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Atmospheric Chemistry and Physics Discussions

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Interactive comment

Interactive comment on "Improvement from the satellite-derived NO_x emissions on air quality modeling and its effect on ozone and secondary inorganic aerosol formation in Yangtze River Delta, China" *by* Yang Yang et al.

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We thank very much for the valuable comments and suggestions from the reviewer, which help us improve our manuscript. The comments were carefully considered and revisions have been made in response to suggestions. Following are our point-by-point responses to the comments and corresponding revisions.

This manuscript has presented a top-down estimate of NOx emissions in the Yangtze River Delta (YRD) region and demonstrated that air quality modeling using the top-

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Discussion paper

down NOx emissions could improve the simulations of ozone and secondary inorganic aerosol (SIA) over this region. A set of sensitivity simulations are conducted to better understand the formation of ozone and SIA under perturbed precursor emission conditions. This manuscript offers some new knowledge on the regional secondary pollution over YRD including an improved estimate of NOx emissions and predicted effectiveness of various emission controls on secondary pollution formation. This study is overall well conducted and analyzed. The manuscript is well written, and fits the scope of ACP. I think the following comments shall be addressed for merit publication.

Response and revisions:

We appreciate the reviewer's positive remarks.

Specific Comments:

1. Sect. 2.1, top-down estimation method: My main concern lies on the top-down method. The present description in this section is not clear. The section states "the a posterior daily emissions were used as the a priori emissions of the next day, and the monthly top-down estimate of the NOx emissions was scaled from the average of the a posterior daily emissions of the last three days in the month". Do you mean the NOx emissions were calculated day by day for each month? In that case, there shall exist strong day-to-day variations in the top-down estimates, reflecting either true emission changes or uncertainties in satellite measurements and model results. It is then not proper to derive the monthly emission estimate based on only daily emissions in the last three days. This needs to be clarified in the manuscript and the daily emission variations if significant should be discussed.

Response and revisions:

We thank the reviewer's important comment. Currently, the inverse model we applied in this work assumed that the daily emissions were similar (Zhao and Wang, 2009; Gu et al., 2014; Cooper et al., 2017). For example, the daily variation was expected to be negligible over most regions of east China (Zhao and Wang, 2009). In our previous work (Yang et al., 2019), we evaluated the robustness of the method, by applying the "synthetic" TVCDs from air quality simulation based on a hypothetical "true" emission inventory, instead of those from satellite observation. We found that sufficient iteration times could result in a relatively constant emission estimate (the top-down estimate) close to the "true" emission input, implying the reliability of the inverse modeling method.

The assumption would bring some uncertainty as the daily variation of emissions did exist. Due mainly to the fair missed values of satellite detection, however, the daily variation could not be precisely captured by the top-down method, particularly at regional scale with relatively high horizontal resolution. Such method was designed for monthly mean of emissions. From a bottom-up perspective, the difference in NOX emissions between weekday and weekend was within 5% in the YRD region (Zhou et al., 2017), indicating an insignificant bias from ignoring the daily variation of emissions. We have added those descriptions in line 166 and lines 171-179 in the revised manuscript.

Reference: Zhao, C., and Wang, Y. X.: Assimilated inversion of NOx emissions over East Asia using OMI NO2 column measurements. Geophys. Res. Let., 2009, 36(L06805): 1-5.

Gu, D.S., Wang, Y.X., Smeltzer, C., Boersma, K.F.: Anthropogenic emissions of NOx over China: Reconciling the difference of inverse modeling results using GOME-2 and OMI measurements, J. Geophys. Res.: Atmosphere, 119, 7732-7740, 2014.

Cooper, M., Martin, R.V., Padmanabhan, A., Henze, D.K.: Comparing mass balance and adjoint methods for inverse modeling of nitrogen dioxide columns for global nitrogen oxide emissions, J. Geophys. Res.: Atmosphere, 122, 4718–4734, 2017.

Yang Y., Zhao Y., Zhang L., Lu Y.: Evaluating the methods and influencing factors of satellite-derived estimates of NOX emissions at regional scale: A case study for Yangtze River Delta, China. Atmos. Environ., 219, 1-12, 2019.

Zhou, Y.D., Zhao, Y.D., Mao, P., Zhang, Q., Zhang, J., Qiu, L.P., Yang, Y.: Development of a high-resolution emission inventory and its evaluation and application through air quality modeling for Jiangsu Province, China. Atmospheric Chemistry and Physics 17, 211-233, 2017.

2. Page 4, Line 94 and Line 110:“0.4 Tg N/yr” and “69.6 x 10ËĘ13 molecules cm-2”. Please also provide relative percentage numbers from the two studies, so that the magnitudes can be better understood.

Response and revisions:

We thank the reviewer’s comment. The relative percentage number for 0.4 Tg N/yr was 5.8% (Gu et al., 2014) and provided in line 96 in the revised manuscript, while that for 9.6 x 10ËĘ13 molecules cm-2 was unavailable in the original paper (Jena et al., 2014).

3. Page 7, Sect. 2.2 Model configuration: What is domain 3 (D3) labelled in Figure 1? Is it used in this study?

Response and revisions:

We thank the reviewer’s reminder. The domain 3 (D3) in the original figure is not used in this study and thus removed in the revised Figure 1.

4. Page 8, Line 204-210: Which year of data is used for the MEIC emission estimates?

Response and revisions:

We thank the reviewer’s reminder. The MEIC emission data for 2015 were used in this study and we have added the information in lines 213 in the revised manuscript.

5. Page 9, Line 228-232: Some previous studies (e.g., Lamsal et al., 2008; Liu et al. ACP 2018) suggested that the NO2 measurements obtained from the molybdenum-catalyzed conversion technique might be overestimated due to interference from other nitrogen species. Would this affect your results?

Lamsal, L. N., et al.: Ground-level nitrogen dioxide concentrations inferred from the satellite-borne Ozone Monitoring Instrument, J. Geophys. Res.- Atmos., 113, D16308, https://doi.org/10.1029/2007JD009235, 2008.

Liu, M., et al.: Spatiotemporal variability of NO2 and PM2.5 over Eastern China: observational and model analyses with a novel statistical method, Atmos. Chem. Phys., 18, 12933–12952, https://doi.org/10.5194/acp-18-12933-2018, 2018.

Response and revisions:

We thank the reviewer's very valuable comment. We agree with the reviewer that the NO2 concentration could be overestimated with the molybdenum-catalyzed conversion technique, while the effect of such overestimation on our results is expected to be limited. On one hand, the top-down estimates of NOX emissions were derived from satellite observation instead of ground observation. The observed ground NO2 concentrations were only used to evaluate the model performance with the bottom-up and top-down estimates of NOX emissions. On the other hand, as shown in Figure 4 in the revised manuscript, the simulation of NO2 concentration with the top-down estimates were improved by 30%-100% (indicated by the NMBs) compared to that with the bottom-up emission data, substantially larger than the common overestimation in NO2 observations with the measure around 15%. Therefore, the overestimation in ground-level NO2 concentrations could hardly change the basic judgment of this study that application of top-down estimates in NOX emissions would improve the model performance of NO2 concentration in the YRD region.

6. Page 10, Line 258: According to Figure 5 and 6, peaking ozone concentrations in YRD are also shown in the July month, and many previous studies have suggested more active ozone formation in summer. Some sentences are needed here to explain why this study focused on April and did not discuss July.

Response and revisions:

We thank the reviewer's comment. In the YRD region, on one hand, the peaking time of O3 concentrations has gradually moved forward from summer to late spring. In this work, for example, the mean observed O3 concentration of YRD in April was 72.5 μg/m3, even larger than that (71.9 μg/m3) in July. On the other hand, the model performance of O3 in this work was better for April than that for July (Fig. 6 in the revised manuscript). Therefore, we selected April to explore the sensitivity of O3 formation to precursor emissions. The corresponding revision was shown in lines 264-269 in the revised manuscript.

7. Page 10, Sect. 3.1: The spatial distribution of top-down vs. bottom-up NOx emission changes in YRD as shown in Figure S2 is an important finding of this study for explaining and supporting improvements in the top-down estimates. I suggest move Figure S2 to the main manuscript, e.g., combine with the present Figure 2.

Response and revisions:

We thank the reviewer's comment. We replot Figure S2 in the original submission and improve the figure quality. We move the figure to the main manuscript (Figure 3 in the revised manuscript).

8. Page 13, Line 363 and 364: Here "Fig. 5" should be "Fig. 4"

Response and revisions:

We are sorry for the error and thank the reviewer's reminder. "Fig. 5" should be "Fig. 4" in the original submission. As we add a figure in the revised manuscript (see our response to Question 7), now it should be Fig. 5 again. Hopefully this explanation is not confusing.

9. Page 17, Line 465-469: The decreases in the nitrate aerosol concentration in July with the top-down NOx emissions are interesting and worth further discussion. Reductions in NOx emissions would lead to increases in the nitrate aerosol concentration in other months (January, April, and July). Can you explain why the response in July is dif-

ferent from those in other months? Is it because the percentage reduction of top-down NOx emissions in July is much larger?

Response and revisions:

We thank the reviewer's important comment. It could result from two factors. First, the reduction of top-down NOx emissions in July was much larger, as suggested by the reviewer. Second, the VOC-limit mechanism in O3 formation was found weaker in summer than winter (see Fig. 7e and Fig. 7g), resulting in less O3 formation and thereby nitrate aerosol through oxidation. The corresponding revision was shown in lines 491-496 in the revised manuscript.

10. Page 19, Line 538: Should "Fig. 9b" here be "Fig. 9c"?

Response and revisions:

We are sorry for the error and thank the reviewer's reminder. "Fig. 9b" is now corrected to "Fig. 10c" (we add a new figure in the revised manuscript).

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