Anonymous Referee #2

A very interesting approach is shown to analyse the ratio of CO/NOx and SO2/NOx spatially over megacities and its development over time. The manuscript is basically well-written but it contains some carelessness, which I will mention below.

Response: We thank the reviewer for his/her helpful comments on improving the manuscript. We have carefully studied the comments and carried out the revisions accordingly. We believe we have addressed all of them completely. Below is a point-by-point response to the reviewer's comments. We have also provided a copy of the track-change manuscript as well as a clean copy of the revised manuscript.

Page 1, Line 15-19 [Our results ....relative to 2005]: This sentence is very confusing and ambiguously written. A range of ratios is given, but 4 cities are mentioned, it is relative to 2005 and the sentence is ending with an dependent clause. I suggest to split-up this sentence and give a some more explanation.

Response: Thank you. We have rephrased the sentence to:
"*We present results for four Chinese cities (Shenyang, Beijing, Shanghai, and Shenzhen) representing four levels of urban development. Our results show a robust coherent progression of declining-to-growing $\Delta CO/\Delta NO_2$ relative to 2005 (-5.4±0.7 to +8.3±3.1%), and slowly-declining ΔSO2/ΔNO2 (-6.0±1.0% to -3.4±1.0%) across the four cities. The coherent progression we found is not evident in the trends of emission ratios reported in Representative Concentration Pathway (RCP8.5) inventory.*"

Page 1, Line 20 [...sectors in Shanghai and Shenzen...]: Only Shanghai and Shenzhen are mentioned. What about Shenyang and Beijing?

Response: We have changed the sentence:
"*This progression is likely due to a shift towards cleaner combustion from industrial and residential sectors in Shanghai and Shenzhen, which is presently obfuscated by China's still relatively higher dependence on coal.*"
To

"*This progression is likely due to a shift towards cleaner combustion from industrial and residential sectors in Shanghai and Shenzhen that is not yet seen in Shenyang and Beijing. This overall trend is presently obfuscated by China's still relatively higher dependence on coal.*

Page 4, line 21, Page 5, line 14: If you are looking at a 2 x 2 degree area around cities in China, does this not lead to overlap, for instance, in the case of Guangzhou and Shenzhen.

Response: Using 2˚×2˚ area to represent cities does lead to slight overlap over Guangzhou and Shenzhen, Beijing and Tianjin. This does not affect our analyses of emission inventories because we apply geopolitical maps of city boundaries to calculate emissions for each city. This does have an impact on our analyses of satellite observations because we use all the grids in the 2˚×2˚ area to conduct the spatial regression. However, we do not expect the overlap to significantly change

our results because (1) the overlapped area is relatively small; (2) the overlapped cities are sometimes considered together as a whole region because of their similarities and connections (for example, the Jing-Jin-Ji megalopolis and the Pearl River Delta), and (3) the overlapped cities are in the same classes with similar patterns based on our analyses (i.e., Beijing and Tianjin are both in class 2, while Guangzhou and Shenzhen are both in class 4; Table 2).

We appreciate the reviewer for pointing this issue out and have included a similar statement above in Section 2.2.1 (Page 5 Line 8 – Line 17 of the track-changed manuscript).

Page 7, line 18: The results start with Figure 3, while Figure 2 is mentioned later. This is unusual, but moreover I think the storyline of your paper becomes clearer if you start with explaining Figure 2 first.

Response: We have restructured Section 3.1 to start with discussion on Figure 2, followed by discussion and analysis of Figure 3. Please see the revised manuscript for details.

Page 7, line 24-28: When I compare the numbers of the given ratios with Figure 3, the unit reads %/year instead of %. The rate is in fact an annual rate.

Response: Thank you for pointing it out. The rate is indeed an annual rate and we have changed "%" to "%/year" in the revised manuscript.

Page 7, line 31 (also on Page 10, line 7): Which four levels of development do you mean? These developments within cities is a very important aspect of the paper, nevertheless the four levels are not discussed nor defined.

Response: Thank you. The four levels in this study are defined using broad clustering between the average GDP per capita per year and the rate of change in $\Delta CO/\Delta NO_2$ derived from satellite observations. This is shown in Table 2, where a general rule resulting from this analysis would be a classification mainly based on GDP per capita per year, except Harbin and Wuhan.

We have added this statement to Section 3.1 (Page 8 Line 9 – Line 12 of the track-changed manuscript).

Page 8, line 14: Here a reference to Figure 3a is made. However, Figure 3a is not defined while the first subfigure is about Shenyang.

Response: Thank you. We have added names for each subfigure in Figure3, and changed "Figure 3a" to "Figure 3e" to refer Los Angeles in the text.

Page 11, line 17: The reference has been forgotten here.

Response: We thank the reviewer for pointing it out. We have added Shindell et al. (2011), Zhang et al. (2012), Kheirbek et al. (2014), Yang et al. (2016), and Paulot et al. (2017) to the sentence as references.

Appendix A, page 15, line 17: "..the fractional contribution of x emission sector f."
Change to "..the fractional contribution of emission sector f for species x."

Response: We have changed *"... the fractional contribution of x emission sector f."* to *"... the fractional contribution of emission sector f for species x."*

Appendix B: The estimation of H depends on a-prior information because it is an underdetermined problem. Can you also give an indication how much information is coming from the measurements and how much of the a-priori ?

Response: Since **H** is drawn based on Monte-Carlo sampling, we do not have a diagnostic for the relative contributions of the prior and the data on **H**. We chose the mean across 100 **H** values resulting to estimates of $\mathbf{H}\hat{\mathbf{x}}$ with the lowest RMSEs relative to the data. The changes in $\widehat{\mathbf{H}}$ relative to the $\mathbf{H_a}$ can be explored in the sectoral changes shown in Figure 4. This is especially the case for Shanghai and Shenzhen where the change in **H** is larger than the change in **x**.

We have added the statement above in the revised manuscript (Page 18 Line 16 – Line 20 of the track-changed manuscript).

Figure 3: - The grey area is very hard to see in this Figure. - It would also be helpful if the underlying data points of the fit are plotted in the Figure as has been done in Figure 2. - The error bars mentioned in the caption are missing.

Response: Thank you. We have adjusted the color of the grey area in Figure 3 (as well as Figure S1) to make it clearer.

We also added underlying data points of the fit of satellite trend in Figure 3 as well as Figure S1. We have deleted the descriptions of the error bars (already had been deleted from the figure), and added descriptions of the grey areas instead.