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Supplement of

Ozone and NO_x chemistry in the eastern US: evaluation of CMAQ/CB05 with satellite (OMI) data

T. P. Canty et al.

Correspondence to: T. P. Canty (tcanty@atmos.umd.edu)

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Section 5 mentions two additional model simulations performed in support of the manuscript which we describe in further detail here. Mollner et al. (2010) reported an update to the kinetics that govern the reaction rate of



Figure S1 shows a comparison of the rate of this reaction based on the recommendations of IUPAC (Atkinson et al., 2006), JPL (Sander et al., 2006), and Mollner et al., (2010). The newer study gives a slower rate constant compared to the prior recommends. We have modified the CB05 chemical mechanism to use the results from Mollner et al. (2010). This change is in addition to the modification of alkyl nitrate (NTR) chemistry, a 50% reduction in emissions of NO_x from mobile sources, and an update to biogenic emissions, as described in the manuscript. This scenario is called CMAQ_{OH+NO2} and was performed for a 20 day period spanning the end of July 2007 to the beginning of August 2007. To facilitate a comparison with the results from the CMAQ_{TOT} simulation, we regenerate the scatter plots presented in Figure 3 of the manuscript for this time period. In Figure S2 we show the results from model runs CMAQ_{TOT} and CMAQ_{OH+NO2} for both the DOMINO and GSFC retrievals. Overall, this change has little effect on the model output with, perhaps, a slight improvement in the comparison of modeled and observed column NO₂ for rural regions.

The accommodation coefficient of N₂O₅ (called $\gamma_{\text{N}_2\text{O}_5}$) is most likely overestimated within CB05 (Vinken et al., 2014, Yegorova et al., 2011) and therefore NO₂ may be improperly suppressed. In CMAQ, the calculation of gamma is based on Davis et al. (2008). We have performed a simulation, CMAQ_{N2O5}, with $\gamma_{\text{N}_2\text{O}_5}$ equal to zero and include all of the changes made to the model in the CMAQ_{TOT} scenario. Results from CMAQ_{N2O5} are presented in Fig S2. With this NO_x loss mechanism turned off, we would expect modeled column NO₂ to increase. Even though column NO₂ does rise, this scenario shows very little overall difference compared to CMAQ_{TOT}. This is in agreement with prior studies (Han et al., 2015, Stavrou et al., 2013).

References:

- Atkinson, R., Blauch, D. L., Cox, R. A., Crowley, J., Hampson Jr., R. F., Hynes, R. G., Jenkin, M. E., Rossi, M. J., and Troe, J.: Evaluated kinetic and photochemical data for atmospheric chemistry: Volume I – gas phase reactions of Ox, HOx, NOx and SOx species, *Atmos. Chem. Phys.*, 4, 1461–1738, 2004, Updated at <http://www.iupac-kinetic.ch.cam.ac.uk>, 2006.
- Davis, J. M., Bhawe, P. V., and Foley, K. M.: Parameterization of N₂O₅ reaction probabilities on the surface of particles containing ammonium, sulfate, and nitrate, *Atmos. Chem. Phys.*, 8, 5295– 5311, 2008, <http://www.atmos-chem-phys.net/8/5295/2008/>.
- Han, K. M., Lee, S., Chang, L. S., and Song, C. H.: A comparison study between CMAQ simulated and OMI-retrieved NO₂ 5 columns over East Asia for evaluation of NOx

- emission fluxes of INTEX–B, CAPSS, and REAS inventories, *Atmos. Chem. Phys.*, 15, 1913–1938, doi:10.5194/acp-15-1913-2015, 2015.
- Mollner, A. K., Valluvadasan, S., Feng, L., Sprague, M. K., Okumura, M., Milligan, D. B., Bloss, W. J., Sander, S. P., Martien, P. T., Harley, R. A., McCoy, A. B., and Carter, W. P. L.: Rate of Gas Phase Association of Hydroxyl Radical and Nitrogen Dioxide, *Science*, 330, 646–649, doi:10.1126/science.1193030, 2010.
- Stavrakou, T., Müller, J.-F., Boersma, K. F., van der A, R. J., Kurokawa, J., Ohara, T., and Zhang, Q.: Key chemical NO_x sink uncertainties and how they influence top-down emissions of nitrogen oxides, *Atmos. Chem. Phys.*, 13, 9057–9082, doi:10.5194/acp-13-9057-2013, 2013.
- Vinken, G. C. M., Boersma, K. F., van Donkelaar, A., and Zhang, L.: Constraints on ship NO_x emissions in Europe using GEOS–Chem and OMI satellite NO₂ observations, *Atmos. Chem. Phys.*, 14, 1353–1369, doi:10.5194/acp-14-1353-2014, 2014.
- Yegorova, E. A., Allen, D. J., Loughner, C. P., Pickering, K. E., and Dickerson, R. R.: Characterization of an eastern US severe air pollution episode using WRF/Chem, *J. Geophys. Res.*, 116, D17306, doi:10.1029/2010JD015054, 2011.

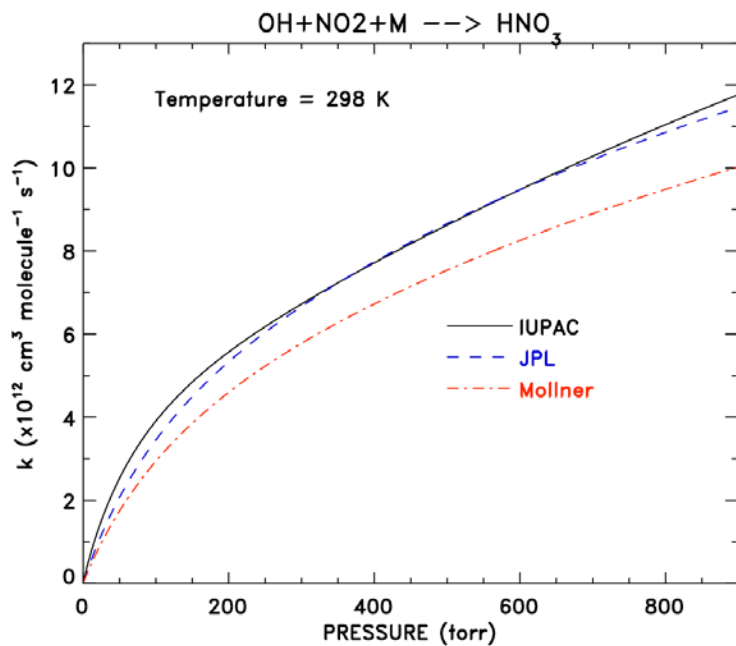


Fig S1. Reaction rate of OH+NO₂+M at 298K based on the recommendations from IUPAC (black), JPL (blue, dashed) and Mollner et al. 2010 (red, dashed dot).

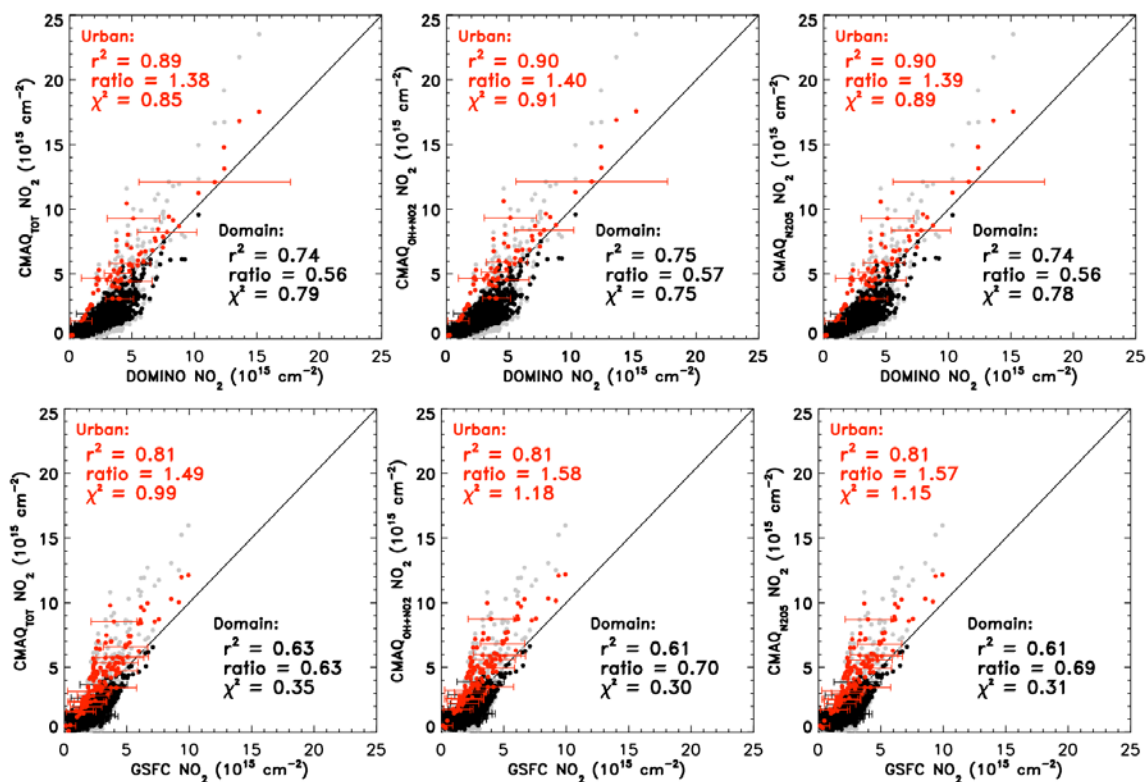


Fig S2. Scatter plots of model output to both the DOMINO (top panels) and GSFC (bottom panels) retrievals of tropospheric column NO₂. Results span late July/early August 2007. Gray points represent the results shown in Fig. 2 of the manuscript. Red points represent areas where the model is at least 25% greater than observations. Results from model scenario CMAQ_{TOT} (left panels) for this time period are shown for comparison with results presented in the manuscript. CMAQ_{OH+NO2} (center panels) includes the modifications made to CMAQ_{TOT} as well as a change in the reaction rate of OH+NO₂. CMAQ_{N2O5} (right panels) has the heterogeneous loss of N₂O₅ turned off and includes the modifications made to CMAQ_{TOT}.