A letter of Response to Reviewers

We thank the three reviewers for their valuable comments. This manuscript has been improved significantly by addressing these comments. Below are the point-to-point responses to all of the comments. The revision in the manuscript related to the comments was marked in yellow color in the copy of the revised manuscript.

Response to the Reviewer 1:

This work describes the long-term emission inventories for seven pollutants throughout Guangdong Province, one of most developed regions in China, which is highly significant. However, the paper still suffers for two detail problems. 1) The single and plural forms for source categories in the article are not very uniform. For example, the emission source –"dust sources" in line 147 is a plural form, but "dust source" is presented in Figure 4. There is an example taken here, which have also been the same problems in the paper. 2) The abscissa of figure 4(d) $PM_{2.5}$ is diagonal. However, those for the other pollutants are vertical.

Response: Thanks for the positive feedback. For the two helpful comments, we accepted and revised on the manuscript.

1) Accepted. We unified single and plural forms for source categories. The major categories normalized as power plants, industrial combustion, residential combustion, on-road mobile source, non-road mobile source, dust source, industrial process source, industrial solvent use, non-industrial solvent use, storage and transportation, agricultural source, biomass burning, and other sources. In response to the comment, we revised the description in the manuscript. For example, source categories were revised in Lines 250-253 as "including power plants, industrial combustion, residential combustion, on-road mobile source, non-road mobile source, dust source, industrial process source, industrial solvent use, non-industrial solvent use, storage and transportation, agricultural source, biomass burning, and other sources, were estimated."
2) Accepted. The figure 4(d) PM2.5 was replaced with the vertical abscissa in the manuscript (Fig. 4).

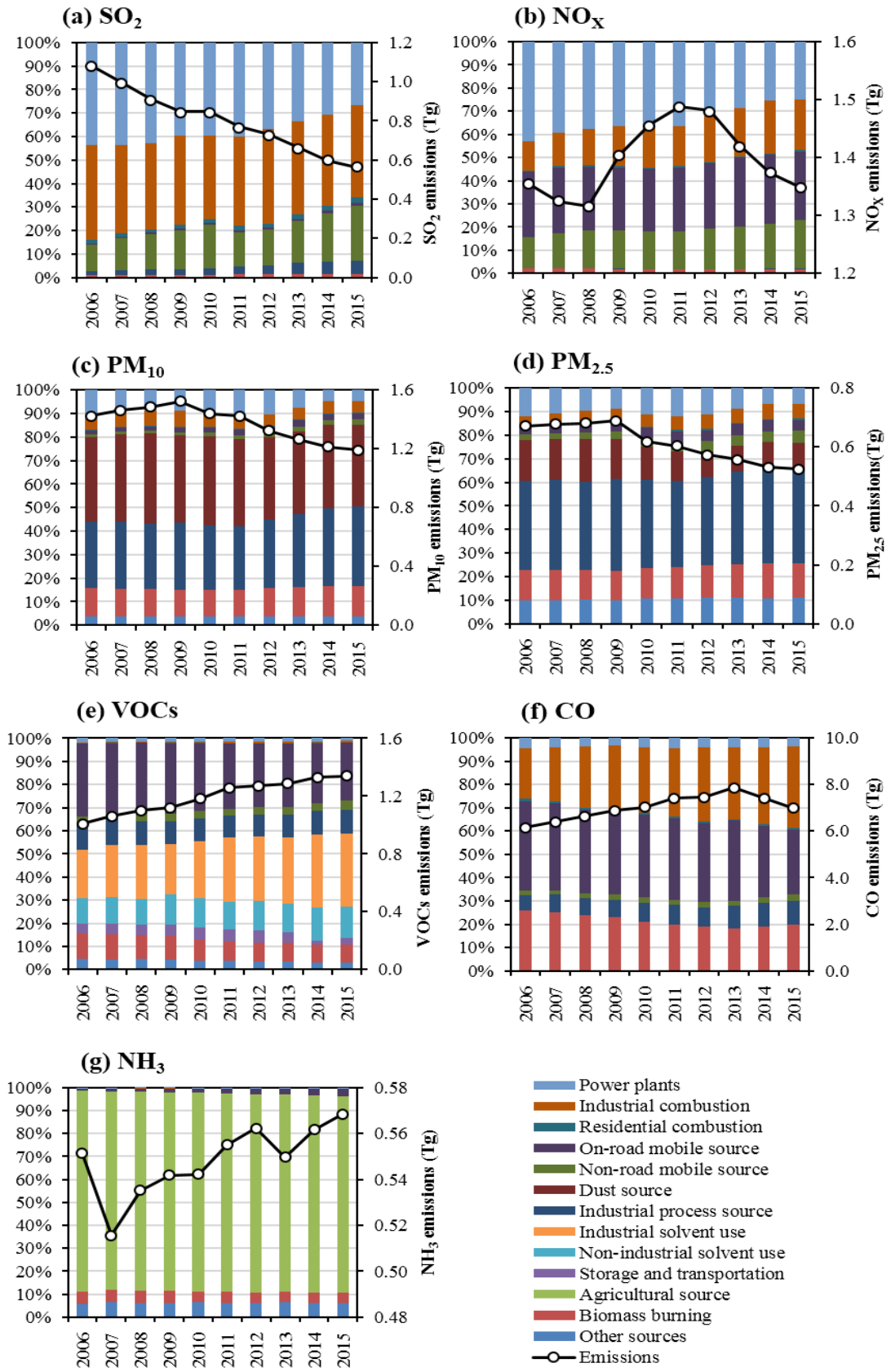

Figure 4. Source emission evolutions in Guangdong Province for **(a)** SO_2, **(b)** NO_X, **(c)** PM_{10}, **(d)** $PM_{2.5}$, **(e)** VOCs, **(f)** CO and **(g)** NH_3 from 2006 to 2015.

Response to the Reviewer 2:

General comments： This paper calculated a multi-year emission inventory (from 2006 to 2015 and a future year 2020) using the method of activity data and the emission factors. And then the author discusses the emission trends, source contributions and pick out some examples to explain the background reasons. This is a good try to show the audience a data set and comparison. However, this paper is more like a government report instead of a top scientific research paper. There are no sound scientific findings in the manuscript. The results of the manuscript are almost a repeat or quite similar to previous publications:

(1). Zhong Z. (2018) Recent developments of anthropogenic air pollutant emission inventories in Guangdong province, China, Science of Total Environment.

(2). Yin, X. H., Huang, Z. J., Zheng, Y., Yuan, Z. B., Zhu, W. B., Huang, X. B., and Chen, D.H.: Source contributions to PM2.5 in Guangdong province, China by numerical modeling: Results and implications, Atmospheric Research.

Zhong's paper is comparing with 2006, 2010 and 2012 GuangDong emission inventory while the current manuscript is analyzing the data from 2006 to 2015. The source categories of these two papers are more or less the same, although the author claimed in the manuscript that the source categories follows another publication: Pan et al. 2015 (line 144). The unabated emission factors of current manuscript are more or less the same with that in Zhong's paper. Moreover, there is inconsistency in factor values or total emission amount for a corresponding source/pollutants using the same model such as IVE model for mobile source. In terms of the emission validation using observation and satellite data, it is a good way to validate the bottom-up emission inventory utilizing this kind of data. However, the sparsity of the satellite needs to be considered. The carelessness comparison in this manuscript is very simple, not much extra information. Finally, although the manuscript is evaluating the control measures, The main results (figure 9) is more like a repeat of section 2.3 Evolution of source emissions in Guangdong Province for the 2 year 2006 to 2015. To sum up, I recommend a direct rejection because no meaningful results or new scientific findings can be found through the manuscript.

Response: We would like to thank the reviewer's valuable time and effort in reviewing the manuscript.

1) "*This paper is more like a government report instead of a top scientific research paper. There are no sound scientific findings in the manuscript*". Such an impression might be raised when we introduced the emissions and their trends in Section 2.3 and 2.4, which is somehow inevitable in a data-intense paper. However, it does not necessarily mean that it cannot be a top scientific research paper. The importance of long-term historical emission inventories in air pollution research have been widely recognized by the academic community (Li et al., 2017; Zhang et al., 2018; Hosely et al. 2018; Jin et al. 2017; Li et al. 2017; Lei et al. 2011; Ohara et al. 2017). Here, we advance the understanding of long-term air pollutant emissions in China. The study area, Guangdong province, is one of the three largest city clusters in China. It plays an important role in China's air pollution research frontiers and mitigation efforts. Surprisingly, no attempts have been made to estimate its historical emissions. Some studies have estimated the emissions of the Pearl River Delta (PRD) region, but the emissions of PRD cannot fairly represent Guangdong as a whole. To our best

knowledge, this work provides the first long-term record of anthropogenic air pollutant emissions of Guangdong. It provides robust data to assess the effectiveness of control measures and unveil the primary cause of air quality change and fills an important gap in China's air pollution research. In fact, our team had used the long-term emission data and numerical simulation to reveal the dominant cause of ozone changes in PRD (not published yet). Thus, it is potentially highly cited by future work in South China. Moreover, this work observes the phenomenon of 'emission leakages' from developed cities to less developed ones. Emissions in Non-PRD (the other less-developed cities in Guangdong) are becoming increasingly important with some emerging sources such as industrial processes. A new clue or an alarm is putting forward, in sharp contrast to the current mitigation efforts focusing on developed regions. Even so, we still carefully revised the manuscript and enhanced the discussions and analysis regarding the evolution of source emission in GD (section 2.3).

See Lines 551-593 for instance: “In PRD, SO2 emissions steadily declined, from 0.788 Tg in 2006 to 0.292 Tg in 2015. The decline was dominated by power plants and industrial combustion, accounting for 50.0% and 46.8% of the total decrease, respectively (Fig. 5a). The decline of SO2 emissions from power plants and industrial combustion is highly associated with the control measures enacted during the 11th FYP and 12th FYP. In response to the SO2 emission cap in 11th FYP, Guangdong province required the elimination of small thermal power units with high energy consumption and outdated combustion technology in 2007. By the end of 2011, 12.2 million kilowatts of small thermal power units were eliminated during the 11th FYP. In addition, action plan for air pollution prevention and control in Guangdong province (2014-2017) (GDEP, 2014) released in the 12th FYP further strengthen the emission control of power plants. As a result, the flue gas desulfurization (FGD) penetration in coal-fired power plants rose to 87% in 2013. For the industrial combustion, its fuel consumption in PRD decreased by 47.5% during 2006-2015, which can be explained by energy structural adjustment regulations, including the “total amount control of coals” and “changing fuel from coal to natural gas” control measures (Fig. S7). The detailed regulations on energy structural adjustment are summarized in Table S1.

In NPRD, on the contrary, SO2 emission increased until 2010, then saw a downturn. Before 2010, the SO2 emission growth was mainly associated with the increase of industrial combustion and non-road mobile source (Fig. 5b). These two sources still maintained a slight rise after 2010, but their increased emissions were offset by a plunge of emissions from power plants. To reveal the cause of emission growth from industrial combustion in NPRD, we compared the standard fuel-consumption trends from industrial combustion in PRD and NPRD (Fig. 6). In PRD, the fuel consumption from industrial combustion in PRD steadily dropped by 47% during 2006-2015, with an average annual descending rate of 6.9%. In NPRD, by contrast, the fuel consumption increased by 99%.In fact, the average annual growth rates of fuel consumption in NPRD was only 3.2% before 2010, but after that, the increase accelerated, with an average annual growth rate of 11.9% during 2011-2015. Particularly, the growth rate peaked in 2011 (17.1%) and 2012 (14.5%). Meanwhile, the descending rates of fuel consumption in PRD also reached its peak in 2011 (9.9%) and 2012 (11.4%), when the shift of industries implemented. This fact indicates the existence of emission leak from PRD to NPRD due to the policy of “vacate the cage and change birds” that brought many energy-intensive industries from PRD to NPRD. Statistical data showed that the NPRD region had undertaken 33 industrial parks that migrated from PRD, as shown in Table S10 (GDEI, 2014). Consequently, the industrial shift might, in turn, promote the emission of power plants in NPRD to

some extent."

2) "*The results of the manuscript are almost a repeat or quite similar to previous publications (Zhong et al., 2018; Yin et al., 2017)".* We appreciate reviewer's efforts in literature review, but we have to point out that our work is quite different from them regarding methodologies, study areas and findings. Zhong et al. (2018) summarized recent updates of regional emission inventories in Guangdong in terms of the emission source supplement, spatiotemporal distribution refinement and estimation method. Yin et al (2017) used an atmospheric model to quantify the source contributions to ambient $PM_{2.5}$ other than $PM_{2.5}$ emissions while the 2012 Guangdong emission inventory was used as model input. In comparison, our study applied a dynamic technology-based methodology to develop a long-term (2006-2015) emission inventory in Guangdong and analyzed its emission trends, spatial variations, source-contribution variations, and emission reduction potentials. The study area is another difference. While Zhong et al. (2018) compared the PRD emission inventories using different estimation methods as an example (not the Guangdong emission inventories as claimed by the reviewer: *Zhong's paper is comparing with 2006, 2010 and 2012 Guangdong emission inventory*), this study estimated long-term emissions for Guangdong using self-consistent methodologies. Also, this study compared the emission evolution between the PRD and Non-PRD regions, and found that the emission evolution in the Non-PRD is quite different from their counterparts in PRD, which are not revealed by two previous studies. Therefore, it is arbitrary and hasty to conclude that our work is a 'repeat' of Zhong et al (2018) and Yin et al (2017).

3) *"The source categories of these two papers are more or less the same although the author claimed in the manuscript that the source categories follows another publication: Pan et al. 2015 (line 144). The unabated emission factors of current manuscript are more or less the same with that in Zhong's paper. Moreover, there is inconsistency in factor values or total emission amount for a corresponding source/pollutants using the same model such as IVE model for mobile source".* In fact, Zhong et al. (2018) updated the source categories based on Pan et al. (2015). Therefore, it is not surprising that the source categories of our study and Zhong et al. are similar. However, I am afraid that the classification similarity does not mean these papers are repetitive. Instead, a similar source category is beneficial to the evaluation of our study with previous studies. In response to the comment, we made a clarification in the method section 1.1 (Lines 253-257). "On the basis of Zhong et al. (2018), and 70 sub-categories based on Pan et al. (2015) and the guidelines for the development of an air-pollutant emission inventory for Chinese cities (MEPC, 2017), the 13 major categories were further broken down into 70 sub-categories to improve emission estimation." Regarding the vehicle emission factors, the inconsistency arises because the base years of unabated emission factors in this study is 2007, rather than 2006 in Zhong et al. (2018).

4) *"In terms of the emission validation using observation and satellite data, it is a good way to validate the bottom-up emission inventory utilizing this kind of data. However, the sparsity of the satellite needs to be considered. The carelessness comparison in this manuscript is very simple, not much extra information."* In this study, the evaluation of a long-term emission inventory focus on its emission trend, rather than the amount or high-resolution spatial pattern. Therefore, the sparsity of the ground-level observations is not a concern in the evaluation. For satellite data, it is gridded and has the same domain coverage with emissions. Therefore, there is no issue regarding the sparsity

in satellite data. In fact, similar evaluation was also adopted by Zhang et al. (2018), which compared the trends in $PM_{2.5}$ precursor emissions in China with satellite and ground-based $PM_{2.5}$ concentrations, indicating the evaluation is acceptable.

In response to the questioning, we explained the revised the method section 1.2 (Lines 436-444) "We focused on the evaluation of emission trends. Therefore, we mainly compared annual observations averaged over the PRD/GD region with annual emissions in PRD/GD. Also, emissions and satellite observations in PRD and NPRD region were compared. Since emissions mainly concentrated in urban areas, where ground-level observations located, the sparsity of ground-level observations was ignored in the evaluation."

5) "*the main results (figure 9) is more like a repeat of section 2.3 Evolution of source emissions in Guangdong Province*". We're afraid the conclusion is improper. Section 2.3 compares how the emissions in PRD and non-PRD are evolving in different ways. Figure 9 studies how might the emission changes in the absence of control measures and reduction potentials if actions have been taken, under the future projection of activity level. Combining section 2.3 and Figure 9 could provide a better understanding of emission trends. For example, we observed a slight change of mobile VOC emissions in Section 2.3. In fact, control measures were effective in reducing mobile VOC emissions in the past decade. Nevertheless, the emission reduction was compensated by the growth of the vehicle population as revealed by Figure 9. Emission potentials are also quantified in Figure 9, which is not included in Section 2.3. For clarification, we revised the description of method in section 2.4 (Lines 805-809) "To evaluate the efficiencies of the control measures enacted in GD and to provide implications for future policies, we decomposed the emission changes into two categories: (1) changes resulting from change of activity level (activity-driven emission) in the absence of control measures and (2) changes due to the implementation of pollution controls (control-driven emission reduction) (Zheng et al., 2018)."

References: Zheng, B., Tong, D., Li, M., Liu, F., Hong, C., Geng, G. N., Li, H. Y., L., X., Peng, L. Q., Qi, J., Yan, L., Zhang, Y. X., Zhao, H. Y., Zheng, Y. X., and He, K. B.: Trends in China's anthropogenic emissions since 2010 as the consequence of clean air actions, Atmos. Chem. Phys., 18, 14095-14111, doi: 10.5194/acp-18-14095-2018, 2018

Detailed comments:

1. **Abstract section:** Line 39-43: "The declines of SO2, NOx, PM2.5, and PM10 emissions are mainly attributed to the control-driven emission reductions in the Pearl River Delta (PRD) region, especially from power plants, industrial combustion, on-road mobile sources, and fugitive dust, and partly to the shift of industries from the PRD to the non-PRD (NPRD) region in GD." It does not show any new findings here, especially the idea of "shift of industries from the PRD to the non-PRD (NPRD) region in GD" comes from a published paper on Atmospheric Research (AR) titled: "Source contributions to $PM_{2.5}$ in Guangdong province, China by numerical modeling: Results and implications" by X. Yin (2017).
   Response: We are afraid that this is a misunderstanding. In Yin et al. (2017), the atmospheric modeling results showed that emissions in NPRD could contribute to the $PM_{2.5}$ pollution in

PRD. Therefore, they suggested that relocating industries from PRD to NPRD region cannot help ease the pollution in PRD. In short, Yin et al. (2017) discussed one possibility of emission control and proved that it might not work. In our study, emission leakages from PRD to NPRD are observed through a multi-year record of emissions, which is not revealed by Yin et al. (2017).

Line46-48: "this might be one of the reasons that led to the slight upward trends of ozone concentrations in GD." is this finding a reliable result or just a guess? It is not appropriate to put an ambiguous answer in the central part of the abstract.

Response: Thanks for the comment. Ou et al. (2016) revealed that O_3 formation in most parts of PRD was VOC-limited in autumn and winter. This indicates that the growing VOC emissions and the decreasing NO_X emissions, which was observed in this study, might contribute to the growth of ozone concentrations in PRD. However, this does not mean that the emission change is the dominant cause of ozone growth. Further studies using numerical simulations are required to verify the guess. In fact, our preliminary simulations confirm that the growing VOC emissions and the decreasing NO_X emissions account for about 90% of the ozone growth in PRD in the past decade. Since this result has not yet been published, we removed this sentence from abstract. Also, we revised the abstract in the revised manuscript.

Lines 40-87: "Guangdong province (GD), one of the most prosperous and populous regions in China, still experiences haze events and growing ozone pollution despite of substantial air quality improvement in recent years. Integrated control of fine particulate matter ($PM_{2.5}$) and ozone in GD calls for a systematic review of historical emission. In this study, emission trends, spatial variations, source-contribution variations, and reduction potentials of sulfur dioxide (SO_2), nitrogen oxides (NO_X), $PM_{2.5}$, inhalable particles (PM_{10}), carbon monoxide (CO), ammonia (NH_3), and volatile organic compounds (VOCs) in GD from 2006 to 2015 were examined using a dynamic methodology, taking into account economic development, technology penetration, and emission controls. The relative change rates of anthropogenic emissions in GD during 2006-2015 are -48% for SO_2, -0.5% for NO_X, -16% for $PM_{2.5}$, -22% for PM_{10}, 13% for CO, 3% for NH_3, and 13% for VOCs. The declines of SO_2, NO_X, $PM_{2.5}$, and PM_{10} emissions in the whole province are mainly resulted from the stringent emission control in the Pearl River Delta (PRD) region, where most previous control measures focused on, especially from power plants (SO_2 and NO_X), industrial combustion (SO_2, $PM_{2.5}$ and PM_{10}), on-road mobile source (NO_X), and dust source ($PM_{2.5}$ and PM_{10}). Emissions from other areas (the non-PRD, NPRD), nevertheless, remain relatively stable due to the lax control measures and rapidly growing energy consumption. In addition, emission leaks of SO_2 and NOx from industries are observed from the PRD to NPRD in 2010 and 2011. As a result, emissions in NPRD are increasingly important in GD, particularly those from industrial combustion. Contribution of NPRD to the total SO_2 emissions in GD, for example, increased from 27% in 2006 to 48% in 2015. On-road mobile source and solvent use are the two key sources that should receive more effective control measures in GD. Current emission reductions from on-road mobile source are neutralized by the substantial growth of vehicle population, while VOC emissions in GD steadily increase due to the growth of solvent use and the absence of effective control measures Besides, future work could focus on power plants and industrial combustion in GD and industrial process source in NPRD, which still have large emission reduction potentials. The historical emission inventory developed in this study not only helps to

understand the emission evolution in GD, but also provides robust data to quantify the impact of emission and meteorology variations on air quality and unveil the primary cause of significant air quality change in GD in the recent decade."

Reference: Ou, J. Y., Yuan, Z. B., Zheng, J. Y., Huang, Z. J., Shao, M., Li, Z. K., Huang, X. B., Guo, H., and Louie, P. K. K.: Ambient Ozone Control in a Photochemically Active Region: Short-Term Despiking or Long-Term Attainment? Environmental Science & Technology, 50(11):5720-5728, doi: 10.1021/acs.est.6b00345, 2016.

2. **Introduction section**:

Overall review: For a publication in a top journal such as ACP, the research motivation of the current manuscript is not strong enough. The literature review is too weak. Too many self-cited references were present.

Response: Thanks for the comment. The study area, Guangdong province, is one of the three largest city clusters in China. It plays an important role in China's air pollution research frontiers and mitigation efforts. Subsequently, Guangdong has experienced significant air quality improvement, particularly the Pearl River Delta region, which is the first region to meet China's national 35 $\mu g/m^3$ $PM_{2.5}$ standard for four consecutive years. Understanding the emission evolution and assessing the effectiveness of control measures call for a systematic review of historical emissions in Guangdong. Surprisingly, no attempts have been made to estimate Guangdong's historical emissions. In this study, we developed a long-term historical emission inventory in Guangdong. To our best knowledge, this work provides the first long-term record of anthropogenic air pollutant emissions of Guangdong.

In response to the comment, we rewrote the Introduction to highlight the research motivation of this study and removed some self-cited references. The new introduction is shown below.

Lines 91-240: "Guangdong Province (GD), comprising nine major cities in the Pearl River Delta (PRD) region and eleven less developed cities in the Non-PRD (NPRD), is the most prosperous and populous regions adjacent to the coast of southern China (Fig. S1 in the Supplementary Material). With only 1.9% of China's land coverage, GD contributed 10.0% of China's gross domestic product (GDP) and 8.0% of the population in 2017 (GDPBS, 2016-2017). Particularly, the PRD region, dubbed 'the world factory' and embracing a wide variety of industries, represented about 80.0% and 53.4% of GD's total GDP and population, respectively (GDPBS, 2016-2017). Benefited from China's opening-up policies, GD has experienced rapid economic growth accompany with serious and complex air pollution issues.

To improve air quality, great efforts to have been made to formulate various control measures and policies, especially after 2013, when the Action Plan on the Prevention and Control of Air Pollution (PCAP, 2013-2017) was launched. The PCAP required the PRD region to reduce $PM_{2.5}$ concentration by 15% by the year 2017 compared with the 2013 levels. These policies and control measures, which were summarized in Table S3 of SI, partly alleviated regional air pollution in GD. Ground-level observations showed that GD saw an obvious air quality improvement, with SO_2 and PM_{10} concentrations decreasing by 63.3% and 17.7%, respectively, from 2006 to 2017 (EPGD, 2006-2017). In particular, the PRD region is the first region to meet China's national standard of $PM_{2.5}$ standard (35 $\mu g/m^3$) for three consecutive years (34 $\mu g/m^3$ in 2015, 32 $\mu g/m^3$ in 2016 and 34 $\mu g/m^3$ in 2017).

However, air pollution in GD is still a major concern. The annual $PM_{2.5}$ levels still far exceed WHO's $PM_{2.5}$ guideline value of 10 μg/m^3 (World Health Organization, 2006) and haze events frequently occur in winter (Tao et al., 2017). Also, the ambient ozone concentrations have been growing in recent years. The 90th percentile of the maximum 8-hour average ozone concentration (90%-8h-ozone) in the PRD region increased by 24% since 2015, reaching 165 μg/m^3 in 2017 (GDEMC, EPDHK, EPBMC, 2018).

On contrast to the need for further air quality improvement, the unclear causes of recent air quality change in GD hinders the development of evidence-based air quality control strategies. Although several studies had assessed the impact of emission and meteorology change on air quality, their results are inconsistent with each other. For instance, Lin et al. (2018) suggested that emission controls helped improve local air quality in the PRD region, according to a high consistency of ambient $PM_{2.5}$ concentrations and emissions. However, Mao et al. (2018) argued that meteorological and climate conditions rather than PM emissions dominate the interannual variabilities and trends of winter haze days in PRD based on an observation-based approach. Yang et al. (2019) concluded that the inappropriate NOx/VOC control ratio within the PRD over the past years is likely responsible for the ozone increase, while Li et al. (2018) argued that the decrease of $PM_{2.5}$ concentration is the main driver.

Further mitigation of air pollution in GD calls for a systematic review of historical emissions. First, the long-term historical emission data could help policymakers understand the evolution of emissions, quantify the cuts in emissions that have been achieved by control measures, and identify those sources with the greatest potential for large future emission reductions (Gurjar et al., 2004; Ohara et al., 2007; Zhong et al., 2013). This is particularly important for GD whose potential for further emission reduction are shrinking as control measures tighten. Second, with the use of atmospheric chemical transport models (CTMs), the historical emission can be used to examine the linkages between air quality improvements and control measures, and thus identify the main causes of air quality change and key control measures. All this information is crucial to guide future air-quality management and formulate robust evidence-based policies.

GD is one of the first areas to compile its own emission inventories in China (Zhong et al., 2013). However, no attempts have been made to estimate the historical emissions of GD. Most of the previous emission studies mainly focused on the PRD region, but ignored the NPRD region that also has increased emissions. For instance, Zheng et al. (2009) developed the first high-resolution emission inventory for the PRD region in 2006, followed by emission inventories for speciated volatile organic compounds (VOCs) (Zheng et al., 2009), biogenic VOCs (Zheng et al., 2010), ammonia (NH_3) (Yin et al., 2015), biomass burning (He et al., 2011, Zhang et al., 2013), sea salt (Liu et al., 2015) and multiple-year anthropogenic sources (Lu et al., 2013). Due to the strengthened emission controls in PRD, relocation of industries are in courses for those sectors that are energy intensive, or highly polluting, or have excess production capacity from the PRD region to non-PRD (NPRD) areas (Chun, 2012; Yin et al., 2017) (Fig. S2). As a result, emissions in the PRD and NPRD regions have experienced substantial changes in recent years. This means that the emissions of PRD cannot fairly represent Guangdong as a whole. Although several emission inventories in GD were developed recently (Huang et al., 2015; Pan et al., 2015; Liu et al., 2017; Zhong et al., 2018), they were carried out in a single year and limited in spatial coverage, and source categories. Therefore,

there is a need to develop a long-term historical emission inventory in GD using a consistent methodology and the same underlying driver data to fill the data gaps and to assist with future air pollution control measures.

Line 84-91: I do not see the long-term emission inventory comparison could resolve the controversial issue of whether the emission or the meteorology plays the main role of pollution. Please explain how the long-term emission inventory could help differentiate the causes of the emission or the meteorology?

Response: Thanks for the comment. A long-term emission inventory alone could not identify the cause of air quality change. However, combining with atmospheric chemical transport models (CTMs), the impact of emission change and meteorological variation on air quality can be quantified. For instance, by combining a long-term emission and the WRF-CMAQ (Weather Research and Forecasting Model and Community Multiscale Air Quality) model, Cheng et al (2018) found that the rapid decrease in $PM_{2.5}$ concentrations in Beijing during 2013-2017 was dominated by local and regional emission reductions, rather than meteorology variation. For clarification, we revised manuscript in the Lines 154-157"Second, with the use of atmospheric chemical transport models (CTMs), the historical emission can be used to examine the linkages between air quality improvements and control measures, and thus identify the main causes of air quality change and key control measures."

Reference: Cheng, J., Su, J. P., Cui, T., Li, X., Dong, X., Sun, F, Yang, Y. Y., Tong, D., Zheng, Y. X., Li, Y. S., Li J. X., Zhang. Q, and He, K, Dominant role of emission reduction in $PM_{2.5}$; air quality improvement in Beijing during 2013-2017: a model-based decomposition analysis, Atmospheric Chemistry and Physics Discussions, 19, 6125-6146. https://doi.org/10.5194/acp-2018-1145, 2019

Line 96-112 introduces the history of emission inventory development in PRD and GD. Too many self-cited papers are included. Due to differences in source categories or data sources for different geometry, using a unified method to calculate a multi-year emission inventory is fine, but what is new here? Is the technique original or are the data sources new? What are the advantages/Strength of the method/data in this manuscript comparing with the previous publications? Only calculation of a multi-year EI seems too weak to be published in a top journal ACP.

Response: Thanks for the comment. Following previous long-term emission papers (Lu et al. 2012, Zheng et al. 2018), we applied a dynamic technology-based methodology that considers economic development, technological penetration, and emission controls to estimate the long-term emission inventory in Guangdong in this study. It is true that the methods and data sources are not new. However, these methods satisfy the requirement of developing a reliable long-term emission inventory. Although new data sources, such as AIS data and satellite data, can promote emission inventories, obtaining new data spanning a long-term period is challenging. For this study, like most long-term emission inventory papers, the scientific value lies in revealing the long-term emission evolution pattern, assessing the effectiveness of past control measures and

providing guidelines for future control measure development. Also, this study provides the first long-term record of anthropogenic air pollutant emissions of Guangdong, the three largest city clusters in China, and advances our understating of air pollutant emissions and control measures in Guangdong. For instance, this study observes the phenomenon of 'emission leakages' from the PRD region to the Non-PRD region, which received less attention of air quality control compared with the PRD, indicating that emissions in Non-PRD (the other less-developed cities in Guangdong) are becoming increasingly important in Guangdong. In addition, this study found that past control measures for vehicle source and solvent use source are not stringent enough because emission reduction driven by these measures cannot cover the emission growth due to the increasing vehicle population and solvent use. Finally, combining with atmospheric chemical transport models (CTMs), the long-term emission data developed in this study could help identify the dominant cause of air quality change in Guangdong. In fact, our team had used the long-term emission data and numerical simulation to reveal the dominant cause of ozone changes in PRD (not published yet).

In response to the comment, we clarified why we used the top-down method in the revised manuscript (Lines 263-273). Also, we highlighted the scientific significant in the revised abstract and introduction. "this study provides the first long-term record of anthropogenic air pollutant emissions of Guangdong, the three largest city clusters in China, and advances our understating of air pollutant emissions and control measures in Guangdong.", "The historical emission inventory developed in this study not only helps to understand the emission evolution in GD, but also could also help to reveal the dominant causes of air-quality change in PRD." and so on.

Lines 263-273: "Unlike the single-year emission inventory, the estimation of a long-term emission inventory is generally more complicated since it requires the data sources, estimation method and source category for all years are consistent. Although top-down methods or on-line estimation methods based on big data can promote the estimation, obtaining the long-term activity data, such as Automatic Identification System (AIS) data for ship emission and Global Positioning System (GPS) data for on-road mobile source, is challenging. Therefore, most studies (Streets et al., 2006; Zhang et al., 2007; Lu et al., 2012; Zheng et al., 2018), still generally applied the top-down method to develop long-term emission inventories. Following previous studies, we applied a dynamic technology-based methodology that considers economic development, technological penetration, and emission controls to estimate the anthropogenic emission trends in GD."

References:

1. Streets, D. G., Zhang, Q., Wang, L. T., He, K. B., Hao, J. M., Wu, Y., Tang, Y. H., and Carmichael, G. R.: Revisiting China's CO emissions after the Transport and Chemical Evolution over the Pacific (TRACE-P) mission: Synthesis of inventories, atmospheric modeling, and observations, Journal of Geophysical Research Atmospheres. 111, doi: 10.1029/2006JD007118, 2006.
2. Zhang, Q., Streets, D. G., He, K. B., Wang, Y. X., Richter, A., Burrows, J. P., Uno, I., Jang, C. J., Chen, D., and Yao, Z. L., NOX emission trends for China, 1995–2004: The view from the ground and the view from space, Journal of Geophysical Research: Atmospheres, 112, doi: 10.1029/2007JD008684, 2007.

3. Lu, Q., Zheng, J. Y., Ye, S. Q., Shen, X. L., Yuan, Z. B., and Yin, S. S.: Emission trends and source characteristics of SO_2, NO_X, PM_{10} and VOCs in the Pearl River Delta region from 2000 to 2009, Atmospheric Environment, 76, 11-20, doi: 10.1016/j.atmosenv.2012.10.062, 2013.
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3. Methodology section.

General review: No new method or new data sources were found in this manuscript comparing with the previous Emission Inventory publications in the PRD region. The calculation of marine emission is outdated, and its uncertainty is considerable. The validation method of the emission totals is too simple to be believed.

Response: Thanks for the comment. Unlike the single-year emission inventory, the estimation of a long-term emission inventory is generally more complicated since it requires the data sources, estimation method and source category for all years are consistent. Although new estimation methods and new data sources can promote emission inventories, obtaining new data span a long-term period is challenging. We understand that the bottom-up estimation method based on AIS data can generate reliable ship emission inventories. However, obtaining 10-year AIS data is a considerable work. Moreover, the spatial coverage of AIS data in China before 2012 is limited. Instead, long-term ship fuel consumption data or cargo volumes can be easily obtained. Although the method based on fuel consumption might underestimates marine emissions, due to the absence of passing ships that traveled through the study domain but did not call at any port in mainland China. However, the underestimation is slight since the passing ships only accounted for about 7% of the total fuel consumption in China (Li et al.2018). Therefore, the top-down method based on fuel consumption or cargo volumes is more feasible than the bottom-up method based on AIS data in the long-term emission estimation. Another example is the satellite data for biomass burning. For the satellite-derived method, the burned area or fire radiative power (FRP) are key parameters used to estimate the amount of fuel combustion. These parameters can be rapidly updated and better reflect temporal and spatial variations. However, satellite data generally have larger uncertainty since they overlook small fires due to the low spatial resolution of the burned area product or the limitation of overpass time of the orbiting satellite. Moreover, long-term emission inventories generally focus on emission trend, sectoral evolution and emission projection, rather than the high-resolution spatial distribution. Therefore, most studies (Streets et al. 2006, Zhang et al, 2007, Lu et al. 2012, Zheng, et al. 2018) still generally applied the top-down method to develop long-term emission inventories. In this study, we applied a dynamic technology-based methodology that considers economic development, technological penetration, and emission controls to estimate the long-term emission inventory in Guangdong. Also, we used interpolation method and fuel consumption estimation method to estimate some missing data, make sure all activity data during 2006-2015 are comparable. For clarification, we added the explanation why the method was used in this study, as shown below.

Lines 263-273: “Unlike the single-year emission inventory, the estimation of a long-term emission inventory is generally more complicated since it requires the data sources, estimation method and source category for all years are consistent. Although top-down methods or on-line estimation methods based on big data can promote the estimation, obtaining the long-term activity data, such as Automatic Identification System (AIS) data for ship emission and Global Positioning System (GPS) data for on-road mobile source, is challenging. Therefore, most studies (Streets et al., 2006; Zhang et al., 2007; Lu et al., 2012; Zheng et al., 2018), still generally applied the top-down method to develop long-term emission inventories. Following previous studies, we applied a dynamic technology-based methodology that considers economic development, technological penetration, and emission controls to estimate the anthropogenic emission trends in GD.”

Lines 1059-1078: “This study applied a top-down method to develop long-term emission inventories. This method is feasible for two reasons. On one hand, long-term emission inventories generally focus on emission trend, sectoral evolution and emission projection, rather than the high-resolution spatial distribution. On the other hand, obtaining long-term activity data for top-down method is more readily accessible. Therefore, most previous studies (Streets et al. 2006, Zhang et al, 2007, Lu et al. 2012, Zheng et al., 2018) also applied the same method to develop long-term emission inventories.”

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6.

Table S5: is the table for PM10? Please be specific.

Response: Yes. We revised the typo error in supplementary information "Table S5. PM_{10} emission factors for power plants, industrial combustion and dust source"

Line 212 – 215: The dealing method of marine may under-estimate the marine emissions, which are quite important for Guang Dong coastal cities.

Response: Thanks for the comment. It is true that the top-down method based on fuel consumption from statistic yearbook might under-estimate the marine emissions due to the absence of passing ship that traveled through the study domain but did not call at any port in mainland China (Li et al., 2018). These passing ships accounted for about 7% of the total fuel consumption in China. To fix the underestimation, the AIS data can be used to calibrate the total fuel consumption in GD. In response to the comment, we discussed the underestimation of marine emissions in the conclusion section. (Lines 1066-1071) "However, this study can be further improved in following aspects. First, a method based fuel consumption might underestimates marine emissions, due to the absence of passing ships that traveled through the study domain but did not call at any port in mainland China (Li et al., 2018). In China, the passing ships accounted for about 7% of the total fuel consumption. To fix the underestimation, the AIS data can be used to calibrate the total fuel consumption in GD."

References: Li, C., Jens, B., Zheng, J. Y., Yuan, Z. B., Ou, J. M., Li, Y., Wang, Y. L., and Xu, Y. Q.: Decadal evolution of ship emissions in China from 2004 to 2013 by using an integrated AIS-based approach and projection to 2040, Atmo. Chem. Phys., 18, 6075-6093, doi: 10.5194/acp-18-6075-2018, 2018.

Line 222-223: Please specify how typical are the annual average vehicle kilometers? How many cities and what types of roads in GuangDong were involved in the field survey?

Response: Thanks for the comment. We obtained eight cities and 111 roads by types in Guangdong province in the field survey. The road type includes arterial road, secondary arterial road, and branch, covering most road types in Guangdong. For clarification, we revised the manuscript in lines 348-353 "For on-road mobile sources, population data of different vehicle types (i.e., passenger trucks, buses, taxis, and motorcycles) and the gross weight (heavy and light duty) from the 2006-2015 statistical yearbooks, and annual average vehicle kilometers traveled from a field survey of 8 cities and 111 roads by types (i.e., arterial road, secondary arterial road and branch) in GD were used to characterize the annual change of activity level."

Line: 225-227: The vehicle ratios of fuel use used in this manuscript is too outdated to be used. Moreover, the vehicle ratios should be differentiated for PRD and NPRD. The pattern of the vehicle population in GD changed quite a lot from 2009 to 2015.

Response: Thanks for the comment. In fact, we considered the annual change of vehicle ratios of fuel use based on the field survey of vehicle ratios that covered eight cities in Guangdong for the years of 2010, 2012, 2014, and 2015 and published data from Che et al. (2009) for the year of 2006. For other years, we estimated the vehicle ratios using an interpolation method. We agree that the vehicle ratios between PRD and NPRD are different. However, due to the limited survey data, we used the same vehicle ratio for PRD and NPRD regions, which could bring

uncertainty in emission estimation. In response to the comment, we clarified the data and discussed the limitation in the revised manuscript.

Lines 353-359: "We further divided the vehicle type into diesel and gasoline vehicles to obtain a more accurate estimate, based on the vehicle ratios (diesel/gasoline) from our previous study (Che et al., 2009) in 2006 and the field survey covered 8 cities in the years of 2010, 2012, 2014, and 2015. The vehicle ratios in other years (2007-2009, 2011, and 2013) were estimated using an interpolation method."

Lines 1059-1078: Second, the annual average vehicle kilometers traveled and the ratio of diesel vehicles to gasoline vehicles were obtained from a field survey, which only cover 8 cities and 111 roads in GD. These survey data cannot well represent each city in GD and thus might bring uncertainties in characterizing emission evolution of on-road mobile source. To better reflect the temporal pattern and differentiate emissions between different cities in GD, more field survey of vehicle types, fuel consumption, and emission standard should be conducted to better differentiate vehicle emissions between different cities in GD.

Reference: Che, W. W., Zheng, J. Y., and Zhong, L. J.: Vehicle Exhaust Emission Characteristics and Contributions in the Pearl River Delta Region, Research of Environmental Sciences, 22, 456-461, 2009.

4. **Results section**

General review: It is just the description of the changes and the sectoral contribution of the multi-year emission inventory. Relevant policy measures were used to explain the sharp drops or increases. It is more like a government report instead of a research paper published in a top journal.

Response: Thanks for the comment. Such an impression might be raised when we introduced the emissions and their trends in Section 2.3 and 2.4, which is somehow inevitable in a data-intense paper. However, it does not necessarily mean that it cannot be a top scientific research paper. The importance of long-term historical emission inventories in air pollution research has been widely recognized by the academic community (Li et al., 2017; Zhang et al., 2018; Hosely et al. 2018; Jin et al. 2017; Li et al. 2017; Lei et al. 2011; Ohara et al. 2017). Here, we advance the understanding of long-term air pollutant emissions in China. The study area, Guangdong province, is one of the three largest city clusters in China. It plays an important role in China's air pollution research frontiers and mitigation efforts. Surprisingly, no attempts have been made to estimate its historical emissions. Some studies have estimated the emissions of Pearl River Delta (PRD), but the emissions of PRD cannot fairly represent Guangdong as a whole. To our best knowledge, this work provides the first long-term record of anthropogenic air pollutant emissions of Guangdong. It provides robust data to assess the effectiveness of control measures and unveil the primary cause of air quality change and fills an important gap in China's air pollution research. Thus, it is potentially highly cited by future work in South China. Moreover, this work observes the phenomenon of 'emission leakages' from developed cities to less developed ones. Emissions in Non-PRD (the other less-developed cities in Guangdong) are becoming increasingly important with some emerging sources such as industrial processes. A

new clue or an alarm is putting forward, in sharp contrast to the current mitigation efforts focusing on developed regions. In response to the comment, we revised the introduction and summary section to highlight scientific significance (see previous response). Also, we added more discussions and analysis in Section 2.3 to highlight the "emission leakages" from PRD to NPRD and the cause of emission evolution, as marked in yellow color in the revised manuscript.

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Line 343: I did not see "emission trends in PRD and NPRD are shown in Fig. 2", Fig 2 only show the trend of PRD instead of NPRD. Please specify.

Response: Thanks for pointing out this. The wrong expression for NPRD was removed in the manuscript. We revised the expression in the line 510 "emission trends in PRD are shown in Fig. 5".

Line 346-348: PM2.5 was not shown in figure 2. Please specify.

Response: Thanks for pointing out this. We made a typo error and changed "PM2.5" to "PM10" in the revised manuscript. We revised the expression in the lines 512-514"For PM_{10} emissions, the declining trend also closely followed the fluctuant downward trend of ambient PM_{10} concentrations and AOD".

Line 349-365: Spatial maps of differences between different years (Figure 3) for the validation are better to see the changes. In term of calculating the emission changes, did you count all the grids or just the typical points in PRD and NPRD region? Please specify.

Response: Accepted, we replotted the spatial maps accordingly. Regarding the calculation of

emission changes, we count all the grids in the PRD and NPRD region. We clarified the calculation method in the manuscript (lines 411-413) "In this study, satellite observations, including SO_2, NO_2 and AOD column, and ground-level observations (SO_2, NO_2 and $PM2.5$ concentrations) in PRD were used to evaluate the long-term total emission in GD area." Also we provided the maps in Figure 3 as below.

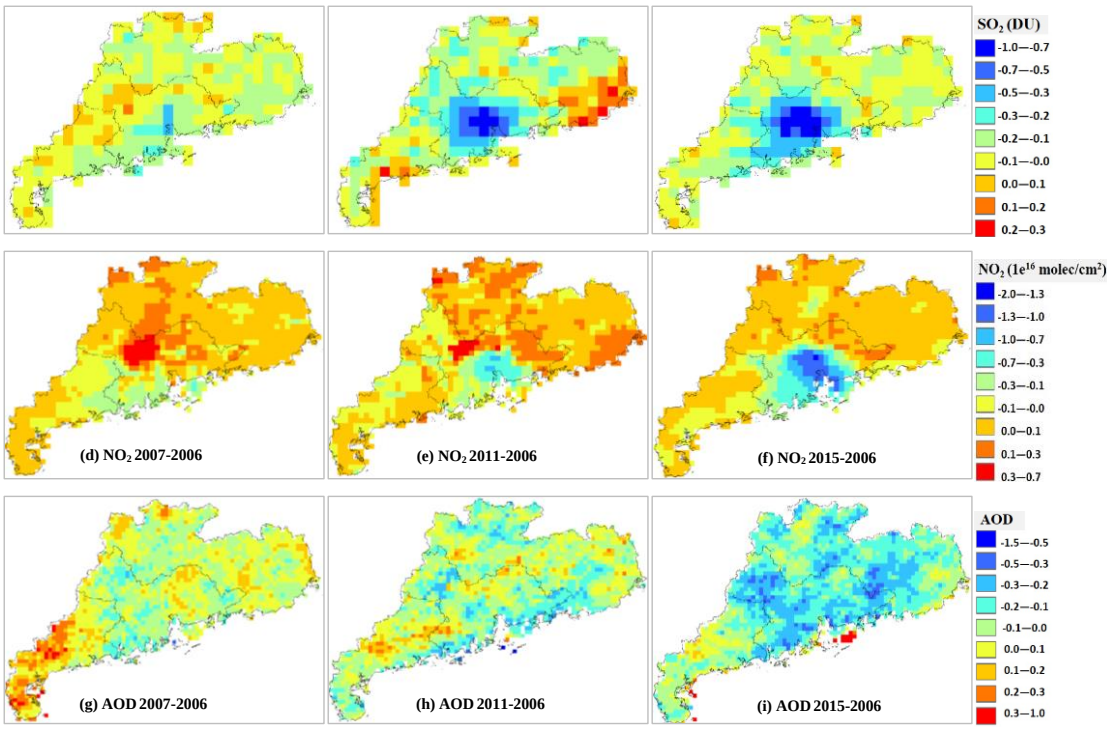

Figure 3. Changes in spatial patterns of satellite observations over GD in 2007, 2011 and 2015 base on 2006 for **(a)-(c)** SO_2, **(d)-(f)** NO_2, and **(g)-(i)** AOD. The legends represent a gradual increase in emissions from top (blue) to bottom (red). Central zone inside the inner black line presents the PRD region, outside presents the NPRD region.

Line 395: figure 5a is showing the result of PRD, not GD. Please specify.

Response: Accepted. We changed the wrong ordinal number in the sentence (line 599) "The source contribution of to SO_2 emissions in GD also changed (Fig. 4a)."

Line 437: "On-road mobile was also a major contributor to NOX emissions in GD (Fig. 5b)." Fig. 5b is showing results of SO2 for NPRD. Please double check.

Response: Thanks for pointing this. We changed the wrong ordinal number in the sentence (line 650) "On-road mobile was also a major contributor to NOX emissions in GD (Fig. 4b)."

Line 673: it should be 2.4.4

Response: Accepted. We corrected the number in the manuscript (Line 927) "2.4.4 VOCs".

Response to the Reviewer 3:

This paper shows the long-term evolution (2006~2015) of anthropogenic emissions in Guangdong Province of China, to reveal the causes of source-contribution changes during this periods. The

conclusions obtained in this paper are of great significance for the deep understanding of regional pollution emission characteristics, and have important guiding for co-control strategies on regional atmospheric pollution. The datasets produced by the article are conducive to the in-depth study of scientific issues in related fields. Therefore, this paper is strongly recommended for publication.

Response: Thanks for your positive comments.

Supplement of

Evolution of Anthropogenic Air Pollutants Emissions in Guangdong Province, China from 2006 to 2015

Evolution of Anthropogenic Air Pollutant Emissions in Guangdong Province, China, from 2006 to 2015

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Abstract

Guangdong province (GD), one of the most prosperous and populous regions in China, still experiences haze events and growing ozone pollution, despite of although it has seen substantial air quality improvement in recent years. Co- Integrated control of fine particulate matter ($PM_{2.5}$) and ozone in GD calls for a systematic review of historical emission patterns. In this study, emission trends, spatial variations, source-contribution variations, and reduction potentials of sulfur dioxide (SO_2), nitrogen oxides (NO_X), $PM_{2.5}$, inhalable particles (PM_{10}), carbon monoxide (CO), ammonia (NH_3), and volatile organic compounds (VOCs) in GD from 2006 to 2015 are were revealed examined using a dynamic methodology, taking into account economic development, technology penetration, and emission controls. The relative change rates of anthropogenic emissions in GD during 2006-2015 were are -48% for SO_2, -0.5% for NO_X, -16% for $PM_{2.5}$, -22% for PM_{10}, 13% for CO, 3% for NH_3, and 13% for VOCs. The declines of SO_2, NO_Xx, $PM_{2.5}$, and PM_{10} emissions are in the whole province are mainly attributed to resulted from the control-drivenstringent emission reductions control in the Pearl River Delta (PRD) region, where most previous control measures focused on, especially from power plants (SO_2 and NO_Xx), industrial combustion (SO_2, $PM_{2.5}$ and PM_{10}), on-road mobile source (NO_Xx)s, and fugitive dust source ($PM_{2.5}$ and PM_{10}). Emissions from the other areas (the non-PRD, NPRD), nevertheless, remain relatively stable Unlike the PRD region, the non-PRD (NPRD) region see relatively small emission declines, due to the lax control measures and rapidly growing energy consumption. AlsoIn addition, SO_2 and NOx emission leaks of SO_2 and NOx from power plants and industries werearе observed from the PRD to the NPRD were observed duringin 2010 and 2011. As a result, Consequently, eemissions in NPRD, are increasingly important in GD, particularly those from industrial combustionsources. Contribution of NPRD to the total NOx SO_2 emissions in GD, for example, has increased– from *3272% in 2006 to *481% in 2015, taking NOx emissions as an example. On-road mobile source and solvent use are the two key sources that should receive more effective control measures in GD. Current Although plenty of control measures were released in the past decade, some of them are not effective, or not enough. Control-driven eemission reductions from on-road mobile source in GD are neutralized by the substantial growth of light-duty gasoline vehicles population, increased by * times during 2006-2015, while VOC emissions in GD steadily increase ., and partly to the shift of industries from the PRD to the non-PRD (NPRD) region in GD. NPRD also contributed to part of the emission decline, but it was only

effective until 2011 when GD's Clean Air Action of *12th Five Year* was implemented. Ddue to the growth of solvent use and the absence of effective control measures Besides, f, VOC emissions in PRD and NPRD both steadily increased, and this might be one of the reasons that led to the slight upward trends of ozone concentrations in GD. To further reduce emissions, future work should could focus on power plants and industrial combustion in GD and industrial process source in NPRD, which still have large emission reduction potentials. The historical emission inventory developed in this study not only helps to understand the emission evolution in GD, but alsofor emissions of SO_2, NO_x, and particulate matter, and on solvent use and on-road mobile sources for VOC emissions. This study provides solid scientific support for further air quality improvement in GD. In addition, it provides robust data to quantify the impact of emission and meteorology variations on air quality and unveil the primary cause of significant air quality change in the PRD – GDregion in the recent decade.

Keywords: emission trends; source contribution; Guangdong Province; emission reductions

Introduction

Guangdong Province (GD), comprising nine major cities in the Pearl River Delta (PRD) region and eleven ~~under-development~~less developed cities in the Non-PRD (NPRD), is the most prosperous and populous regions ~~embedded in~~adjacent to the coast of southern China (Fig~~ure~~. S1 in the Supplementary Material). ~~In 2017, occupying~~ With only 1.9% of China's land coverage, GD contributed 10.0% of China's gross domestic product (GDP) and 8.0% of the population in 2017 (GDPBS, 2016-2017). Particularly, the PRD region, dubbed 'the world factory' and embracing a wide variety of industries, represented about 80.0% and 53.4% of GD's total GDP and population, respectively (GDPBS, 2016-2017). Benefited from China's opening-up policies, GD has experienced rapid economic growth ~~but also along~~accompany with serious and complex air pollution issues.

~~In order to~~To improve ~~curb the deterioration of its~~ air quality, ~~GD's government agencies have made~~ great efforts to have been made to formulate various control measures and policies, especially after 2013, when the Action Plan on the Prevention and Control of Air Pollution (PCAP, 2013-2017) was launched. The PCAP required the PRD region to reduce $PM_{2.5}$ concentration by 15% by the year 2017 compared with the 2013 levels. These policies and control measures, which were summarized in Table S~~3~~1 ~~of SI~~, ~~helped~~partly alleviated regional air pollution ~~problems~~ in GD. Ground-level observations show~~n~~ed that GD saw an obvious air quality improvement, with SO_2 and PM_{10} concentrations decrease~~ing~~ by 63.3% and 17.7%, respectively, from 2006 to 2017 (~~DEPG~~EPGD, 2006~~,~~–201~~8~~7). In particular, the PRD region is the first region to meet China's national standard of ~~35 μg/m^3~~ $PM_{2.5}$ standard (35 μg/m^3) for three consecutive years (34 μg/m^3 in 2015, 32 μg/m^3 in 2016 and 34 μg/m^3 in 2017).~~China, the world's most prolific emitter of anthropogenic air pollutants, has been working on ways to curb the deterioration of its air quality in recent decades. After the launch of the "Clean Air Action Plan" (CAAP) in 2013, China has seen a dramatic reduction in emissions, mainly driven by control measures in power plants and industrial sources. In 2017, the Ministry of Environmental Protection declared that China had achieved the desired targets of CAAP. Average fine particulate matter ($PM_{2.5}$) concentrations fell 35% in 74 cities across China from 2013 to 2017 (Zheng et al., 2017). China's Guangdong Province (GD), which is one of the most prosperous and populous provinces in China (Fig. S1), is one of the regions that has experienced significant air quality improvement in recent years. Particularly the Pearl River Delta (PRD) region, known as the hub of the "World Factory," is the first region to meet China's national 35 μg/m^3 $PM_{2.5}$~~

~~standard for three consecutive years (34 μg/m^3 in 2015, 32 μg/m^3 in 2016 and 34 μg/m^3 in 2017).~~

However, air pollution in GD is still a major concern. ~~First, t~~The annual $PM_{2.5}$ levels still far exceed WHO's $PM_{2.5}$ guideline value of 10 μg/m^3 (World Health Organization, 2006) and~~far exceed stricter air quality standards, such as the WHO IT-2 (25 μg/m^3). Also,~~ haze events frequently occur in winter (Tao et al., 2017). ~~Second~~Also, the ambient ozone concentrations have been growing in recent years~~, . a phenomenon also observed in northern China.~~ The 90th percentile of the maximum 8-hour average ozone concentration (90%-8h-ozone) in the PRD region increased by 24% since 2015, reaching~~was~~ 165 μg/m^3 in 2017~~, 14% and 9% higher, respectively, than those in 2015 and 2016~~ (GDEMC, EPDHK, EPBMC, 2018).

On ~~the~~ contrast to the need for further air quality improvement, the unclear causes of recent air quality change in GD ~~Moreover, the main causes of air quality change in GD is not clarified yet, which might~~hinder~~ins~~g the development of evidence-based air quality control strategies~~measures~~. Although several studies had assessed the impact of emission and meteorology change on air quality, their results are inconsistent with each other. For instance, Lin et al. (2018) suggested that emission controls helped ~~to~~ improve local air quality in the PRD region, according to a high consistency of ambient $PM_{2.5}$ concentrations and emissions. However, Mao et al. (2018) argued that meteorological and climate conditions rather than PM emissions ~~are in control of~~dominate the interannual variabilities and trends of winter haze days in PRD based on an observation-based approach. Yang et al. (2019) concluded that the inappropriate NOx/VOC control ratio within the PRD over the past years is likely responsible for the ozone increase, while Li et al. (2018) argued that the decrease of $PM_{2.5}$ concentration is the main driver.

Further mitigation of air pollution in GD calls for a systematic review of historical emissions. First, the long-term historical emission data could help policymakers understand the evolution of emissions, quantify the cuts in emissions that have been achieved by control measures, and identify those sources with the greatest potential for large future emission reductions (Gurjar et al., 2004; Ohara et al., 2007; Zhong et al., 2013). This is particularly important for GD whose~~that have less~~ potential for further emission reduction are shrinking as control measures tighten. Second, with the use of atmospheric chemical transport models (CTMs), the historical emission can be used to examine the~~establish~~ linkages between air quality improvements and control measures, and thus identify the main causes of air quality change and key control measures. All this information is crucial to guid~~ing~~e future air-quality management and formulat~~ing e~~ robust evidence-based policies.

As the pioneer of air pollution research in China, GD is one of the first areas to compile its own emission inventories in China (Zhong et al., 2013). However, no attempts have been made to estimate the historical emissions of GD. Most of the previous emission studies mainly focused on the PRD region, but ignored the NPRD region that also has increased emissions. For instance, Zheng et al. (2009) developed the first high-resolution emission inventory for the PRD region in 2006, followed by emission inventories for speciated volatile organic compounds (VOCs) (Zheng et al., 2009), biogenic VOCs (Zheng et al., 2010), ammonia (NH_3) (Yin et al., 2015), biomass burning (He et al., 2011, Zhang et al., 2013), sea salt (Liu et al., 2015) and multiple-year anthropogenic sources (Lu et al., 2013). Due to the strengthened emission controls in PRD, relocation of industries are in courses for those sectors that ~~and the shift of industries that are~~ are energy intensive, or highly polluting, or have excess production capacity from the PRD region to non-PRD (NPRD) areas (Chun, 2012; Yin et al., 2017) (Fig. S2)~~,~~. As a result, emissions in the PRD and NPRD regions have experienced substantial changes in recent years. This means that the emissions of PRD cannot fairly represent Guangdong as a whole. Although several emission inventories in GD were developed recently~~,~~ (Huang et al., 2015~~;~~; Pan et al., 2015~~;~~; Liu et al., 2017~~;~~; Zhong et al., 2018), they were carried out in a single year ~~or~~and ~~in a~~ limited in spatial coverage~~region~~, ~~or~~and ~~considering limited~~ source categories~~, so that they vary in methodology and source classification~~. Therefore, there is a need to develop a long-term historical emission inventory in GD using a consistent methodology and the same underlying driver data to fill the data gaps and to assist with future air pollution control measures.~~Further mitigation of air pollution in GD calls for a systematic review of historical emissions, which could help policymakers understand the evolution of emissions, quantify the cuts in emissions that have been achieved by control measures, and identify those sources with the greatest potential for large future emission reductions (Gurjar et al., 2004; Ohara et al., 2007; Zhong et al., 2013). This is particularly important for those regions that have less potential for further emission reduction as control measures tighten. In addition, it is essential to achieve a long-term emission inventory to reveal the main causes of changes in air quality, a step that is controversial at present. For instance, Lin et al. (2018) suggested that emission controls helped to improve local air quality in the PRD region, since there was a high consistency of ambient $PM_{2.5}$ concentrations and emissions in that region. However, Mao et al. (2018) argued that meteorological and climate conditions rather than PM emissions are in control of the interannual variabilities and trends of winter haze days in PRD based on an observation-based approach. Using atmospheric chemical transport

models (CTMs) and long-term emission data, the impacts of emission changes and control measures on air quality can be quantified. Coupled with other models, the corresponding impacts on climate change and population exposure can also be assessed. All this information is crucial to guiding future air-quality management and formulating robust air-quality policies.

Guangdong province was one of the first areas in China to compile emission inventories (Zhong et al., 2013). The local government has published PRD regional emission inventories for the base years of 1997, 2001, and 2003 using a top-down approach to support air-quality management. Zheng et al. (2009) developed the first high-resolution emission inventory for the PRD region in 2006. This inventory included six major categories and seven pollutants. Subsequently, emission inventories for black carbon (BC), organic carbon (OC), ammonia (NH_3), and biogenic volatile organic compounds (VOCs) in PRD were developed for different base years (Yu et al., 2011; Yin et al., 2015; Li et al., 2016). Huang et al. (2015) expanded the 2006-based emission inventories in PRD to provincial inventories in GD. Pan et al. (2015) updated the GD emission inventories by advancing the base year to 2010 and including additional emission sources. More recently, Zhong et al. (2018) updated the 2012-based GD emission inventories by source classification, emission methods, emission factors, and spatial-temporal surrogates. Regarding the emission trend, Lu et al. (2013) characterized anthropogenic emission trends and their variations in the PRD region from 2000 to 2009. Liu et al. (2017) developed long-term vehicle emissions in GD from 1994 to 2014.

One limitation of the above-mentioned studies is that most of them were carried out in a single year or in a limited region, or considering limited sources, so that they vary in methodology and source classification. Furthermore, most of these studies focused mainly on the PRD region due its notable economic growth and urbanization, but ignored the NPRD region that generally had less emissions. However, due to the strengthened emission controls in PRD and the shift of industries that are energy intensive, or highly polluting, or have excess production capacity from the PRD region to non-PRD (NPRD) areas (Chun, 2012; Yin et al., 2017) (Fig. S2), emissions in the PRD and NPRD regions might have experienced substantial changes in recent years. Therefore, there is a need to develop a long-term historical emission inventory in GD using a consistent methodology and the same underlying driver data to fill the data gaps and to assist with future air pollution control measures.

In this study, we developed a multi-year anthropogenic emission inventory for SO_2, NO_X, PM_{10}, $PM_{2.5}$, CO, VOCs, and NH_3 for the years from 2006 to 2015 using a

dynamic methodology that considers economic development, technological penetration, and emission controls. The emission trends were validated by ground-based measurements and satellite observations. Based on the long-term historical inventory, the emission changes, contribution variations, possible causes for the observed air quality improvement, and reduction potentials in 2020 in PRD and NPRD were analyzed and compared, which could provide scientific evidence for future air quality regulations in GD. ~~Also, t~~The long-term emission inventories developed in this study are also essential ~~data~~ to evaluate the effectiveness of emission control measures and identify the dominant cause of significant air quality change in ~~the PRD region~~GD.

1 Methodology and data

1.1 Methods for emission estimations

In this study, ~~we applied a dynamic technology-based methodology that considers economic development, technological penetration, and emission controls to estimate the anthropogenic emission trends in GD, following previous studies on emission trends (Streets et al., 2006; Zhang et al., 2007; Lu et al., 2013). We estimated~~ emissions of 7 pollutants (SO_2, NO_X, PM_{10}, $PM_{2.5}$, VOCs, CO, and NH_3) from 13 major categories, including power plants, industrial combustion, residential combustion, on-road mobile source, non-road mobile source, dust source, industrial process source, industrial solvent use, non-industrial solvent use, storage and transportation, agricultural source, biomass burning, and other sources, were estimated. On the basis of Zhong et al. (2018), ~~and 70 sub-categories based on~~ Pan et al. (2015) and the guidelines for the development of an air-pollutant emission inventory for Chinese cities (MEPC, 2017), the 13 major categories were further broken down into 70 sub-categories to improve emission estimation. Detail categories applied in this study are listed in Table S~~1~~2 ~~of SI~~. ~~The major categories include power plants, industrial combustion, residential combustion, on-road mobile source, non-road mobile source, dust sources, industrial processes~~industrial process source, ~~industrial solvent use, non-industrial solvent use, storage and transportation, agricultural sources~~agricultural source, ~~biomass burning, and other sources (Table S1).~~

Unlike the single-year emission inventory, the estimation of a long-term emission inventory is generally more complicated since it requires the data sources, estimation method and source category for all years are consistent. Although top-down methods or on-line estimation methods based on big data can promote the estimation, obtaining

the long-term activity data, such as Automatic Identification System (AIS) data for ship emission and Global Positioning System (GPS) data for on-road mobile source, is challenging. Therefore, most studies (Streets et al., 2006~~,~~; Zhang et al., 2007~~,~~; Lu et al., 2012~~,~~; Zheng~~,~~ et al., 2018), still generally applied the top-down method to develop long-term emission inventories. Following previous studies, we applied a dynamic technology-based methodology that considers economic development, technological penetration, and emission controls to estimate the anthropogenic emission trends in GD~~,~~ ~~following previous studies on emission trends (Streets et al., 2006; Zhang et al., 2007; Lu et al., 2013).~~

Except for on-road mobile and construction dust source~~s~~, emissions of most anthropogenic sources were calculated as follows:

$$E_{i,n} = \sum_{i,j,k} A_{i,j,k,n} \sum_{m} (X_{i,j,k,m,n} EF_{j,k,m,n}) \sum_{z} [C_{z,n}(1-\eta_z)] \quad (1)$$

where *i, j, k, m, n,* and *z* represent the city, the emission source, the type of fuel or product, the production technology, the year, and the control technology, respectively, *A* represents the activity level (such as the fuel consumption or material production), *X* represents the percentage of fuel or production for a sector consumed or produced by a specific technology *m*, *EF* is the unabated emission factor, $EF \sum_z [C_z(1-\eta_z)]$ is the net EF after applying control technology, *C* is the penetration of the control technology *z*, η is the removal efficiency of the control technology *z*, and *S* and *SR* represent the sulfur content in fuel and the sulfur retention in ash, respectively. For fuel combustion, the EF of SO_2 was estimated using the following equation:

$$EF = 2 \times S \times (1 - SR) \quad (2)$$

For on-road mobile source~~s~~ and construction dust, emissions were estimated by Eqs. (3) and (4), respectively. The methods from previous studies employed for the other emission sources are listed in detail in Table S2 in the Supplementary Material (SI).

$$E_{i,n} = \sum_{i,j} (P_{i,j,n} \times VKT_{i,j,n} \times EF_{j,n}) \quad (3)$$

$$E_{i,n} = \sum_{i} \left(S_{i,n} \times T_{i,n} \times EF_{i,n}\right) \quad (4)$$

where *i, j,* and *n* represent the city, the vehicle type, and the year, respectively, *P* is the vehicle population, and *VKT* is annual average vehicle kilometers traveled. *i* and *n* represent the city and the year, respectively. *S* is the construction area, and *T* is the construction cycle.

~~The annual emission mainly depends on activity data, emission factors, and~~

~~removal efficiencies of emission controls. Therefore,~~ An accurate representation of the annual change of activity data and emission factors is critical for estimating long-term ~~characterizing the~~ emission ~~s~~trend. Here, we provided a detailed description of activity data and emission factors applied in this study.

1.1.1 Activity data

~~The estimation of a multiyear emission inventory is complicated since it requires consistent and accurate activity data and EFs (Zhang et al., 2007).~~ Most of the activity data during 2006-2015 ~~in this study~~ were obtained from officially released statistics or relevant reports. Either surrogate data or data interpolation was used to fill in the data for some sources that lack continuous and consistent long-term activity data. Notably, the activity data that specifies an individual industry or power plant, defined as point data, were preferentially used, since these data generally have detailed information on the location, technical level and control measures. Otherwise, activity data at the city level, known as areal data, were adopted as a second choice. In this study, for power plants and industrial combustion, we used a combination of point data and areal data to characterize the activity level, while the other sources all relied on areal data. The detailed data sources are summarized in Table ~~S2~~S3. Here, we describe the processing of activity data for some major sources, e.g., industrial combustion, construction dust, marine and on-road mobile source~~s~~.

For industrial combustion, we used the total consumption of different energy types during 2006-2015 from the GD Statistical Yearbook (GDPBS, 2007-2016) to represent the activity level of each city. Also, we used a detailed dataset from GD pollutant statistical reports to estimate consumption value of different energy types, the averaged sulfur contents, and removal efficiencies of industrial combustion in each city. This dataset, which records the annual fuel consumption, sulfur contents, control devices, removal efficiencies, product output, and the geographic location of each plant, contains about 85% of the plants in GD and covers the years of 2006, 2010, 2012, 2014, and 2015. For the years that lack a detailed dataset, the averaged sulfur contents and removal efficiencies were estimated by linear interpolation and emission control policy.

For construction dust source~~s~~, we used the total annual construction area and construction cycle time to represent the activity level. The construction area data were derived from the GD city statistical yearbook (GDPCSY, 2007-2016), and the construction cycle time was determined by the time ~~requirement~~ required for different construction phases, i.e., earthwork excavation, foundations, earthwork backfill, and general construction. Considering the effect of rainfall in suppressing dust source~~s~~, we

revised the construction cycle time by combining our previous study (Yang, 2014) with rainfall information for each year obtained from the GD Meteorological Service (GDMS, 2007-2016).

Regarding marine sources, the characterization of activity level was based on heavy and diesel fuel consumption. However, it is challenging to acquire detailed consumption of various fuel types directly. Thus, we used the method described in Li et al. (~~2017~~2018) to convert the cargo volumes and transport distances of major navigation routes into fuel consumption data via fuel consumption rates. Fuel consumption rates were taken from the IMO report (IMO, 2009). The cargo volumes in each city were obtained from GD Statistical Yearbooks (GDPBS, 2007-2016). Transport distances of major navigation route data were measured by the historical AIS-based digital map.

For on-road mobile source~~s~~, population data of different vehicle types (i.e., passenger trucks, buses, taxis, and motorcycles) and~~,~~ the gross weight (heavy and light duty) from the~~—~~ 2006-2015 statistical yearbooks, and annual average vehicle kilometers traveled from a field survey of ~~some~~ 8 cities and 111 roads by types (i.e., arterial road, secondary arterial road and branch) in GD were used to characterize the annual change of activity level. We further divided the vehicle type into ~~differentiated~~ diesel and gasoline vehicles to obtain a more accurate estimate, based on the ~~, but this information was not available from the official statistics. Not only~~ vehicle ratios (diesel/gasoline) from our previous study (Che et al., 2009) in 2006 and ~~were considered, but also~~ the field survey covered 8 cities in the years of 2010, 2012, 2014, and 2015 ~~were carried out to obtain those information~~. The vehicle ratios in other years (2007-2009, 2011, and 2013) were estimated using an interpolation method. ~~However, subjected to the availability and continuity of data, vehicle ratios in some absent years were deduced according to the known years. Therefore, we distinguished the vehicle population based on test results regarding the vehicular ratios of fuel use in GD (Che et al., 2009) following a method in our previous study (Lu et al., 2013).~~

1.1.2 Emission factors (EFs)

EFs could have changed with the implementation of emission controls in GD during 2006-2015 ~~(Table S3)~~, which involve technological penetration and evolution. To deal with that possibility, we developed a dynamic method to reflect the response of EFs to control measures and technological penetration. First, we established the unabated EF of each source to represent ~~what~~ that the emission level would have been without any treatment. The unabated EFs for most emission sources of various

pollutants (i.e., NO_X, PM_{10}, $PM_{2.5}$, VOCs, CO, and NH_3) used in this study are listed in Tables S4-9, and are based mainly on the latest research results and values recommended in related manuals of air-pollutant emission coefficients. Next, we estimated the net EFs of each source according to the corresponding processing technologies, control technologies, and removal efficiencies that might vary with years. In this study, we applied the dynamic method to all emission sources. In the following subsection, we mainly describe their application to major sources that have received intensive control measures in the past decade: including power plants, industrial combustion, VOCs-related sources, and on-road mobile sources.

The net EFs of SO_2 for industries and power plants in GD were determined based on removal efficiencies and fuel sulfur content. The annual removal efficiencies and sulfur content in 2006, 2010, 2012, 2014, and 2015 were obtained from GD pollutant statistical reports (GDPSR, 2006, 2010, 2012, 2014, 2015) for industries; those parameters for power plants in 2006-2014 were obtained from power-plant reports (CSPG, 2006-2014). For years without these documented data, an interpolation method that considers newly released regulations of SO_2 emission controls and expert judgment were used to estimate the removal efficiency and sulfur contents (2007-2009, 2011, and 2013 for industries and 2015 for power plants). For instance, the sulfur content of coal and oil in industrial sources can be estimated as <0.7% and 0.8%, respectively, according to the *Guangdong industrial boiler pollution remediation program (2012-2015)* released in 2012.

For VOCs-related sources, such as industrial solvent use, non-industrial solvent use, and industrial process sources, the net EFs of VOCs were determined based on the installation rate of VOC control technologies and the removal efficiencies, which were acquired by an on-site investigation of VOCs-related industries in GD. Additionally, the new VOC emission standards were also used to determine in which year VOC control technologies were implemented. For example, emission standards for furniture surface coating and shoemaking were implemented in 2010 (*Emission standard of volatile organic compounds for furniture manufacturing operations (DB44/814-2010), Emission standard of volatile organic compounds for shoe-making industry (DB44/817-2010)*). Thus, we estimated the net VOC EFs for furniture surface coating and shoemaking with the consideration of VOC removal efficacies since 2010. For the vehicular EFs, we used the same method employed in our previous study (Lu et al., 2013). The vehicular EFs were calculated based on the 2007 International Vehicle Emissions (IVE) model (ISSRC, 2008), while the EFs for other years were derived from 2007-based EFs in consideration of emission standards, fuel standards, and vehicle

lifespans.

1.2 Data for validation

In this study, satellite observations, including SO_2, NO_2 and AOD column, and ground-level observations (SO_2, NO_2 and $PM_{2.5}$ concentrations) in PRD were used to evaluate the long-term total emission in GD area ~~(the total emission in GD area).~~ ~~To validate the multi-year emissions in GD, we compared the emission trends with satellite-based data.~~ The SO_2 column amount (OMSO2e v003) and the NO_2 tropospheric column (OMNO2d v003) were retrieved from the Ozone Monitoring Instrument (OMI) with a spatial resolution of 0.25°×0.25° (available at https://giovanni.gsfc.nasa.gov/giovanni/). Aerosol optical depth (AOD) data were taken from the Moderate-Resolution Imaging Spectroradiometer (MODIS) aerosol product MOD04 with a high resolution of 10 km (available at https://ladsweb.modaps.eosdis.nasa.gov/). The ground-level observations in PRD during 2006-2015 were obtained from the PRD Regional Air Quality Monitoring Network (RAQMN) (GDEMC, EPDHK, EPBMC, 2007-2016)~~In addition, ground-level observations obtained from the PRD air quality monitoring network (GDEMC, EPDHK, EPBMC, 2007-2016) were also used to validate emission trends in PRD~~. The RAQMN, which came into operation at the end of 2005 and has provided accurate air quality data to local governments and the public, was adopted because of the high reliability of the operating procedures on quality assurance and quality controls (QA/QC). We also collected the ground-level observations averaged over the GD from annual reports of Guangdong Provincial environment published by the department of environmental protection of Guangdong province. However, this data is less representatives since the ground-level stations in NPRD are much sparser and the QA/QC procedures are not well implemented in NPRD.

~~The air quality monitoring network came into operation at the end of 2005 and has provided accurate air quality data to local governments and the public. These data from the PRD air quality monitoring network were adopted because of the high reliability of the operating procedures on quality assurance and quality controls.~~ We focused on the evaluation of emission trends. Therefore, we mainly compared annual observations averaged over the PRD/GD region with annual emissions in PRD/GD. Also, emissions and satellite observations in PRD and NPRD region were compared.~~Also, emissions and satellite observations in PRD and NPRD region were compared to briefly evaluate the spatial variation of emissions in GD.~~

Since emissions mainly concentrated in urban areas, where ground-level observations in RAQMN located, the sparsity of ground-level observations was ignored in the evaluation.

2 Results and discussion

2.1 Overall emission trends

The overall emission trends of SO_2, NO_X, PM_{10}, $PM_{2.5}$, VOCs, CO, and NH_3 in GD during 2006-2015 are presented in Fig. 1. From 2006 to 2015, anthropogenic emissions decreased by 48% for SO_2, 0.5% for NO_X, 16% for $PM_{2.5}$, and 22% for PM_{10}, but increased for CO, NH_3, and VOCs, by 13%, 3%, and 33%, respectively. Specifically, SO_2 emissions fell steadily during 2006-2015 ~~, which might be~~ due to the strict controls on SO_2 emissions implemented in the 11th Five Year Plan (FYP) (2006-2015~~01~~) and 12th FYP (2011-2015). These two FYP all required the total national SO_2 emission to be cut by 1~~0~~5%, relative to the 2005 and 2010 level, respectively. NO_X emissions overall showed an upward trend in the early period, reaching a peak in 2011. After the implementation of *the Planning for Guangdong province environmental protection and ecological construction in 12th FYP* (PGGP, 2011) in 2011, in which NO_X emission caps of all industrial sectors were proposed, NO_X emissions decreased, declining by 9% in 2015. The PM_{10} and $PM_{2.5}$ emissions showed an increasing trend during 2006-2009 but then decreased steadily. Similarly, CO emissions showed a small rise during 2006-2013, followed by a sharp decline. NH_3 emissions changed a little, while VOC emissions steadily increased over the 2006-2015 period, mainly fueled by the absence of effective emission-control measures.

Although emissions of SO_2, NO_X, PM, and CO declined in recent years, the per capita GDP, fuel consumption, and vehicle population in GD, which account for most anthropogenic pollution activity, saw growth from 2006 to 2015, as shown in Fig. 1. From 2006 to 2015, the per capita GDP and vehicle population significantly increased, by 135% and 66%, respectively. Obviously, anthropogenic emissions in GD were decoupling from economic and energy consumption growth. This means that the emission regulations and control measures enacted in GD have alleviated emissions despite the growth of economic activity. For instance, NO_X emissions are closely related to fuel consumptions because a large proportion of their emissions are from industries or power plants that consume a great deal of fuel. However, the trends of NO_X emissions and fuel consumption have deviated from each other since 2011 when low NO_X-

combustion (LNB) control measures and flue gas denitrification technology (i.e., selective catalytic reduction (SCR) and selective non-catalytic reduction (SNCR)) in power plants were enacted.

We have also compared emission trends in PRD and NPRD (Fig. S3). Since emissions in GD were mainly concentrated in the PRD region, emission trends in PRD were similar to those in GD, except NO_X emissions, which started off steady until 2012 and then fell slightly. Compared with 2006, the 2015 emissions of SO_2, NO_X, $PM_{2.5}$, and PM_{10} in PRD decreased by 63%, 14%, 35%, and 27%, respectively, while CO and VOCs emissions increased by 2% and 35%, respectively. In NPRD, SO_2, NO_X, $PM_{2.5}$, and PM_{10} emission trends differed from those in PRD. Compared with 2006, the 2015 emissions of SO_2, $PM_{2.5}$, and PM_{10} in NPRD decreased by only 8%, 8%, and 5%, respectively, but emissions of VOCs, NH_3, CO, and NO_X significantly increased ~~—~~by 30%, 10%, 31%, and 29%, respectively. The discrepancy of emission trends between PRD and NPRD ~~was expected. This is~~raised because these two regions achieved different levels of progress on air quality management in the past decade ~~due to the discrepancy in economic development~~. Overall, most previous control strategies ~~(Table S3)~~ still focused ~~mainly~~ on PRD (Table S~~3~~1) ~~while the NPRD region received little attention~~. In Table S~~3~~1, all of these 11 control strategies involve PRD, while only 6 of them cover NPRD. ~~–~~Moreover, ~~initiated by~~ due to the policies of "vacate the cage and change birds" (in Chinese, Teng Long Huan Niao) initiated by the Guangdong provincial government in 2008 (Li & Fung, 2008; Yang, 2012), many low-tech industries in PRD were relocated to NPRD. As a result, emissions of NPRD is becoming more and more obvious in GD. For instance, the contribution of NPRD to SO_2 emissions in GD has increased from 27.1% in 2006 to 48.2% in 2015 (Table 1). For $NO_{~~x~~X}$, $PM_{2.5}$ and PM_{10} emissions, the proportions increased from 31.6% to 40.6%, from 49.5% to 56.0% and from 50.3% to 54.0%, respectively. ~~The statistical results showed 33 industrial parks were undertaken in NPRD until 2014 (GDEI, 2014).~~ Detailed emission evolutions and the corresponding causes are discussed in Sec. 2.3.

2.2 Validation of emission trends

In this section, we validate emission trends in PRD using ground observations and satellite data (Fig. 2). The comparison of emission trends with measurements in GD also presented in Fig. S4. In general, these two data sources are consistent for emission trends of SO_2, NO_X, and PM_{10}. During 2006-2015, SO_2 emissions in PRD decreased by 63% (emission trends in PRD ~~and NPRD~~ are shown in Fig. ~~2~~5), slightly less than the 68% and 86% observed in ground-level and satellite data, respectively. NOx emissions

and observations all showed a declining trend during 2006-2015. For PM_{10} emissions, the declining trend also closely followed the fluctuant downward trend of ambient $PM_{2.5}$ concentrations and AOD. The fluctuations of observations were associated with $PM_{2.5}$ formation influenced by annual variations of meteorology.

The change in the spatial variation of emissions in GD was also evaluated using satellite measurements. Here, the relative change of column concentrations of SO_2, NO_2, and AOD in 2007, 2011, and 2015 to 2006 were illustrated in Fig. 3. The annual column ~~changes based on 2006 in column concentrations of SO_2, NO_2, and AOD for GD in the years 2007, 2011, and 2015 were applied, as illustrated in Fig. 3 (the changes of column~~ concentrations ~~in the remaining years~~during 2006-2015 are all displayed in Fig. S45. ~~The spatial patterns of satellite observations of SO_2, NO_X, and AOD are displayed in Fig. S5 and. eEmission spatial variation of SO_2, NO_X, and $PM_{2.5}$ during 2006-2015 are displayed in Fig. S4 and Fig. S57, respectively).~~ Overall, the spatial patterns of the satellite measurements also reveal the different emission trends between PRD and NPRD. For example, SO_2 column concentration in PRD decreased by 71% from 2007 to 2011, but in NPRD increased by 26%. This agrees with the emission trends, in which SO_2 emissions in PRD decreased by 39% while those in NPRD increased by 15%. From 2011 to 2015, SO_2 column concentration decreased by 31% and 42% in PRD and NPRD, respectively, and SO_2 emissions decreased by 32% and 20% in PRD and NPRD, respectively. From 2007 to 2011, NO_2 column concentration decreased by 16% in PRD but increased by 16% in NPRD. These trends also coincided with emission changes, which NO_X emissions in PRD decreased by 13% but increased by 36% in NPRD. From 2011 to 2015, NO_2 column concentration and NO_X emissions both decreased in PRD and NPRD. AOD displayed a decrease of approximately 23% in PRD and NPRD from 2007 to 2015. A similar pattern was also found in $PM_{2.5}$ emission trends, with decreases of 35% in PRD and 9% in NPRD.

Overall, the above-mentioned validations using satellite observations and ground measurements demonstrate that emission trends of SO_2, NO_X, and PM estimated in this study are reliable.

2.3 Evolution of source emissions in Guangdong Province

To further understand the evolution of emissions in GD, we estimated the changes of anthropogenic emissions by emission category in PRD and NPRD during 2006-2015. Here, we examine the PRD and NPRD regions separately since these two regions may have experienced diverse emission evolutions due to their different levels of progress on air-quality management. The annual source contributions during 2006-2015 are also

presented in Fig. 4 (GD) and Fig. ~~S5~~ S6~~7~~ (PRD and NPRD).

2.3.1 SO_2

In PRD, SO_2 emissions steadily declined, from 0.788 Tg in 2006 to 0.292 Tg in 2015. The decline was dominated by power plants and industrial combustion, accounting for 50.0% and 46.8% of the total decrease, respectively (Fig. 5a). The decline of SO_2 emissions from power plants and industrial combustion is highly associated with the control measures enacted during the 11th FYP and 12th FYP. ~~which decreased by 0.247 Tg and 0.232 Tg, respectively, in this period (Fig. 5a).~~In response to the SO_2 emission cap in 11th FYP, Guangdong province required the elimination of small thermal power units with high energy consumption and outdated combustion technology in 2007. By the end of 2011, 12.2 million kilowatts of small thermal power units were eliminated during the 11th FYP. In addition, *action plan for air pollution prevention and control in Guangdong province (2014-2017)* (GDEP, 2014) ~~the "Action plan for air pollution prevention and control in Guangdong province"~~ released in the 12th FYP further strengthen the emission control of power plants. As a result, the flue gas desulfurization (FGD) penetration in coal-fired power plants rose to 87% in 2013. For the industrial combustion, its fuel- consumption in PRD decreased by 47.5% during 2006-2015, which can be explained by energy structural adjustment regulations, including the "total amount control of coals" and "changing fuel from coal to natural gas" control measures (Figure. S8~~7~~). The detailed regulations on energy structural adjustment are summarized in Table S1.

~~This indicates that flue-gas desulfurization, the control of sulfur content of fuels, and boiler renovation of SO_2 emission controls in the 11th FYP were effective.~~ In NPRD, on the contrary, SO_2 emission increased until 2010, ~~when it~~then saw a downturn. Before 2010, the SO_2 emission growth was mainly associated with the increase of industrial combustion and non-road mobile source~~s~~ (Fig. 5b). These two sources still maintained a slight rise after 2010, but their increased emissions were offset by a ~~sharp~~ plunge of emissions from power plants~~, which resulted mainly from tightening desulfurization technology~~. To reveal the ~~reason for~~ cause of ~~the~~ emission growth from~~of~~ industrial combustion in NPRD, we compared the standard fuel-consumption trends from industrial combustion in PRD and NPRD (Fig. 6). ~~As shown in Fig. 6, t~~In PRD, the fuel consumption from industrial combustion in PRD steadily dropped by 47% during 2006-2015, with an average annual descending rate of 6.9%. ~~By contrast,~~ In NPRD, by contrast, the fuel consumption ~~it~~ increased by 99%~~, mainly after 2010~~. ~~In~~ In fact, the average annual growth rates of fuel consumption in NPRD was only 3.2% before 2010,

but after that, the increase accelerated, with an average annual growth rate of 11.9% during 2011-2015. Particularly, the growth rate peaked in 2011 (17.1%) and 2012 (14.5%). Meanwhile, the descending rates of fuel consumption in PRD also reached its peak in 2011 (9.9%) and 2012 (11.4%), when the shift of industries implemented. This fact indicates the existence of emission leak from PRD to NPRD due to the policy of "vacate the cage and change birds" that brought many energy-intensive industries from PRD to NPRD. Statistical data showed that the NPRD region had undertaken 33 industrial parks that migrated from PRD, as shown in Table S10 (GDEI, 2014). Consequently, the industrial shift might, in turn, promote the emission of power plants in NPRD to some extent.

~~This might be closely associated with the policy of "vacate the cage and change birds" that brought many energy-intensive industries from PRD to NPRD. Emissions and emissions per unit GDP of industrial waste gases increased by 63.1% and 53.3% in NPRD during 2008-2013, respectively, due to industrial relocation in GD, far more than those in PRD (Liu, 2016).~~

The source contribution ~~of~~ to SO_2 emissions in GD also changed (Fig.~~5a~~4a). In GD, the contribution of power plants dropped from 43% to 27%. By contrast, the contribution of industrial combustion remained stable, in the range of 36% to 41%. Therefore, industrial combustion ~~took over from~~replaced power plants ~~to~~ and ~~became~~ became the largest SO_2 emission source in GD. The PRD and NPRD regions also had similar changes (Fig. ~~S5a~~S~~7~~6a-b). Note that the contribution of non-road mobile source~~s~~ to SO_2 emissions both increased ~~both~~ in PRD (from 13% to 29%) and NPRD (from 8% to 17%), due to the absence of effective emission control measures. Also, the contribution of industrial process source slightly increased in NPRD, from 5% to 11%. ~~Therefore, future work should be focused on non-road mobile source to further reduce SO_2 emissions in GD. In addition, in NPRD, industrial combustion also might have potential for reduction, since its contribution slightly increased, from 5% to 11%.~~

2.3.2 NO_X

As with SO_2 emissions, the evolution of NO_X emissions also differed between PRD and NPRD. In PRD, NO_X emissions dropped overall from 0.926 Tg in 2006 to 0.797 Tg in 2015, especially after ~~2012~~ 2011, when $NO_{~~x~~X}$ was put on the list of pollutant control, NO_X emissions saw a noticeable downturn (Fig. 5c). Power plants were the ~~principal~~ primary sources leading to emission reduction. This expected because the local government had released ~~"~~ ~~"~~*the implementation plan for nitrogen reduction and denitrification project for power plants in Guangdong province*~~"~~ ~~"~~ ~~in~~to respond

~~response~~ to the ~~explicit~~ reduction target of ~~reducing~~ NO_X emission~~s~~ in the 12th FYP (2011-2015). As a result, ~~,~~ a large number of power plants ~~had~~ installed NO_X removal equipment since 2011. ~~Thus, i~~By the end of~~n~~ 2015, NO_X emissions from power plants declined by 54.0% compared with~~were 0.214 Tg less than in~~ 2006 due to the increasing installment of flue gas denitration facility since 2011. However, a part of the NO_X emission reductions ~~–~~from power plants in PRD was ~~still~~ canceled out by the ~~growth in~~ emission growth from non-road mobile source, which increased by 65.7% during 2006-2015~~s, an increase of 0.452 Tg; and on-road mobile sources, with a slight increase of 0.012 Tg~~. Despite the intensive control measures for urban vehicle exhaust, NOx emissions from on-road mobile source still kept a slowly growing trend, indicating that these control measures were ~~Recent control measures have focused on vehicle exhaust, but it is~~ unlikely to offset the increasing vehicle population ~~in PRD~~, which increased by 88% during 2006-2015. Consequently, on-road mobile source~~s~~ (growing from 32% in 2006 to 38% in 2015) overtook power plants (growing from 43% to 23%) as the largest contributor to NO_X emissions in PRD since 2010 (Fig. ~~S5c~~S~~67~~c).

Unlike in PRD, NO_X emissions in NPRD rebounded in 2008 and grew sharply until 2012. The emission changes were mainly from industrial combustion, non-road mobile source~~s~~, and power plants (Fig. 5d). Among these three sources, NO_X emissions from industrial combustion and non-road mobile source~~s~~ both showed an upward trend, increasing by 159% and 84%, respectively, during 2006-2015. These trends can be explained by the shift of energy-intensive industries from PRD to NPRD and the absence of catch-up emission controls for industries and non-road mobile source~~s~~. In fact, most of the control measures for non-road mobile source were released after 2015. Since 2011, possibly due to the implementation of denitrification technology, NO_X emissions from power plants steadily went down and offset the slight increase in emissions from industrial combustion and non-road mobile source~~s~~. Consequently, the contribution of power plants to the total NOx emissions in NPRD, which was once a large contributor, decreased from 44% in 2006 to 28% in 2015. By contrast, contributions of industrial combustion and non-road mobile source~~s~~ increased from 14% to 29% and from 16% to 23%, respectively (Fig. S~~67~~5~~6~~d).

On-road mobile was also a major contributor to NO_X emissions in GD (Fig. ~~5b~~4b). Although the total NO_X emissions of on-road mobile source~~s~~ changed slightly in 2006-2015, their sectoral contribution showed significant change, especially in PRD (Fig. S~~56~~76c). Here, we further analyze the trends of NO_X emissions from on-road mobile source~~s~~ and the vehicle population from 2006 to 2015 for PRD (Fig. 7a) and NPRD (Fig. 7b). Overall, on-road mobile ~~NO_x~~ NO_X emissions in PRD were approximately

three times higher than those in NPRD, but their trends were similar. ~~In both PRD and NPRD,~~ Although the heavy-duty diesel truck (HDDT) population slightly grew until 2014 in both PRD and NPRD, NO_X~~x~~ emissions from HDDT still dropped by 10.0% and 30.8% during 2006-2015, respectively, which ~~NO_X emissions from heavy-duty diesel truck (HDDT) slightly decreased although the HDDT population showed growth before 2014. Apparently, this~~ was closely related to the improvement in vehicular emission and fuel standards. During the study period, the emission standard for vehicles raised from China III~~–III~~ (2008) to China IV~~–IV~~ (2013), while the oil standard improved from China III~~–III~~ (2009) to China V~~–V~~ (2014). The proportion of China III HDDT vehicles increased by over 50% in both PRD and NPRD (Liu et al. 2017). NO_X emissions from heavy-duty diesel vehicle (HDDV) also dropped, partly due to the ~~reduced~~ decreased HDDV vehicles~~s~~ population. Unlike the HDDT and HDDV, the population of light-duty gasoline vehicle (LDGV) increased significantly, by a factor of 5 and 6 in PRD and NPRD, respectively. This inevitably led to the obvious growth of NO_X~~x~~ emissions from LGDV, despite of the enhancement of vehicle emission standard and weed out of yellow label vehicles ~~The upsurge of the LDGV population outpaced the new standards on vehicle emission enacted since 2009~~ (PGGP, 2009)~~, leading to growth of NO_X emissions from LGDV~~.

~~Consequently, the contribution of LGDV to NO_X emissions in PRD surged from 12% in 2006 to 34% in 2015.~~

Based on the above analysis, it can be concluded that the NO_X emission trend in GD is dominated by the decline of power plants in PRD and the increase of non-road mobile source and industrial combustion in PRD and NPRD. Particularly, the contribution of non-road mobile source and industrial combustion to ~~NO_x~~–NO_X emissions in GD increased from 13% in 2006 to 23% in 2015 and from 12% to 20%, respectively (Fig. 4b), indicating that these two sources should receive more attention in future emission-control measures, especially industrial combustion in NPRD. Regarding on-road mobile source~~s~~, the largest contributor to NO_X~~x~~ emissions in PRD, LDGVs, should require more attention in the future.

2.3.3 PM_{10}/$PM_{2.5}$

As shown in Fig. ~~4c~~5e-~~d~~h, the main sectoral changes between PM_{10} and $PM_{2.5}$ emission were somewhat similar to each other. Therefore, we focused mainly on the analysis of PM_{10} emissions. PM_{10} emissions in PRD and NPRD showed similar ~~PM_{10} emission~~ variations regarding emission trends and source contributions. They both topped out in 2009 and then decreased monotonically. Compared with 2006, PM_{10}

emissions in 2015 dropped by 27% and 6% in PRD and NPRD, respectively. Dust source~~s~~, power plants, and industrial process source~~s~~ were the major contributors to the change of PM_{10} emissions in PRD and NPRD. However, emission trends of these three sources in PRD and NPRD were slightly different, particularly the ~~industrial processes~~industrial process source. In PRD, PM_{10} emissions from ~~industrial processes~~industrial process source ~~–~~ steadily declined after 2010, while in NPRD, PM_{10} emissions from ~~industrial processes~~industrial process source ~~–~~kept an upward trend during 2006-2015. One possible reason for the difference is that control measures for PM_{10} emissions in PRD were stricter than those in NPRD. In 2010, the local government issued two air action plans and regulated emission control measures for non-metallic minerals industries (Table S1), one of the major $PM_{2.5}$ and PM_{10} emission sources in GD. For instance, all cement plants, glass manufacturers and other non-metallic minerals industries that still outdated production processes were eliminated by the end of 2012.~~–~~

~~Speed up the elimination of high energy consumption and seriously polluted old-fashioned production processes. By the end of 2012, eliminate completely backward cement production capacity, and "horizontal sheet process" backward flat glass production capacity.~~

~~—~~However, these control measures are limited to the PRD region. ~~–~~

In PRD and NPRD, ~~fugitive~~ dust source increased during 2006-2010 and showed a decrease during 2011-2015. The downturn in the later years was due to the implementation of emission control technologies of dust source~~s~~ in response to the release of the *clean air action plan for the Pearl River Delta in Guangdong province* in 2010.

As emissions from ~~industrial processes~~industrial process source, dust source~~s~~, and power plants changed dramatically, the major sources contributed to $PM_{2.5}$ and PM_{10} emissions also changed accordingly (Fig. 4c-d, S~~5676g-j~~c-h). For $PM_{2.5}$ emissions in GD, contributions from dust source~~s~~ and power plants declined slightly, from 17% in 2006 to 11% in 2015, and from 12% to 7%, respectively. The contribution from ~~industrial processes~~industrial process source, the largest contributor, also slightly decreased from 38% to 33%. For PM_{10} emissions in GD, the contribution of ~~industrial processes~~industrial process source increased, from 28% to 34%. Particularly in NPRD, it replaced dust source~~s~~ as the largest contributor since 2012. In PRD, fugitive dust was still the largest PM_{10} emission source. Based on the above-mentioned analysis, PM emission controls for dust source~~s~~, especially in PRD, and for ~~industrial~~

~~processes~~industrial process source, especially in NPRD, should be a priority of the agenda in the next stage of emission controls. Also, $PM_{2.5}$ emissions from cooking cannot be neglected in GD; this is because the removal of cooking oil fumes from homes and restaurants was not strictly enforced, although some regulations and emission standards regarding cooking emissions were enacted gradually.

2.3.4 VOCs

As shown in Fig. 5i-j, the sectoral changes of VOC emissions in PRD and NPRD were similar. The total emissions in these two regions both showed a rising trend during 2006-2015, increasing by 35% and 30% in PRD and NPRD, respectively. The steady increase mainly originated from the growth of industrial-solvent use and non-industrial solvent use, whose emissions in GD respectively increased by 99% and 69% during 2006-2015 (Fig. 4e). Industrial solvent use was a large increasing source, especially in PRD where most industrial sources are concentrated. This is expected because solvent use required by industrial sources was growing, but control measures were insufficient. Several VOC control technologies had been adopted since 2010. For instance, the use of low VOC-containing raw materials for printing, shoemaking, furniture manufacturing, and other industries was first proposed at the *clean air action plan for the Pearl River Delta in Guangdong province* in 2010. Although these measures slowed an increasing trend of VOC emissions in PRD (VOC emissions from industrial solvent use in PRD increased by 18% during 2006-2010 while they increased by 6% during 2011-2015 ~~(Fig. S76)~~), the control efficiencies were still low~~. , According to a field survey,~~ only 40% of VOC-emitting industries had removal equipment in GD in 2014 according to a field survey (Wang et al., 2018). ~~Noted that t~~

The contribution from industrial solvent used in ~~the~~ NPRD increased from 11% in 2006 to 17% in 2015, ~~and two of significant increase was from~~partly driven by the electronics coating (0.5% of industrial solvent in 2008 rise to 17% in 2011) and appliance coating (14% to 30%) (Fig. ~~S~~78). ~~Which~~This coincided with ~~yang~~Yang[2] ~~s~~ at al. (2014) study that electronic equipment and appliance manufacturing accounted for 23% of 457 transferred enterprise until 2011~~(Yang et al., 2014).~~ The increasing VOC emissions from industrial-solvent use made it become the largest contributor to VOC emissions in GD in 2015 (Fig. 4e), with a percentage of 32%. Therefore, the implementation of policy and upgrade of control technologies are still required to reduce VOC emissions~~, especially in PRD~~. In NPRD, non-industrial solvent use was also a major contributor to the increase of VOC emissions. In particular, it became the largest contributor to VOC emissions in NPRD in 2015, with a percentage of 22%,

slightly larger than on-road mobile source~~s~~ (21%) (Fig. S~~56~~76j).

Since on-road mobile source~~s~~ w~~ere~~as also a major contributor to VOC emissions, the evolution of their VOC emissions is also discussed here (Fig. ~~8~~9). In both PRD and NPRD, VOC emissions from motorcycles, the largest contributors to VOC emissions from on-road mobile, went down in the past decade due to the relatively strict ban on motorcycles. They decreased by 55% and 38% in PRD and NPRD, respectively. By contrast, VOC emissions from LDGV increased by 118% and 197% in PRD and NPRD, respectively, likely due to the upsurge of the LDGV vehicle population. Particularly in PRD, LDGV's became the largest contributor to vehicle-related VOC emissions since 2008, which might also happen in NPRD according to the current trend.

2.3.5 CO/NH_3

As shown in Fig. 5k-l, CO emissions in PRD and NPRD both increased steadily during 2006-2013, and then decreased after 2013. However, the sectoral changes were different in these two regions. In PRD, the growth of CO emissions during 2006-2013 ~~is~~ was mainly attributed to industrial combustion and on-road mobile source~~s~~, while in NPRD, it ~~is associated principally with~~was from the industrial combustion and ~~industrial processes~~industrial process source. The difference exists because on-road mobile source~~s~~ ~~were~~ was primarily concentrated in PRD while iron and steel sectors, the largest CO emitters among industrial process source~~s~~, were located mainly in NPRD. Notably, production of the iron and steel sectors soared during 2006-2015, increasing by almost 95% in GD (GDPBS, 2007-2016), but emission controls fell behind. As to the decline of CO emissions during 2013-2015 in PRD, on-road mobile was the major reason. By contrast, the slight downturn in NPRD was mostly due to declining emissions from on-road mobile source~~s~~ and biomass burning. All these sectoral changes made industrial combustion (35% in 2015) become the largest contributor to CO emissions in GD (Fig. 4f). In NPRD, the contribution of industrial process source~~s~~ also increased. In contrast, the contribution of on-road mobile to CO emissions in NPRD decreased by 19% in 2015 compared with that in 2006.

As shown in Fig. 4g and Fig. 5m-n, ~~agricultural sources~~agricultural source constituted most to the change of NH_3 emissions, as they accounted for 86%-87% of the total NH_3 emissions in GD. However, their annual changes were different in PRD and NPRD (Fig. 5m-n). In NPRD, NH_3 emissions by ~~agricultural sources~~agricultural source increased by 11% during 2006-2015, partly as the result of the growth of fertilizing and livestock to meet the increasing demand for food. Another reason was the absence of effective emission controls on ~~agricultural sources~~agricultural source in

China. By contrast, in PRD, NH_3 emissions by ~~agricultural sources~~agricultural source remained stable.

2.4 Evaluation of emission control measures and policy implications

To evaluate the efficiencies of the control measures enacted in GD and to provide implications for future policies, we decomposed the emission changes into two categories: (1) changes resulting from change of activity level (activity-driven emission) in the absence of control measures and (2) changes due to the implementation of pollution controls (control-driven emission reduction) (Zheng et al., 2018)~~Emission changes can be divided into two categories: (1) changes resulting from change of activity level (activity-driven emission) in the absence of control measures and (2) changes due to the implementation of pollution controls (control-driven emission reduction) (Zheng et al., 2018). In this study, these two categories of emission changes were quantified to help evaluate the efficiencies of the control measures enacted in GD and to provide implications for future policies.~~ We estimated the unabated emissions if pollution control had been frozen at the 2006 level. In other words, we assumed that there were no new control measures adopted since 2006. Then the control-driven emission reduction was estimated by comparing the unabated emissions and the actual emissions, and the activity-driven emission was estimated by calculating the annual changes of unabated emissions. Also, we projected the actual emission to 2020 to help understand the potential for more emission control. Here, the planned emission controls before 2020 were assumed to be completely implemented in 2020. These related regulations for SO_2, NO_X, PM_{10}, and VOC emissions controls are summarized in Table ~~S3~~S1. The control-driven and activity-driven emissions of SO_2, NO_X, PM_{10}, and VOCs in 2007, 2009, 2011, 2013, and 2015 in addition to the predicted emissions in 2020 in PRD and NPRD are presented in Fig. ~~9~~10.

~~Figure 9 studies how might the emission changes in the absence of control measures and reduction potentials if actions have been taken, under the future projection of activity level.~~

2.4.1 SO_2

During 2007-2011, the decline of SO_2 emissions in PRD was driven by emission controls. The control-driven SO_2 emission reductions in PRD dramatically grew from 0.120 Tg in 2007 to 0.568 Tg in 2011 (Fig. ~~9a~~10a), mainly attributable to stringent SO_2 emission control regulations on industrial combustion and power plants—e.g., shutting down small coal-fired thermal power units, phasing out small boilers, installing flue-gas desulphurization (FDG) equipment and limiting the sulfur contents of fuel. SO_2

emission reductions from industrial combustion and power plants account for 57%-75% and 25%-37% of the total emission reductions, respectively. The activity-driven emissions increased by 0.178 Tg during 2007-2011, but their increments were far less than the control-driven emission reductions. The control-driven emission reductions flattened out in recent years. During 2013-2015, the control-driven emission reductions only increased by 4%, which owes much to the effectiveness of SO_2 emission controls in earlier years and the shrinking of control measures in recent years. Nonetheless, SO_2 emissions in PRD still steadily declined, partly due to the decrease of activity-driven emissions. By contrast, activity-driven emissions in NPRD kept rising, similar to Fig.6, which ~~might~~ also be associated with the transfer of energy-intensive industries from PRD to NPRD ~~(~~(Fig. ~~9b~~10b). Even so, the control-driven emission reductions dramatically increased and outweighed the activity-driven emissions since 2011, when stricter control measures were implemented. Similar to the situation in PRD, pollution control-driven emission reductions in NPRD were mainly attributed to industrial combustion and power plants.

Although SO_2 emissions dramatically decreased since 2006, there is potential for further reduction in 2020. On the basis of control-driven emission reductions in 2015, SO_2 emission reduction potentials in PRD and NPRD in 2020 are projected to be 0.10 Tg (34% of the total SO_2 emissions in 2015) and 0.29 Tg (approximately equal to the total SO_2 emissions in 2015), respectively. These reductions can be achieved by technical innovations, including ultra-low-emission measures in power plants, a series of actions regarding boiler management, sulfur content controls in fuels, and flue-gas desulfurization in industries. Most of these emission reductions are from industrial combustion and power plants. Particularly for NPRD, 60% of the reductions can come from industrial combustion, more than that in PRD. This is because SO_2 removal efficiencies in industries are ~~still~~ relatively low in NPRD. SO_2 emission contribution from non-road mobile source~~s~~ in GD previously presented an increasing trend (Fig. 4a). This reminds us that non-road mobile source~~s~~ still ~~have~~ has a high potential for SO_2 emission reduction. In PRD, it could account for approximately 20% of the total SO_2 emission reductions in 2020. Thus, future measures should be focused on industrial combustion and non-road mobile source~~s~~ for controlling SO_2 and NO_X emissions (NO_X also presented a similar result in the subsequent analysis).

2.4.2 NO_X

In PRD, the decline of NO_X emissions was driven by emission controls, and was significantly enhanced in 2011 when the ~~11~~th 12th Five Year Plan was enacted in GD,

including the application of technology for flue-gas denitrification and low NO_X combustion (LNB) in industries and power plants (Kurokawa et al., 2013), the elimination of yellow-label cars, and progressive advancements in vehicle emission and fuel standards. These mitigation measures yielded 0.315 Tg NO_X emission reductions in 2011, and offset the growth of activity-driven emissions. During 2007-2015, power plants and on-road mobile source~~s~~ were the two major contributors, accounting for 34%-59% and 38%-60% of the total control-driven emission reductions, respectively. Industrial combustion also contributed 2%-14% of the total control-driven emission reductions. Unlike in PRD, NPRD's new clean-air actions ~~only~~ mostly focused on power plants, and these measures were not stringent enough to cover the growth of NO_X emissions before 2011. After 2011, the control measures of NO_X emissions from power plants in NPRD were strengthened, leading to a significant increase of control-driven NO_X emission reductions. Consequently, the total NO_X emissions slightly declined. Apart from power plants, on-road mobile source~~s~~ and industrial combustion also partly contributed to the control-driven emission reductions in NPRD in recent years.

On the basis of control-driven emission reductions in 2015, NO_X emissions in 2020 could be further reduced by 0.24 Tg both in PRD (30% of the total NO_X emissions in 2015) and NPRD (43% of the total NO_X emissions in 2015). Most of these projected reductions could come from industrial combustion and power plants as a result of the implementation ~~implement~~ of stricter regulations, e.g., ultra-low emissions for power plants, boiler management, and flue-gas denitrification for industries. In PRD, on-road and non-road mobile sources also have a relatively high potential for NO_X emission reduction. Particularly, on-road mobile source~~s~~, especially LDGV, require~~s~~ more effective control measures. Although current control measures have alleviated the amount of vehicle emissions in recent decades, it still cannot cover the increased emissions driven by the rapid growth of vehicle population, as shown in Fig. ~~5~~5c~~b~~. Further reduction of on-road mobile NO_X emissions can be achieved by the implementation of control regulations.

2.4.3 PM_{10}

The activity-driven emission of PM_{10} in PRD and NPRD both steadily increased during 2006-2015 due to growth in activity. From 2006 to 2015, the gross industrial output value increased by 48% (GDPBS, 2007-2016), but control measures were not implemented until 2009, leading to a slight increase of PM_{10} emissions during 2006-2009. After 2009, the installation of dust removal equipment dramatically increased with the stricter implement of PM control measures, such as special requirements

limiting soot emission in power plants, boiler management with smaller capacity, and a series of pollution controls for non-metallic minerals industries. These measures boost the control-driven emission reductions, which can counterbalance the growth of activity-driven emission of PM_{10} in PRD and NPRD. In PRD, the control-driven emission reductions dramatically improved, from 0.024 Tg in 2009 to 0.318 Tg in 2015, while in NPRD, they improved from 0.014 Tg in 2009 to 0.258 Tg in 2015. In both the PRD and NPRD region, industrial combustion, power plants, and dust sources were the three major contributors to the control-driven emissions reductions.

Compared with control-driven emission reductions in 2015, PM_{10} emissions in 2020 in the PRD and NPRD could be further reduced by 0.31 Tg (60% of the total PM_{10} emissions in 2015) and 0.33 Tg (49% of the total PM_{10} emissions in 2015), respectively. Fugitive dust is the most significant contributor, accounting 45% and 31% of the total reductions in PRD and NPRD, respectively. This can be achieved by applying online monitoring technology for supervising construction dust (Sun et al., 2016) and more advanced measures, such as achieving a "6 100%" target for construction sites and increasing machine cleaning ratio for road dust. Industrial process sources, power plants, and industrial combustion also have major potential to achieve emission reduction, ~~in emissions~~ especially industrial process sources in the NPRD.

2.4.~~3~~ 4 VOCs

For VOCs, control-driven emission reductions in PRD and NPRD were slight in the past decade. Although VOC emission-control measures, such as promoting emphasis on strict end-of-pipe controls and leak detection and repair (LDAR) technology in VOC-emitting industries, and strengthening oil and gas recovery in gas stations, have been gradually highlighted since 2014 when *action plan for air pollution prevention and control in Guangdong province (2014-2017)* (GDEP, 2014) was released, the regulation has not been well executed. Emission reductions from solvent sources, the largest contributor to VOC emissions in PRD (Fig. ~~S5i~~S~~6~~76i), were 0.075 Tg in 2015, highly associated with the use of low-VOC products and environmental-friendly paints that contain low or even no VOCs. However, these emission reductions only accounted for 10%-35% of the total VOC emission reductions. In fact, 65%-86% of the control-driven VOC emission reductions were from on-road mobile sources, which is mainly attributed to the improvement of emission standards and oil quality for vehicles, management of yellow label cars, and the popularization of green traffic. Even so, the control-driven VOC emission reductions (from 0.016 Tg in 2007 to 0.294 Tg in 2015) were far outweighed by the activity-driven growth in emissions (from 0.699 Tg in 2007

to 1.172 Tg in 2015), resulting from the growth of vehicle populations and increasing use of solvents, which consequently drove up VOC emissions in PRD and NPRD (Fig. 5i-j).

In 2020, if the existing emission control regulations were fully implemented, VOC emissions in PRD would decrease by 30% relative to the emission level in 2015. The emission reduction potentials are 0.61 Tg in PRD (69% of the total VOC emissions in 2015) and 0.56 Tg in NPRD (0.2 times higher than total VOC emissions in 2015), respectively, much larger than the emission reduction potentials of SO_2, NO_X, and PM_{10}. Reduced solvent use is the largest factor. This is because current VOC end-of-pipe removal efficiency in GD is still low. Therefore, VOC emissions from solvent use could be greatly reduced by improving end-of-pipe removal efficiency. In fact, VOC emission controls on solvent use and industrial process source were particularly prioritized during the 13th FYP (2016-2020). If the VOCs end-of-pipe removal efficiencies achieve their control targets in the 13th FYP, VOCs emission reductions from solvent use will be 0.388 Tg and 0.257 Tg in PRD and NPRD, respectively, accounting for 43% and 38% of the total VOC emission reductions. Another source with large potential for emission is on-road mobile sources.

3 Summary and conclusions

This study provided the first long-term record of anthropogenic air pollutant emissions in GD, the three largest city clusters in China, and advanced our understating of air pollutant emissions and control measures in Guangdong. ~~In this study, we provide a detailed examination of anthropogenic emission of a wide variety of pollutants in GD from 2006 to 2015 using a technology-based methodology.~~ The emission trends and their spatial variation were validated by ground-based observations and satellite data. Anthropogenic emissions of most pollutants in GD generally saw downward trends over the 2006-2015 decade, with NH_3 and VOC emissions being the exceptions. In that decade, emissions of SO_2, PM_{10}, $PM_{2.5}$, and NO_X decreased by 48%, 22%, 16%, and 0.5%, respectively, despite the significant growth of economic and anthropogenic activity. The decoupling of anthropogenic emissions from economic and energy consumption growth means that emission regulations and control measures on power plants, industrial combustion, on-road mobile sources, and dust sources enacted over the past decade have alleviated emissions. By contrast, because of the absence of effective control measures, NH_3 emissions remained stable while VOC emissions steadily increased by 33% during 2006-2015.

Because of their ~~differences~~ discrepancies in ~~economic and industrial structure and in their implementation of~~emission control ~~measures~~progresses, PRD and NPRD showed different emission ~~trends~~evolutions. Overall, emissions of SO_2, NOx, $PM_{2.5}$, and PM_{10} in the PRD region showed significant downward trends during the period of 2006-2015, while emission from NPRD remained relatively stable due to the lax control measures. Specially, industrial combustion and industrial process presented opposite trends between PRD and NPRD. In PRD, emissions from industrial combustion declined consistently during 2006-2015 while in NPRD, these emissions continued to show an upward trend. Similar to the industrial combustion, emissions from industrial process source also declined in PRD but increased in NPRD during 2006-2015. As a result, emissions in NPRD were increasingly important in GD. The contribution of NPRD to SO_2 emissions in GD dramatically increased from 27% in 2006 to 48% in 2015. By the end of 2015, emissions of PM_{10} (56%), $PM_{2.5}$ (54%) and NH_3 (68%) in NPRD already accounted for more than 50% of the total emissions in GD. Particularly, industrial emission leaks from PRD to NPRD were observed in this study, which partly enhanced the significance of emissions in NPRD. Although the shift of industries reduced PRD emissions, it might not effectively alleviate the air quality in PRD since industrial emissions also have a certain influence on the RPD region, particularly in winter season (Yin et al., 2017). ~~In PRD, SO_2 and NO_X emissions exhibited a downward trend during 2006-2015, but in NPRD, these emissions grew before 2010. Most of the increased emissions were from power plants, industrial combustion, and non-road mobile sources, highly associated with the shift of industries and power plants from PRD to NPRD and the lack of stringent emission control measures in NPRD. The evolution of sources also showed differences between PRD and NPRD. In PRD, emissions from industrial combustion declined consistently during 2006-2015 owing to stringent control measures e.g. phasing out small boilers, installing flue gas desulphurization (FDG) equipment, and limiting the sulfur contents of fuel while in NPRD, these emissions continued to show an upward trend, even though some emissions from industrial combustion had been reduced by control measures. Similar to the situation with industrial combustion, emissions from industrial processes~~industrial process source ~~also declined in PRD but increased in NPRD during 2006-2015. The above-mentioned trends inevitably changed the relative contribution of different sources. Industrial combustion surpassed power plants as the largest contributor to SO_2 emissions in PRD and NPRD. For NO_X emissions, on-road mobile sources were was still the largest source in PRD, but in NPRD, the contribution of industrial combustion steadily increased and might replace power plants as the key~~

~~contributor in the future. As to VOC emissions, industrial solvent use was the largest contributor in PRD, but in NPRD, the contribution of non-industrial solvent use increased and became the largest contributor.~~

~~The historical emission inventory developed in this study not only helps to understand the emission evolution in GD, but also can help to develop robust control measures for the co-control of $PM_{2.5}$ and ozone.~~ In GD, future emission control works should focus on power plants, industrial combustion, and non-road mobile sources to further reduce emissions of SO_2, NO_X, and particulate matter. This can be achieved by technical innovations consisting of ultra-low emissions in power plants, a series of actions regarding boiler management, control of sulfur content in fuels, flue-gas desulfurization in industries, and special pollution controls for non-metallic minerals industries. In addition, control measures on ~~agricultural sources~~agricultural source, the largest contributors of NH_3 emissions, should be highlighted. As revealed by Yin et al. (2018), the chemical region in PRD might transit to an ammonia-rich region with ~~the~~ a decrease of SO_2 and NO_X emissions. In this case, a larger reduction in NH_3 emissions would be required to further decrease ambient $PM_{2.5}$ levels in GD. This is feasible since NH_3 emissions in GD still have great potential for further reductions. VOC emission reduction is another concern~~In order~~ to achieve co-control of $PM_{2.5}$ and ozone, future work should also focus on VOC emissions. In fact, the reduction of VOCs emissions is promising since stringent controls on solvent use was released in *Volatile organic compounds (VOCs) remediation and emission reduction work plan in Guangdong Province (2018-2020)* (GDEP, 2018). Apart from regulating solvent use, control measures for on-road mobile source~~s~~ should be enhanced to cover the growth of emissions induced by the increase of vehicle population.

The historical emission inventory developed in this study not only helps to understand the emission evolution in GD, but also~~ A long-term historical emission inventory~~ could also help to reveal the dominant causes of air-quality change~~. I~~ in PRD. ~~, t~~The annual averaged $PM_{2.5}$ concentrations in PRD showed a decrease in the 2006-2015~~ decade~~, from 58 μg/m^3 in 2007 to 34 μg/m^3 in 2017. By contrast, the 90th-percentile daily max 8-h average ozone showed a fluctuating increase, from 146 μg/m^3 in 2007 to 165 μg/m^3 in 2017. Our proposed a long-term historical inventory might be able to explain the change of $PM_{2.5}$ and ozone concentrations. As shown in Fig. 5, emissions of SO_2, NO_X, and $PM_{2.5}$ in PRD all steadily fell in this decade. Particularly, SO_2 and NO_X emissions, the two major precursors of $PM_{2.5}$ formations, decreased by 63% and 14%, respectively, during 2006-2015. This trend of precursor emissions

agreed with the declining trend of ambient $PM_{2.5}$ concentrations. VOC emissions in PRD showed a rising trend, increasing by 35% during 2006-2015. Ou et al. (~~2017~~2016) had revealed that most parts of PRD formed a VOC-limited region in autumn and winter. This suggests that the growing VOC emissions and the decreasing NO_X emissions might contribute to the growth of ozone concentrations in PRD. However, this does not mean that emission changes are the dominant cause. Using numerical simulations and the long-term historical emission inventory developed in this study, we can quantify the effectiveness of emission control measures and the impact of meteorological change on air quality in PRD. Consequently, the dominant ~~dominate~~ cause of the increase of ambient ozone concentrations and the downward trend of $PM_{2.5}$ concentrations in PRD in the recent decade can be identified.

This study applied a top-down method to develop long-term emission inventories. This method is feasible for two reasons. On one hand~~First~~, long-term emission inventories generally focus on emission trend, sectoral evolution and emission projection, rather than the high-resolution spatial distribution. ~~Seconds,~~On the other hand, obtaining long-term activity data for top-down method is more readily accessible. Therefore, most previous studies (Streets et al. 2006, Zhang et al, 2007, Lu et al. 2012, Zheng~~,~~ et al., 2018) also applied the same method to develop long-term emission inventories. However, this study can be further improved in following aspects.– First, a method based fuel consumption might underestimates marine emissions, due to the absence of passing ships that traveled through the study domain but did not call at any port in mainland China (Li et al., 2018). In China, the passing ships accounted for about 7% of the total fuel consumption. To fix the underestimation, the AIS data can be used to calibrate the total fuel consumption in GD. Second, the annual average vehicle kilometers traveled and the ratio of diesel vehicles to gasoline vehicles were obtained from a field survey, which only cover 8 cities and 111 roads in GD. These survey data cannot well represent each city in GD and thus might bring uncertainties in characterizing emission evolution of on-road mobile source. To better reflect the temporal pattern and differentiate emissions between different cities in GD, more field survey of vehicle types, fuel consumption, and emission standard should be conducted to better differentiate vehicle emissions between different cities in GD.

Authorship Contribution Statement

Zheng J. Y., and Huang Z. J. provided writing ideas with Shao M. support. Huang Z. J., Bian Y. H., and Ou J. M. carried them out. Huang Z. J., Zheng J. Y., and Ou J. M.

revised and polished the article. Bian Y. H., Zhong Z. M., Xiao X., Ye, X. and Wu Y. Q. developed the decadal emission inventories and contributed to discussions of results. Chen, L. F., Xu, Y. Q., Zhang, Z. W. and Yin, X. H. helped with verification of satellite data. All authors have made substantial contributions to the work reported in the manuscript.

Competing interests

The authors declare that they have no conflict of interest.

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Appendix A. Supplementary information

Attached please find supplementary information associated with this article.

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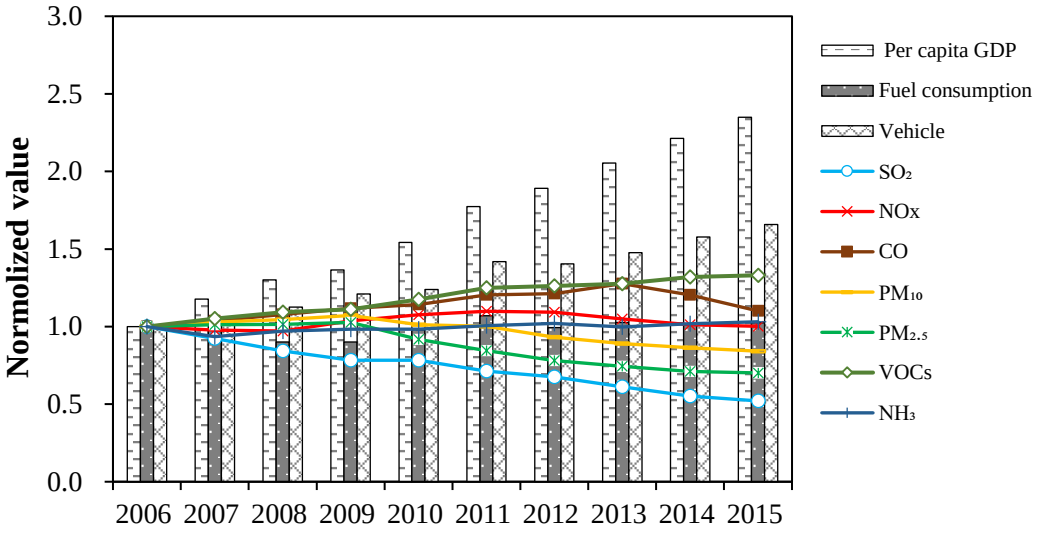
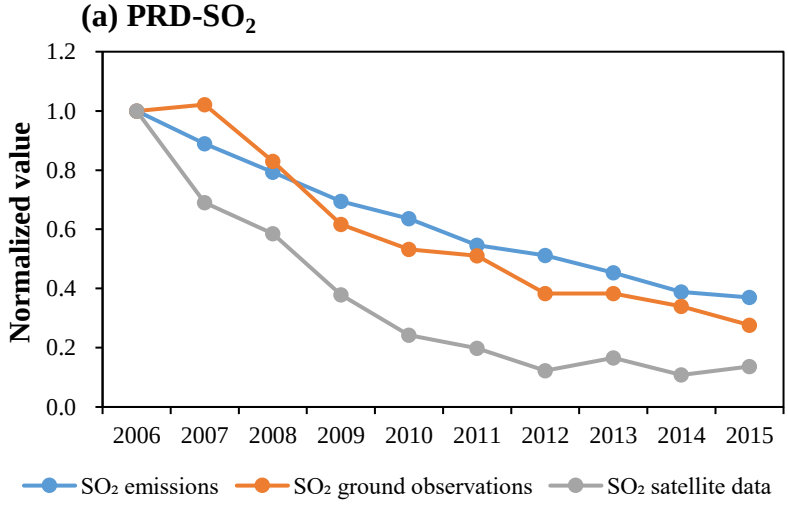
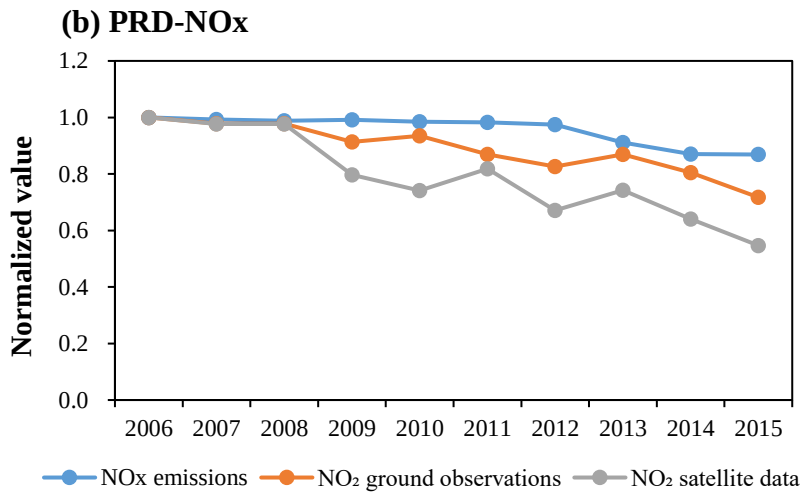
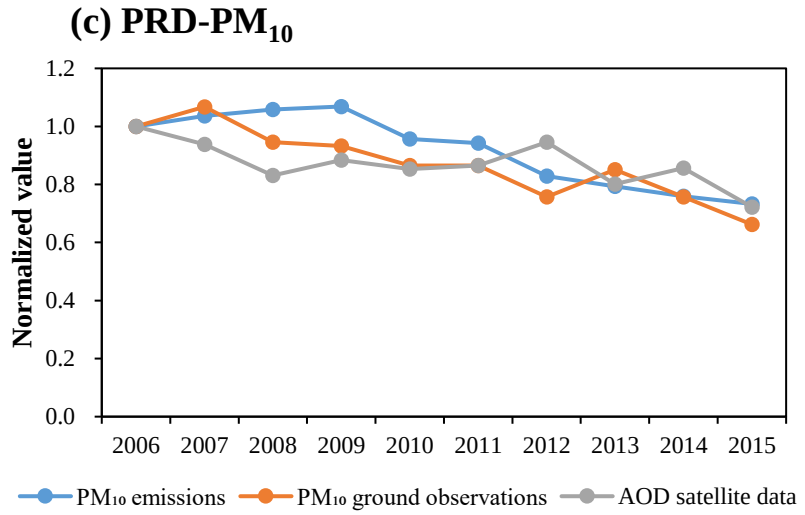


Figure 1. Trends in the air pollutant emissions, per capita GPD, fuel consumption and vehicle population in Guangdong Province from 2006 to 2015 (all of data are normalized to the year 2006).







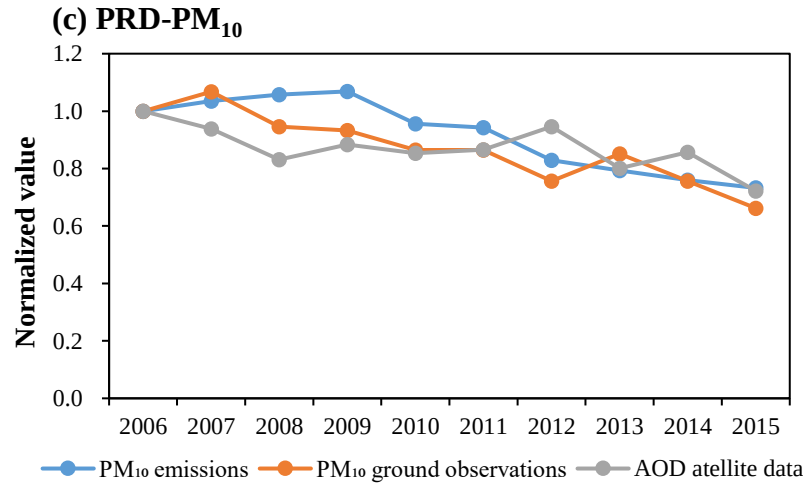
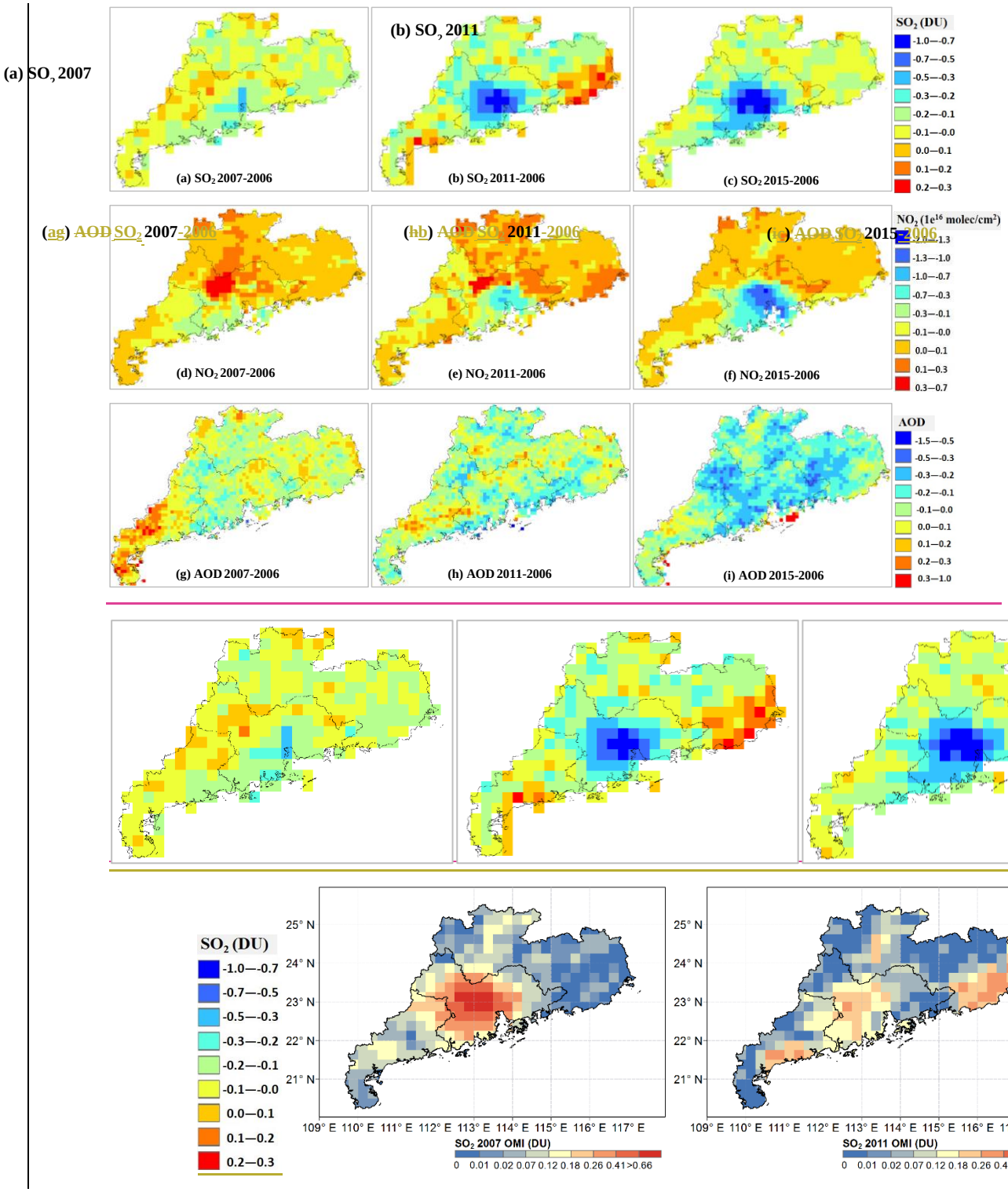
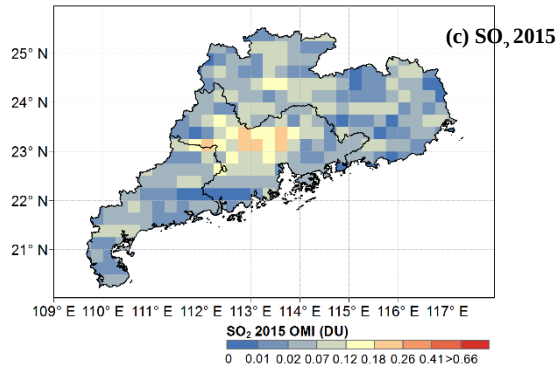
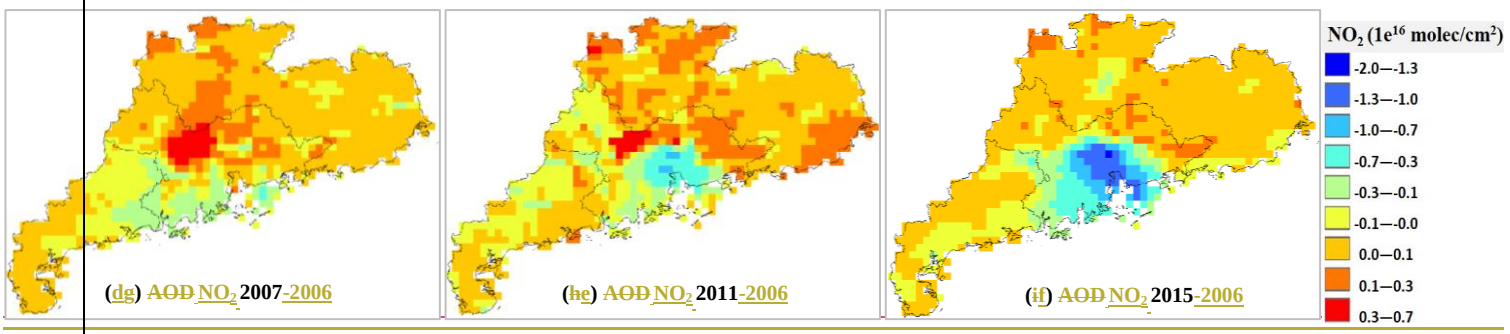
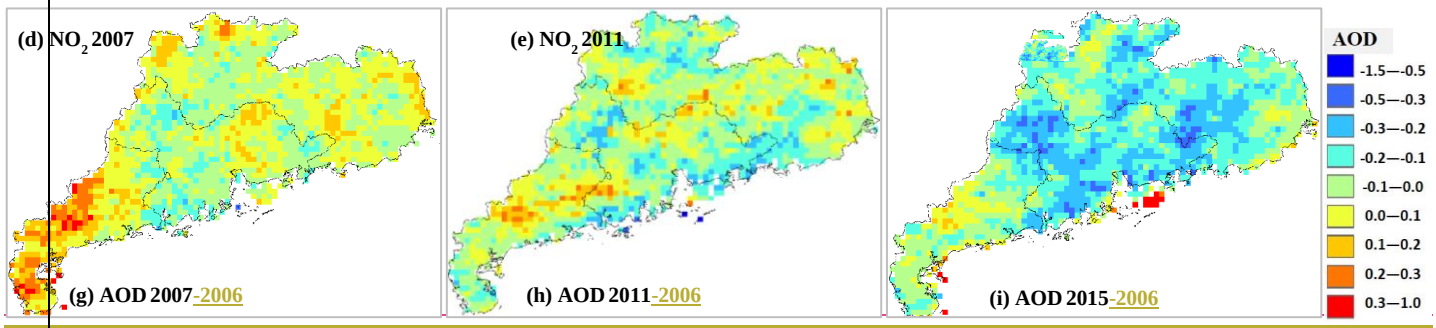


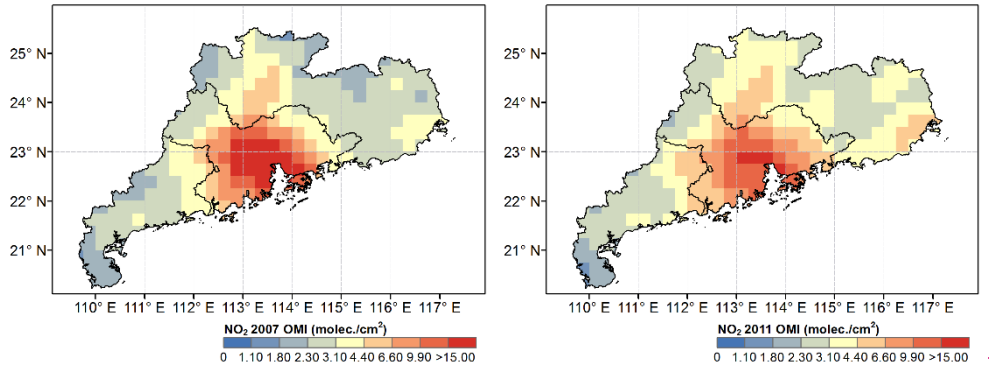
Figure 2. Comparison of emission trends with measurements in the PRD from 2006 to 2015. **(a)** SO_2 **(b)** NO_X **(c)** PM_{10} (all of data are normalized to the year 2006).

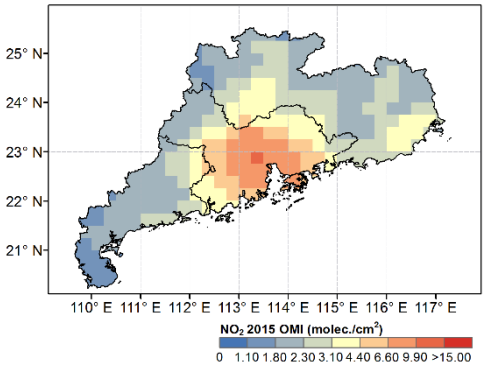

(a) SO, 2007
(b) SO, 2011
SO2 (DU)
-1.0—-0.7
-0.7—-0.5
-0.5—-0.3
-0.3—-0.2
-0.2—-0.1
-0.1—-0.0
0.0—0.1
0.1—0.2
0.2—0.3
(a) SO2 2007-2006
(b) SO2 2011-2006
(c) SO2 2015-2006
(ag) AOD SO2 2007-2006
(hb) AOD SO2 2011-2006
(ic) AOD SO2 2015-2006
NO2 (1e16 molec/cm2)
-1.3—-1.0
-1.0—-0.7
-0.7—-0.3
-0.3—-0.1
-0.1—-0.0
0.0—0.1
0.1—0.3
0.3—0.7
(d) NO2 2007-2006
(e) NO2 2011-2006
(f) NO2 2015-2006
AOD
-1.5—-0.5
-0.5—-0.3
-0.3—-0.2
-0.2—-0.1
-0.1—-0.0
0.0—0.1
0.1—0.2
0.2—0.3
0.3—1.0
(g) AOD 2007-2006
(h) AOD 2011-2006
(i) AOD 2015-2006
SO2 (DU)
-1.0—-0.7
-0.7—-0.5
-0.5—-0.3
-0.3—-0.2
-0.2—-0.1
-0.1—-0.0
0.0—0.1
0.1—0.2
0.2—0.3
25° N
24° N
23° N
22° N
21° N
109° E 110° E 111° E 112° E 113° E 114° E 115° E 116° E 117° E
SO2 2007 OMI (DU)
0 0.01 0.02 0.07 0.12 0.18 0.26 0.41>0.66
SO2 2011 OMI (DU)
0 0.01 0.02 0.07 0.12 0.18 0.26 0.41>0.66


(c) SO2 2015
25° N
24° N
23° N
22° N
21° N
109° E 110° E 111° E 112° E 113° E 114° E 115° E 116° E 117° E
SO2 2015 OMI (DU)
0 0.01 0.02 0.07 0.12 0.18 0.26 0.41 >0.66


NO2 (1e16 molec/cm2)
-2.0—-1.3
-1.3—-1.0
-1.0—-0.7
-0.7—-0.3
-0.3—-0.1
-0.1—-0.0
0.0—0.1
0.1—0.3
0.3—0.7
(dg) AOD NO2 2007-2006
(he) AOD NO2 2011-2006
(if) AOD NO2 2015-2006


(d) NO2 2007
(e) NO2 2011
AOD
-1.5—-0.5
-0.5—-0.3
-0.3—-0.2
-0.2—-0.1
-0.1—0.0
0.0—0.1
0.1—0.2
0.2—0.3
0.3—1.0
(g) AOD 2007-2006
(h) AOD 2011-2006
(i) AOD 2015-2006


25° N
24° N
23° N
22° N
21° N
110° E 111° E 112° E 113° E 114° E 115° E 116° E 117° E
NO2 2007 OMI (molec./cm2)
0 1.10 1.80 2.30 3.10 4.40 6.60 9.90 >15.00
NO2 2011 OMI (molec./cm2)
0 1.10 1.80 2.30 3.10 4.40 6.60 9.90 >15.00


25° N
24° N
23° N
22° N
21° N
110° E 111° E 112° E 113° E 114° E 115° E 116° E 117° E
NO2 2015 OMI (molec./cm2)
0 1.10 1.80 2.30 3.10 4.40 6.60 9.90 >15.00

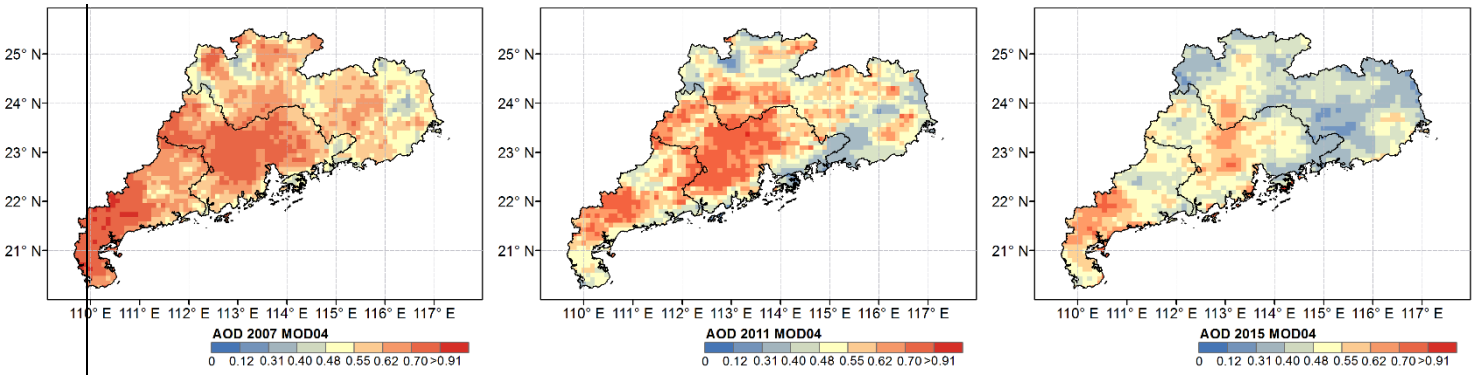
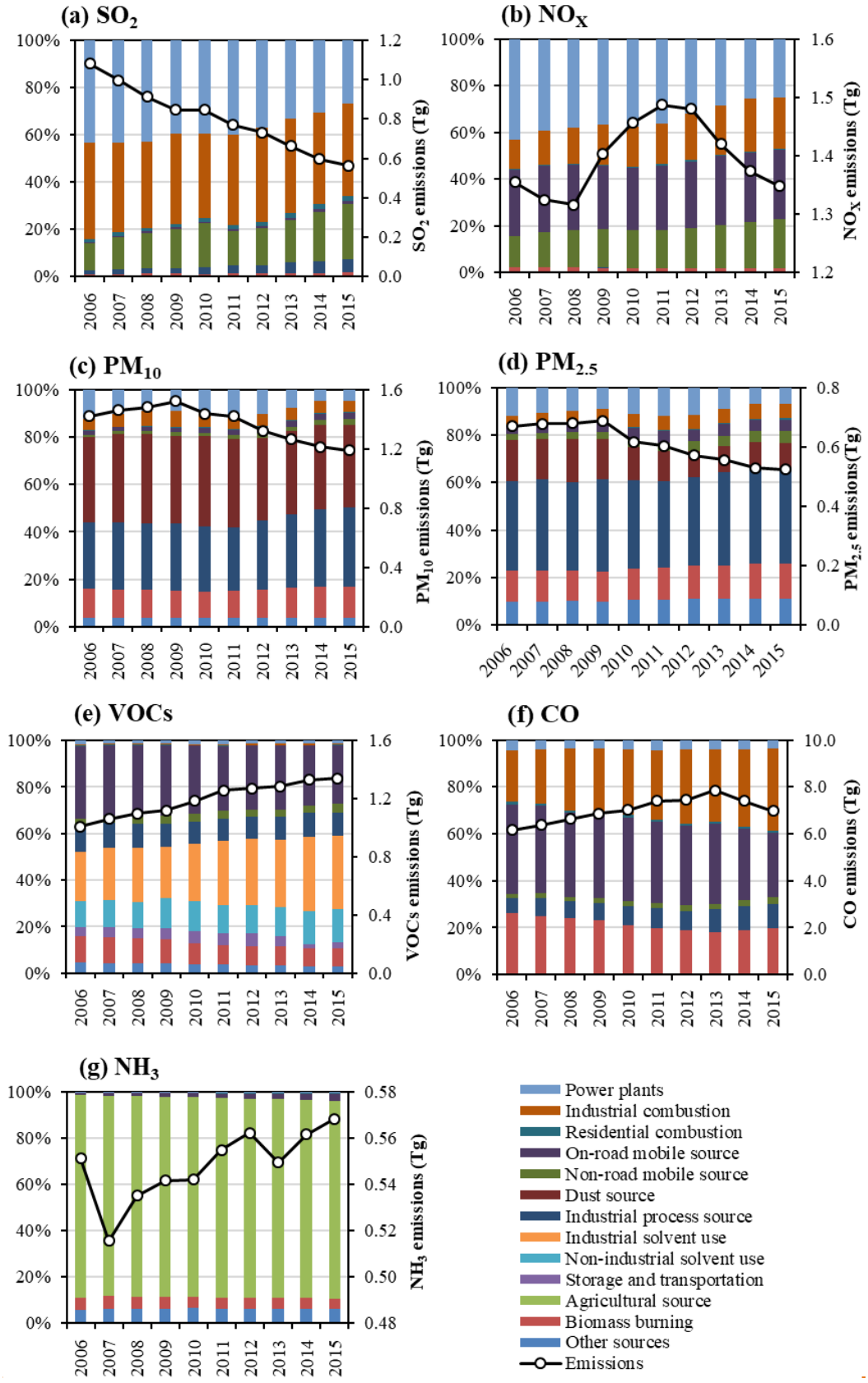


Figure 3. ~~The~~ Changes in spatial patterns of satellite observations over GD in 2007, 2011 and 2015 base on 2006 for **(a)-(c)** SO_2, **(d)-(f)** NO_2, and **(g)-(i)** AOD. The legends represent a gradual increase in emissions from ~~right~~top (blue) to ~~left~~bottom (red). Central zone inside the inner black line presents the PRD region, outside presents the NPRD region.


(a) SO2
(b) NOX
(c) PM10
(d) PM2.5
(e) VOCs
(f) CO
(g) NH3
SO2 emissions (Tg)
NOX emissions (Tg)
PM10 emissions (Tg)
PM2.5 emissions(Tg)
VOCs emissions (Tg)
CO emissions (Tg)
NH3 emissions (Tg)
Power plants
Industrial combustion
Residential combustion
On-road mobile source
Non-road mobile source
Dust source
Industrial process source
Industrial solvent use
Non-industrial solvent use
Storage and transportation
Agricultural source
Biomass burning
Other sources
Emissions

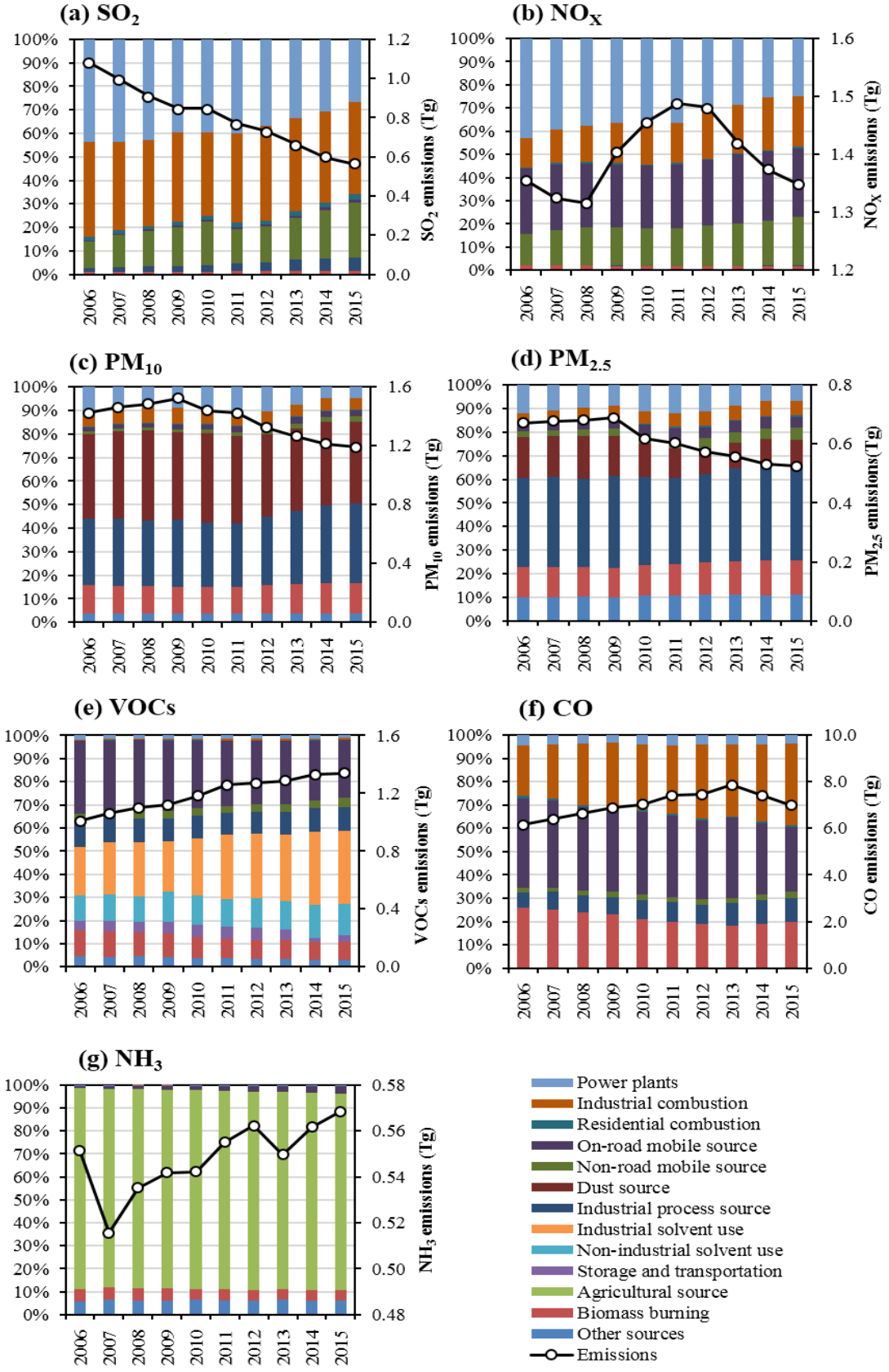
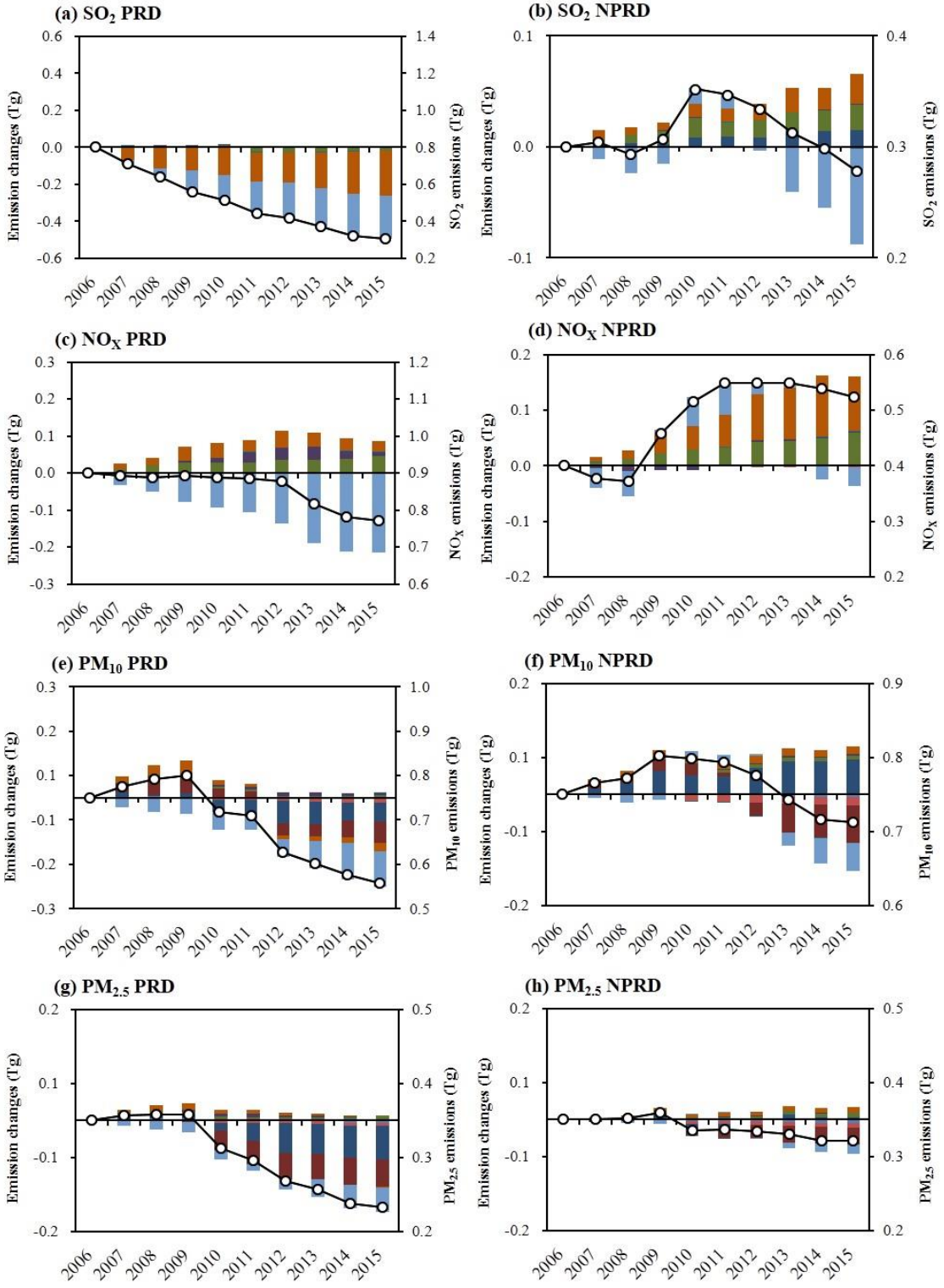


Figure 4. Source emission evolutions in Guangdong Province for **(a)** SO_2, **(b)** NO_X, **(c)** PM_{10}, ~~VOCs,~~ **(d)** $PM_{2.5}$, ~~PM_{10},~~ **(e)** ~~$PM_{2.5}$,~~ VOCs, **(f)** CO and **(g)** NH_3 from 2006 to 2015.


(a) SO2 PRD
(b) SO2 NPRD
(c) NOX PRD
(d) NOX NPRD
(e) PM10 PRD
(f) PM10 NPRD
(g) PM2.5 PRD
(h) PM2.5 NPRD
Emission changes (Tg)
SO2 emissions (Tg)
NOX emissions (Tg)
PM10 emissions (Tg)
PM2.5 emissions (Tg)
2006
2007
2008
2009
2010
2011
2012
2013
2014
2015

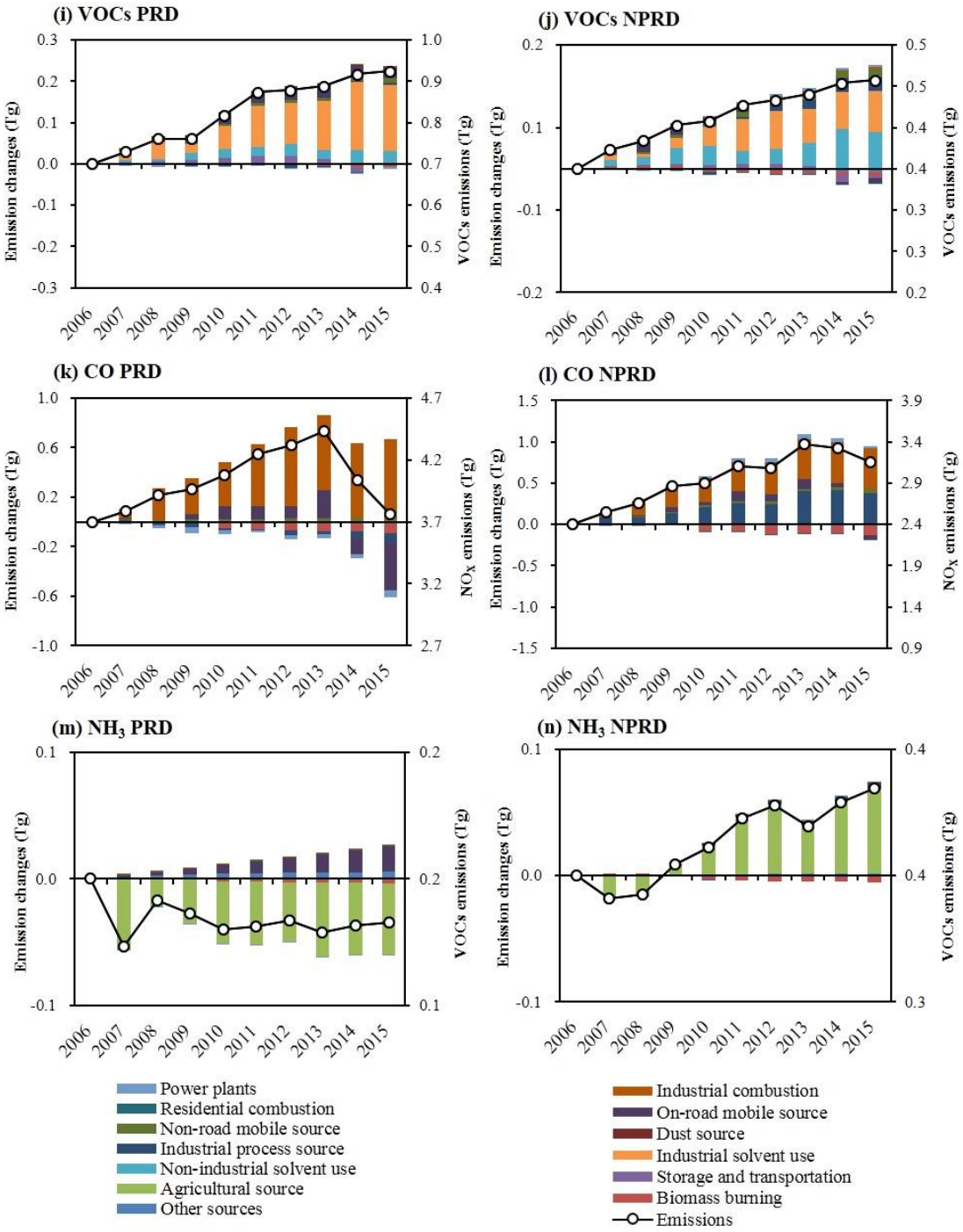


Figure 5. Emission evolutions by source in the PRD and NPRD for **(a)-(b)** SO_2, **(c)-(d)** NO_X, **(e)-(f)** PM_{10}, **(g)-(h)** $PM_{2.5}$, **(i)-(j)** VOCs, **(k)-(l)** CO and **(m)-(n)** NH_3 from 2006 to 2015. Source emissions in 2006 were subtracted from total emissions for each year to exhibit the additional emissions compared to 2006 (left axle). The total emissions by pollutant during 2006-2015 was also reflected in right axle.

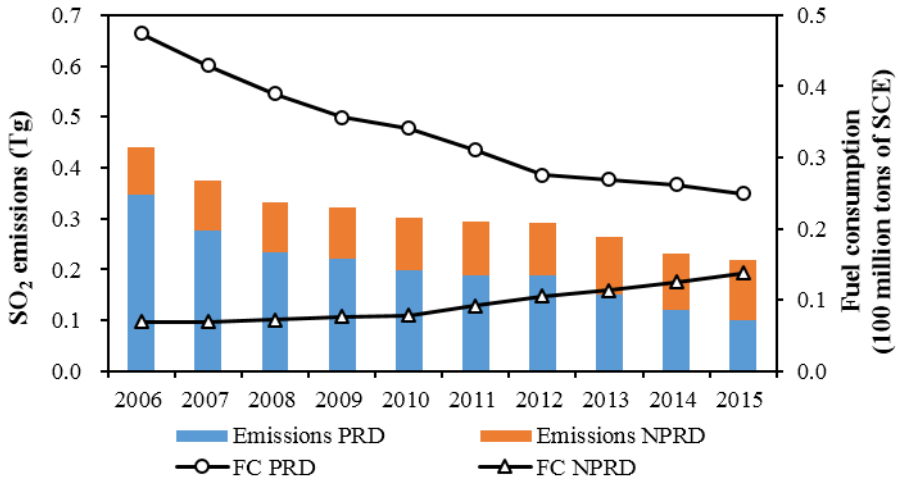


Figure 6. Trends of SO_2 emissions and fuel consumption from industrial combustion from 2006 to 2015. ~~(~~SCE~~:~~ represents standard coal equivalent; FC represents~~:~~ fuel consumption (i.e., coal, fuel oil, coke, and nature gas),~~).~~ and all of FC data are normalized to the year 2006.

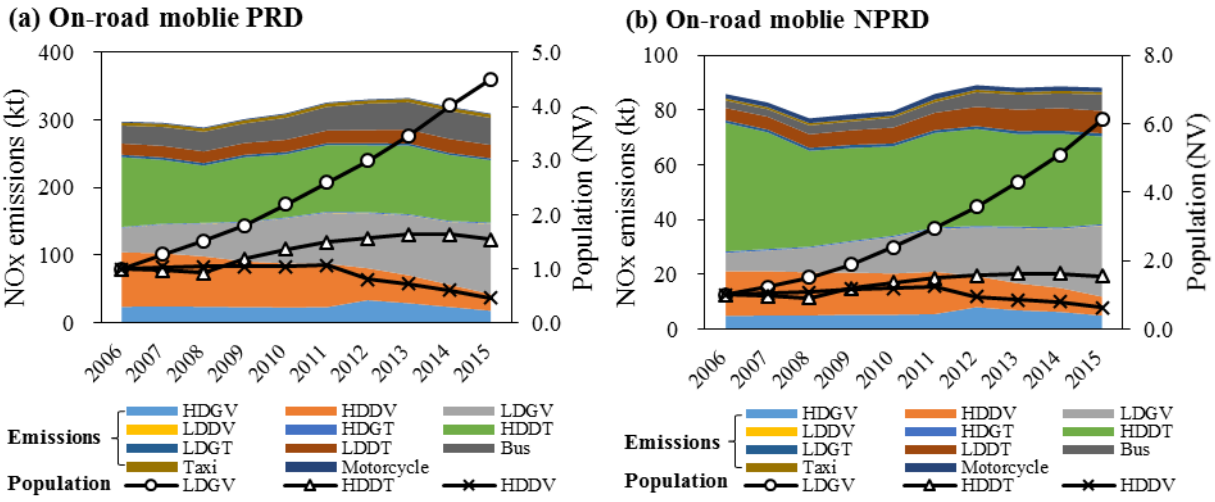


Figure 7. Trends of NO_X emissions from on-road mobile source and its activity data from 2006 to 2015 in the **(a)** PRD and **(b)** NPRD.

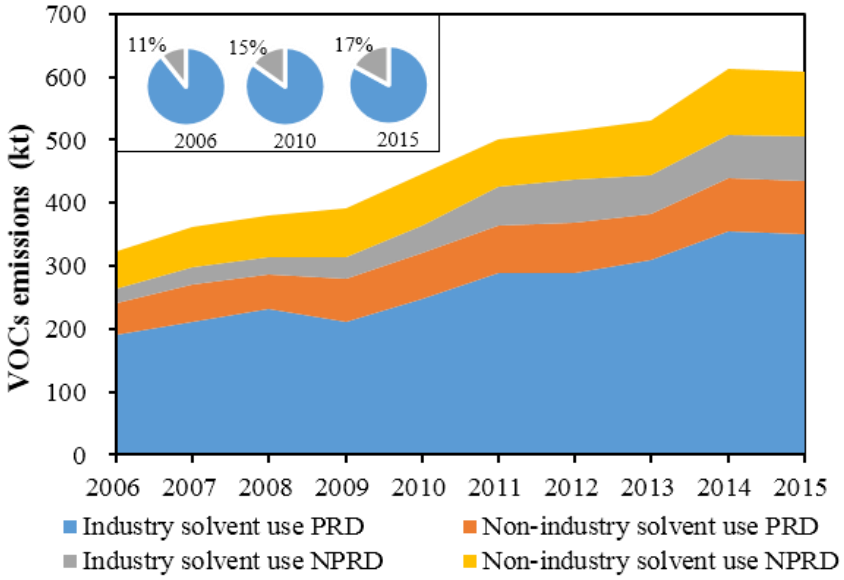


Figure 8. Trends of VOC emissions from solvent use (both industrial and non-industrial) and the contributions of industrial solvent use in the PRD and NPRD from 2006 to 2015.

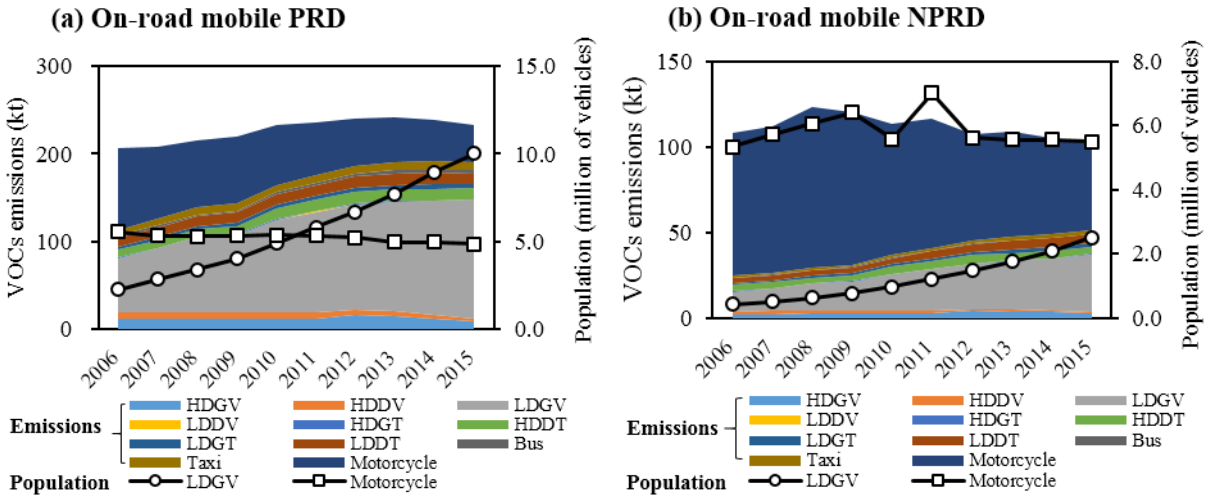


Figure 89. Trends of VOCs emissions from on-road mobile source and its activity data from 2006 to 2015 in the **(a)** PRD and **(b)** NPRD

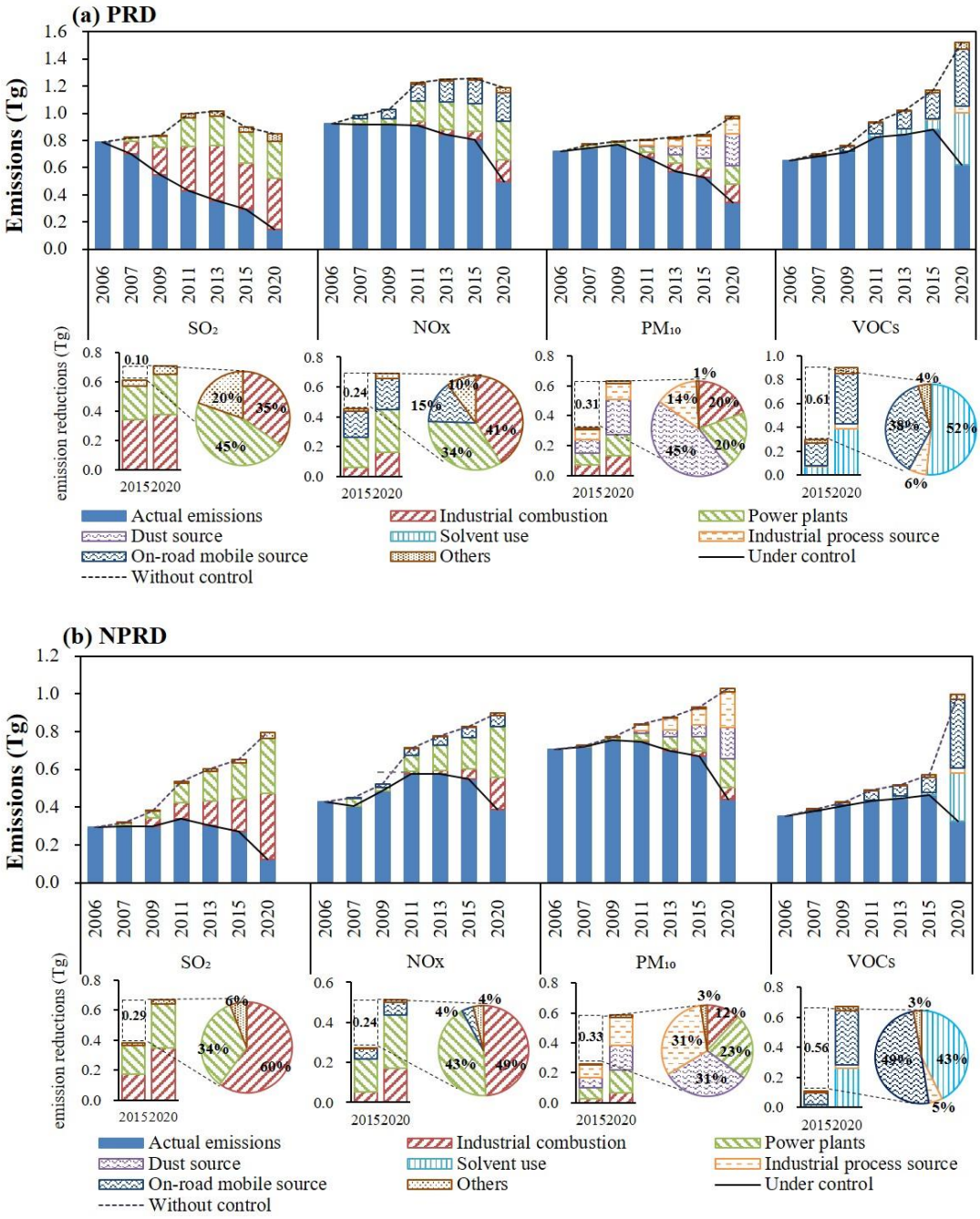


Figure 9 10. Control- and activity-driven emissions of SO_2, NO_X, PM_{10} and VOCs in 2007, 2009, 2011, 2013, and 2015 in addition to emission predictions in 2020 for the **(a)** PRD and **(b)** NPRD. Solvent use here includes industrial solvent use and non-industrial solvent use. The solid black line and the solid blue bar denotes the actual emissions we estimated under control (i.e., the results under the interaction of control- and activity-driven emissions; if control-driven emission dominated, the actual emissions would drop, and vice versa), and the dotted black line denotes the hypothetical emissions without control (i.e., activity-driven emission; if no new control measures were adopted after 2006). The non-solid chromatic bars and pies illustrate the emission reductions for multiple sources (i.e., control-driven emission). The dotted box

represents extra emission reductions in 2020 compared to 2015.

Table 1. Proportions of emissions in NPRD to the total emissions in GD.

Years	SO_2	NOx	CO	PM_{10}	$PM_{2.5}$	VOCs	NH_3
2006	27.1%	31.6%	39.4%	49.5%	50.3%	35.2%	64.0%
2007	29.7%	30.5%	40.2%	49.2%	49.8%	35.6%	66.7%
2008	31.3%	30.5%	40.4%	48.9%	49.8%	35.3%	64.5%
2009	35.3%	34.6%	41.9%	49.7%	50.4%	36.3%	65.9%
2010	40.7%	37.3%	41.5%	52.3%	51.9%	34.8%	67.1%
2011	44.1%	38.8%	42.2%	52.5%	50.6%	34.3%	67.7%
2012	44.7%	39.0%	41.6%	55.1%	52.0%	34.5%	67.7%
2013	46.0%	40.6%	43.2%	55.0%	52.0%	34.6%	67.7%
2014	48.7%	41.3%	45.1%	55.6%	53.5%	34.5%	68.1%
2015	48.2%	40.6%	46.9%	56.0%	54.0%	34.5%	68.2%