Response to reviewer #1

Thank you for the careful and thorough reading of this manuscript and your thoughtful comments and suggestions. Our responses follow the reviewer's comments (in *Italics*).

General comments:

There are a number of recent papers on the topic of NOx emission trends in the United States as observed from space and as compared to predictions from models. The papers raise issues about the emission models, about the resolution of measurements and models needed to derive accurate trends, about interpretation of satellite observations including whether and how the regional background is included in the trend analysis, and whether the lifetime of NOx is also changing with time affecting interpretation of temporal trends. The analysis in this paper focuses on nonlinearities in chemistry which is related to the question of chemical lifetime. The analysis in the paper seems solid and the discussion and conclusions try to put the paper in context of the recent literature.

I recommend the abstract be revisited in light of the discussion and conclusions as nowwritten.

Reply:

Thank you for your suggestion. We listed the factors affecting the nonlinear relationships among anthropogenic NO_x emissions, NO_2 surface concentrations, and NO_2 TVCDs in Lines 17 - 18 in the revised manuscript. Not only chemistry and background sources but also physical processes, such as transport, contribute to the nonlinearities.

I also recommend the authors consider whether they can make some more general conclusions about the role of nonlinearities that are the focus of their work as a guide to future research. For example does this research help push forward the conversation about the model resolution needed to describe NOx to a specified accuracy? Other papers suggest that 36km might not be sufficient for the absolute accuracy the authors are trying to achieve. On the other hand there might be cancellation of errors in computation of trends that allows use of lower resolution for questions about trends?

Reply:

We think 36 km is sufficient for the regional analysis in this study. A higher resolution model result will not change the nonlinearity discussion in section 3.1. The low-anthropogenic-NO_x emission regions are more sensitive to various factors, such as lighting and soil NO_x emissions and transport, than high-anthropogenic-NO_x emission regions. The critical thing in trend analysis using satellite data directly is therefore to use the data over high-anthropogenic-NO_x emission regions and avoid low-anthropogenic-NO_x emission regions. Therefore, the favorable resolution depends on the emission distributions of the study area. The model analysis used here is only to show the problems associated with using data over the low-anthropogenic-NO_x emission regions. Silvern et al. (2019) used modeling results with a resolution of 0.5° × 0.625° and shown that the tropospheric NO_x lifetime decreased from 8.1 hours to 7.7 hours from 2005 – 2017. When using high-resolution simulations, suggested by Valin et al. (2011), the required accuracy on the anthropogenic emission distribution is much higher than 36 km. Our model results using 4- and 36-km resolutions indicate that the errors of 4-km

NO_x emission distribution are significant and need to be accounted for in modeling analysis. It is beyond what is of interest in this study.

References

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Valin, L. C., Russell, A. R., Hudman, R. C., and Cohen, R. C.: Effects of model resolution on the interpretation of satellite NO_2 observations, Atmos. Chem. Phys., 11, 11647-11655, 10.5194/acp-11-11647-2011, 2011.