Anonymous Referee #1

General Comments

1. The manuscript entitled ‘High-resolution (0.05°×0.05°) NO_x emissions in the Yangtze River Delta inferred from OMI’ focuses on developing a method to inverting NO_x emissions at a high resolution in major urban areas by using the long-term satellite measurements of nitrogen dioxide. The results show that the inverted NO_x emission dataset can reveal the features which are not well represented or not included widely used Multi-scale Emissions Inventory of China. Overall, though the topic is important and the methods are technically, the manuscript need be restructured and rephrased. I recommend to reconsider its publication pending the following concerns satisfactorily addressed.

The manuscript has been overhauled considering the comments from both referees.

A brief review has been made about the inventories at similar resolutions, including Zhao et al. (2015) and CAMS-reg (Granier et al., 2019) on page 3 line 8 (see the revised manuscript) based on bottom-up methods. Top-down estimates can be further combined with bottom-up inventories and spatial proxies to increase the spatial resolution, by downscaling and/or source sector apportionment (e.g., MarcoPolo on page 4 line 1-4). MarcoPolo emissions can reach higher resolutions than 0.05°×0.05°, i.e., 0.01°×0.01°, given the detailed information of the location of the emission sources which ask for lots of efforts to collect and are absent or inaccurate at times. Top-down emissions including our work offer an important supplement and reference at high resolutions.

The PHLET model has been upgraded and re-built on the FEniCS platform, the necessary citations of which have also been included. Based the FEniCS platform, we improve the calculation efficiency of the PHLET and A-PHLET largely. Now, the inversion calculation takes less than one hour as stated on page 6 line 14.

We have also fixed a bug to correctly account for the effect of S_0 in Eq. 3. The corresponding results and discussions have been revised including NO_x emissions, lifetimes and the uncertainties. After processing the error covariance properly, the derived the lifetime of NO_2 due to deposition becomes longer (30.4 h), which is more consistent with our knowledge about NO_x chemistry.

We have shortened the study time period from summer 2012-2016 to summer 2012-2015. According to the National Bureau of Statistics of China (http://data.stats.gov.cn/), NO_x emissions have dropped substantially from 2015 to 2016. Thus, including summer 2016 may not be the best practice to derive emissions.

To substantiate the emission distribution, more discussion has added in Sect. 4.2 based on the distributions of proxies such as nighttime light, population density, marine shipping routes, coal power plant locations and land use indicated by a satellite photo from Google Earth. Sect. 4.3 compares our emissions with other inventories besides MEIC.

In the conclusion section, we give a summary of the limitations and shortcomings of our method.

Most of the figures have been re-arranged. Some figures have been added, considering comments from both referees.

We have substantially improved the structure of the manuscript to accommodate both reviewers' suggestions. A flowchart has been added to Sect. 2.1 in order to illustrate the procedures of our inversion method. Section 2.3 has been divided into 5 subsections for clarification. Section 2.3.1-2.3.3 describe the model setting and assumptions. Sect. 2.3.4 shows how the SCM matrix is applied to PHLET simulated VCDs, with the detailed procedures shown in Appendix B. Section 2.3.5 summarizes the uncertainty estimates. The part (former Appendix D) about solving the observation error covariance matrix and the adjoint model has been moved to Sect. 2.4, supplemented with an extended discussion on assuming the covariance to be diagonal. The OSSE-like test (former Appendix E) based on GEOS-Chem simulated NO_2 data has been moved to a new Sect. 5.

Specific comments

1. Why the shortest lifetime of NO_2 has the advantage to better relate NO_x emissions to NO_2 VCDs at the 0.05°×0.05° resolution?

Due to the short lifetime of NO_2, the effect of transport and diffusion is rather local. Therefore, the distribution of NO_2 VCDs can better reflect that of NO_x emission at high-resolution; and the effect of transport errors on emission estimate is smaller.

2. Page 6, Line 1-7: What's the relation between the NO_2 retrieval with the AOD? The description is needed.

The NO_2 retrieval becomes unreliable when the loading of aerosol gets too high. We have added necessary citation to this description.

3. Section 2 is generally messy and lack of logics. What's the relation between the PHLET model and PHLET-A model? I suggest the authors rephrase the part 'data and method' more logically.

In order to clarify our method, we have added a flowchart and additional descriptions to illustrate the procedures in Sect. 2. See our response to general comment 1 for the detailed structural changes.

4. The main of this manuscript includes two parts: part one is to show the distributions of NO_2 basing on the retrieved emission data, part two is to evaluate above emission data. Thus, showing more explicit analyses are needed.

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5. In Figure 1, why the NO_x emission and local net source are somewhat related to the lifetime of NO_2? The good relationship between the NO_2 VCDs and lifetimes of NO_2 can be understood well, however, the relations with NO_x emission and local net source are not taken for granted.

We have clarified the methodology; see our response to general comment 1.

As shown in Eq. (2), the local net source is the difference between emission and loss.

Sect. 2.5 and Appendix C presents how to calculate emission and lifetime from the local net source.

6. Figure 1 and Figure 2 should be rearranged. Fig. 2a-d can be combined into Fig.1a-d; Fig. 2e-f and Fig. 1f can be combined into one graph. The current arrangement is messy to describe.

More figures are included in the revised manuscript. The figures are also re-arranged taking the comments from both of the referees into consideration.

7. Page 17, Line 6, what does ‘Figure 3ows’ mean?

Typing error. Changed.

8. How do the authors define ‘anthropogenic’ emission? Including what?

As now clarified in Sect. 2.3.2 (page 10 line 13-21):

“Lightning emissions, biomass burning emissions, aircraft emissions, transport from neighboring regions, and convection can lead to NO_2 at higher altitudes over the YRD area. However, the amount of NO_2 aloft is much smaller than near-ground NO2 due to large ground sources (Lin, 2012). Thus, we regard NO_2 aloft as the regional background, and do not include it in Eq. 1. Also, for near-ground NO_2 over the YRD area, the contribution of downward vertical transport is negligible compared to the contribution of ground sources. Aircraft emissions contribute little to the total ground source, because 78% of aircraft emissions occur at the high altitudes (9–12 km) (Ma and Xiuji, 2000). Therefore, PHLET only accounts for near-ground NO_2 from ground soil, biomass burning and anthropogenic sources (energy, industry, transportation, and residential)."

And in Sect. 4.3 (page 21 line 11-13):

"Our emission data and the DECSO inventory are top-down estimates and include the contributions of soil and biomass-burning sources. Thus, we estimate soil and biomass burning emissions from independent sources, and then subtract these emissions from our and DECSO emission datasets" (to obtain anthropogenic emissions.)

9. What's the reason of inconsistent difference of total anthropogenic NOx emission in each city for summer inverted by this study versus from the MEIC inventory? Otherwise, the difference should be same for each city, that is to say, systematically higher or lower.

Both our and MEIC inventories are gridded, and their differences are grid cell independent and vary from one city to another.

10. The tile of Section 4.3 should be 'Comparing our inverted emission dataset with the MEIC inventory', or more exactly, it should be 'Comparison between our inverted emission dataset with the MEIC inventory'.

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