Responses to Reviewer 3's comments

Thank you so much for providing valuable comments. We have improved our manuscript following your suggestions and comments. Please find our responses below. Your comments are highlight in blue.

C1) The analysis is based primarily on one set of model physics (Morrison microphysics and Grell 3-D convection). The authors do test the sensitivity of the results to a second microphysics scheme (Thompson) and found little difference. However, the simulation is for summer conditions (June to September), when a significant amount of cloudiness is due to convection. Therefore, there should be a sensitivity test also run with a second convective scheme. I would suggest running the relatively new Grell-Freitas scheme. From what I have seen, this scheme will produce more clouds.

Following reviewer's suggestion, we have performed sensitivity tests with Grell-Freitas scheme. As done for microphysics scheme, a period of 10 days (3–12 July 2013) was considered. An example showing spatial distribution of cloud optical depth from the two cumulus parameterization schemes are presented in Fig. T1. In general, the spatial patterns and the location of large systems are similar to each other. The Grell-Freitas scheme produces more and/or thicker clouds in some regions such as the north Michigan and the south Ohio than the Grell-3D scheme. However, the Grell-Freitas scheme produces fewer and/or thinner clouds in other regions such as the east Texas and North Carolina. In Fig. T2, the histograms of cloud optical depth obtained for the 10-day period from Grell-Freitas scheme (left) and from Grell-3D scheme (right) show that the distributions of cloud optical depth are in general similar to each other. The Grell-Freitas scheme tends to produce fewer clouds with small or moderate cloud optical depth. Figure T3 shows that the degree of cloud correction in reducing O_3 bias is larger in VOC-limited regimes than in NO_X-limited regimes in the simulation with Grell-Freitas scheme, and thus the conclusions originally drawn remain unchanged.

We included the summary of this discussion above in the revised manuscript and figures (Figs. T2 and T3) in the supplementary materials.

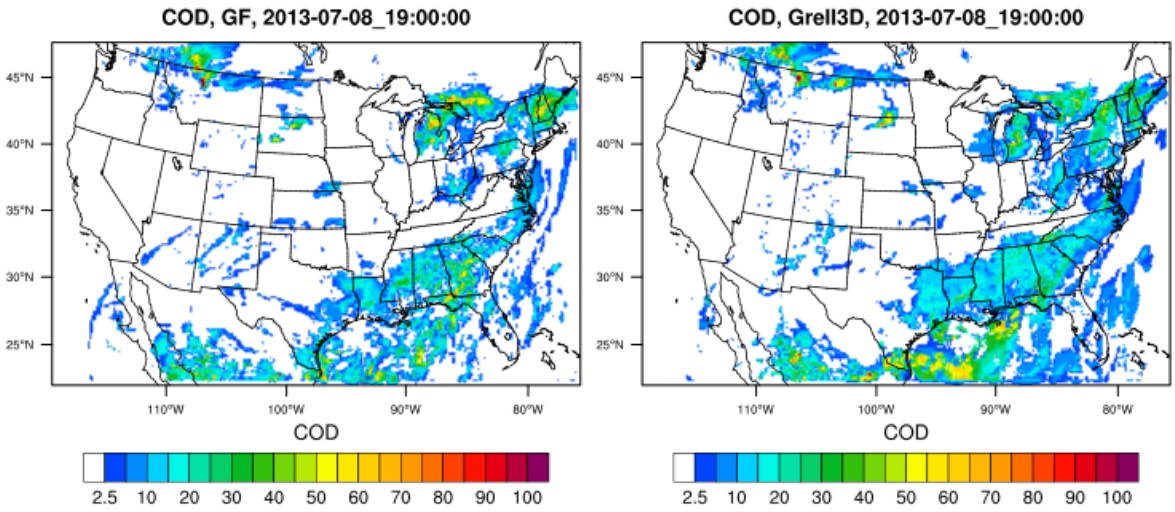

Fig. T1. Cloud optical depth (COD) at 19 UTC 8 July 2013 using the (left) Grell-Freitas scheme and (right) Grell-3D scheme. Both simulations use the Morrison microphysics scheme.

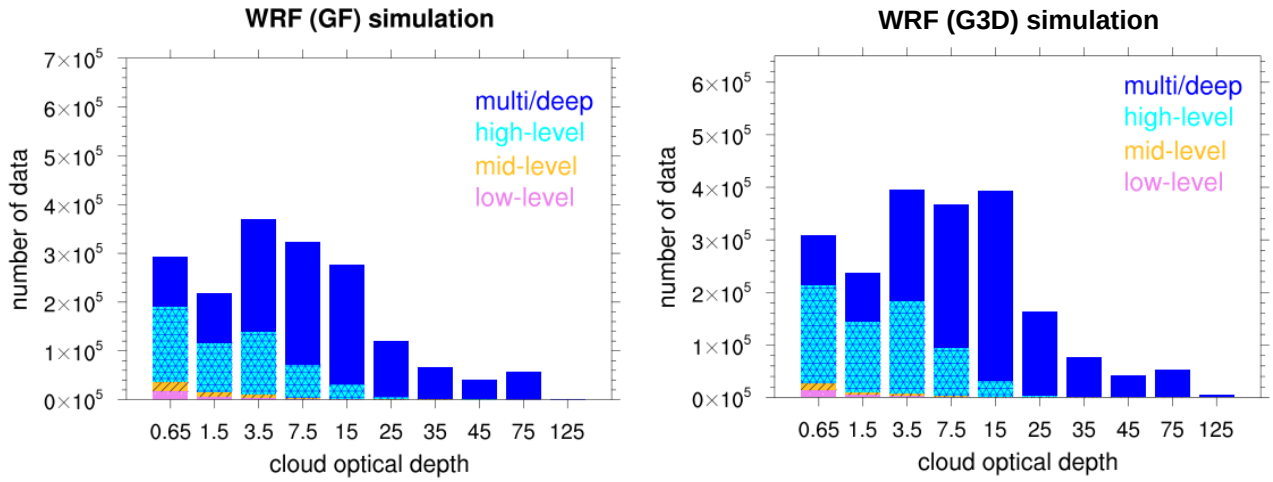

Fig. T2. Histogram of hourly cloud optical depths during the daytime (16–23 UTC) over CONUS (land only) for the period of 3–12 July 2013 from simulations with the (left) Grell-Freitas scheme and (right) Grell-3D scheme.

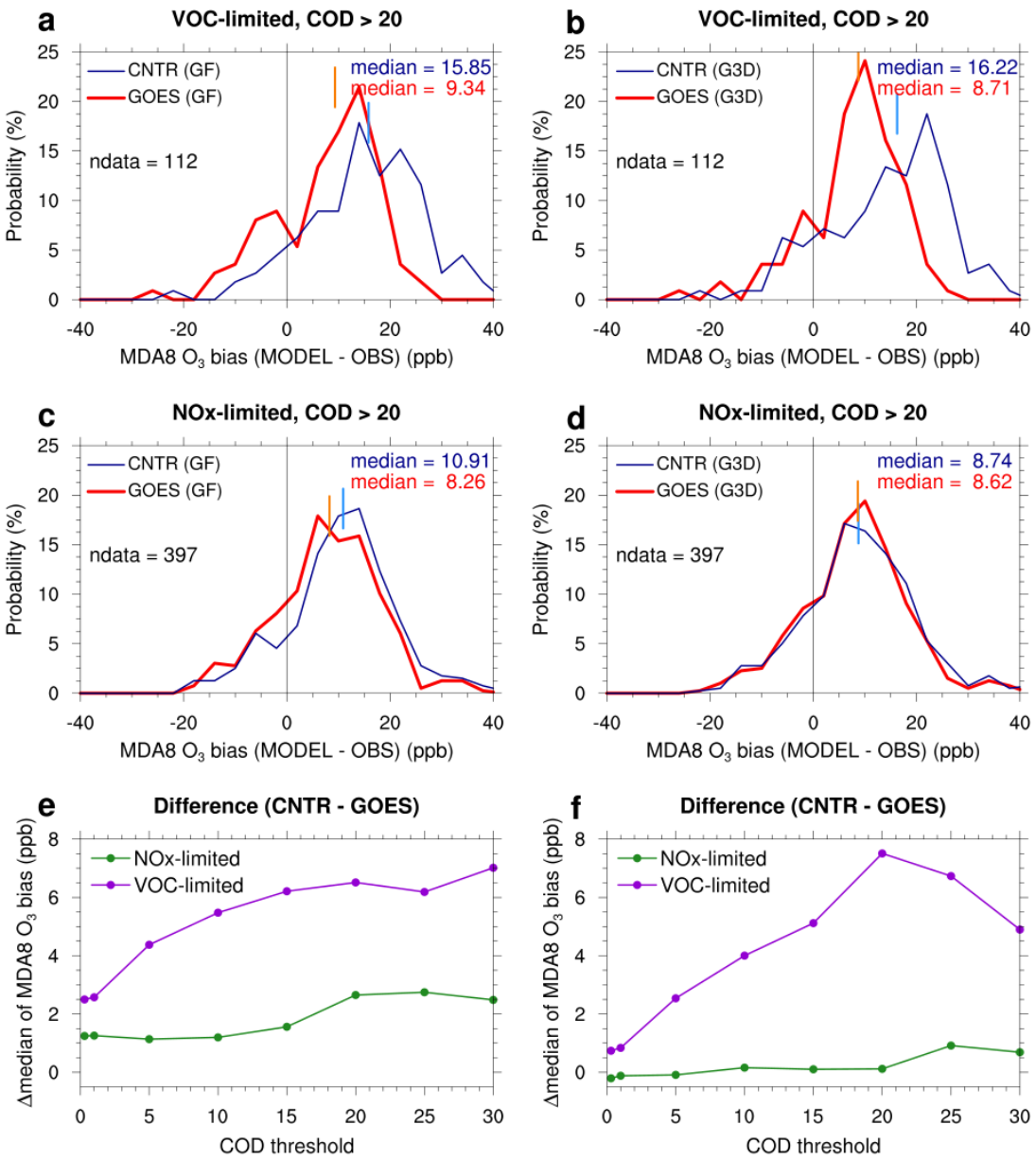

Fig. T3. (Left column) The results of 3–12 July 2013 WRF-Chem simulations with Grell-Freitas scheme. (a/c) Probability density function of MDA8 O_3 bias (model value minus observation value) for VOC/NO_X-limited regime under cloudy sky conditions defined with COD threshold of 20 in the simulations with the Grell-Freitas scheme. (b/d) Same as (a/c), but for the simulations with the Grell-3D scheme. (e and f) Difference in median values of MDA8 O_3 bias between the two simulations with respect to COD threshold (i.e., CNTR minus GOES) for the simulations with the Grell-Freitas and with the Grell-3D schemes, respectively.

C2) In Section 2.3 the authors use the delta O3 to delta NOy ratio to determine VOC-limited and NOx-limited conditions. How is delta NOy determined at EPA monitoring sites? NOy is not

routinely measured at these sites. Even true NOx is measured at only some small fraction of the O3 monitoring sites. This issue needs explanation or substantive revision.

NO_y used in this study is the modeled NO_y and O_3 is also modeled O_3. As you indicated, NO_y is not routinely measured, so the sites having NO_y measurements are very limited. Therefore, we could not rely on NO_y observations. We included the following sentence in the revised manuscript.

"*Note that modeled O_3 and NO_y in the CNTR simulation are used to determine whether an EPA site is in VOC-limited or NO_X-limited regime because NO_y measurements are available for limited sites*."

In addition, we included examples showing how to determine VOC-limited or NO_X-limited sites in the supplementary materials (Fig. S1).

Minor comments:

C3) line 127: Which year NEI NOx was too high? Did Travis et al. indicate all NOx emission types were overestimated, or was it primarily mobile sources?

Travis et al. (2016) used 2011 NEI emissions and adjusted to 2013. They reduced NOx emissions from mobile and industrial sources (all sources except for power plants). Based on the references mentioned in Travis et al. (2016), several local studies reported that NEI NOx emissions for mobile sources are high by a factor of 2 or more (Castellanos et al, 2011; Fujita et al., 2012; Brioude et al., 2013; Anderson et al., 2014).

In our present study, we reduced NO_X emission from all anthropogenic sources by 40% based on the analysis of Travis et al. (2016), and this is mentioned in the revised manuscript.

Reference

Castellanos, P. Marufu, L. T., Doddridge, B. G., Taubman, B. F., Schwab, J. J., Hains, J. C., Ehrman, S. H., and Dickerson, R. R.: Ozone, oxides of nitrogen, and carbon monoxide during pollution events over the eastern United States: An evaluation of emissions and vertical mixing, J. Geophys. Res., 116, D16307, doi:10.1029/2010JD014540, 2011.

Fujita, E. M., Campbell, D. E., Zielinska, B., Chow, J. C., Lind-hjem, C. E., DenBleyker, A., Bishop, G. A., Schuchmann, B. G., Stedman, D. H., and Lawson, D. R.: Comparison of the MOVES2010a, MOBILE6.2, and EMFAC2007 mobile source emission models with on-road traffic tunnel and remote sensing measurements, J. Air Waste Manage., 62, 1134–1149, doi:10.1080/10962247.2012.699016, 2012.

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Anderson, D. C., Loughner, C. P., Diskin, G., Weinheimer, A., Canty, T., P., Salawitch, R. J., Worden, H. M., Fried, A., Mikoviny, T., Wisthaler, A., and Dickerson, R. R.: Measured and modeled CO and NOy in DISCOVER-AQ: An evaluation of emissions and chemistry over the eastern US, Atmos. Environ., 96, 78–87, doi:10.1016/j.atmosenv.2014.07.004, 2014.

C4) lines 255 to 260: I don't follow this description of cloud fraction. Please clarify.

This part originally explained the results without showing figures that are relevant to the cloud fraction, but without showing figures we concluded that this part was too confusing to reader, and we decided to remove it. Please see the comment 12 of the first reviewer and our responses.

C5) Section 5.5 describes in detail how the box model calculations show that OH is less sensitive to changes in radiation in the NOx-limited regime. Some statements also need to be made about the effect on P(O3) in the box model.

Figure T4 shows the net chemical production of O_3 in the box model, and the result is consistent with that is found in the WRF-Chem simulations: larger sensitivity of $P(O_3)$ to cloudiness in VOC-limited regimes than NO_X-limited regimes. We briefly included this result in the revised manuscript as follows.

"Note that the net chemical production of O_3 obtained from the box model results also shows a larger sensitivity to cloudiness in VOC-limited regimes than in NO_X-limited regimes (not shown)."

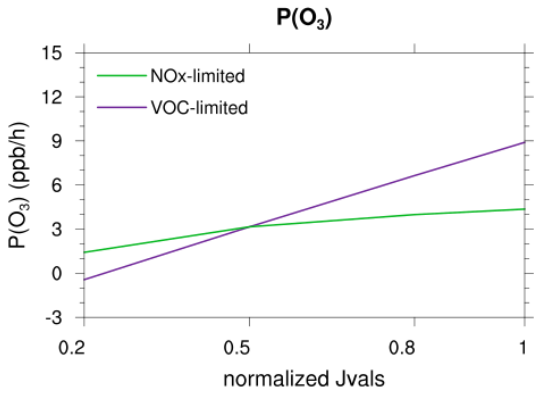

Fig. T4. The net chemical production of O_3 from the box model simulations.