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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Advantages of 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" *by* Y. Zhao et al.

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We thank very much for the valuable comments from the reviewer, which help us improve the quality of our manuscript. Following is our point-by-point responses to the comments and corresponding revisions.

Reviewer #1

0. The authors' survey approach to estimate stationary emissions is a significant and

novel contribution to modelling air pollution in China, and highlights that more work is needed to improve emissions characterization of Chinese power generation and industrial activity. My main critique relates to some of the emissions and observational comparisons made in the analysis (detailed below), as well as highlighting areas where additional documentation would be helpful. Overall, the manuscript reads well and the presentation is clear (except where noted). I recommend that with revision, this manuscript be considered for publication in ACP.

Response and revisions:

We thank the reviewer's positive comment on the paper.

1. Some summary tables or figures are needed on activity levels (2010-2012) used in this analysis either in the supplemental material or main text, especially for coal-fired power plants, and other major industrial sectors. The authors highlight significant uncertainties in energy statistics for China (Section 4.6), so it is important that activity estimates are clearly summarized for reference.

Response and revisions:

We thank the reviewer's important comment. We have added a new Table S1 in the revised supplement, summarizing the activity levels for the continuous three years 2010-2012 in Nanjing by sector. We also added a paragraph in Section 4.6 (lines 872-891, Pages 33-34 in the revised manuscript) to discuss the uncertainty by source. In the paper, however, we cannot fully provide the detailed activity levels plant by plant in Nanjing, as we had a agreement with local Environmental Protection Agency, that the detailed unit/plant-based information should not be published without permission.

2. Captions are needed for all figures in the supplemental material.

Response and revisions:

We thank the reviewer's comment. The captions of all the figures in the supplement are provided in Pages S2-S3.

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3. Abstract. The horizontal resolution of the emissions inventory should be included. It is not clear what the authors mean by “high-resolution” for a reader just glancing at the manuscript.

Response and revisions:

We thank the reviewer’s comment. In lines 4-6, Page 2 of the revised manuscript, the horizontal resolution information has been included: “In this paper, a high-resolution emission inventory (with a horizontal resolution at 3×3 km) of air pollutants and CO2 for Nanjing, ...”

4. Section 2.1. This is where a table on activity of point and area sources (like Table S3 for EF) would be helpful.

Response and revisions:

We thank the reviewer’s suggestion and have added a new Table S1 in the revised supplement, which summarizes the annual activity levels for the main point and area sources for 2010-2012.

5. Page 18699, Line 13. How comprehensive is the spatial coverage of the Intelligent Traffic Violation Monitoring System? Does it cover all roads, or mostly highways? Could some traffic be missing from the monitoring system, which would contribute to underestimating emissions from motor vehicles? Also, can the authors provide a short description of how the monitoring system derives traffic flows (e.g. loop detectors, cameras, mobile cellphone data, etc.)?

Response and revisions:

We thank the reviewer’s important comment and admit that the relevant information was not clearly stated in the original manuscript. Cameras are used in the Intelligent Traffic Violation Monitoring System to continuously record the real-time traffic flows for most arterial roads and highways, and part of residential roads in the city. The system does not cover all the roads (particularly for residential roads), and the traffic

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flows in those roads were simulated from the real-time monitoring data for similar roads that are covered by the system. Since the emissions from on-road vehicles were first calculated with CORPERT based on the registered vehicle population and investigated mileages traveled for the whole city, and then spatially distributed to the road net of the city according to the monitored and simulated traffic flow intensities, there is little underestimation on the total emissions resulting from the missed traffic by Intelligent Traffic Violation Monitoring System. However, uncertainty does exist in the spatial distribution of emissions, particularly for areas with fewer cameras installed. We have described the system in lines 187-196, Pages 8-9 in the revised manuscript.

6. Section 3.1. What explains the growth and then decrease in NOx emissions shown in Figure 1a?

Response and revisions:

We thank the reviewer for the comment. In general, the inter-annual variation of emissions results from the combination of varied activity levels (e.g., coal consumption) and emission factors that are significantly influenced by emission control measures. In this case, the coal consumption in Nanjing increased largely from 31 million metric tons in 2010 to 35 in 2011, and it dominated the growth of NOX emissions, even with improved use of SCR in power sector. From 2011 to 2012, the growth in coal consumption was limited while the average removal efficiencies of SCR are significantly improved (as shown in Table 1), leading to reduced NOX emissions for the whole city. We have clarified this in lines 279-284, Page 12 in the revised manuscript.

7. Figure S2. A caption with acronym meanings would be helpful here. The color coding scheme is also hard to follow? It may help to color major source categories a single color (e.g. transportation, power plant, industrial, area, etc.), and different shades of that color for subcategories (e.g. cement, iron & steel production, etc.).

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We thank the reviewer's reminder. We have added the acronym meanings in the caption of Figure S2 in Page S2 of the revised supplement. The figure has been modified as suggested.

8. Section 3.1. If the penetration and removal efficiency dramatically increased for NOx control at coal-fired power plants (Table 1) between 2010 and 2012, why did the NOx emissions distribution among sources not change much year-to-year (Figure S2)?

Response and revisions:

We thank the reviewer for the important comment. The small inter-annual variation in sector distribution of NOX emissions is attributed mainly to following issues. First, as discussed in the response to Question 6, the variation in annual emissions from power sector resulted from the combination of the varied emission control levels and coal consumed by the sector. The growth in coal consumption of power plants partly offset the benefits of improved penetration and removal efficiency of SCR on NOX control, and the NOX emissions from power sector did not vary much for the three years (97, 112 and 94 Gg for 2010, 2011, and 2012, respectively). Second, besides power sector, emission controls were also improved for other sources, including the increased use of retrofitted low-NOX burner for industrial boilers, and the implementation of strict emission standards for new gasoline and diesel vehicles. For example, the emissions from other industrial combustion (OIN) were estimated to decrease from 22 to 14 Gg from 2010 to 2012, and the emissions from on-road vehicles did not change a lot despite of vehicle population growth. As a result, the fractions of NOX emissions from power plants, industrial boilers and on-road vehicles to total were estimated to change slightly from 2010 to 2012.

We have clarified this in lines 300-312, Page 13 in the revised manuscript.

9. Figures S3a and S3b. It is not clear in the figure which line refers to MEIC and which refers to emissions from this study. Also, a caption is needed. What does A and B refer to in panel a?

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Response and revisions:

We thank the reviewer's reminder. A and B in panel (a) refer to MEIC and this study, respectively. We have modified the caption and added the information in Page S2 of the revised supplement.

10. Section 4.1. I have some questions with regards to the inter-annual comparison of the emissions inventory with satellite NO2 column data, and the statement that NOx levels in the atmosphere decreased along with decreases in emissions (Line 24). First, what explains the large fluctuations in the vertical column densities in the beginning of 2010, 2011, and 2012 that are not seen in the preceding years? To what extent do year-to-year changes in meteorology affect the trend? Second, what are the uncertainties bars on the satellite-derived trends in NO2, and are these uncertainties larger than the changes in NOx emissions? If there is a decreasing trend after 2011, the analysis would be strengthened if additional years of satellite data were added beyond 2012.

Response and revisions:

We thank the reviewer's important comment, and admit that uncertainties exist in the comparison between the inter-annual variations in NOX emissions and NO2 VCDs and, and that the results should be interpreted cautiously. First, we have clarified in lines 432-434, Page 17 in the revised manuscript that each data point in Figure 4 is the 12-month moving averages, calculated as the means of the data for the previous and subsequent six months. Thus the VCD fluctuations at 2010, 2011 and 2012 were not at the beginning but the middle of the years. The fluctuations come mainly from inclusion or exclusion of extremely high monthly mean VCDs in winter. In particular, the lacking of data for certain months in the moving average calculation would exaggerate the fluctuations. The inter-annual changes in meteorology in Nanjing are summarized in a new Figure S5 in the revised supplement. During the study period (2010-2012), the variations were small for most parameters except for precipitation. Although varied precipitation has limited direct impacts on ambient NO2 level, it could change the data

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sampling of retrieval. Therefore we admit that the NO2 VCD from OMI could partly be influenced by the meteorology (particularly precipitation) besides the varied emissions, and we have discussed this in lines 456-460, Page 18 in the revised manuscript.

Second, the biases of NO2 VCD retrieval from OMI could reach 40% (Boersma et al., 2007; 2011), and the uncertainties are larger than the inter-annual changes in NOX emissions in the three years. However, since the emissions in this work are not directly compared with absolute values of VCDs but the relative trends, the uncertainties from NO2 VCD observation could be less influential on the reliability of result. We have added the discussion in lines 450-456, Page 18 in the revised manuscript. We have agreed with the reviewer that the period for emission and VCD comparison is short, and have modified Figure 4 with the latest information on NO2 VCDs included. As can be seen in the revised figure, the NO2 level in Nanjing did not increase after 2012. Analysis on long-term trend in emissions from the bottom-up method is further recommended for better understanding the NOX pollution in the area.

Reference:

Boersma, K. F., Eskes, H. J., Dirksen, R. J., van der A, R. J., Veefkind, J. P., Stammes, P., Huijnen, V., Kleipool, Q. L., Sneep, M., Claas, J., Leitao, J., Richter, A., Zhou, Y., and Brunner, D.: An improved tropospheric NO2 column retrieval algorithm for the Ozone Monitoring Instrument, Atmos. Meas. Tech., 4, 1905-1928, 2011.

Boersma, K. F., Eskes, H. J., Veefkind, J. P., Brinksma, E. J., vander A, R. J., Sneep, M., van den Oord, G. H. J., Levelt, P. F., Stammes, P., Gleason, J. F., and Bucsela, E. J.: Near-real time retrieval of tropospheric NO2 from OMI, Atmos. Chem. Phys., 7, 2103–2118, 2007.

11. Section 4.1, last paragraph. Could atmospheric chemistry and transport of urban emissions also be important for NOx enhancements seen over the Yangtze River? Are these areas downwind of Nanjing?

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Response and revisions:

We thank the reviewer's important comment. The area with high NO2 VCDs along the Yangtze River was located in eastern Nanjing. Given the dominating east wind of the city, it is upwind instead of downwind of Nanjing. However, we agree with the reviewer that sources other than ships (e.g., factories located along the river) could contribute to the high VCDs, and we have added this in lines 518-519, Pages 20-21 in the revised manuscript.

12. Figure 5. For clarity, the caption should mention that panel (a) refers to city-scale emissions from "this study".

Response and revisions:

We thank the reviewer's reminder and have revised the caption of Figure 5 as suggested.

13. Section 4.2. Given that NOx has a relatively short atmospheric lifetime, why is the slope in the correlation of emissions and concentrations so flat? What explains the large y-intercept? A more direct comparison of local emissions is to plot emissions versus enhancements in NOx, SO2, and CO above regional background levels.

Response and revisions:

We thank the reviewer's important comment and admit that the limitation of the correlation analysis was not fully discussed in the original manuscript, particularly for NO2 concentrations versus NOX emissions. In urban areas with plenty of emission sources and relatively large emission density, NO was found to account for large fraction of NOX in the ambient atmosphere (Zhou et al., 2008). As the dominating component of primary NOX directly from emission sources, NO is more difficult to be oxidized to NO2 in urban than in rural or remote regions, attributed to less resident time for chemistry conversion in urban. Given the 9 state-operated monitoring sites are all located in urban/suburban Nanjing, the observed NO2 concentrations were less sensitive or

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correlated to the NOX emissions, compared to the case of SO2 or CO. That partly explains the small slope of ambient NO2 levels to NOx emissions in Figure 7(a), and the lowest correlation coefficient among all the concerned species.

The y-intercepts can be considered as the approximation of regional background levels of corresponding air pollutants. As pointed by the reviewer, the value for NO2 is relatively high compared to SO2. Since the YRD is a developed region with large economy, high energy consumption, and, particularly, large vehicle population and intensive transportation, the background levels of NO2 could be enhanced, and the local emissions in Nanjing thus have a less significant impact on NO2 concentrations. We agree with the reviewer that comparisons between the emissions and enhancements in pollutants above regional background levels are more useful. Attributed to the limitation of observation sites (all of which are in urban areas), however, the background levels for the region have not been sufficiently observed and reported, and are thus currently unavailable for the analysis. (Note the y-intercepts are rough estimates as mentioned in the beginning of this paragraph, and the correlation coefficients between the emissions and the concentration enhancements above the y-intercepts would remain the same as those between the emissions and the original observed concentrations.) We are currently conducting the simulation on the regional background levels of certain air pollutants (SO2, NO2 and PM2.5) for the city using chemistry transport model, and more accurate estimates could be expected in near future. We have added the relevant discussions in lines 539-554, Pages 21-22 in the revised manuscript.

Reference:

Zhou, W., Wang X., Zhang, Y., Su, H., and Lu, K.: Current status of nitrogen oxides related pollution in China and integrated control strategy, Acta Scientiarum Naturalium Universitatis Pekinensis, 44, 323-330, 2008 (in Chinese).

14. Section 4.3.2. What was the basis for why the authors used the lowest daily OC/EC value observed as a surrogate for the primary emissions ratio? Also a figure with time

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series data of the OC/EC ratio should be included in supplemental material.

Response and revisions:

We thank the reviewer's important comment. The principle of determining the primary OC to EC, i.e., (OC/EC)pri, in the EC-tracer method is to exclude the effects of SOC formation as much as possible (Castro et al., 1999). (OC/EC)pri can be estimated by various ways, including the OC to EC ratio from emission inventory, OC to EC concentration ratio from observation when SOC formation is weak and thus the concentrations are dominated by emissions, or the lowest OC to EC concentration ratio during the observation. Given the relatively small offline sampling size in Li et al. (2015), they chose the lowest daily OC/EC as (OC/EC)pri to estimate the SOC level using the EC-tracer method. In this work, therefore, we have adopted their assumption and results. We admit the uncertainty from the limited data availability and recommend further improvement on observation. We have discussed this in lines 647-656, Page 25-26 in the revised manuscript.

The time series data of OC/EC ratios are taken directly from Li et al. (2015) and illustrated in a new Figure S7 in the revised supplement as required by the reviewer.

Reference:

Castro, L. M., Pio C.A., Harrison, R. M., and Smith. D. J. T.: Carbonaceous aerosol in urban and rural European atmospheres: estimation of secondary organic carbon concentrations, Atmos. Environ., 33, 2771-2781, 1999.

Li, B., Zhang, J., Zhao, Y., Yuan, S. Y., Zhao, Q. Y., Shen. G. F., and Wu, H. S.: Seasonal variation of urban carbonaceous aerosols in a typical city Nanjing in Yangtze River Delta, China, Atmos. Environ., 106, 223-231, 2015.

15. Section 4.3.2, Line 15. McDonald et al. also found factor of 2 higher OC/EC ratios from Los Angeles filter samples that did not use backup filters to correct for a positive OC artifact. Given the large uncertainty in the OC measurements used in this study,

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how confident can the authors be that their emissions ratio is closer to the artifact corrected ambient OC/EC ratio than MEIC (Line 4)? It is possible that the true ambient OC/EC ratio is lower than the two emission inventories. McDonald, B.C., A.H. Goldstein, and R.A. Harley, Long-term trends in California mobile source emissions and ambient concentrations of black carbon and organic aerosols. Environmental Science & Technology, 2015. DOI: 10.1021/es505912b.

Response and revisions:

We thank the reviewer's crucial comment and agree that different sampling/analytical methods can lead to considerable uncertainties in observation of ambient carbonaceous aerosol concentrations, and that such uncertainty could be important for evaluation of emission inventories. We have cited the study suggested by the reviewer, and have stated that the artifacts of OC measurement can vary between cities. In Nanjing, such artifacts have not been well quantified, and it is thus difficult to make a conclusive evaluation or comparison on the results from different bottom-up emission inventories, as indicated by the reviewer. Therefore, we have revised the text and deleted the words that imply a better estimate by this work over MEIC in lines 672-680, Page 26 in the revised manuscript. Improved analysis on artifacts of OC observation has also been recommended for the city (lines 680-685, Page 26-27 in the revised manuscript).

16. Section 4.3.3. The authors suggest that oxidation of NMVOCs could be an important source of secondary CO (Page 18716, Line 17). To control for chemistry, the authors could also see how ambient ratios of CO2/CO compare during morning periods (i.e., when photochemistry is slower).

Response and revisions:

We thank the reviewer's important comment. Following the reviewer's suggestion, we have checked the observation data during the night till early morning (i.e., from 6pm to 6am) for the subset (2), and the ambient CO2 to CO ratio is estimated at 89.2, larger than the value based on daily average concentrations, at 91.9. The results thus confirm

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the reviewer's judgment that photochemistry is slower at relatively lower temperature, leading to less secondary CO formation in the atmosphere, and thereby larger CO2 to CO ratio. If the CO concentrations are further revised based on the description in Section 4.4, the modified ambient CO2 to CO ratio is calculated at 70.7 for 6pm to 6am period. Compared to the ratio based on daily average concentrations at 66.8, the ratio for 6pm to 6am period is closer to the result from city-scale emission inventory at 76.1. We have added the discussions in lines 738-742, Page 29 and lines 800-802, Page 31 in the revised manuscript.

17. Table 1. What does "Gas release ratio" mean?

Response and revisions:

It means the fraction of flue gas that is not recycled or collected for purification treatment, and is thus directly released to the atmosphere. We have added the information as a note of Table 1.

18. Figure 1(b). The points are hard to see when they overlap. Maybe a bar chart would be better here, similar to panel (a).

Response and revisions:

We thank the reviewer's suggestion and have revised the figure accordingly.

19. Table S1. What do stages mean?

Response and revisions:

"Stages" mean the staged emission standards for vehicles. China I-IV are equivalent to Euro I-IV, respectively, as stated in line 184, Page 8 in the revised manuscript. To avoid confusion, we have changed to words to China I-IV in the revised Table S2.

20. Table S3. For completeness, emission factors for coal-fired power plants and cement facilities should be added to the table.

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Response and revisions:

We thank the reviewer's comment, and have added the information on emission factors for coal-fired power plants and cement facilities in Table S4 in the revised supplement.

21. Title: Should read "Advantages of *a* city-scale emission inventory".

Response and revisions:

We thank the reviewer's reminder and have revised the title as suggested.

22. Figure 8. "Original" observation is misspelled in the figure.

Response and revisions:

We thank the reviewer's reminder and have revised the figure as suggested.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 18691, 2015.