Anonymous Referee #1

This paper reported the decadal changes in the efficiency and cleanness of bulk combustion over large cities in mainland China using satellite observations. The authors have done a lot of works, which are very impressive. It is very interesting to see the temporal variations of SO2/NO2 and CO/NO2. The driving forces of the variations have not been well explained in the text, even though many details are provided. I recommend publishing the paper after reorganizing the parts about driving forces.

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.

General comments:

1. The influence of inter-annual variations due to meteorology. The authors mentioned that analyzing molar ratios rather than absolute molar concentrations contributes to decrease the effects of meteorology. I'm wondering how it works to account for the temporal variation in lifetimes of air pollutants associated with meteorology.

Response: As the co-emitted species (i.e., CO, SO_2, and NO_2) are subject to the same meteorological conditions (affecting transport, dilution/mixing, and lifetime), their enhancement ratios are expected to be less sensitive to meteorology compared to the absolute molar concentrations. This is supported by the fact that decadal $\Delta\mathrm{CO}/\Delta\mathrm{NO}_2$ as well as $\Delta\mathrm{SO}_2/\Delta\mathrm{NO}_2$ for different seasons have similar trends (Figure 2 of the manuscript). Previous studies have also proven that the ratios compared to the concentrations themselves are relatively immune to changing meteorological conditions, and can provide insights into the magnitude and temporal trends of the emissions (Parrish et al, 2002, 2006, 2009, Silva et al., 2013, Hassler et al. 2016). In addition, they can be directly compared to the corresponding emissions ratios under certain circumstances.

However, we note that even though the ratios derived from satellite observations are relatively less sensitive to meteorology, this methodology cannot eliminate all the impacts from meteorology. The enhancement ratios may be impacted by the meteorological conditions because lifetimes of different air pollutants may respond to meteorological conditions different as the reviewer succinctly pointed out. Nevertheless, we believe such impact should not influence our main conclusions for the following two reasons: (1) Our analysis focuses on decadal trends instead of short-term trends. As shown by previous study, meteorology also plays an important role on relatively short time scales, but meteorology probably plays a lesser role in the longer-term trends (Krotkov et al. 2016); (2) The satellite retrieval samples are taken over the megacities (right above strong emission sources) instead of downwind of the pollution sources, making them more representative of megacity sources.

We thank the reviewer for this helpful comment. We have incorporated the discussion above in the manuscript (Section 3.1, Page 8 Line 27 – Page 9 Line 2 of the track-changed manuscript).

2. The analysis focusing on the differences of SO2/NO2 between the US and China needs substantial improvements. The authors listed the possible reasons for the differences in Page 8. However, no quantitative analysis at urban scale has been performed. For instance, the shares of fuel usage and emissions contributions from different sources for typical cities are expected, which suggest the different emissions characteristic between the US and China. Additionally, the declining SO2/NOx is most likely caused by the de-SO2 procedure in China (Li et al., 2018). The related discussion is missing. The recent reduction in NOx emissions (van der A., et al, 2017; Liu et al., 2016) has not been discussed as well.

Response: We have revised and extended the analysis of the differences of $\Delta SO_2/\Delta NO_2$ between the US and China to address the reviewer's concern. We have also included additional references in this part to strengthen the analysis and discussion (Bhattacharya et al., 2015; Hassler et al., 2016; Liu et al., 2016; van der A., et al, 2017; Wu et al., 2017; Li et al., 2018; Sun et al., 2018; Zheng et al., 2018).

The following part:

"*Here, we postulate that the absence of an apparent shift in* $\Delta SO_2/\Delta NO_2$ *across the four Chinese cities is due to continuing heavier reliance of these cities (and China) on coal burning relative to United States (Wang and Hao, 2012; Qi et al., 2016; Yang et al., 2016). In 2005, it was estimated that coal accounts for about 69% and 23% of the total primary energy consumption in China and U.S., respectively. While there are on-going activities regulating coal-related emissions, including usage of low-sulfur coals, installation of flue gas desulfurization (FGD) facilities, and closing of small units, coal consumption in China remains to increase in the past decade (Qi et al., 2016; Yang et al., 2016). In terms of mass, it has increased by 70% from 2005 to 2014 (Korsbakken et al., 2016). On the other hand, the use of coal in U.S. has been found to be slightly decreasing along with previous adoption of* SO_2 *control technologies (Taylor et al., 2005).*"

have been changed and extended to

"*Here, we postulate that the absence of an apparent shift in* $\Delta SO_2/\Delta NO_2$ *across the four Chinese cities is due to continuing heavier reliance of these cities (and China) on coal burning relative to United States (Wang and Hao, 2012; Bhattacharya et al., 2015; Qi et al., 2016; Yang et al., 2016; Sun et al., 2018; Zheng et al., 2018). In terms of the sectoral share, the majority of NOx emissions over Los Angeles basin is from transport according to a recent fuel-based inventory (Hassler et al., 2016), whereas fossil fuel combustion (from power generation and industry) is the most dominant NOx source in China (Sun et al., 2018). In terms of the energy share, it was estimated that coal accounts for about 69% and 23% of the total primary energy consumption in China and U.S. in 2005, respectively. Actions including usage of low-sulfur coals, installation of flue gas desulfurization (FGD) facilities, and closing of small units, have been taken to reduce coal-related emissions in China. The aforementioned de-*SO_2 *procedure in China is most likely to be the dominant driving factor of the declining* $\Delta SO_2/\Delta NO_2$ *(Li et al., 2018; Zheng et al., 2018). While there are on-going activities regulating coal-related emissions, coal consumption in China remains to increase in the past decade (Qi et al., 2016; Yang et al., 2016). In terms of mass, it has increased by 70% from 2005 to 2014 (Korsbakken et al., 2016). On the other hand, the use of coal* *in U.S. has been found to be slightly decreasing along with previous adoption of SO_2 control technologies (Taylor et al., 2005). In addition, previous studies have reported recent reduction in NOx emissions over China since 2011 based on satellite observations and emission inventories (Liu et al., 2016; van der A., et al, 2017). The installation of selective catalytic reduction (SCR) equipment at power plants and new emissions standards for vehicles both contribute to the NOx emission reduction (Liu et al., 2016; van der A., et al, 2017; Wu et al., 2017). On the other hand, based on our analysis of decadal trends (2005-2014), only NO_2 over Shenzhen overall decreased in the decade, while 10-year average changes of NO_2 over Shenyang, Beijing, and Shanghai were overall positive (Table 2). Intradecadal changes as reported in Liu et al. 2016 (from increasing to decreasing NOx emissions around 2011) do not contradict the derived 10-year trend in this work, especially over Shenyang, and Beijing where NOx emissions are still rapidly increasing during the first half of the decade (2005-2011). The changes in SO_2 emissions and NO_2 emissions together contribute to the trends of $\Delta SO_2/\Delta NO_2$ that we found. Positive ΔNO_2 and negative ΔSO_2 produce negative $\Delta SO_2/\Delta NO_2$ over the three cities; while negative ΔSO_2 and negative ΔNO_2 (albeit smaller in magnitude) still produce negative $\Delta SO_2/\Delta NO_2$ but smaller magnitude over Shenzhen than $\Delta SO_2/\Delta NO_2$ over the other cities (Table 2). This indicates a stronger influence of the changes in SO_2 emissions (as reflected in ΔSO_2) in the decreasing trends of these ratios."*

3. Too many very lengthy sentences. The authors preferred long sentences through the manuscript. However, those sentences are too long to understand sometimes. I would suggest the authors to go through the text and to simplify some sentences when necessary.

Response: We thank the reviewer for point this out. We have gone through the whole manuscript to shorten and/or rephrase the long sentences. Please see the revised manuscript for details.

4. section 3.3. This section is trying to explain the driving forces of the trend. It contains many details and the readers may be easily lost. I would recommend the authors to summary the main findings and storyline somewhere in the beginning or at the end of the section, and to reorganize this section based on the summary.

Response: We have divided section 3.3 into three subsections: 3.3.1 Inverse Analysis of the Ratios, 3.3.2 Combustion Emission Pathway, and 3.3.3 Traces in Sectoral Emission Ratios. We have also added the following summary in the beginning of Section 3.3 (Page 11 Line 25 – Line 31 of the track-changed manuscript):

"*We define combustion emission pathway as a trajectory in time of the overall changes in emissions due to combustion with respect to socioeconomic development (e.g., Riahi et al., 2011; Steinberger et al., 2012; Li et al., 2016; Marangoni et al., 2017). In this section, we identify a common combustion emission pathway across these four levels of development and associate them to sectoral changes through inverse analysis. We will briefly describe the inverse analysis of the ratios in section 3.3.1, present our findings on combustion emission pathway in section 3.3.2, and elucidate the driving factors by means of time traces in sectoral emission ratios in section 3.3.3.*"

Specific comments:

1. Page 1, line 14, the phrase of "mature satellite instruments sounds not fine. What is the definition of mature? Which instruments are not mature?

Response: We have changed the phrase “mature” to “widely used”.

2. Page 1, line 31. The English seems to be incorrect.
Response: We have changed the following sentence
“*This is especially problematic since it is in these large cities where anthropogenic activities are most intense, accompanied by immense energy consumption mainly in the form of fossil fuel combustion (Mage et al., 1996; Kennedy et al., 2015)*”
to
“*Anthropogenic activities are most intense in megacities, accompanied by immense energy consumption mainly in the form of fossil fuel combustion (Mage et al., 1996; Kennedy et al., 2015)”*.

3. Page 6, line 8. “Here, we treat emissions of these species across the entire extent of the megacity as a point source” As far as my understanding, the authors discarded all the CO and SO2 measurements where there are no NOx measurements. Could you please clarify how do you set up the criteria and why does the criteria make the entire urban areas to a point source?

Response: Thank you. In this study, we only use co-located CO and NO_2 observations to derive $\Delta CO/\Delta NO_2$, and co-located SO_2 and NO_2 observations to derive $\Delta SO_2/\Delta NO_2$.

As described in the Section 2.2.1, each city is represented by a 2°×2° area around each city center. And within each city (2°×2° area), there are 400 of 0.1°×0.1° grids. In another words, during our analyses, a city is represented by 400 grids instead of one single point. And the spatial regression is conducted using 400 grids within each city to obtain enhancement ratios over the city. There is only one enhancement ratio derived from the spatial regression for each city every time. And the enhancement ratios represent bulk characteristics of spatially heterogeneous combustion sources within the megacity. By analyzing the enhancement ratios derived from the spatial regression over the city, we analyze the bulk characteristics of the whole city, which is a complex and mixed signal of all the emission sources and sectors within the city. Therefore, the sentence “*we treat emissions of these species across the entire extent of the megacity as a point source*” only corresponding to the aforementioned methodology of analyzing the bulk characteristics of the whole city using regression ratios. However, we understand the reviewer’s concern and realize this sentence could be confusing, so we have deleted it from the manuscript.

In addition, to further understand the bulk characteristics, we do analyze the individual emission sectors contributing to and driving factors of the enhancement ratios that represent the bulk characteristics of the whole city in Section 3.

4. Could you please give the definition of “Combustion Emission Pathway” somewhere?

Response: We have added “*We define combustion emission pathway as a trajectory in time of the changes in emissions from combustion in relation to socioeconomic development (e.g., Riahi et al., 2011; Steinberger et al., 2012; Li et al., 2016; Marangoni et al., 2017).”* at the beginning of Section 3.3 (Page 11 Line 25 – Line 27 of the track-changed manuscript). Please also see our response to General Comment 4.

In our case, it includes decadal trend and change of combustion emissions across the megacities in mainland China. Specifically, we found a robust coherent progression of declining-to-growing $\Delta CO/\Delta NO_2$ (-5.4±0.7% to +8.3±3.1%), and slowly-declining $\Delta SO_2/\Delta NO_2$ (-6.0±1.0% to -3.4±1.0%) from Shenyang, Beijing, Shanghai, to Shenzhen relative to 2005. Such progression is well-correlated with economic development, and traces a common emission pathway that resembles evolution of air pollution in more developed cities (Figure 4).