Responses to Reviewer 1's comments

We thank the reviewer for providing valuable comments. We have improved our manuscript following his/her suggestions and comments. Please find our responses below. Reviewer's comments are highlight in blue.

Major comments:

C1) Evaluate vertical profiles of O_3, NO_X, NO_Z from field campaigns.

Figure R1 shows the average vertical profiles of O_3, NO_X, HNO_3 (top panels) and their root-mean-square-error (RMSE) from SEAC^{4}RS campaign. NO_Z species except for HNO_3 are not saved in the WRF outputs along the flight tracks (at 1-min time intervals), and thus only HNO_3 is compared here. The modeled vertical profiles of O_3, NO_X, and HNO_3 are in a reasonable agreement with observations. The large deviations in O_3 near the surface were also reported in previous studies such as Travis et al. (2016). The campaign average differences in vertical profiles of O_3 between CNTR and GOES simulations are small as the aircraft measurements are mostly made in rural environments or high altitudes where O_3 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. However, it is seen that the cloud corrections slightly reduce O_3 RMSE in general particularly below ~1 km altitude. Some examples from SEAC^{4}RS and NOMADSS flights show that the effects of cloud correction can be considerable if the aircraft flew over relatively high-NO_X regions under cloudy conditions (please see Figs. P3 and P4 in the responses to Dr. Kasibhatla's comments). Even though clouds were present during some flights, the cases allowing to estimate their effects are sparse as aircrafts usually avoid flying on heavily cloudy days. So, when all the data are averaged, the effects of cloud correction are expected to be small. The average profiles and RMSE of NO_X and HNO_3 from CNTR and GOES simulations are also very similar to each other.

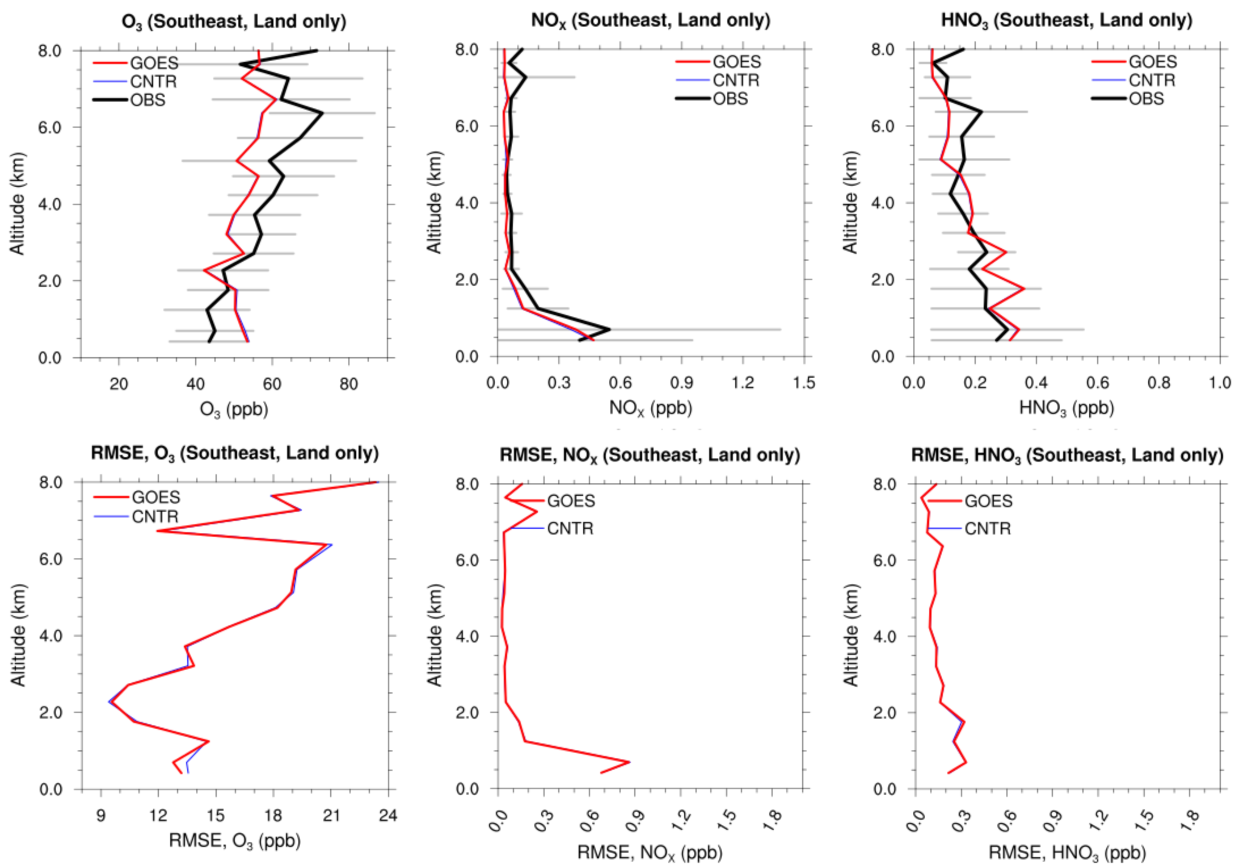

Fig. R1. (Top, from left to right) Averaged vertical profiles of O_3, NO_X, and HNO_3, respectively, for SEAC^{4}RS measurements. The aircraft data over land within the southeast region (latitude: 25–40°N, longitude: 95–70°W) are only used for the averages. (Bottom, from left to right) The corresponding root-mean-square-error (RMSE) of O_3, NO_X, and HNO_3, respectively.

C2) I have some reservations concerning the analyses involving NOx and VOC limited regimes in Sections 5.4 and 5.5 (although I like the last paragraph in Section 5.5). This manuscript has specific conclusions for VOC and NOx limited regimes. There are urban areas that are NOx limited. I suspect the NOx limited conclusions are heavily weighted toward rural areas and don't accurately represent polluted urban and suburban areas. I suggest binning sites based on ozone concentrations and then performing the analyses described in Sections 5.4 and 5.5 so the reader can compare VOC and NOx limited sites with similar ozone concentrations as well as VOC limited sites over a range of ozone concentrations and NOx limited sites over a range of ozone concentrations. Perhaps this can be done by binning the sites based on the peak maximum 8 hour average ozone concentration throughout the year (i.e., bin 1: peak MDA8>75, bin 2: peak MDAO3 between 70-75, …). It may be interesting to include the sites that fall into the transitional zone in your analysis. Include a figure showing delta O3 / delta NOy to identify NOx and VOC limited regimes.

Thanks for providing these valuable suggestions. We agree that analyses for the VOC- and NO_X-limited sites that have similar ranges of O_3 concentration would provide more fair comparisons. Therefore, we performed additional analyses of the sensitivity of maximum daily 8-h average (MDA8) O_3 bias to cloud correction in VOC- and NO_X-limited regimes that have similar peak MDA8 O_3 values. As O_3 concentration is high in summertime, we only consider the period of June through September 2013. All the EPA sites are sorted into several bins based on peak (maximum) MDA8 O_3 concentration during the period of June–September 2013. Figure R2 shows the same analysis as done in Fig. 7 but for various MDA8 bins. Please note that the COD threshold of 30 is not shown here because the number of data with this threshold is too small (generally less than ~50) when the sites are grouped into bins. It is clearly seen that the effects of cloud correction on reducing O_3 bias are greater in VOC-limited regimes than NO_X-limited regimes for all the bins although the degree is somewhat different among the bins. For the NO_X-limited sites that have peak MDA8 O_3 > 75 ppb, the maximum decrease in O_3 bias due to cloud correction is ~3.5 ppb and this value is similar to that (~3 ppb) found in the analysis for all the sites (Fig. 7d in the manuscript). The NO_X-limited sites with peak MDA8 O_3 > 75 ppb are mostly located near the major US cities or the state of California (Fig. R3). Those sites are likely characterized by polluted urban or suburban areas. For the NO_X-limited sites with peak MDA8 O_3 of 60–65 ppb that are mostly located in rural environments, for example, the effects of cloud correction on reducing O_3 bias (maximum value of ~2 ppb) are smaller than those seen for the sites with peak MDA8 O_3 > 70 ppb (maximum value of ~4 ppb). So, even for NO_X-limited regimes it can be said that the effects of cloud correction are larger in more polluted areas. Still, however, the effects of cloud correction are larger in VOC-limited regimes than NO_X-limited regimes. Therefore, our conclusions originally drawn in the manuscript remain unchanged. We mentioned the results of this analysis in the manuscript as follows.

"*We performed additional analysis by dividing VOC- and NO_X-limited sites into groups that have similar ranges of peak MDA8 O_3 concentration during the period of June–September 2013 (Fig. S3). All sites are grouped into bins with peak value of MDA8 O_3 ranging from larger than 75 ppb, 70–75 ppb, 65–70 ppb, 60–65 ppb, to smaller than 60 ppb. The maximum reduction in O_3 bias due to cloud corrections is obtained for the VOC-limited sites with peak MDA8 O_3 of 65–70 ppb and reaches ~8 ppb. The maximum reduction for NO_X-limited sites, on the other hand, is ~4 ppb and is found for the sites with peak MDA8 O_3 of 70–75 ppb. Although the* degree *of the O_3 bias reduction varies somewhat among the bins for a given ozone regime, the effects of cloud correction on O_3 bias reduction remain larger in VOC-limited regimes than NO_X-limited regimes*."

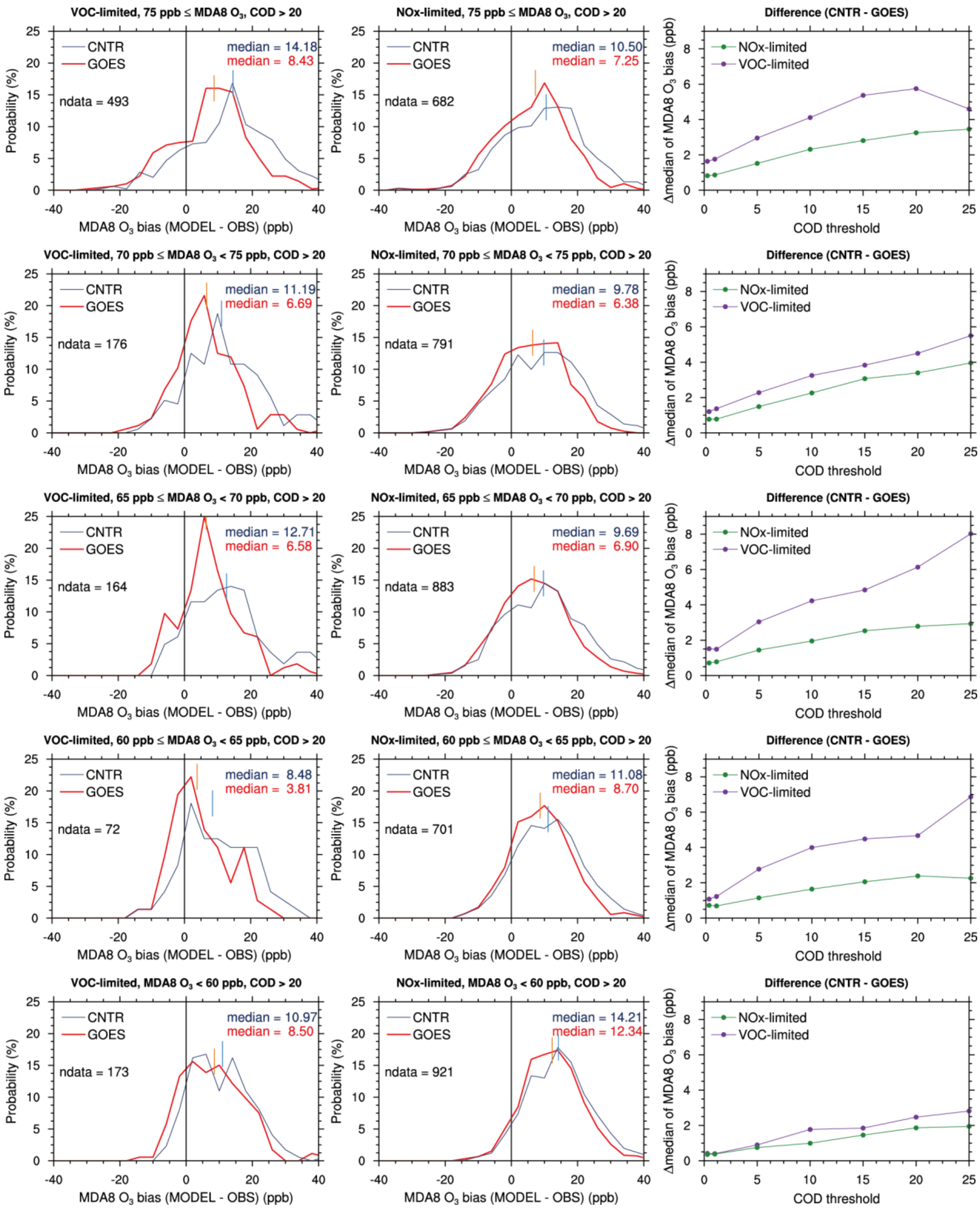

Fig. R2. Similar to Fig. 7 in the revised manuscript but for several bins with different peak MDA8 O_3 ranges.

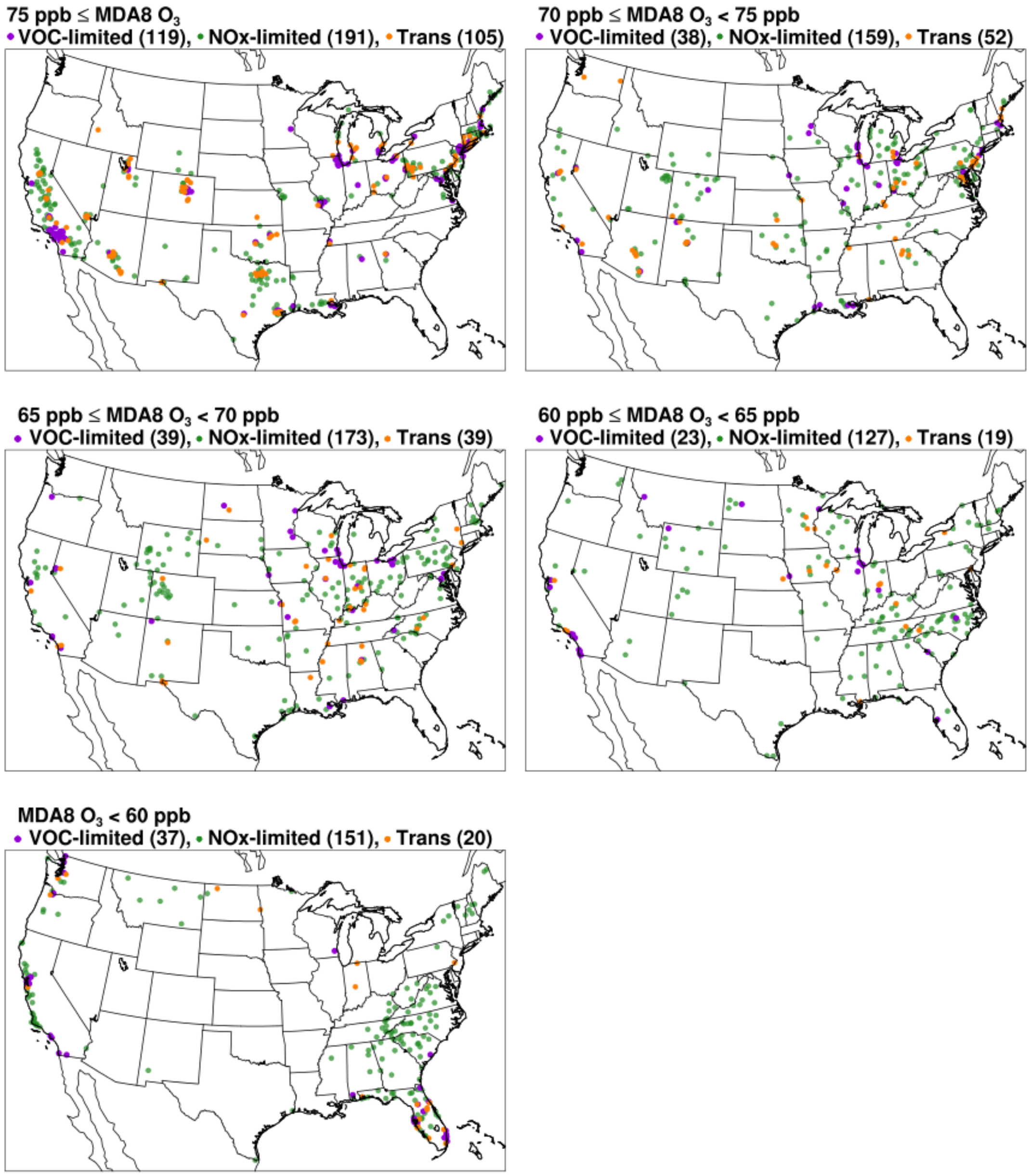

Fig. R3. Maps showing the sites that belong to each peak MDA8 O_3 bin. The number in parenthesis indicate the number of sites in each ozone range. For example, the number of VOC-limited sites with peak MDA8 O_3 > 75 ppb is 119.

In addition, the sites that fall into the transitional zone are added in the analysis (Fig. R4). The effects of cloud correction on O_3 bias reduction for the transition sites are in-between those for the VOC-limited regimes and NO_X-limited regimes. This is now explained in the revised manuscript.

“*Note that the results for the sites in transitional zone (the slope of $\Delta O_3/\Delta NO_y$ is 4–6) showed that the effects of cloud in the transitional zone are intermediate; that is, larger than those for NO_X-limited regimes but smaller than those for VOC-limited regimes (not shown).*”

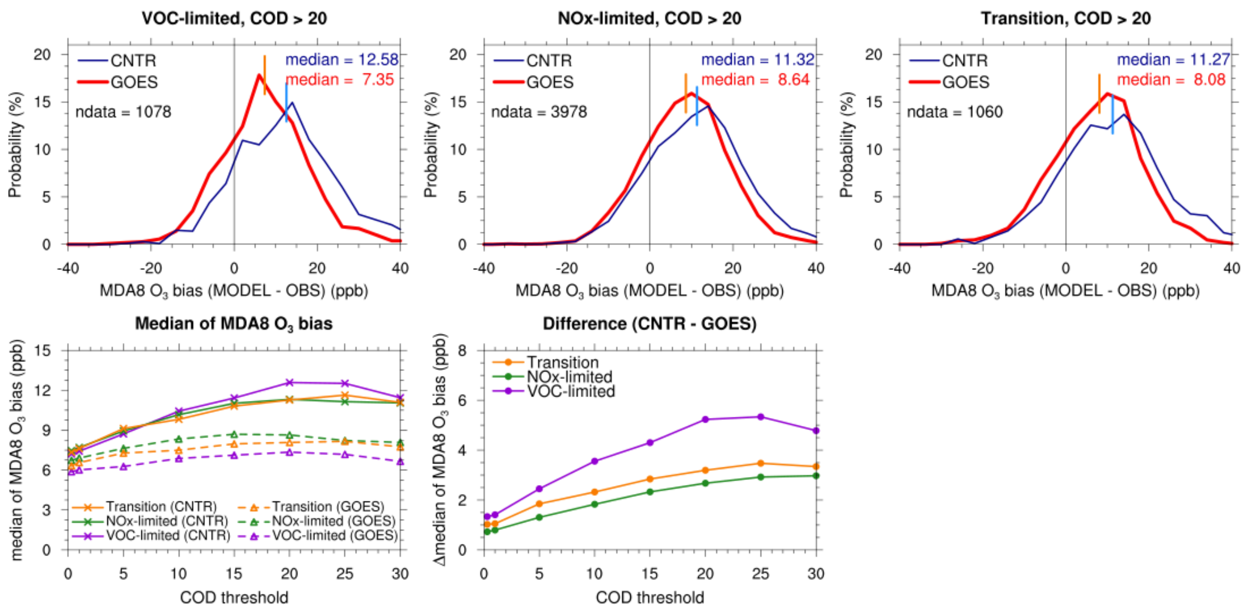

Fig. R4. Same as in Fig. 7 but with the results for transitional zone.

Examples of scatter plots of O_3 and NO_y, which are used to identify VOC- or NO_X-limited sites, are shown in Fig. R5 and following the reviewer’s comment we included this in the supplementary material (Fig. S1).

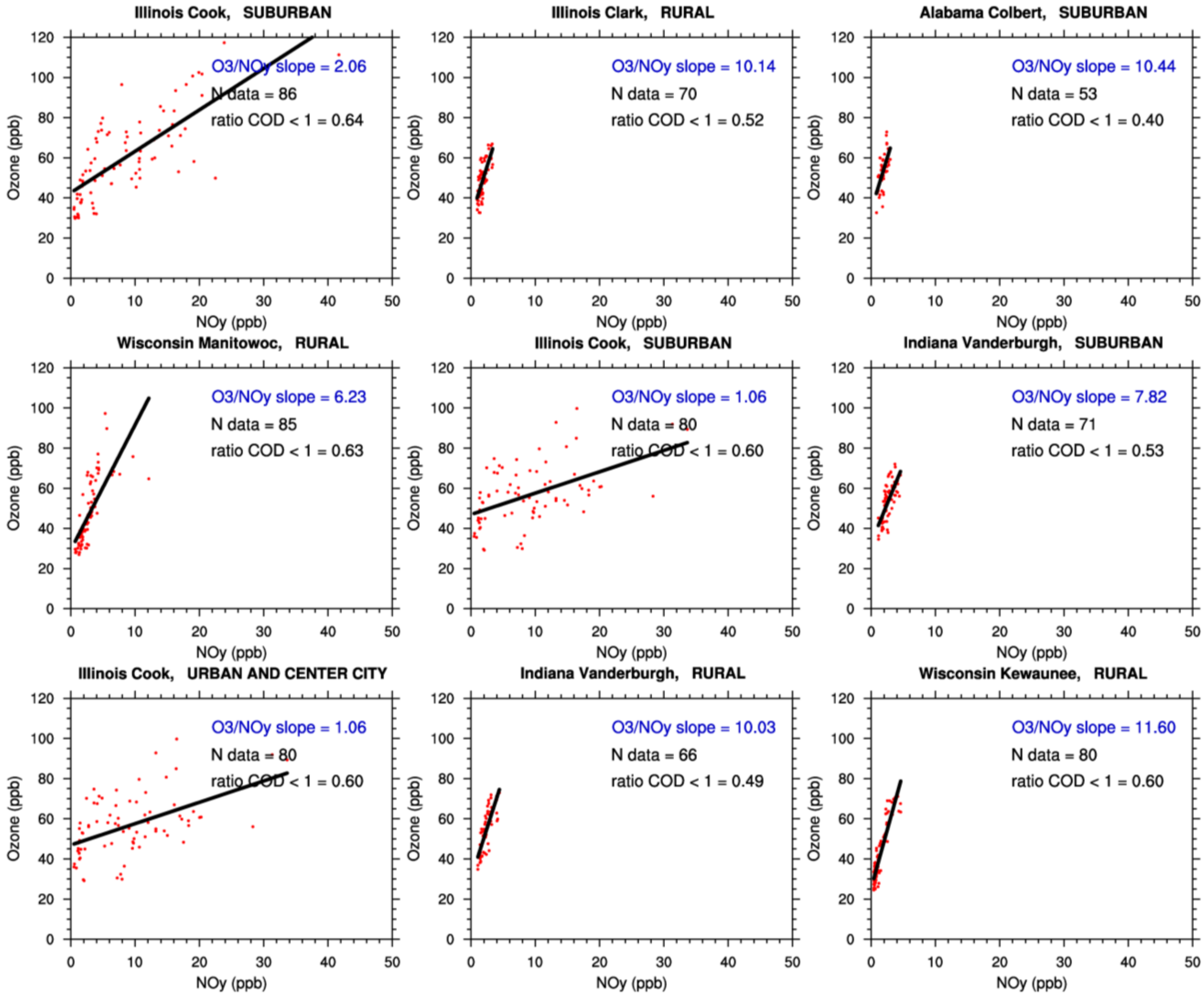

Fig. R5. Scatter plots of O_3 and NO_y. The thick black line indicates the linear regression coefficient. The modeled O_3 and NO_y concentrations at 15–16 local time under clear sky conditions (hourly COD < 1) in the CNTR simulation are used for analysis. On the title heading, the first and second words indicate the state and the county of the site. The third one indicates the type of the site defined by EPA.

Minor comments:

C3) Abstract, line26: Remove mention of "robust with respect to the choice of the microphysics scheme." Only 2 microphysics schemes were tested.

We have removed that part following the reviewer's comment.

C4) Page 5, lines 89-91: Why skip pixels to create an 8km product? Why not leave the product at 4 km?

The 8-km products are sampled every other pixel (4-km pixel) to save processing time. It does not affect the statistics of the analysis.

C5) Page 9, line 181: Change “and with fire” to “and fire”
It is changed.

C6) Page 10, line 189: Change “(Sillman and He (2002)” to “Sillman and He (2002)”
It is corrected.

C7) Page 11, lines 203-204: Change “wrong clouds (that are not present in reality)” to “clouds that are not present in reality”
It is changed.

C8) Page 11, lines 204-205: Re-word this sentence.
It is revised as follows.
“*The overall bias, (A+B)/(A+C), is 0.789 and this means that the WRF underestimates the frequency of cloudy skies.*”

C9) Page 11, line 207: change “except for the mountain regions and northwestern US” to “except for parts of the Rocky Mountains and the Pacific Northwest.”
It is changed.

C10) Page 11, line 208: Change “in the central” to “in central”
It is changed.

C11) Page 13, lines 252-253: Change “This is” to “These reductions are”. Provide a further explanation of this claim.
It is changed following the reviewer’s suggestion. This claim was based on the histograms separating the cloud conditions into below, above, and inside cloud conditions (Fig. R6), which are not shown in the manuscript. The reductions of larger errors with model-to-observation ratio of greater than 2 are due to the reductions under below- and inside-cloud conditions. We elaborate the reasons in the revised manuscript as follows.
“*This is because the number of data influenced by considerably thick clouds is larger in SEAC^{4}RS than in NOMADSS and the measurements in the presence of those thick clouds were mostly made under below-cloud or inside-cloud conditions.*”

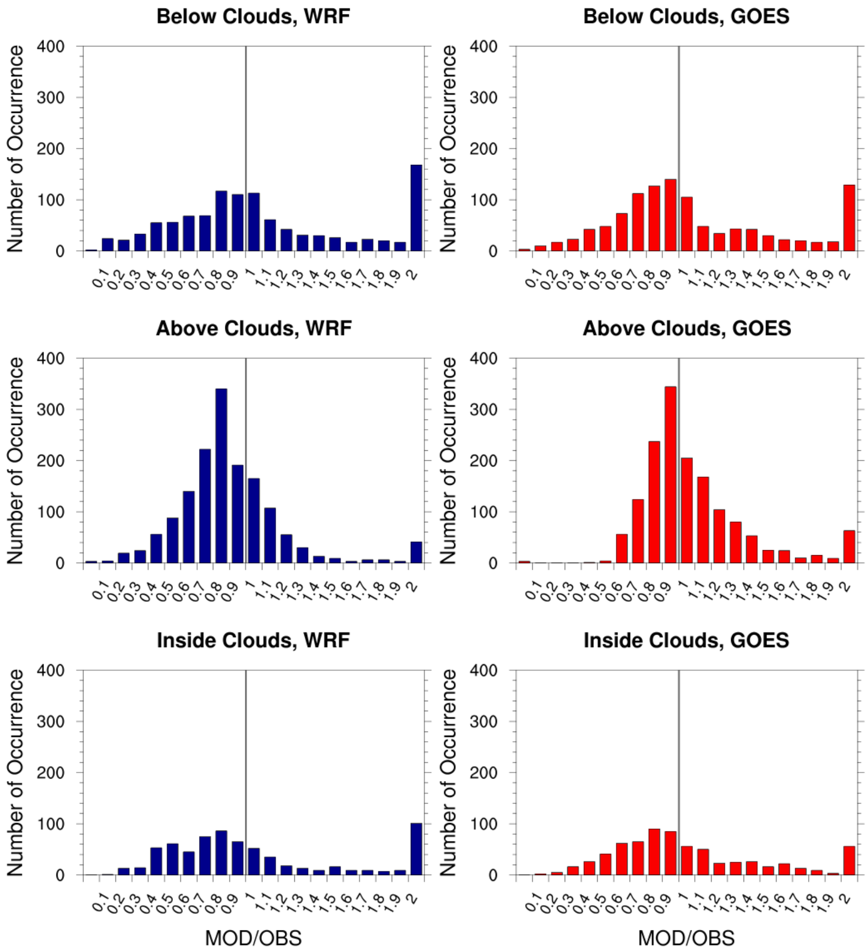

Fig. R6. Histogram of model-to-observation JNO_2 ratio for SEAC4RS under (top) below, (middle) above, and (bottom) inside cloud conditions.

C12) Page 13, lines 254-260: This text states that NOMADSS has a larger mean model-to-observation ratio than SEAC4RS. This is not the case based on Figure 3.

The text was intended to indicate the above cloud conditions. As Fig. R7 shows, the performance in the GOES simulation is not greatly improved even though the satellite clouds are used. The effects of cloud correction for above-cloud conditions for NOMADSS are different from those for SEAC^{4}RS (Fig. R6 middle row). Given that the histograms of Fig. R7 are not included in the manuscript, we have deleted this part in the revised manuscript to avoid confusion.

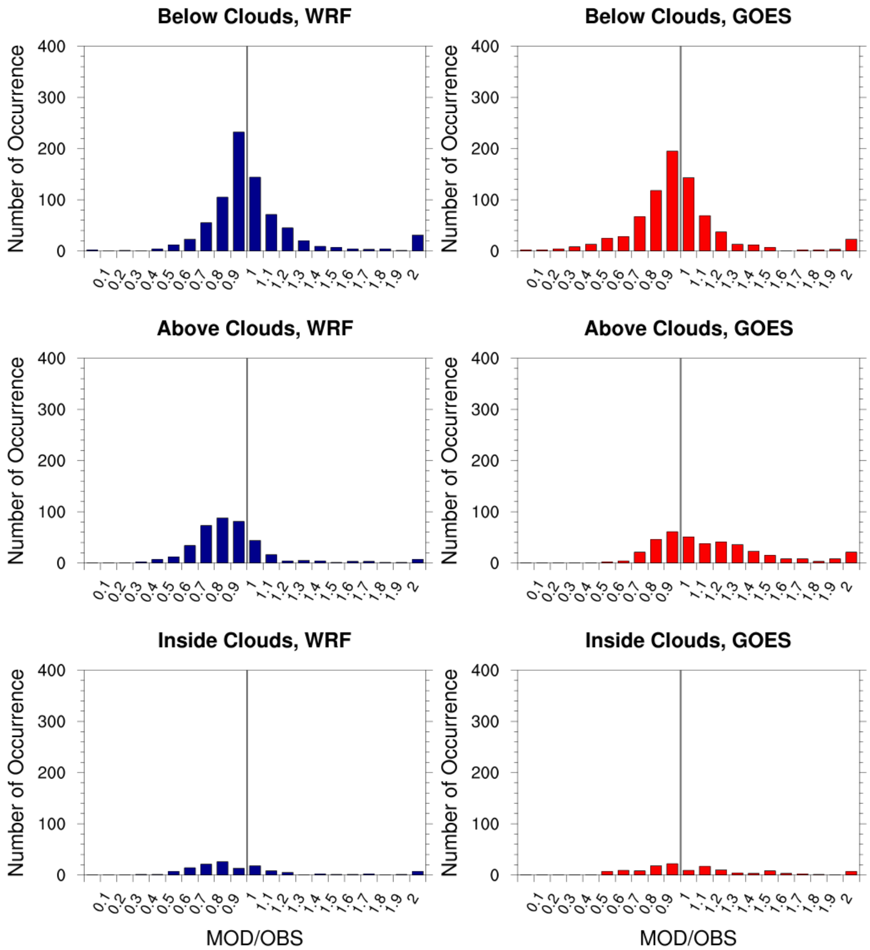

Fig. R7. Histogram of model-to-observation JNO_2 ratio for NOMADSS under (top) below, (middle) above, and (bottom) inside cloud conditions.

C13) Section 5.2: Calculate and discuss model-observations comparison statistics. Use maximum daily 8 hour average O_3 (MDAO3) instead of 8hr average ozone between 10-17 LST.

We have added a discussion of the statistics in the manuscript as requested by the reviewer. Indeed, the root-mean-square-error (RMSE) and correlation coefficient are compared. Both the RMSE and correlation coefficient show a better performance when satellite clouds are used (GOES simulation) than when model clouds are used (CNTR simulation). The RMSE of MDA8 O_3 in the GOES (CNTR) simulation is 13.2 ppb (16.9 ppb) and the correlation coefficient of MDA8 O_3 in the GOES (CNTR) simulation is 0.5 (0.4). This is now explained in the manuscript: *"The performance of the GOES simulation is found to be better than that of the CNTR simulation as compared to observations: for example, under cloudy conditions (COD > 20, see section 5.4 for the criterion), the root-mean-square error of MDA8 O_3 in the GOES (CNTR) simulation is 13.2 ppb (16.9 ppb) and the correlation coefficient of MDA8 O_3 in the GOES (CNTR) simulation is 0.5 (0.4)."*

The spatial ozone distribution map averaged over the study period (Fig. 5) is replaced with MDA8 O_3 in the revised manuscript. The result with MDA8 O_3 is very similar to that shown with daytime 8-h average (10–17 LST) O_3.

C14) Section 5.3: If you have a simulation with "photolysis with WRF clouds and PAR with GOES clouds", this would be interesting to include in this section.
We agree with that. Unfortunately, the current model does not have capability to simulate the setup proposed by the reviewer.

C15) Page 16, lines 316-318 and Figure 6: Difficult to see the relative differences between Figure 6c and 6d. A figure of the absolute value of 6d divided by the absolute value of 6c may be helpful.
Following the reviewer's suggestion, we replaced Fig. 6d with a plot showing the ratio of difference in O_3 between EMIS_BVOC and GOES (previously Fig. 6d) to difference in O_3 between CNTR and GOES (Fig. 6c). The description of this figure is as follows.
"*Figure 6d shows the relative O_3 difference between EMIS_BVOC and GOES simulations to O_3 difference between CNTR and GOES simulations (Fig. 6c).*"

C16) Page 16, lines 318-320: Ozone difference of a simulation with photolysis with WRF clouds and PAR with GOES clouds minus GOES may or may not be 80% of CNTR-GOES. I suggest rewording this sentence to "The contribution of changes in BVOC emissions is ~20% compared to changes of BVOC emissions and photolysis rates using GOES observations."
It is revised based on the reviewer's suggestion:
"*The average contribution of changes in BVOC emissions over land is ~20% compared to changes of BVOC emissions plus photolysis rates using GOES satellite clouds.*"

C17) Figure 4: Use EST or CST, not LST. Map shows areas in the eastern and central time zone.
LST is changed to CST.

C18) Figure 5: Show 3 panels with a CNTR, GOES, and difference plot (CNTR-GOES). Include observations overlayed on-top of the CNTR and GOES plots.
The reason why we show only the result of CNTR simulation is that the spatial distribution of average O_3 in GOES simulation is similar to that in CNTR simulation although the ozone levels are different (Fig. R8). We mentioned the reason why the result of GOES simulation is not shown here in the revised manuscript. In addition, adding observations on the map makes the plot very complicated as the number of sites is ~1300. Lots of sites are closely located to each other as shown in Fig. R3. When all the sites in the bins in Fig. R3 are plotted and overlayed on a map, readers will not be able to see the values of observations.

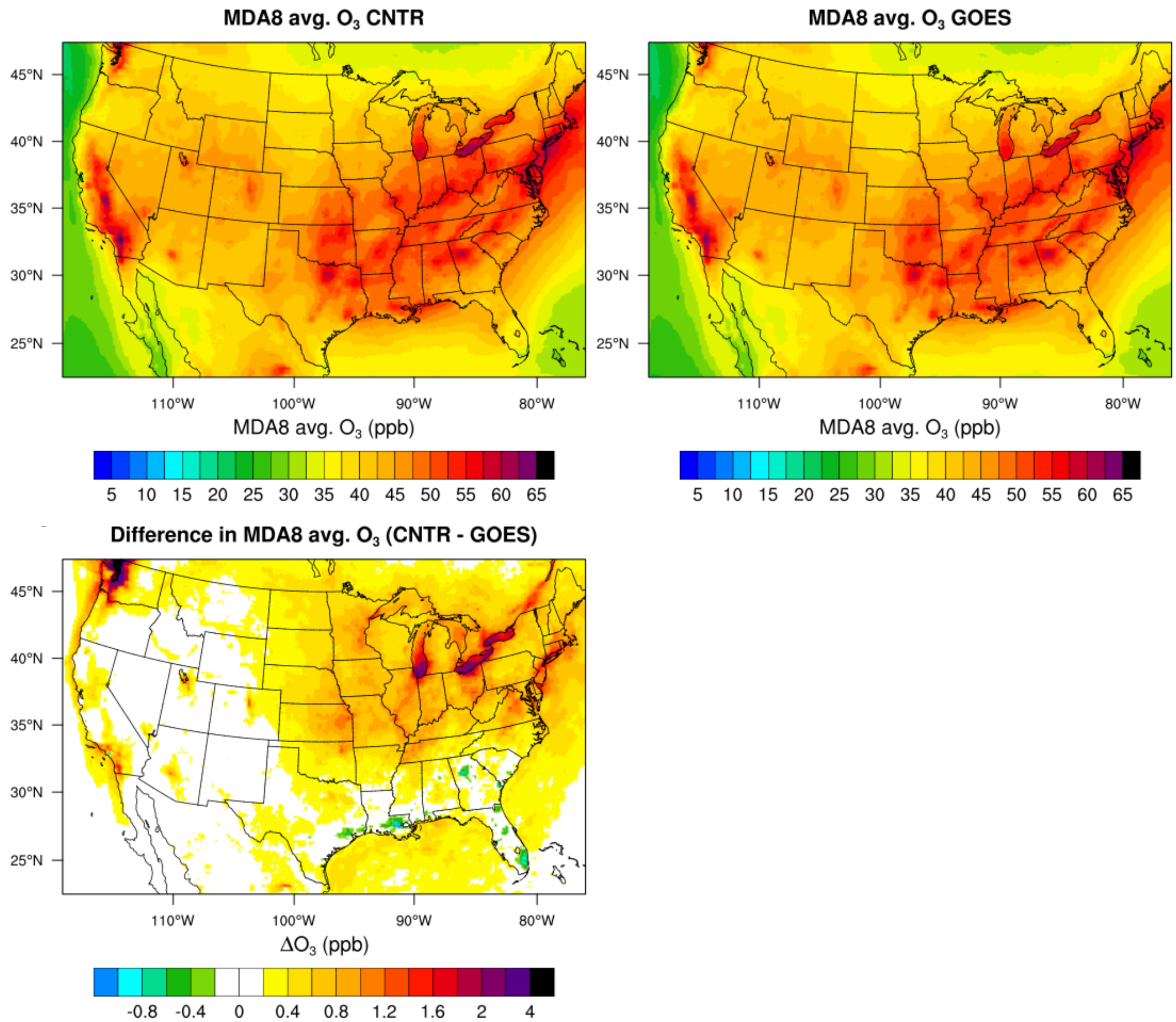

Fig. R8. Spatial distribution of MDA8 O_3 at the lowest model level averaged over the study period (top, left) in the CNTR simulation and (top, right) in the GOES simulation. (Bottom, left) Difference in MDA8 O_3 between CNTR and GOES simulations.