95–70W) are used for the averages. Unlike the vertical profiles of JNO₂, which shows considerable improvements when satellite cloud corrections are applied, the vertical profiles of O₃ do not show significant differences between CNTR and GOES simulations even though the histograms of model-to-observation O₃ ratio show slight improvements in the GOES simulation than in the CNTR simulation. This is likely because the aircraft measurements are mostly made in rural environments or high altitudes where O₃ precursor concentrations are low. As shown in the manuscript, the effects of cloud correction are larger under high-NO_x environments than low-NO_x environments. An example on 21 September 2013 shows that GOES simulation better captures the attenuation of JNO₂ under below cloud conditions (~1830–1940 UTC) (Fig. P3). As the aircraft flew over relatively high-NO_x regions during this time period, O₃ concentration shows a better agreement with observations in GOES simulation than CNTR simulation although both the simulations considerably overpredict O₃ in general. The largest difference in O₃ between the two simulations is 5.6 ppb at 1946 UTC. One of the reasons for the overprediction of O₃ could be the overprediction of NO₂ or misplacement of urban plumes. Other example for NOMADSS, on 7 July 2013 when the aircraft flew mostly over the state of Indiana and Lake Michigan shows similar results (Fig. P4). The sky conditions on that day were characterized by broken clouds, and the coarse resolution of satellite data (the original resolution is 8 km at hourly intervals) is another limitation for capturing the exact locations of small clouds. However, O₃ concentrations along the flight tracks in the two simulations show differences under cloudy conditions and the differences are noticeable only at high NO₂ (e.g., ~1635–1900 UTC). The largest difference in O₃ between the two simulations is 4.4 ppb at 1837 UTC. It should be noted that O₃ concentration in the two simulations is almost the same when NO₂ concentration is low even if JNO₂

values are significantly different (1920–1940 UTC). Thus, even though some cases show that clouds have significant influences on O₃ formation and concentrations above the ground (e.g., within the boundary layer), when all the data points are averaged the effects are hardly noticeable. We anticipate that if airborne measurements of O₃ under cloudy sky conditions are available over cities and/or urban plumes, then we would clearly see the effects of clouds on vertical profiles of O₃. Unfortunately, neither of the campaigns were designed for this purpose, so there are no good airborne observation data to examine the effects of clouds on vertical profiles of O₃.

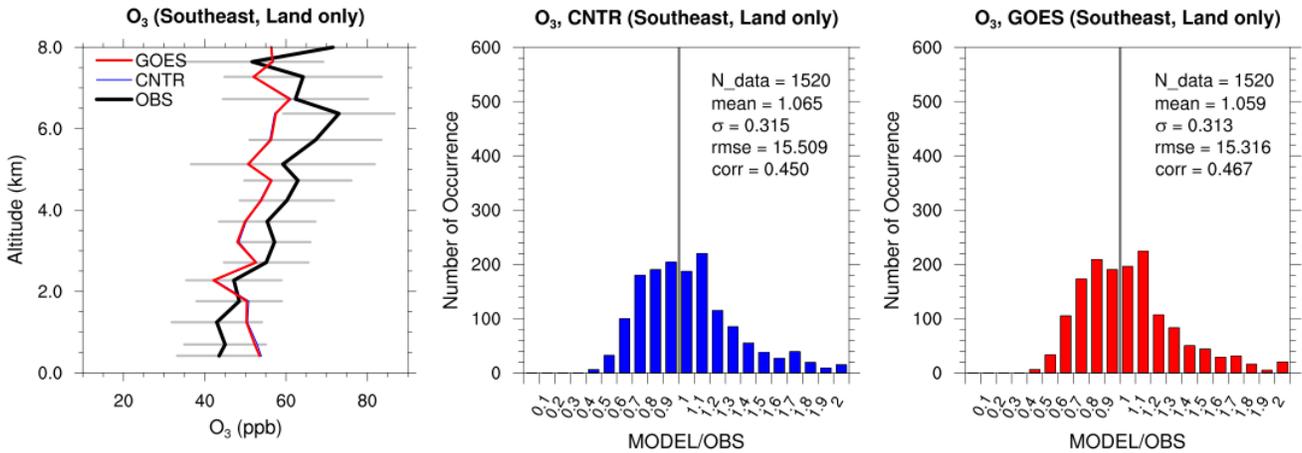


Fig. P1. (Top) (Left) Cloudy-sky averaged vertical profiles of O₃ for SEAC⁴RS observations, CNTR and GOES simulations. (Middle and Right) Histogram of ratio of O₃ simulated by the model to O₃ observed for CNTR simulation and GOES simulation, respectively.

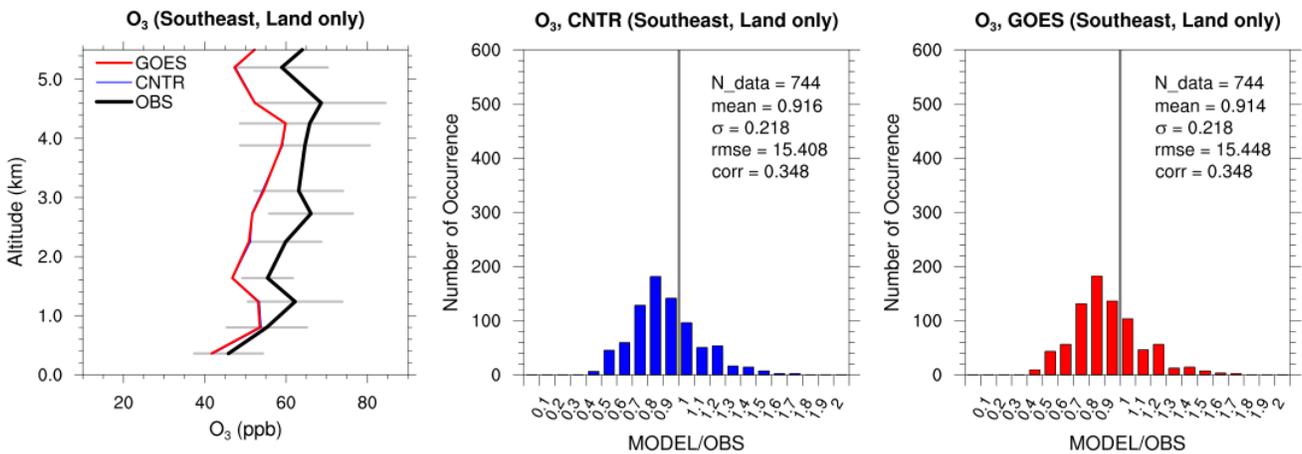


Fig. P2. Same as Fig. P1, but for NOMADSS campaign.

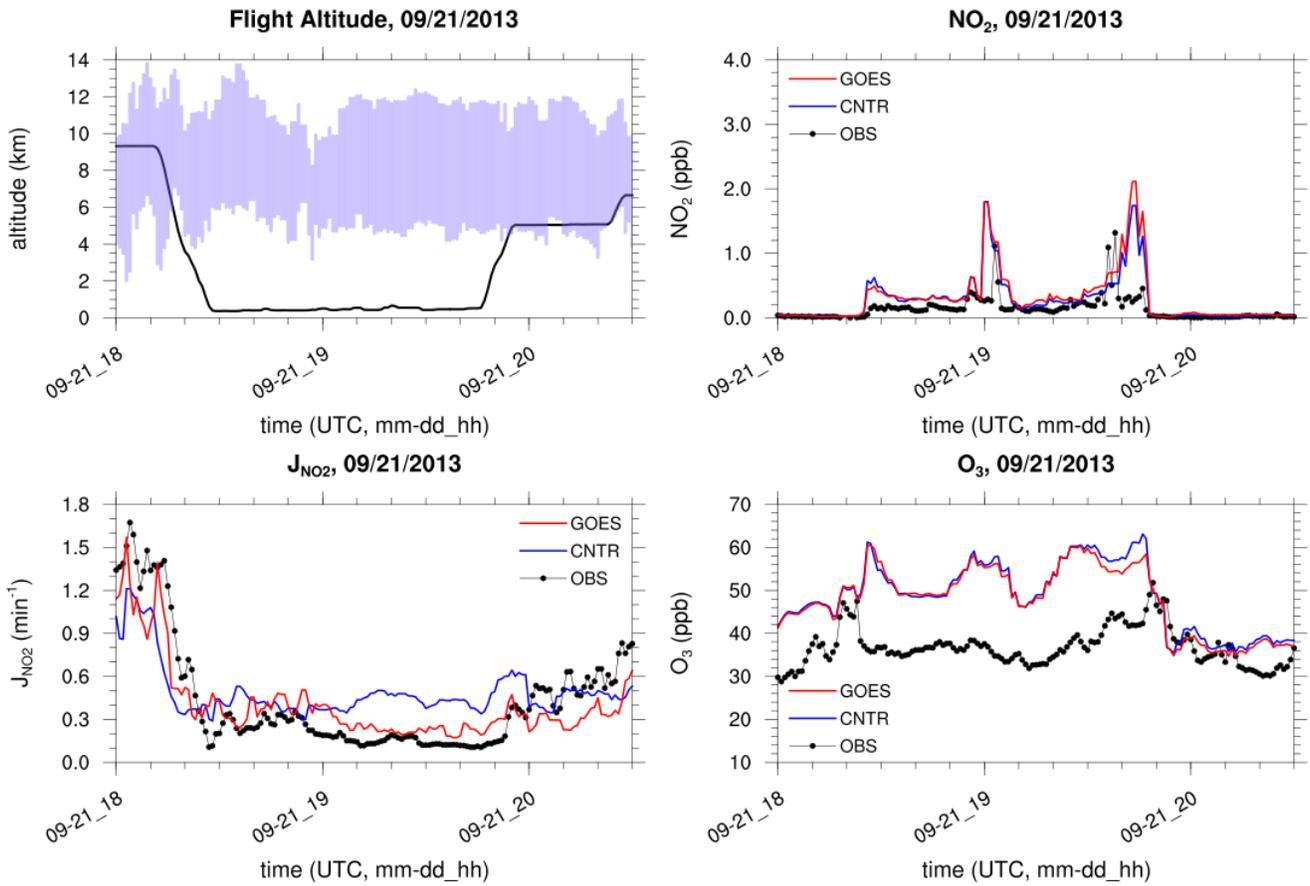


Fig. P3. An example for SEAC⁴RS campaign (21 September 2013). (Top, left) Timeseries of aircraft altitude. Shading indicates cloud boundaries from GOES retrievals. (Top, right) Timeseries of NO₂ concentration. (Bottom, left) Timeseries of JNO₂. (Bottom, right) Timeseries of O₃ concentration. Note that the shorter time period than the whole flight-day time period is shown here to highlight the effects of clouds.

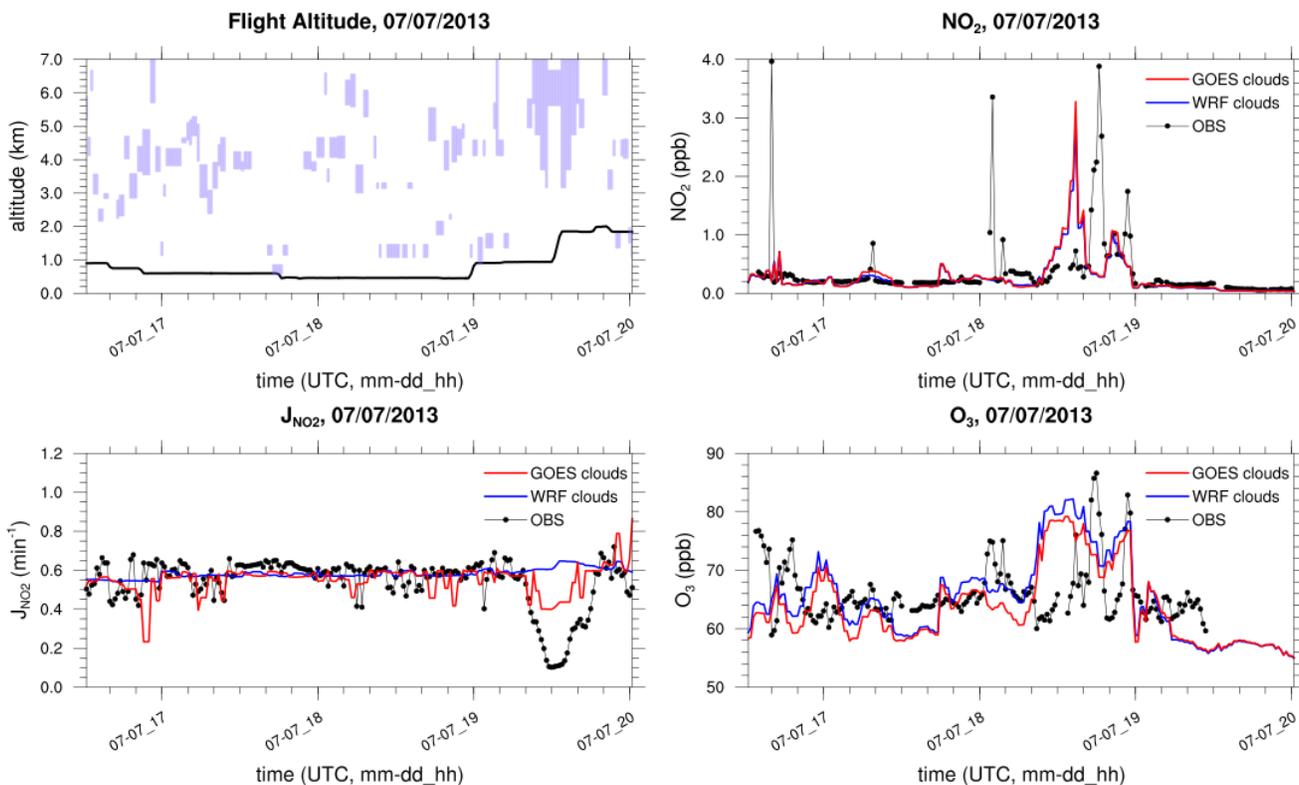


Fig. P4. Same as Fig. P3, but for a NOMADSS example (7 July 2013).

2. Ozone production efficiency evaluation

The ozone production efficiency (OPE) is evaluated against SEAC⁴RS observations over the southeast US (Fig. P5). The OPE from the model (14.3) is similar to that from the observations (14.0), showing a good performance of our model. Both OPE values are smaller than the values shown in Travis et al. (2016); 16.7 for their model and 17.4 for SEAC⁴RS observations. Even though we use the same criteria as in Travis et al. (2016) such as altitudes lower than 1.5 km and $\text{NO}_Z = \text{HNO}_3 + \text{PAN} + \text{aerosol nitrate} + \text{alkyl nitrates}$, we do not exclude urban plumes and open fire plumes because 1) we are interested in urban areas and urban plumes and 2) the condition of filtering out open fire plumes (using CH_3CN) may not be appropriate to apply to our relatively high resolution simulations. When urban plumes and open file plumes are excluded in the SEAC⁴RS observations (not shown), we find a very similar value of observed OPE (17.46) as in Travis et al.'s value (17.4).

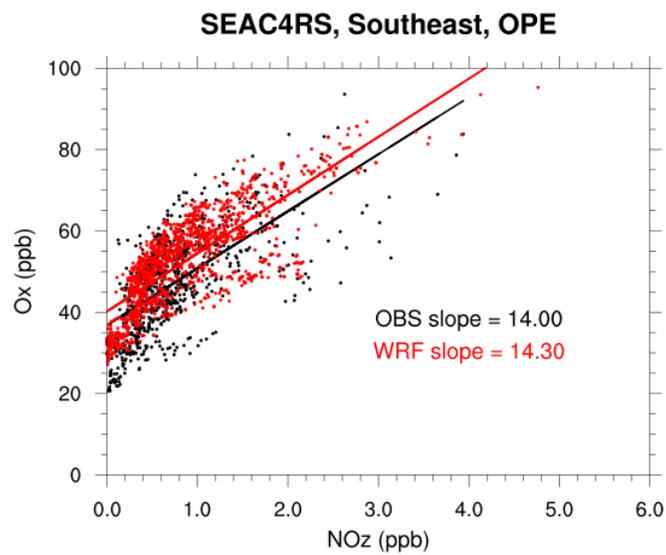


Fig. P5. Ozone production efficiency (OPE) below 1.5 km over the southeast US for SEAC⁴RS campaign. O_X is O₃ + NO₂, and NO_Z is HNO₃ + PAN + aerosol nitrate + alkyl nitrates.