

Anonymous Referee #2

General Comments

1. This inventory is presented as the only high-resolution inventory for the YRD region, but in the MarcoPolo-Panda project, a high-resolution emission inventory of 0.01-degree resolution has been developed for this region (see <http://www.marcopolo-panda.eu/products/toolbox/emission-data/>) Since the affiliations of the authors were also participating in the MarcoPolo-Panda project it is surprising that this inventory is not mentioned or used in their comparisons. Also, Zhao et al. (2015) present a city-scale emission inventory with the resolution of 3kmx3km in Nanjing, in the Yangtze River Delta.: “Zhao, Y., Qiu, L. P., Xu, R. Y., Xie, F. J., Zhang, Q., Yu, Y. Y., Nielsen, C. P., Qin, H. X., Wang, H. K., Wu, X. C., Li, W. Q., and Zhang, J.: Advantages of a city-scale emission inventory for urban air quality research and policy: the case of Nanjing, a typical industrial city in the Yangtze River Delta, China, *Atmos. Chem. Phys.*, 15, 12623-12644, 10.5194/acp-15-12623-2015, 2015.” The authors claim that very high resolution emissions are lacking, but it is not mentioned how “very high” is defined. Several regions have a high-resolution emission inventory with similar resolution as PHLET: CAMS in Europe, GlobEmissions in the Qatar and South Africa, MarcoPolo-Panda in various regions in China.

We have included discussion of these inventories in the revised introduction:

On page 3 line 1-3: “Gridded bottom-up emission inventories typically use spatial proxies (like population and GDP) to allocate provincial-level emission values, which are derived from activity statistics and emission factor data, to individual locations (Zhao et al., 2011; Janssens-Maenhout et al., 2015; Zhao et al., 2015).”

On page 3 line 7-10: “For a small area, emission factors and activity data of the major sources can be collected by on-site surveys to allow construction of a high-resolution inventory (Zhao et al., 2015; Granier et al., 2019), such as Zhao et al. (2015) for Nanjing. However, on-site surveys are extremely time consuming and resource demanding, difficult to be applied to a large domain in a timely manner.”

On page 4 line 1-10: “Top-down estimates can be further combined with bottom-up inventories and spatial proxies to increase the spatial resolution, such as from $0.25^{\circ} \times 0.25^{\circ}$ in the DECSO derived emissions to $0.01^{\circ} \times 0.01^{\circ}$ for 2014 during the MarcoPolo Project (Hooyberghs et al., 2016; Timmermans et al., 2016) and similar inventories over Qatar and South Africa (Maiheu and Veldeman, 2013).”

In this work, high resolution refers to emissions at a resolution equal or higher than $0.05^{\circ} \times 0.05^{\circ}$. We have made this clear on page 2 line 23 and page 4 line 9.

2. The authors mention that their method is not computationally expensive and can be applied world-wide, but a rough calculation shows that their algorithm will take at least 10 year to calculate the emissions for the whole world, which is not faster than many other methods they refer to on page 3.

First, our method is designed for urban and surrounding areas, rather than everywhere of the globe. Second, in this case study, the calculation is completed on only one CPU core, while the CTMs adopted in top-down method generally ask for parallel computing with many cores. For a multi-domain study, our method can easily adopt parallel computation with more cores. Third, we have upgraded the codes on the FEniCS platform, the necessary citations of which are included. Right now, the emission calculation for the YRD takes less than one hour, faster than our previous calculation by a factor of 30-40. Thus, with one computational core, applying our method to the globe on a $0.05^\circ \times 0.05^\circ$ grid for 4 years would take about a few months.

3. The authors say that the methods are limited in time period, spatial domain and horizontal resolution. This is very different for all the referred methods. The methods of Miyazaki et al and of Stavrakou et al. have already been applied on a global scale, while other methods are also not theoretically limited to a certain domain. In general, the methods mentioned can be applied to any time period as long as satellite observations are available.

We have clarified this point in the revised introduction (page 3 line 25 – page 4 line 1):

“These more sophisticated methods have often been applied to relatively short time periods (e.g., Gu et al., 2016 for one month), small spatial domains (e.g., Tang et al., 2013 in Texas), and/or at coarse horizontal resolutions (e.g., Miyazaki et al., 2012 at 2.8° and Stavrakou et al., 2008 at $5^\circ \times 5^\circ$).”

4. The authors suggest that only (Lin et al., 2012) and Stavrakou et al. (2013) provide uncertainties of the CTM, while they are also presented by Miyazaki et al. (2012) and Ding et al. (2017).

The statement and the citations are modified on page 4 line 6-8: “CTM-based studies typically provide an estimate of the overall model error, although Lin et al. (2012) and Stavrakou et al. (2013) present errors in the individual model processes (e.g., key chemical reactions and meteorological parameters).”

5. What are pros and cons of the introduced method? The pros are mentioned, but what is the downside of averaging a time period of 2012-2016. Specifically, in this period strong trends are appearing in NO_2 over China.

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.

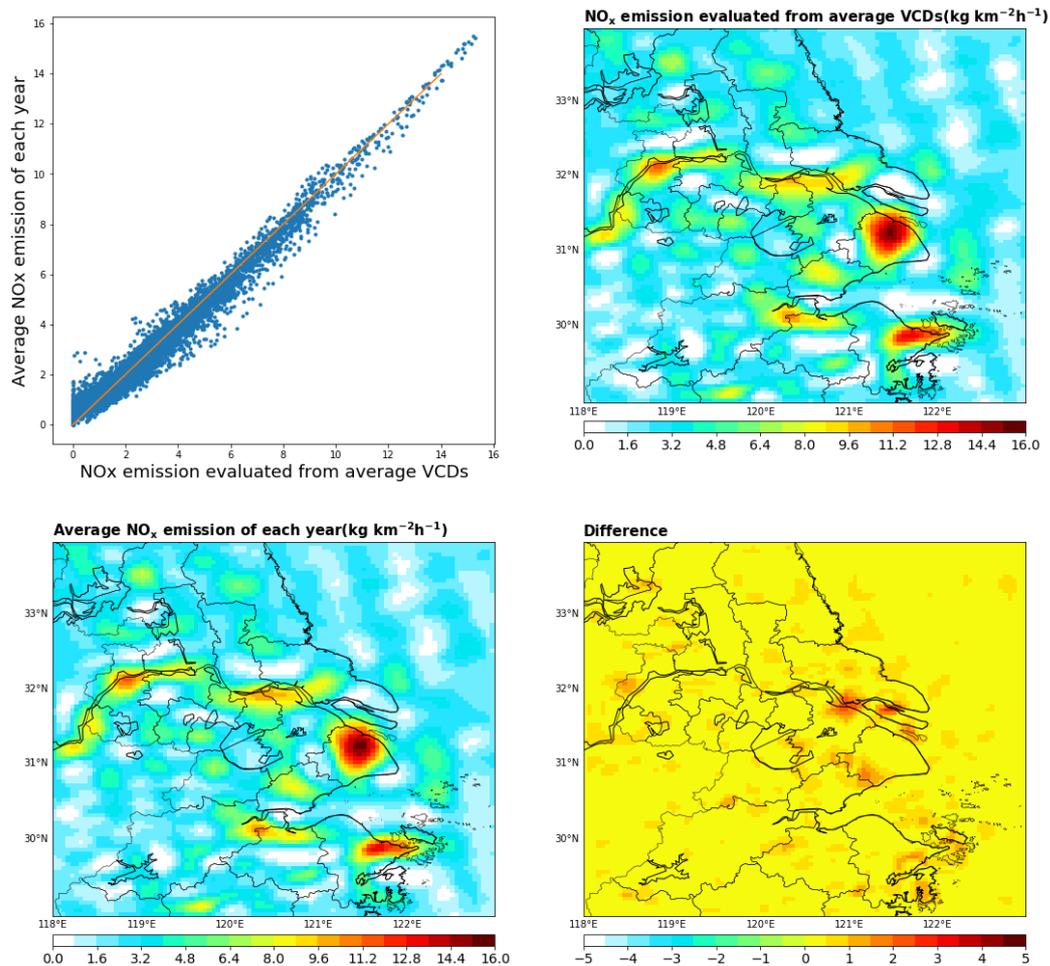
We have also evaluated the emission for each year from 2012 to 2015. The average emissions of those years over 2012-2015 and the emissions evaluated from all VCD data together (as in the main text) are similar. Slope and interception of their linear regression are 0.95 and 0.08, and their correlation coefficient is 0.98 (see the figure below).

We discuss the limitation of our method in Section 2.

On page 9 line 9-11: “Also, combining data from multiple years to derive an averaged NO₂ distribution for simulation (rather than conducting the simulations for individual years and months) leads to an additional uncertainty.”

We have also added the revised conclusion section a paragraph summarizing the limitations of our method:

On page 25 line 11-20: “Our inversion method also has a few shortcomings. The derived emissions do not separate the individual contributions of anthropogenic sectors (i.e., power plants, industry, transportation, and residential). The spatial resolution of the estimated emissions is limited by that of satellite VCD data, although a special oversampling technique has been used to help achieve the highest spatial resolution possible for emissions. The PHLET model is assumed to be 2-dimensional by simplifying the vertical distribution of NO₂ and not accounting for the spatial variability in the vertical shape, similar to previous studies. The adjoint model assumes the observational error covariance matrix to be diagonal, without fully considering the effect of correlations between individual grid cells. Also, we assume a spatially uniform relationship between NO₂ VCDs and NO₂ lifetimes, which may lead to an underestimate in the lifetimes at low-NO₂ locations over the eastern sea.”



6. The period is focusing on the summer time. What are the expected results for the winter-period. Will this change the spatial resolution? Will the magnitude of the emissions change a lot?

To achieve highest spatial resolution possible, we have intended to focus on the summer months, when the lifetimes of NO_x are the shortest.

The lifetimes of NO₂ would be longer in winter, and therefore, the effects of transport and diffusion are more significant. The spatial relation of NO₂ VCDs and NO_x emissions would be lower in winter. Thus, it would be much more difficult to derive the emissions at a high resolution, and the influences of transport errors would be much larger.

7. It should at least be mentioned that there is no sector information in the derived emissions, which is an advantage of bottom-up inventories

We have added on page 25 line 11-12: “Our inversion method also has a few shortcomings. The derived emissions do not separate the individual contributions of anthropogenic sectors (i.e., power plants, industry, transportation, and residential).”

Also note that as stated on page 3 line 14-17: “Top-down inversion typically provides the total emission data, although emissions from individual sources can be further derived by integrating a priori data (often from bottom-up inventories) about source-specific information such as diurnal and seasonal variabilities (e.g., Lin et al., 2010; Lin, 2012) and spatial variabilities (Timmermans et al., 2016).”

8. Although the gridding is on a 0.05° resolution the actual spatial resolution of the resulting emissions seems much lower. An indication of the intrinsic resolution can be obtained from the largest gradients in the emissions. I cannot detect clear structures with a 0.05° resolution. One would at least expect some power plants to show up as clear spots in this region. Maybe the method is still limited by the OMI resolution?

The highest emission is at one grid cell in north Shanghai, and its difference from eight surrounding grid cells are $0.39 \text{ kg km}^{-2} \text{ h}^{-1}$ (2.6%). The mean gradient of the emissions is $0.079 \text{ kg km}^{-3} \text{ h}^{-1}$.

We admit the intrinsic resolution of our derived emissions is limited by the pixel sizes of OMI. We have added in the revised conclusion section that

On page 25 line 12-14: “The spatial resolution of the estimated emissions is limited by that of satellite VCD data, although a special oversampling technique has been used to help achieve the highest spatial resolution possible for emissions.”

Also, in Sect. 4.2 on page 22 line 5-7: “This is because our top-down estimate is limited by the intrinsic resolution of NO_2 VCDs, i.e., our oversampling approach does not fully compensate for the large sizes of OMI pixels. Therefore, the large spatial gradient of NO_x emissions is smoothed to some extent in our dataset.”

We have also discussed emissions related to power plants. As detailed in the revised Sect. 4.2 on page 20 line 10-18:

“The filled circles in Fig. 6g show the locations of coal-fired power plants in 2016 from Carbon Brief (www.carbonbrief.org; last access: 2019/6/27). The radius of a circle denotes the power generation capacity. Figure 6h further shows the GPED v1.0 bottom-up NO_x emissions for power plants on a $0.1^\circ \times 0.1^\circ$ grid in 2016. Coal-fired power plants in the YRD are normally near the urban centers, traffic lines or other sources. Our top-down NO_x emission map shows large emission values near the power plants (Fig. 6b), although it cannot isolate the sole contribution of power plants. At the GPED power plant locations, the correlation between our and GPED emissions reaches 0.26, due to the influence by non-power plant sources; note that the correlation between GPED emissions and POMINO NO_2 VCDs are only about 0.21.”

9. The structure of the paper is somewhat confusing and therefore I suggest moving appendix C and E to the main text. Section 2.1 is too short to understand the method of

determining emissions.

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 NO₂ data has been moved to a new Sect. 5.

10. In section 2.3: The contributions of lightning, biomass burning and aircraft emissions are neglected. The authors explained that the contributions of these emissions are small. In the inversion method, both soil and anthropogenic emissions are derived. In section 4, it is calculated that the soil emissions contribute 0.9% of the inverted emissions. This looks like a very small amount. However, biomass burning is considered as a significant source in the YRD, especially in summer. On a scale of 0.05°×0.05° lots of biomass burning activity will exist. Give some more detailed information and explain why lightning, biomass burning and aircraft emissions are neglected.

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

“Lightning emissions, biomass burning emissions, aircraft emissions, transport from neighboring regions, and convection can lead to NO₂ at higher altitudes over the YRD area. However, the amount of NO₂ aloft is much smaller than near-ground NO₂ due to large ground sources (Lin, 2012). Thus, we regard NO₂ aloft as the regional background, and do not include it in Eq. 1. Also, for near-ground NO₂ 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 NO₂ from ground soil, biomass burning and anthropogenic sources (energy, industry, transportation, and residential).”

11. The model error is set to be the sum of the quadrature of errors contributed by several aspects. However, there is no explanation on how the authors set some errors, for example the treatment of background NO₂ concentrations. The authors use wind fields from ECMWF on a coarse resolution and regridded to a high resolution. The error of regridded wind field on high resolution can be quite large. The authors consider error of wind speed, but how about the wind direction? The error set for the wind looks

optimistic.

Error estimates for individual parameters and processes are based on the literature, our sensitivity tests, and/or expert judgement. For most parameters, the reasoning of choosing specific values is given when the error terms are introduced.

For background NO₂, our choice (0.54×10^{15} molecules cm⁻²) is based on the consideration that background NO₂ would be very small (e.g., smaller than 1×10^{15} molecules cm⁻²; Cui et al., 2016). Doubling the background only has marginal effects on our emission estimate especially at modest- and high-NO₂ locations. Spatially averaged, the error due to the choice of our background value is estimated as 5%.

For errors introduced by winds, we have clarified in Sect. 2.3.3 on page 11 line 17-19 that

“We assess the model errors introduced by the uncertainties in the wind field and effective diffusion coefficients by Monte Carlo simulations in which the wind speeds are changed according to their uncertainties. The resulting relative uncertainty in the modeled NO₂ VCDs is about 20%.”

12. References: All the references should be carefully checked if they are in the correct format, especially the names of authors. Many articles are missing or articles should be removed throughout the whole text. It is advisable to let a native speaker make the necessary corrections.

The references have been checked and some necessary citations have been added.

Specific comments

1. Page 1, Line 14: lacking => missing

Changed.

2. Page 1, Line 17: The inversion => The model used in the inversion

We have change it into ‘the top-down inversion method’ which refers to the whole process.

3. Page 1, Line 18: We construct a model called PHLET (..)

Changed.

4. Page 1, Line 19 Metrix => Matrix

Changed.

5. Page 2, Line 5 tied => related

Changed.

6. Page 2, Line 7 features => structures

Changed.

7. Page 2, Line 8-9: This last sentence is kind of obvious. It should be moved from the abstract to the conclusions/outlook, but I suggest to just remove it.

Removed.

8. Page 2, Line 21: split the sentence into 2 separate sentences to make it more understandable.

Changed. Now on page 2 line 18.

9. Page 2, line 24: on => of

Changed.

10. Page 2, line 24: how is a “very high resolution” defined?

We have made it clear to be $0.05^\circ \times 0.05^\circ$ on page 2 line 23 and page 4 line 9.

11. Page 3, Line 1: Bottom-up emissions do not only use spatial proxies but are also based on gathered statistical information of industrial output, car emissions, etc.

The statement has been modified.

12. Page 3, Line 4: Please define “high”

We have made it clear to be $0.05^\circ \times 0.05^\circ$ on page 2 line 23 and page 4 line 9.

13. Page 3, Line 20-21: What do the authors mean by “low-cost” and “high-resolution”

Low cost refers to the low requirement on computation resources described on page 6 line 13-16:

“With one computational core (Intel® Xeon® Gold 6130 CPU @ 2.10GHz),

derivation of NO_x emissions over the YRD here takes less than one hour after necessary input data are prepared. Applying the framework to multiple areas would take a similar amount of time by using one computational core for each area.”

As for high resolution, we have made it clear to be 0.05°×0.05° on page 2 line 23 and page 4 line 9.

14. Page 3, Line 22: Here it is mentioned that these inventories are important for trends and variability. I agree, but the method presented in this paper do not give the possibility to study trends and variability, which should be mentioned somewhere in the conclusion.

As now clarified in Sect. 6 on page 24 line 22-24:

“Although this study derives the averaged emissions over summer 2012–2015, calculations of emissions at higher temporal resolutions (e.g., every 2 years) is possible to better capture the interannual variability and trends.”

15. Page 3, Line 22-23: Why is it important to understand air pollution with the advent of TROPOMI? I would say it is the other way around: TROPOMI is important for understanding air pollution.

The sentence has been removed. Discussion about TROPOMI is on page 25 line 25-page 2 line 2 now.

16. Page 3, line 24: Constructing => construct

Changed.

17. Page 4, line 1 other 13 => 13 other

Changed. In addition, we have added two more cities which were missed in the original manuscript.

18. Page 4, line 1: explain the acronym POMINO

Changed. On page 6 line 22.

19. Page 4, line 3: change to “a model called PHLET”

Changed.

20. Page 4, line 5: delete “concentration dependent”

Changed.

21. Page 4, line 17: Why is this the finest spatial information possible?

As described in Sect. 2.2 on page 7 line 25 – page 8 line 4, The oversampling approach takes advantage of the fact that the exact location of footprint of the OMI instrument slightly changes from one day to another, so does the exact location of footprint of a satellite pixel at a given VZA. Thus, sampling from multiple days increases the horizontal resolution of data. Besides, the SCM matrix is constructed base on the pixels, and thus the finest spatial information is preserved.

22. Page 4, line 19: What Is SCM? This is explained much later in the text.

We have re-structured the manuscript and have clarified the use of SCM in Sect. 2.3.4, supplemented by Appendix B.

23. Page 4, line 21-22: Without further explanation this does not explain the method.

A flowchart has been added in order to illustrate the procedures of our inversion method on page 5 line 8 -page 6 line 4.

24. Page 4, line 22: Which fixed formula is used?

It refers to Eq. C5 in Appendix C. The statement has been modified.

25. Page 5, line 1: Which nonlinear relationship do the authors mean here? There are 3 quantities mentioned: (1) emissions, (2) lifetimes and (3) VCDs.

Between lifetimes and NO₂ VCDs. Changed now on page 6 line 8.

26. Page 5, line 4: A long time period is mentioned. What do the authors mean, a long time period to average or multiple 5 years periods? And why are these long time periods not presented in this paper?

We meant summer 2012-2016.

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.

27. Page 5, line 5: It is mentioned that the calculation takes about 36 hours after necessary input data? What are the necessary input data? How long does it take to prepare the input data?

The necessary input data are the OMI product (i.e. POMINO) and wind field. The time

it takes to prepare those data depends on the Internet conditions, as in other studies. Running our codes to process these input data takes 30-40 minutes for the YRD domain here.

28. Page 5, line 5: If the inversion takes 36 hours for a 5x5 degree domain, a global calculation will take about 10000 hours, which is about 10 year.

See 'general comments 2'. And as stated on page 6 line 14-16, applying the framework to multiple areas would take a similar amount of time by using one computational core for each area.

29. Page 5, line 8: a reference for OMI is missing.

Added.

30. Page 6, line 2-7: Removing the 30 outer pixel and the row anomaly will strongly reduce the number of pixels used in this research. How may pixels be still used?

22007 pixels. Added.

31. Page 6, line 8: space => grid

Changed. Corresponding texts are now in Sect. 2.3.4

32. Page 6, line 11: The footprint does not change, the location of the footprint changes from one day to another.

Changed.

33. Page 6, line 14: the year of the reference to Fioletov is missing.

The sentence has been changed.

34. Page 6, line 17: For purpose => For the purpose

Changed, now on page 8 line 5.

35. Page 7, Line 7: The assumptions of the PHLET model are not mentioned in Beirle et al. This reference should be removed.

A similar assumption on the vertical shape of NO₂ is taken in Beirle et al. (2011), as their model does not include information about the (horizontal and temporal) changes in the vertical shape of NO₂. In their online supporting information, "At the OMI observation time under cloud free conditions, the megacity emissions undergo rapid

vertically mixing (within some km distance from the source)”.

36. Page 7, Line 10: The transport from neighbouring regions is missing in this list while this is an important contribution.

We consider the transport from outside the study domain as part of the regional background. We write in the revised Sect. 2.3.2 (page 10 line 13-16) that

“Lightning emissions, biomass burning emissions, aircraft emissions, transport from neighboring regions, and convection can lead to NO₂ at higher altitudes over the YRD area. However, the amount of NO₂ aloft is much smaller than near-ground NO₂ due to large ground sources (Lin, 2012). Thus, we regard NO₂ aloft as the regional background, and do not include it in Eq. 1.”

37. Page 7, Line15-16: Can the authors give a reference for this statement.

The reference about aircraft emission is given. The statement has been adjusted since the biomass burning NO_x should be taken into consideration.

38. Page 7, Line 17-20: This is quite some assumption about the background value. What is the basis of this assumption? Why is the uncertainty set to 5%?

For background NO₂, our choice (0.54×10^{15} molecules cm⁻²) is based on the consideration that background NO₂ would be very small (e.g., smaller than 1×10^{15} molecules cm⁻²; Cui et al., 2016). Doubling the background only has marginal effects on our emission estimate especially at modest- and high-NO₂ locations. Spatially averaged, the error due to the choice of our background value is estimated as 5%.

39. Page 8, line 14: What is the source of the wind data?

Described in Sect. 2.3.3. The data are from ERA5.

40. Page 9, Line 20: space => grid

Changed.

41. Page 11, Line 7-8: I would suggest mentioning the average number of iterations (about 60?) needed to reach convergence and remove Fig C1. Is the value of 390 chosen based on this Figure and the fact that it is stable or are there other motivations?

There are 50 times of iterations before the convergence is reached according to the rate of decline of J. J is reduced from an initial value of 6585.2 to a stabilized value of 73.6.

We think the figure is important for the demonstration of how fast J is reduced. Thus,

we have elected to keep the figure (Fig. 2).

42. Page 12, Line 6: It becomes clearer if the short appendix C is just put into the text here.

Adjusted.

43. Page 12, Line 11: “inverted emission” is not the correct term. The concentrations are inverted to get the emissions. This “emission inversion” and “inverted emissions” is appearing in many places in the text.

Changed throughout the text.

44. Page 12, Line 11-12: What is the value of error on the lifetime? I suggest mentioning also the values of the calculated errors in the text.

As described on page 15 line 11-14,

“The error in the lifetime is derived from the errors in NO_x loss (estimated in Appendix C) and NO₂ VCDs, according to the common manner of error synthesis.”

45. Page 14, line 1: inverted => derived

Changed.

46. Page 14, line 14: Since there is a lot of agriculture in the YRD region a soil contribution of 0.9% seems very small and needs some explanation. A discussion on biomass burning emissions (which occur in the agricultural regions) can be helpful here as well.

The soil emissions in GEOS-Chem already account for the effects of both fertilizer and natural soil. We cannot conclude whether the relative contribution of soil emissions (to the total) has been underestimated, because of the dominant emissions from power plants, transportation, industry and residential activities.

We have clarified that biomass burning is part of the sources of our derived emissions. In Sect. 4.2 where we compare our derived “anthropogenic” emissions with other anthropogenic inventories, the GFED v4 biomass burning inventory is adopted to be subtracted from our derived emissions.

47. Page 15, Line 1: Figure 2e is mentioned without discussing 2a-d.

Figure a-d have been discussed before.

Now we have re-arranged the figures. Additional figures have been added, considering comments from both referees.

48. Page 15, Line 13: Please mention the basis of the coloring of Fig 2f.

As stated on page 18 line 20-22, The data points are colored to indicate the different ranges of VCDs at individual grid cells.

49. Page 15, Line 19: Why are the emissions not directly compared to bottom-up inventories instead of these proxies that are used in the bottom-up inventories. For example, a comparison with the MarcoPolo-Panda or the Zhao et al. inventory can give more insights.

In Sect. 4.3 we compare our emissions with other inventories besides MEIC.

50. Page 17, Line 5-6: To separate the anthropogenic emissions, GEOS-Chem is used to calculate soil emissions. What are the uncertainties of the soil emissions calculated by GEOS-Chem?

We have discussed the errors in soil emissions and biomass burning emissions in Sect. 4.2 on page 21 line 13-19:

“Soil emissions are calculated by the nested GEOS-Chem (Fig. 7c), with the uncertainties assumed to be within 50% (Wang et al., 1998; J. Yienger and Ii Levy, 1995). Biomass burning emissions (Fig. 7b) are taken from the Global Fire Emissions Database (GFED4; www.globalfiredata.org/data.html; last access: 2019/7/10) (Giglio et al., 2013), with the uncertainties estimated to be within 10% over the YRD (Giglio et al., 2009; Giglio et al., 2013). Summed over the study domain, the soil sources contribute about 0.5% of our emissions while biomass burning contribute about 5.1%.”

51. Page 17, Line 6: Figure 3ows?

Typo. Corrected.

52. Page 17, Line 8: Comparing Figure 3e and 3f is only useful if they are at the same resolution. Thus, Figure 3e should be regridded to the coarser resolution of Figure 3f.

All data are presented on $0.05^{\circ} \times 0.05^{\circ}$ grid.

53. Page 18, Lines 6-15: There is some repetition of the text of the previous sections

Rephrased.

54. Page 18, Line 19-20: I would remove the last sentence about the programming

language, which is not very relevant in a scientific paper

We have elected to keep the sentence, because we consider that programming with Python, a popular and easily used language, is an important feature that the potential users of our codes may appreciate.

55. Page 22: Line 17: Since one observation of the satellite is used in several grid cells, I doubt if the assumption that covariance matrices are diagonal matrices is correct. A discussion is needed here.

Since we use several pixels to get the mean VCD for each grid cell, and grid cells nearby each other may share the same pixels partly although weight differently. Therefore, we admit that making the covariance matrix to be diagonal may be an imperfect assumption, although a similar assumption has been used in many previous studies (Keiya and Itsushi, 2006; Cao et al., 2018).

To partly account for the uncertainty lead by such approximation, we assume relatively high errors in the VCDs, as shown in Sect. 2.2:

On page 8 line 5-13: “For the purpose of emission estimate, we assume that the error of VCD at a satellite pixel (σ_p) contains an absolute error of half of the mean VCD over the domain (i.e., 1.9×10^{15} molecules cm^{-2}) and a relative error of 30% (Lin et al., 2010; Boersma et al., 2011; Lin et al., 2015a; Beirle et al., 2011). We further add in quadrature an additional error (σ_g) when a satellite pixel is projected to the grid cells at a finer resolution; this error is important in the urban-rural fringe zone. For a given grid cell, σ_g is set to be 50% of the standard deviation of VCDs at its eight surrounding grid cells. Sampling over multiple days reduces the random error by a factor of $s = \left(\sqrt{(1-c)/n + c} \right)$, where c represents the fraction of systematic error (assumed to be 50%) and n the number of days with valid data (Eskes et al., 2003; Miyazaki et al., 2012). Thus, the total error for the temporally averaged VCD at a given grid cell is $\sigma_s = \sqrt{(\sigma_p^2 + \sigma_g^2)} \cdot s$.”

We have also summarized the limitation in the conclusion section:

On page 25 line 16-18: “The adjoint model assumes the observational error covariance matrix to be diagonal, without fully considering the effect of correlations between individual grid cells.”

56. References: Most references contain many spelling errors and omissions.

Checked.

57. Figure 2d: The lifetime is very short over the ocean, contradicting to what is usually

seen in the literature.

We agree that the lifetime over the ocean might be underestimated and the emission there is therefore overestimated. Some discussions are added on page 17 line 6-11 and on page 25 line 12-13.

58. Figure 2e: Although the gridding is on 0.05 degree resolution the actual spatial resolution of this image seems much lower. An indication if the intrinsic resolution can be obtained from the largest gradients in the emissions. I cannot detect clear structures with a 0.05 degree resolution. One would at least expect some power plants to show up as clear spots in this region.

Maybe the method is still limited to the OMI resolution. I would like to see some discussion about this.

Please see our response to general comment 8.

59. Figure 2f: The plot is more logical when the x-axis and y-axis are reversed. I also suggest drawing a line for the 100% relative error in this plot as a helpline to guide the eye.

Changed.

60. Figure 2 caption: What are the magnitudes of POMINO that are mentioned. In Figure 2f it is too small to see.

Changed.

61. Figure 3e: This has to be regridded to the resolution of 2f for comparison.

Adjusted.

62. Figure 3: I miss information on power plants, which are a major source of NO_x .

See our response to general comment 8.

63. Figure 4: The crosses in the plot, indicating the amount of grid cells, are too small.

The crosses have been deleted for simplicity.

64. Figure D1: I do not understand why we need two colors for the dots.

The blue points stand for the grid cells where the VCDs is lower than 5×10^{15} molecules cm^{-2} , and the evaluated local net sources at these grid cells are used to derive

the relation between the NO_x loss and NO_2 VCDs by fitting the fixed formula. An explanation is added on page 30 line 7.

65. Figure D2: The lifetime depends a lot on chemistry, temperature and precipitation. Therefore, the plot seems very simplified.

We agree, but this is the best we can do without involving a computationally much costlier 3-D chemical transport model. We note that previous studies with simplified models have often assumed a single value for lifetime for each city or other emission sources (Beirle et al., 2011; Liu et al., 2016).

We have added a discussion of the limitations of our method, including about the lifetime, in the revised conclusion section on page 25 line 18-20:

“Also, we assume a spatially uniform relationship between NO_2 VCDs and NO_2 lifetimes, which may lead to an underestimate in the lifetimes at low- NO_2 locations over the eastern sea.”