

Response to **Reviewer #2**:

Thank you for the review.

This is an interesting and well-written paper that investigations VOC concentrations and reactivity in North America using an atmospheric model and a comprehensive analysis of aircraft observations from multiple field studies. The model skill in reproducing observed VOC concentrations and reactivity is good in the PBL and poor in the free troposphere. The subjects addressed in the manuscript are appropriate for ACP.

Photochemical reactions are the main sink for VOC. The authors could consider providing more information and critical evaluation of relevant factors that govern VOC removal rates, such as NO_x emissions and O₃ boundary conditions. The boundary conditions are from a global model and they have been accepted and used without much discussion or evaluation in the present manuscript.

Thank you for the positive comments. We have prepared a point by point response to the reviewer's comments below.

Comment 1: Are the ozone levels at the model boundaries and their seasonal variations reasonable and consistent with observational analyses by Parrish and Cooper at NOAA?

Reply to Comment 1: To address this question, we evaluate the modeled boundary/background ozone using ozone observation from the recent Atmospheric Tomography Mission (ATom), which sampled a rich set of data over the Pacific and Atlantic Oceans throughout the troposphere across four seasons. To this end, the model was re-run for the ATom period at 2°×2.5° horizontal resolution and sampled along the ATom flight tracks.

Figure S1 shows ATom ozone observations over the Pacific (100°W-170°E) superimposed on the modeled monthly mean ozone curtain at ~177.5°W. We focus discussion here on the northern hemisphere (NH) as most relevant to the boundary conditions for our study domain. We see that the model captures the overall vertical, latitudinal, and seasonal features seen in the observations. Figure S2 shows the observed and modeled probability density functions for ozone in the northern Pacific. The model generally captures the measured ozone variability with $R^2 = 0.6-0.9$ and normalized mean bias (NMB) of -27% to 5%. The largest discrepancy is seen in spring, likely reflecting insufficient stratosphere-troposphere exchange (also diagnosed by Hu et al. (2017)). Overall, however, the model-measurement comparisons do not point to any persistent, large-scale discrepancies that would alter the conclusions of our paper (particularly given that most campaigns used here, with the exception of DISCOVER-AQ CA, focus on meteorological summer).

We have added Figures S1 and S2 to the Supplement and the following to the Sect. 2: “The Supplement shows an evaluation of these boundary conditions based on Atmospheric Tomography Mission (ATom) (Wofsy et al., 2018) ozone observation in the northern Pacific.”

Comment 2: Please provide analogous maps for NO_x emissions (anthropogenic, soil, lightning, pyrogenic) to match Figures 2a and 3a for VOC. While NO_x is not the focus of the present paper, these emissions are relevant to the analysis as they have strong indirect effects on VOC lifetimes and reactivities.

Reply to Comment 2: Thank you for the comment. As requested we have now added Figures S5 and S6, showing maps of annual NO_x emissions and the fractional contributions by seasons for each emission category.

References:

Hu, L., Jacob, D. J., Liu, X., Zhang, Y., Zhang, L., Kim, P. S., Sulprizio, M. P., and Yantosca, R. M.: Global budget of tropospheric ozone: Evaluating recent model advances with satellite (OMI), aircraft (IAGOS), and ozonesonde observations, *Atmos. Environ.*, <https://doi.org/10.1016/j.atmosenv.2017.08.036>, 2017.

Wofsy, S. C., Afshar, S., Allen, H. M., Apel, E., Asher, E. C., Barletta, B., Bent, J., Bian, H., Biggs, B. C., Blake, D. R., Blake, N., Bourgeois, I., Brock, C. A., Brune, W. H., Budney, J. W., Bui, T. P., Butler, A., Campuzano-Jost, P., Chang, C. S., Chin, M., Commane, R., Correa, G., Crounse, J. D., Cullis, P. D., Daube, B. C., Day, D. A., Dean-Day, J. M., Dibb, J. E., DiGangi, J. P., Diskin, G. S., Dollner, M., Elkins, J. W., Erdesz, F., Fiore, A. M., Flynn, C. M., Froyd, K., Gesler, D. W., Hall, S. R., Hanisco, T. F., Hannun, R. A., Hills, A. J., Hints, E. J., Hoffman, A., Hornbrook, R. S., Huey, L. G., Hughes, S., Jimenez, J. L., Johnson, B. J., Katich, J. M., Keeling, R. F., Kim, M. J., Kupc, A., Lait, L. R., Lamarque, J.-F., Liu, J., McKain, K., McLaughlin, R. J., Meinardi, S., Miller, D. O., Montzka, S. A., Moore, F. L., Morgan, E. J., Murphy, D. M., Murray, L. T., Nault, B. A., Neuman, J. A., Newman, P. A., Nicely, J. M., Pan, X., Paplawsky, W., Peischl, J., Prather, M. J., Price, D. J., Ray, E., Reeves, J. M., Richardson, M., Rollins, A. W., Rosenlof, K. H., Ryerson, T. B., Scheuer, E., Schill, G. P., Schroder, J. C., Schwarz, J. P., St.Clair, J. M., Steenrod, S. D., Stephens, B. B., Strode, S. A., Sweeney, C., Tanner, D., Teng, A. P., Thames, A. B., Thompson, C. R., Ullmann, K., Veres, P. R., Vieznor, N., Wagner, N. L., Watt, A., Weber, R., Weinzierl, B., Wennberg, P., Williamson, C. J., Wilson, J. C., Wolfe, G. M., Woods, C. T., and Zeng, L. H.: ATom: Merged Atmospheric Chemistry, Trace Gases, and Aerosols. ORNL DAAC, Oak Ridge, Tennessee, USA, <https://doi.org/10.3334/ormlaac/1581>, 2018.

Figures:

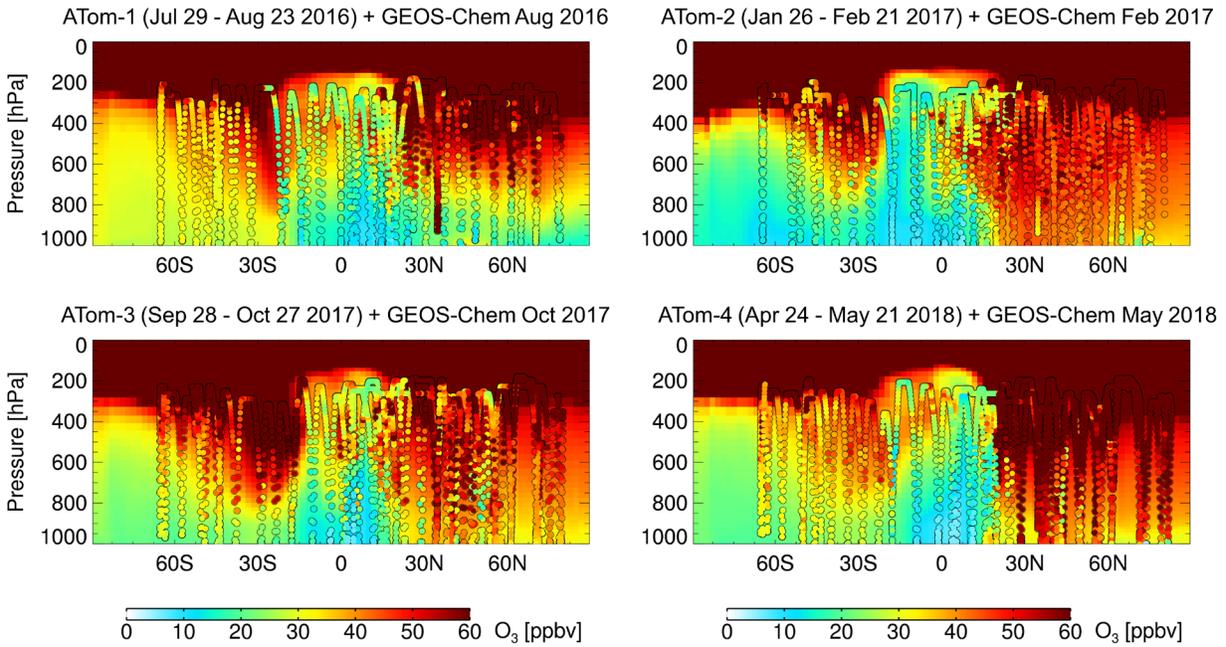


Figure S1. Ozone over the Pacific Ocean. Colored circles show airborne observations from the Atmospheric Tomography Mission (ATom) within 100°W-170°E. Plotted in the background is the monthly mean ozone curtain simulated by GEOS-Chem (global simulation at 2°×2.5°) at ~177.5°W for the corresponding month.

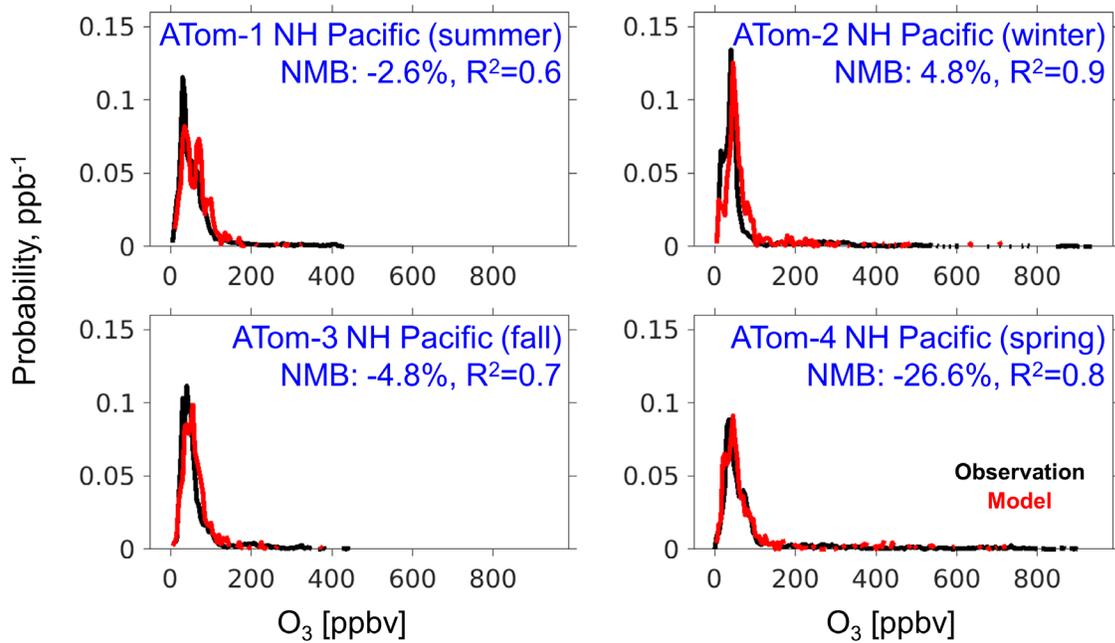


Figure S2. Ozone probability density functions over the northern Pacific (100°W-170°E, 0°-90°N). Plotted are ATom observations (black) and co-located GEOS-Chem model predictions (red), with correlations and normalized mean biases given inset.

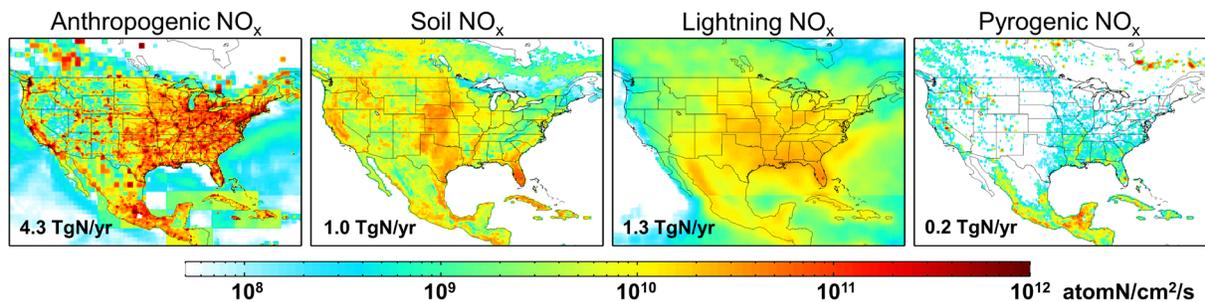


Figure S5. Annual NO_x emission fluxes over North America as simulated by GEOS-Chem for 2013.

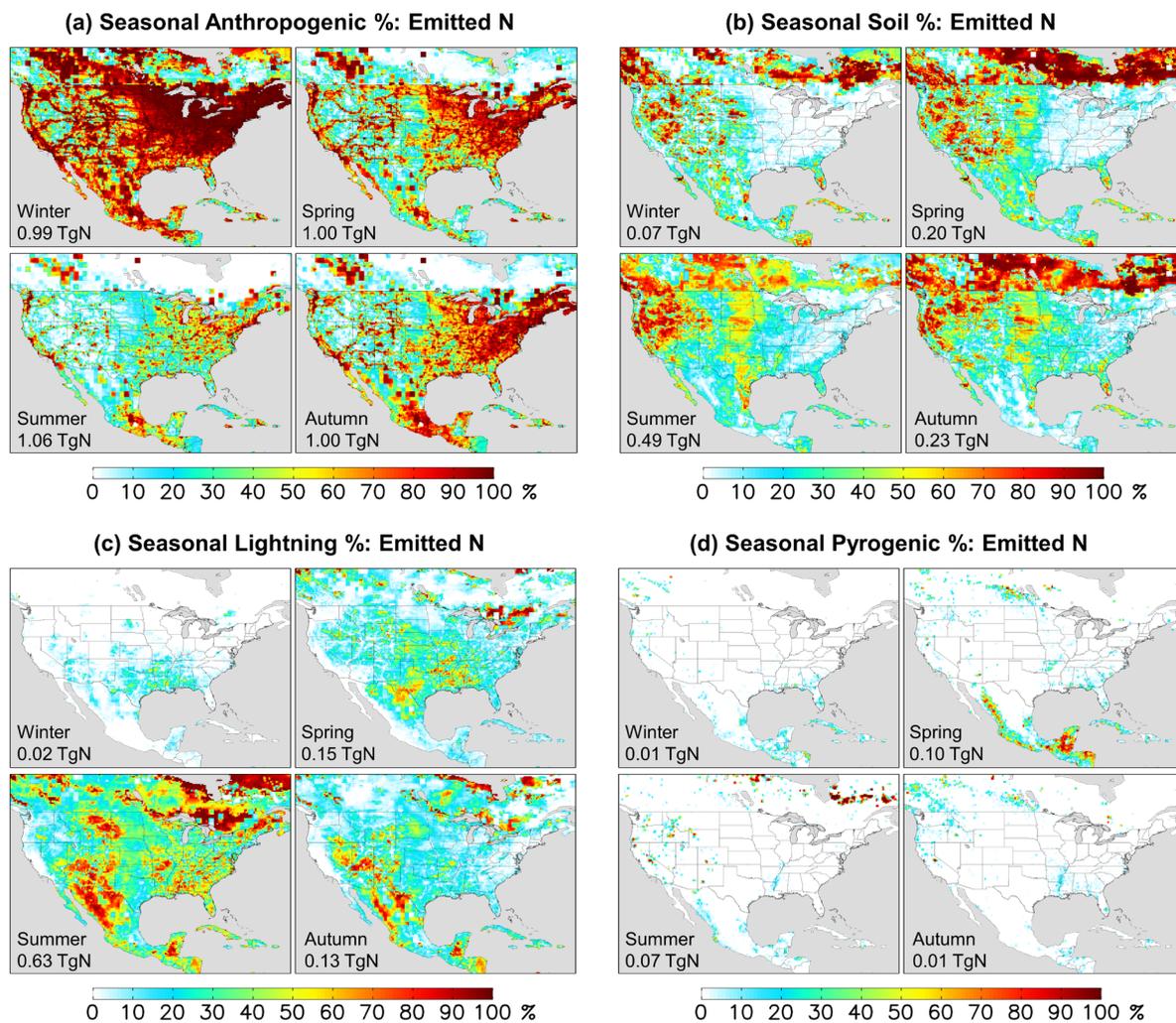


Figure S6. Seasonal contribution of each emission sector to total modeled NO_x emissions.