

Review of “Estimating NO_x emissions of stack plumes using a high-resolution atmospheric chemistry model and satellite-derived NO₂ columns” by Krol et al.

This manuscript proposes a novel method for estimating NO_x emissions from point sources by using a Large-Eddy Simulation (LES) model in conjunction with satellite-derived NO₂ column data. The authors conducted simulations of plumes from four large power plants with NO_x emissions incorporating related chemistry, and compared the model results with TROPOMI tropospheric NO₂ columns. The model columns agreed reasonably with the satellite columns that adopted the improved air mass factors. Considering uncertainties both in the model simulations and satellite retrievals, the level of agreement between the two are quite encouraging. The manuscript introduces several innovative approaches to address the research question and thoroughly discussed the results including uncertainties and limitations of this approach. I would be happy to recommend the publication of this manuscript to ACP after minor revisions. I summarized the points to be revised.

- 1) The abstract of this manuscript is quite distracting. All CO₂ related remarks should go to the later part of introduction or discussion section. The main topic in this manuscript is NO_x emissions from point sources related to LES model simulations and TROPOMI tropospheric NO₂ column observations. Furthermore, the sentence “Moreover, results indicate that common assumptions about the NO₂ lifetime (~4 hours) and NO_x:NO₂ ratios (~1.3) in simplified methods that estimate emissions from NO₂ satellite data (e.g. Beirle et al., 2019) need revision” needs to be revised. This targets only specific studies and does not give broad implications and directions.
- 2) This manuscript deals with the classical nonlinear relationship between NO_x and OH. The authors several times referred to Rohrer et al. (2014) for recycling of OH. Rohrer et al. (2014) is mainly concerning about a new recycling process generating OH under very low NO_x and high biogenic VOC condition. I don't think that this manuscript is closely related to Rohrer et al (2014). There would be better references for this. Meanwhile, NO_x lifetime estimations and related discussions based on satellite observations can be found in Valin et al (2013) and Laughner and Cohen (2019) and references therein.

L. C. Valin et al., *Geophys. Res. Lett.* 40, 1856-1860 (2013).

Laughner and Cohen, *Science* 366, 723-727 (2019).

- 3) It would be beneficial to include the plot for BEL2 from Figure 2 alongside the three plots of the LES model NO₂ columns (projected to the TROPOMI pixels) in Figure 11. This addition would facilitate one-to-one comparison.
- 4) It is not clear that the emission the model used is based on the bottom-up emission inventories and the one measured at each stack. If we consider TROPOMI tropospheric NO₂ columns as a ground truth value, the agreement between the model and satellite gives a confidence in the bottom-up emission from this power plant, probably as shown in this study. While the TROPOMI data appear promising, additional validation and calibration would be necessary, particularly for observations near power plants. If the authors utilized the observed NO_x emission from the power plants for their simulations, the agreement between the model and satellite data provides insight into the accuracy of the TROPOMI columns. Thus, the story changes, depending on the nature of emission in the model.
- 5) The analysis of Is and NO₂ lifetimes for the different power plants is valuable. However, it would be better if Is and NO₂ lifetime are also calculated for the pixels of TROPOMI or larger source box (like 100 km x 100 km) for interpretation of real-world problems. Beirle et al (2019) noted the specific condition for which the assumption of NO_x/NO₂ = ~ 1.3 is valid. It is needed to interpret the results in this study in line with Beirle et al (2019) or similar studies and other research that adopted larger source boxes.
- 6) The recommendations for future calculations of NO_x emissions from stack plumes remain unclear in this study. Should the LES method be applied for all power plants worldwide? Can TROPOMI tropospheric NO₂ columns sufficiently provide NO_x emissions estimations from these sources?