

### Reviewer 3

It is of great significance to know well about the recent trend and future direction of anthropogenic emissions for air pollutants in East Asia, which has recently become the world leader in air pollutant emissions. The study conducted by S.X. Wang and coauthors provides an annual trend for the period from 2005 to 2010 in NO<sub>x</sub>, SO<sub>2</sub>, PM<sub>10</sub>, PM<sub>2.5</sub> and NMVOC emissions in East Asia based on the detail country-specific information about recent control measures (energy-saving measures and end-of pipe control measures) and also compare it with the previous studies and observation in China. In addition the authors projected future emissions up to 2030 for six emission scenarios, considering both energy-saving and end-of-pipe measures. The authors provide recent emission inventory and future emission projections which are more reliable than previous works in East Asia. And also the emission scenarios developed in this work will make a great contribution to the policy making for atmospheric environmental management in East Asia. Consequently, this reviewer believes that the paper is of the interest of ACP and recommends publishing this paper with minor revisions in response to the following questions, comments, and suggestions.

Response: We thank the reviewer for supporting the publication of our manuscript. We also appreciate his or her comments which help us improve the quality of our manuscript. We address the reviewer's comments below. The original comments are in blue and our responses are in black.

1. Page 2617, Line 14: I suggest that "Non-energy related sectors" should be changed to "Solvent use" or other appropriate title because solvent use emissions alone are discussed in this section. Section 3.2.5 is similar.

Response: we agree with the reviewer and have replaced the titles of Section 2.2.5 and Section 3.2.5 with "Solvent use", and "Solvent use and biomass open burning", respectively.

2. Page 2621, Lines 16-17: "Japan's NMVOC emissions decreased by 30% mainly because of the implementation of stringent vehicle emission standards." is correct? Fig. 3e shows that a decrease in solvent use and others is larger than a decrease of transportation.

Response: We appreciate the reviewer's valuable comment. We agree that the decrease in solvent use is even larger than that of transportation. Therefore, we revised the statement as follows in the revised manuscript:

Japan's NMVOC emissions decreased by 30%, mainly attributed to the government's efforts

to reduce the emissions from solvent use and the implementation of stringent vehicle emission standards. (Page 18, Line 9-11 in the revised manuscript)

3. Chapter 3: The authors should add the explanation about activity data for industrial process, such as cement production and cokes production, and for evaporative NMVOC sources, such as solvent use and others.

Response: We thank the reviewer for this valuable comment. Below we describe the method for the projection of the activity data for industrial process, fossil fuel distribution, and solvent use. For industrial process, we just include a brief description in the revised manuscript and refer the readers to Zhao et al. (2013) for detailed information. As for fossil fuel distribution and solvent use, we have added the descriptions below accordingly in the revised manuscript. (from Page 22, Line 15 to Page 23, Line 2 in the revised manuscript)

#### 1) Industrial process

We applied an elasticity coefficient method for the estimation of future production of industrial products, the governing equation of which is as follows:

$$Y_{t1} = Y_{t0} \left( \frac{dv_{t1}}{dv_{t0}} \right)^{\delta} \quad (1)$$

where,  $t0$ ,  $t1$  are time periods, e.g.,  $t0 = 2010$ , and  $t1 = 2030$ ;  $Y$  is the yield of a specific industrial product;  $dv$  is the driving force, namely sectoral value added or population;  $\delta$  is the product specific elasticity coefficient. The values of  $\delta$  are determined through (1) historical trend during 1995-2010; (2) experience of developed countries; (3) projections of industrial associations. Generally speaking, most energy-intensive products for infrastructure construction are expected to increase until 2020, and stabilize or even decline after 2020, whereas the products closely related to everyday life are expected to increase until 2030, though at a declining rate. Future yields in PC scenario are less than those in BAU scenario because of a more conservative life style.

#### 2) Fossil fuel distribution

The increase of fossil fuels stored and distributed is expected to be consistent with the increase of total fuel consumption in the future. The gasoline or diesel sold at service stations is expected to have the same growth rate with fuel consumption of the transportation sector. Therefore, the future activity levels of this sector are derived from the projections of fuel consumption.

#### 3) Solvent use

The activity data for the solvent use sector are the consumption of products containing solvents. The forecast approach, which is consistent with Wei et al. (2011), is illustrated as follows:

$$A_{t1} = \sum_j \left( A_{t0,j} \times \frac{Y_{t1,j}}{Y_{t0,j}} \right) \quad (2)$$

where,  $t0$ ,  $t1$  are time periods, e.g.,  $t0 = 2010$ , and  $t1 = 2030$ ;  $j$  represents the industries using a specific solvent product;  $A_{t1}$  is the consumption of this solvent product in the year  $t1$ ;  $A_{t0,j}$  is the consumption of this solvent product in industry  $j$  in the year  $t0$ ;  $Y_{t0,j}$  and  $Y_{t1,j}$  are the yields of the major products (e.g., crude steel for the iron and steel industry) for industry  $j$  in the year  $t0$  and  $t1$ , respectively. The yields of industrial products were projected using the methodology illustrated by Eq. (1).

## References:

- Wei, W., Wang, S. X., Hao, J. M., and Cheng, S. Y.: Projection of anthropogenic volatile organic compounds (VOCs) emissions in China for the period 2010-2020, *Atmos. Environ.*, 45, 6863-6871, doi: 10.1016/j.atmosenv.2011.01.013, 2011.
- Zhao, B., Wang, S. X., Xu, J. Y., Fu, K., Klimont, Z., Hao, J. M., He, K. B., Cofala, J., and Amann, M.: NO<sub>x</sub> emissions in China: historical trends and future perspectives, *Atmos. Chem. Phys.*, 13, 9869-9897, doi:10.5194/acp-13-9869-2013, 2013.

## 4. Page 2637, Line 8: “NO<sub>x</sub>” is “NMVOC”?

Response: We thank the reviewer for this comment. “NO<sub>x</sub>” has been replaced with “NMVOC” in the revised manuscript. (Page 34, Line 10 of the revised manuscript)

## 5. Page 2639, Line 12: “Zhao et al. (2013d)” is missing in references.

Response: We did include Zhao et al. (2013d) in the manuscript but it was dropped during the typesetting process. We apologize for this. We will make sure it is modified before the final revised paper is published.

## 6. Page 2644, Lines 6-7: The authors should explain the reason why the change from 2009 to 2010 in the SO<sub>2</sub> emissions in this study and SO<sub>2</sub> VCD from satellite observations are quite different.

Response: We thank the reviewer for this comment. Figure R1(a, b) (or Figure 5(a, b) in the revised manuscript) show the comparison of SO<sub>2</sub> vertical column density (VCD) from

satellite observations with emission estimation. Lu et al. (2011) shows a significant increase in SO<sub>2</sub> VCD between 2009 and 2010 (especially that retrieved from SCIAMACHY), while Fioletov et al. (2013) shows a slight increase. Fioletov et al. (2013) implied that the pronounced increase between 2009 and 2010 arises from time-dependent bias in the retrieval algorithms. When the filtering procedure developed in Fioletov et al. (2013) was applied, the pronounced increase turned into only a slight increase. Therefore, we exclude the SO<sub>2</sub> VCD in 2010 in Lu et al. (2011) for the comparison with the temporal trend derived from emission estimation. Actually we have explained this briefly in the original manuscript. In the revised manuscript, we provided a more detailed explanation to this point. (Page 40, Line 1-7 in the revised manuscript)

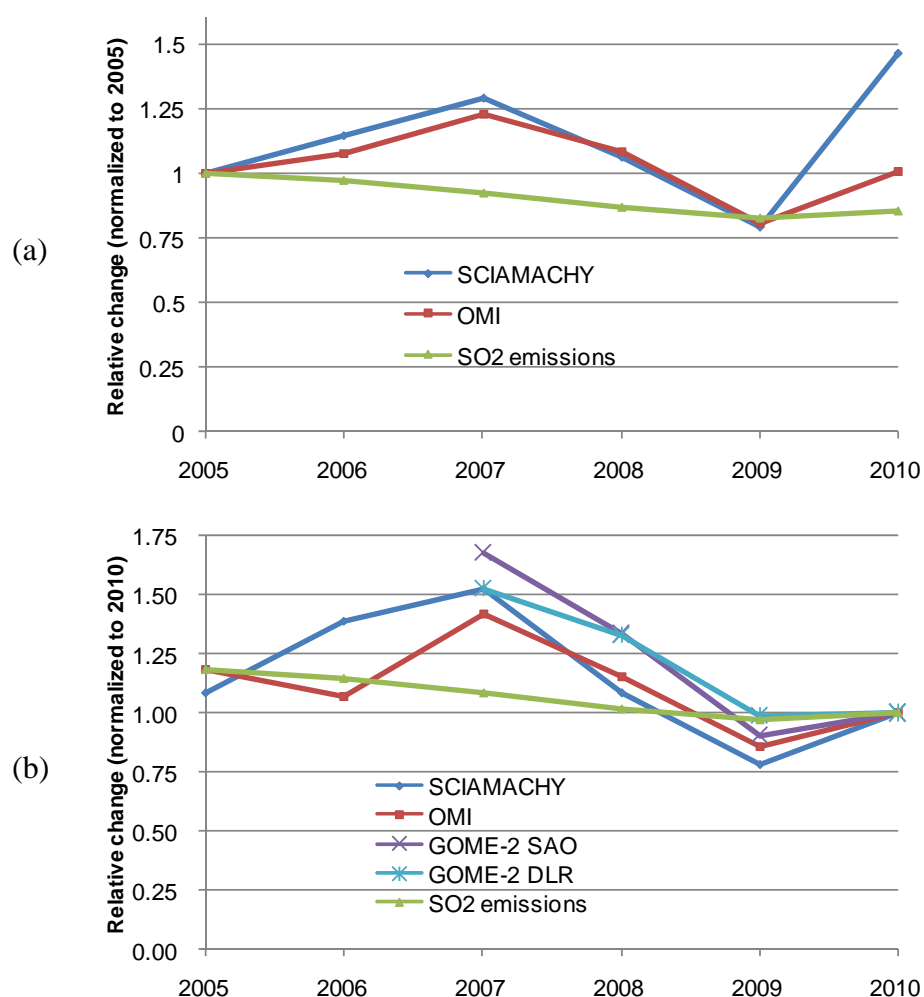


Figure R1 Inter-annual relative changes of SO<sub>2</sub> VCD from satellite observations and emission estimation in this study. (a) Average SO<sub>2</sub> VCD and total SO<sub>2</sub> emissions in Eastern Central China (latitude <45°N, longitude >100°E). SO<sub>2</sub> VCD was derived by Lu et al. (2011). All data

are normalized to 2005. (b) Average SO<sub>2</sub> VCD and total SO<sub>2</sub> emissions over an area of Eastern China (34°N–38°N, 112°E–118°E). SO<sub>2</sub> VCD was derived by Fioletov et al. (2013), in which a filtering procedure was applied to remove local biases, in particular volcanic signals. All data are normalized to 2010 because the data of GOME-2 are only available since 2007. This figure is consistent with Figure 5 (a, b) in the revised manuscript.

#### References:

- Fioletov, V. E., McLinden, C. A., Krotkov, N., Yang, K., Loyola, D. G., Valks, P., Theys, N., Van Roozendael, M., Nowlan, C. R., Chance, K., Liu, X., Lee, C., and Martin, R. V.: Application of OMI, SCIAMACHY, and GOME-2 satellite SO<sub>2</sub> retrievals for detection of large emission sources, *J. Geophys. Res-Atmos.*, 118, 11399–11418, doi:10.1002/jgrd.50826, 2013.
- Lu, Z., Zhang, Q., and Streets, D. G.: Sulfur dioxide and primary carbonaceous aerosol emissions in China and India, 1996–2010, *Atmos. Chem. Phys.*, 11, 9839–9864, doi:10.5194/acp-11-9839-2011, 2011.

#### 7. Page 2645, Line 14: What is the evidence about “PM<sub>2.5</sub> concentration still increased in a large part of China”?

Response: We appreciate the reviewer’s comment. In fact, this conclusion is mainly based on the simulation results in our previous paper (Zhao et al., 2013). The observations of PM<sub>2.5</sub> concentrations were quite rare in China during 2005–2010, and there were few continuous monitoring data for the whole period. Aerosol Optical Depth (AOD) from satellite increased in a large part of China during this period, with especially pronounced increase in the Sichuan Basin and the southern part of the North China Plain. This serves as a circumstantial evidence rather than a direct proof for the increase of PM<sub>2.5</sub> concentrations in some parts of China. To be more accurate, we emphasize that this conclusion is based on the modeling results in the revised manuscript. (Page 41, Line 13–25 of the revised manuscript)

#### References:

- Zhao, B., Wang, S. X., Dong, X. Y., Wang, J. D., Duan, L., Fu, X., Hao, J. M., and Fu, J.: Environmental effects of the recent emission changes in China: implications for particulate matter pollution and soil acidification, *Environ. Res. Lett.*, 8, 024031, doi:10.1088/1748-9326/8/2/024031, 2013a.

#### 8. Table 1: For the definition of BAU [1], the authors should specify that the assumption for end-of-pipe control strategy is for only China.

Response: We thank the reviewer for this comment. In the revised manuscript, we have

modified the definition of the BAU[1] scenario as follows:

The BAU[1] scenario assumes the energy-saving policies of the BAU scenario. For an end-of-pipe control strategy in China, it assumes that new pollution control policies will be released and implemented, representing a progressive approach towards future environmental protection. For the other countries in East Asia, the assumptions of the BAU[1] scenario are exactly the same as the BAU[0] scenario. (Page 20, Line 19-22; Page 24, Line 13-19; and Table 1 of the revised manuscript)

9. Table 6b: For LDB-B and CAR, the vehicle emission standards are incorrect in the sequence and in the number of penetration (%).

Response: We appreciate the reviewer's carefulness. The table in our manuscript was correct but there seems to be a problem with the typesetting process again. We are sorry for this. We will make sure it is modified before the final revised paper is published. The correct table is shown as follows. Note that Table 6 in the original manuscript corresponds to Table 5 in the revised manuscript.

Table 5. Penetrations of vehicle emission standards in China, Japan, and South Korea (%).

(b) Japan

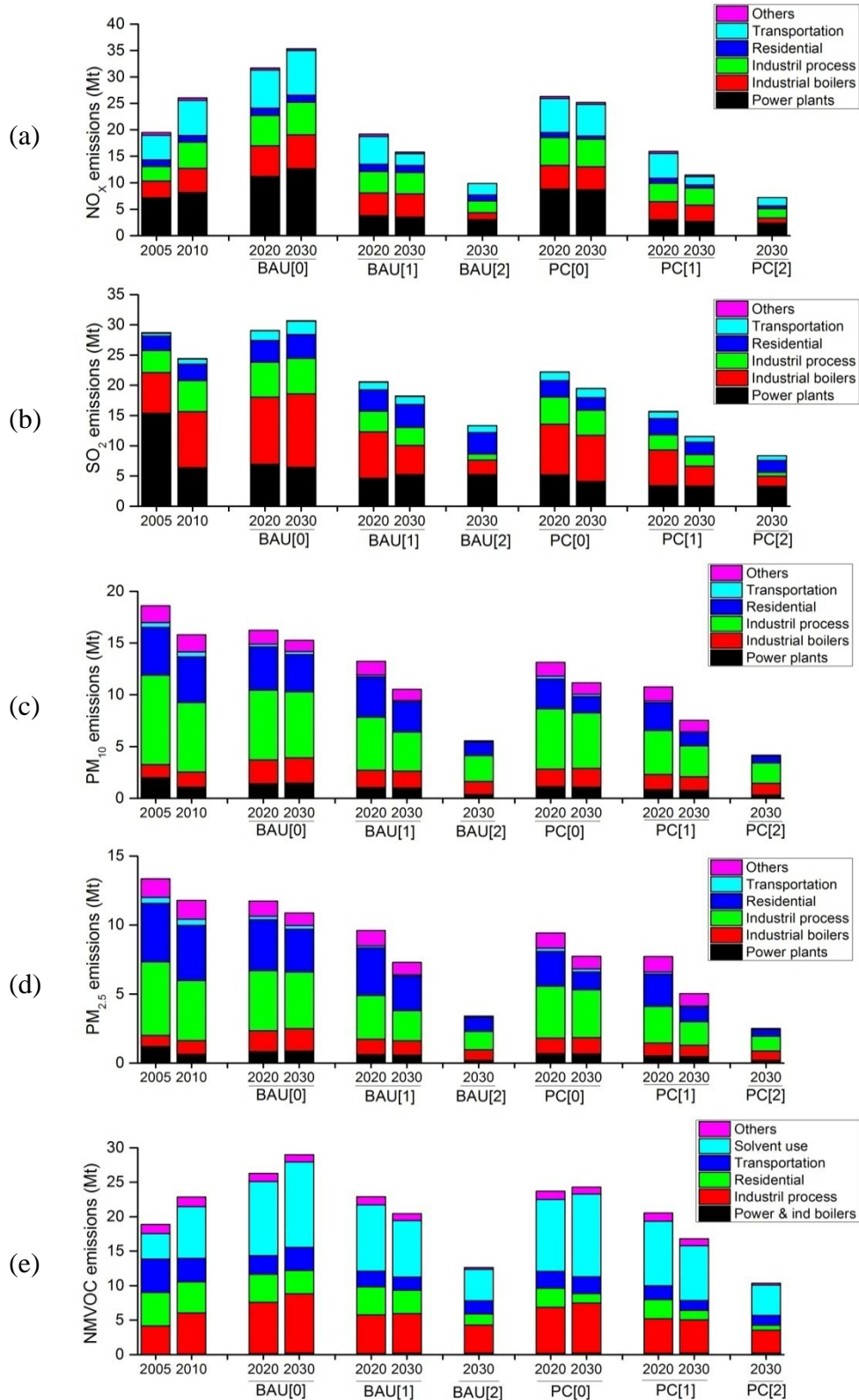
| Vehicle | Standard | BAU[0]/<br>BAU[1]/<br>PC[0]/<br>PC[1] |      |      |      |      | Vehicle | Standard | BAU[0]/<br>BAU[1]/<br>PC[0]/<br>PC[1] |      |      |      |      |
|---------|----------|---------------------------------------|------|------|------|------|---------|----------|---------------------------------------|------|------|------|------|
|         |          | Base year                             |      |      |      |      |         |          | Base year                             |      |      |      |      |
|         |          | 2005                                  | 2010 | 2020 | 2030 | 2030 |         |          | 2005                                  | 2010 | 2020 | 2030 | 2030 |
|         |          | 2005                                  | 2010 | 2020 | 2030 | 2030 |         |          | 2005                                  | 2010 | 2020 | 2030 | 2030 |
| HDT-D   | BST      | 41%                                   | 25%  | 0%   | 0%   | 0%   | LDT-G   | BST      | 38%                                   | 16%  | 1%   | 0%   | 0%   |
|         | ST       | 27%                                   | 19%  | 1%   | 0%   | 0%   |         | ST       | 4%                                    | 2%   | 0%   | 0%   | 0%   |
|         | LT       | 26%                                   | 25%  | 22%  | 0%   | 0%   |         | LT       | 10%                                   | 6%   | 0%   | 0%   | 0%   |
|         | NST      | 7%                                    | 11%  | 10%  | 0%   | 0%   |         | 1998R    | 14%                                   | 10%  | 6%   | 0%   | 0%   |
|         | NLT      | 0%                                    | 20%  | 22%  | 7%   | 0%   |         | NST      | 34%                                   | 31%  | 19%  | 0%   | 0%   |
|         | PNLT     | 0%                                    | 0%   | 44%  | 93%  | 100% |         | NLT      | 0%                                    | 34%  | 24%  | 8%   | 0%   |
| HDB-D   | BST      | 52%                                   | 32%  | 0%   | 0%   | 0%   | LDB-B   | PNLT     | 0%                                    | 0%   | 49%  | 92%  | 100% |
|         | ST       | 19%                                   | 15%  | 2%   | 0%   | 0%   |         | BST      | 12%                                   | 4%   | 0%   | 0%   | 0%   |
|         | LT       | 25%                                   | 24%  | 23%  | 0%   | 0%   |         | ST       | 4%                                    | 1%   | 0%   | 0%   | 0%   |
|         | NST      | 5%                                    | 8%   | 8%   | 0%   | 0%   |         | LT       | 4%                                    | 1%   | 0%   | 0%   | 0%   |
|         | NLT      | 0%                                    | 20%  | 22%  | 8%   | 0%   |         | 1998R    | 16%                                   | 6%   | 3%   | 0%   | 0%   |
|         | PNLT     | 0%                                    | 0%   | 45%  | 92%  | 100% |         | NST      | 63%                                   | 35%  | 17%  | 0%   | 0%   |
| LDT-D   | BST      | 41%                                   | 27%  | 0%   | 0%   | 0%   |         | NLT      | 0%                                    | 52%  | 27%  | 10%  | 0%   |
|         | ST       | 27%                                   | 20%  | 0%   | 0%   | 0%   |         | PNLT     | 0%                                    | 0%   | 53%  | 90%  | 100% |

|      |     |     |     |     |      |     |       |     |     |     |     |      |
|------|-----|-----|-----|-----|------|-----|-------|-----|-----|-----|-----|------|
| LT   | 27% | 23% | 22% | 0%  | 0%   | CAR | 1983R | 72% | 32% | 8%  | 0%  | 0%   |
| NST  | 5%  | 11% | 10% | 0%  | 0%   |     | NST   | 28% | 37% | 24% | 0%  | 0%   |
| NLT  | 0%  | 20% | 23% | 7%  | 0%   |     | NLT   | 0%  | 31% | 23% | 9%  | 0%   |
| PNLT | 0%  | 0%  | 46% | 93% | 100% |     | PNLT  | 0%  | 0%  | 46% | 91% | 100% |

Notes: BST, before short term target; ST, short term target; LT, long term target; NST, new-short term target; NLT, new-long term target; PNLТ, post new-long term target; 1998R, 1998 regulation; 1983R, 1983 regulation.

10. Figs. 2-4: These figures are too small to be visible and should be improved.

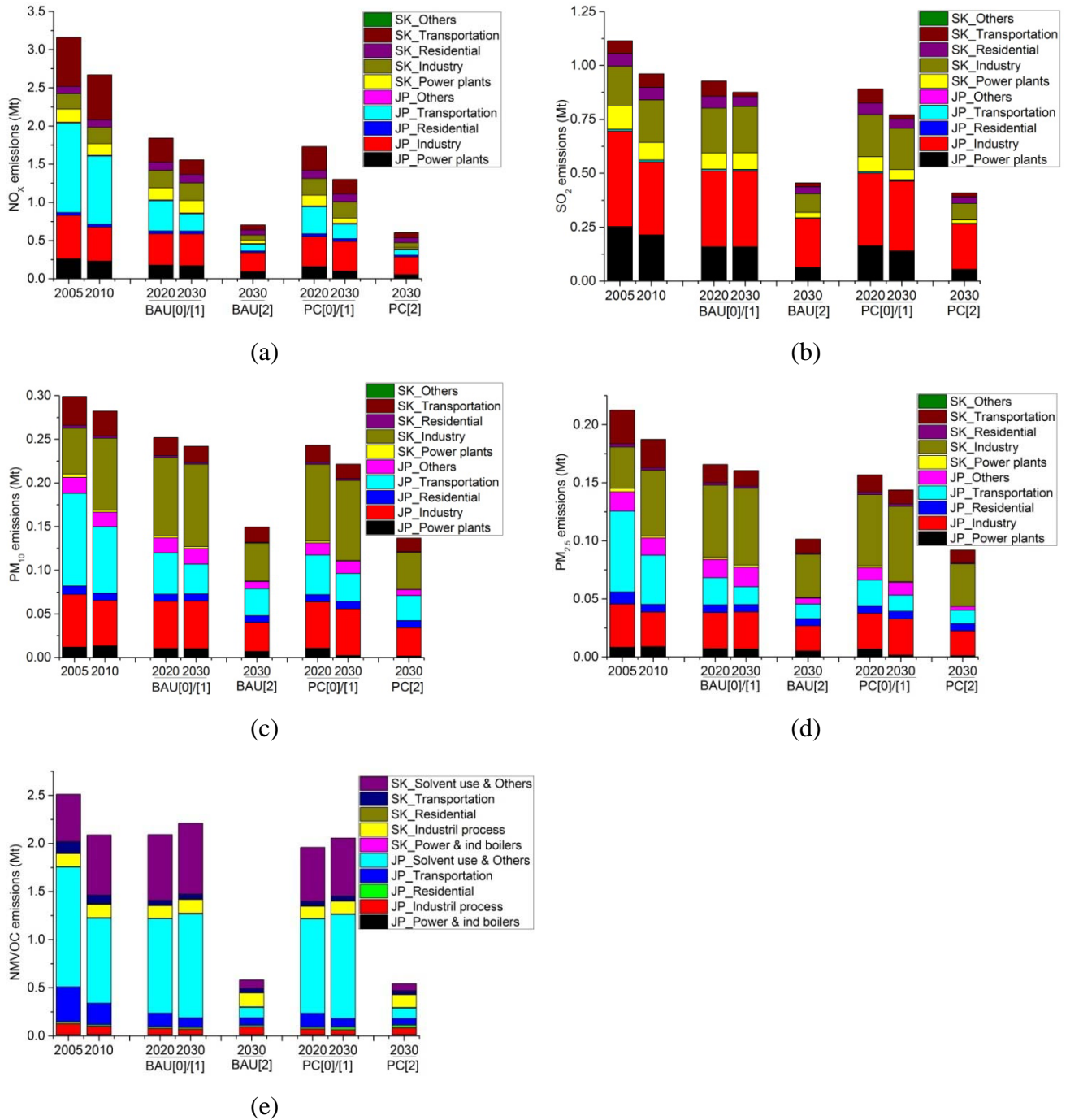
Response: We thank the reviewer for the comment. We have improved the quality of these figures and show the revised figures below. We will also assure that the figures are clear enough when the final revised paper is published.



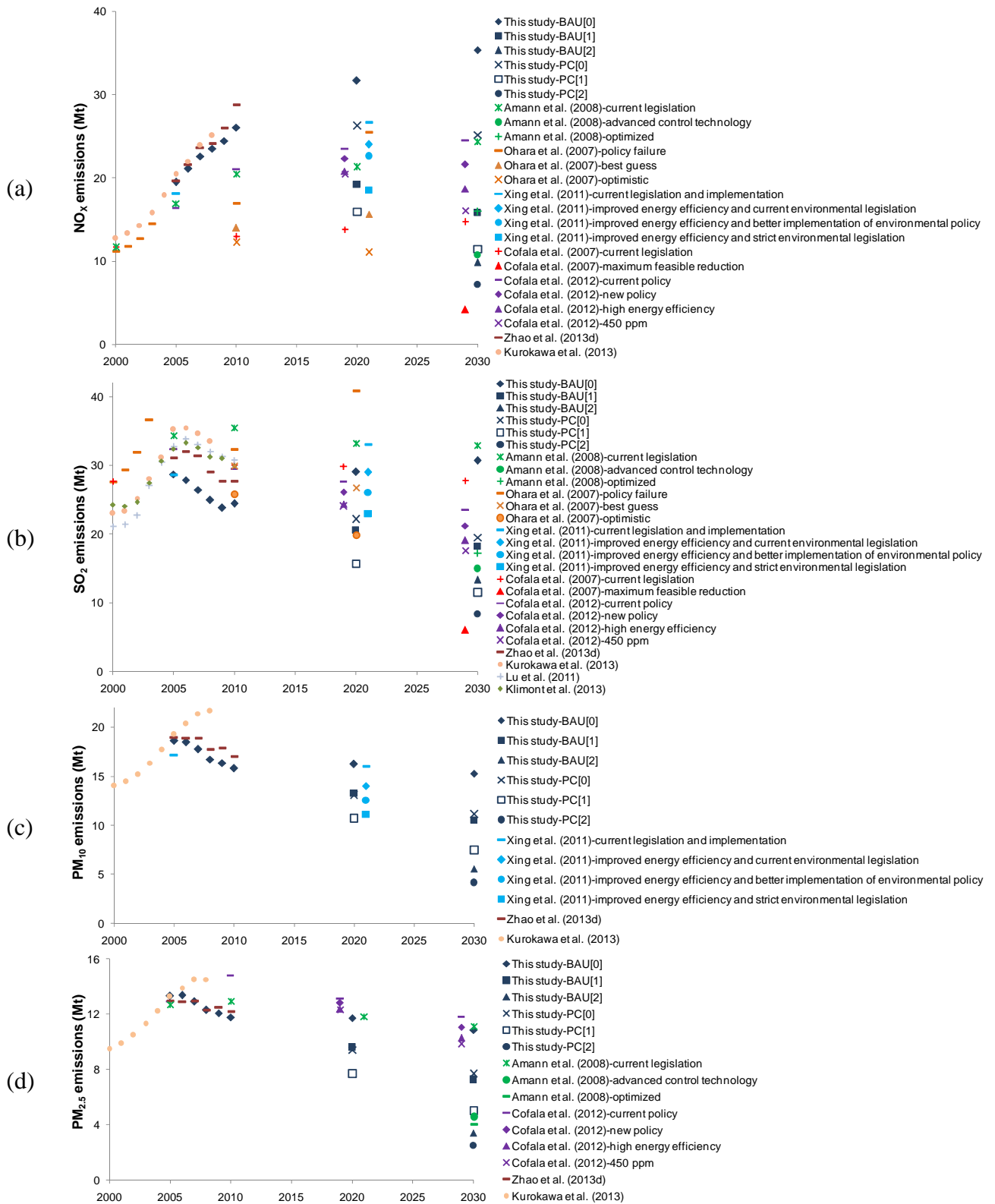
2 Figure 2. Emissions of major air pollutants in China and their sectoral distribution during  
3 2005-2030: (a)  $\text{NO}_x$ ; (b)  $\text{SO}_2$ ; (c)  $\text{PM}_{10}$ ; (d)  $\text{PM}_{2.5}$ ; (e) NMVOC. The sector of “Others”  
4 represents biomass open burning for  $\text{NO}_x$ ,  $\text{SO}_2$ ,  $\text{PM}_{10}$ , and  $\text{PM}_{2.5}$ ; for NMVOC, it includes  
5 biomass open burning, waste treatment, cooking, and smoking, with biomass open burning  
6 contributing over 80% of the total NMVOC emissions of this sector.

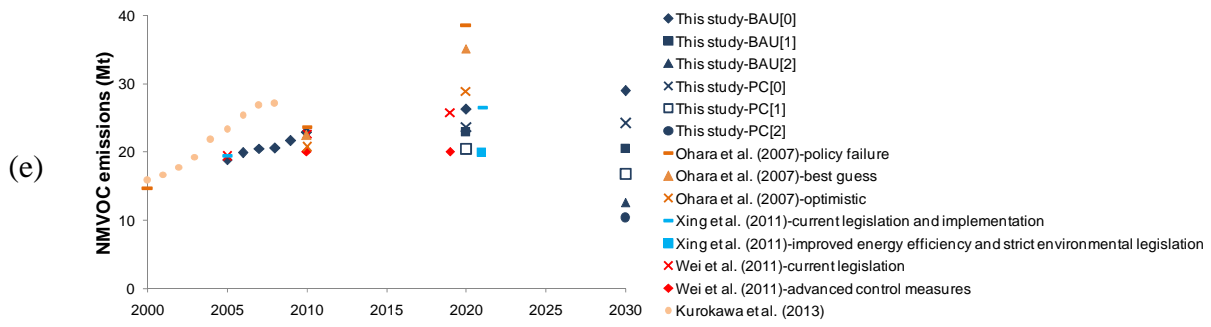


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2 Figure 3. Emissions of major air pollutants in Japan and South Korea and their sectoral  
3 distributions during 2005-2030: (a)  $\text{NO}_x$ ; (b)  $\text{SO}_2$ ; (c)  $\text{PM}_{10}$ ; (d)  $\text{PM}_{2.5}$ ; (e) NMVOC. JP and  
4 SK in the legend represent Japan and South Korea, respectively. The sector of “Others” is  
5 mainly biomass open burning.





1 Figure 4. Comparison of emission estimation in this study with other studies: (a) NO<sub>x</sub>; (b)  
 2 SO<sub>2</sub>; (c) PM<sub>10</sub>; (d) PM<sub>2.5</sub>; (e) NMVOC. The scenarios from the same study are shown with  
 3 symbols of the same colour, and only the historical emissions for the first scenario are shown.  
 4 Some points for the years 2020 and 2030 are shifted a little left or right, in order to avoid  
 5 overlapping. Note that the current legislation scenario in Amann et al. (2008) is consistent  
 6 with the baseline scenario in Klimont et al. (2009), and the historical emission trends of Zhao  
 7 et al. (2013a) is consistent with this study. Therefore, Klimont et al. (2009) and Zhao et al.  
 8 (2013a) are not shown in the figures.