

Response to reviewer

We are very thankful to reviewer for his/her constructive criticisms and valuable comments, which were of great help in improving the quality of the manuscript. We have revised the manuscript accordingly and our detailed responses are shown below.

Referee 1

Response to major comments

1. More detailed descriptions on the development of ROM are needed. For example, what are the assumptions used? What are the uncertainties associated with emission factors? What are the limitations of the ROE that warrant future improvement? How is the ROE developed in this work different from that ITS work of Xiong et al., 2010 over Guangzhou and also by other people over China? What are the innovative features and uniqueness of this work in the context of existing work?

Reply:

Thanks for pointing out this. The very basic assumption of the ROE model was that calculating the street-level emissions from every on-road vehicle on the street segment by using the bottom-up method. The ROE model collected the traffic information from ITS for obtaining the traffic volume and vehicle fleet information from street network. After obtaining the number of each vehicle category, the ROE calculated the emissions of every vehicle category and sum it up for obtaining the total emission for each street segment. In this study, due to the vehicle fleet information (the emission standard information, vehicle category information and fuel type information) is not available from the data source we used, we used a uniform percentage, which is the average value of the Guangzhou city for each segment. This could be update if the street-level fleet information was available in the future. In order to make the model description clearer, we added section 2.1 “*model overview*” in page 3 line 24-30 and added more details in section 2.2.

The uncertainty analysis of emission factors had added in supplementary materials section S2. As shown in Figure S1, the uncertainty range of LDV was the largest for CO, HC and NO_x. Besides, the HDT has the largest uncertainty for PM_{2.5} and PM₁₀, whether it was petrol or diesel. However, more comprehensive emissions factor measurements should be done to analyze the uncertainty and improve the accuracy of the emission factors in the future.

In supplementary materials section S2, *“In order to estimate the uncertainty of emission factors, some results from previous studies had been collected and compared with the emission factors which were applied in this study. As shown in Figure S1, the uncertainties for each pollutant of petrol vehicles were much larger than the diesel vehicles. Overall, the uncertainties of LDVs for CO, HC, and NO_x were largest than other vehicle categories, whether it was petrol- or diesel-fueled. There were maximum 8.9 and 9.8 times higher for CO emission factors, 13.5 and 21.9 for HC, and 10.5 and 2.0 times for NO_x than that of the emission factors applied in this study. And for PM_{2.5} and PM₁₀, the HDTs had the largest uncertainty range, which were maximum 11.9 and 11.3 times higher for*

petrol HDT, and 3.5 and 16.1 times for higher diesel HDT, compared with the emission factors used in this study, respectively. However, it should be indicated more comprehensive emissions factor measurements should be done to analyze the uncertainty and improve the accuracy of the emission factors in the future.”

The limitations of the ROE model had discussed in the section 5 Discussion and conclusions part. In page 10, line 20-23, *“It is worth noting that the ROE model is highly dependent on the ITS traffic data. For economically underdeveloped cities, this aspect may pose a barrier against the use of the ROE model. In addition, China is promoting the CHINA VI emission standards for on-road vehicles. The ROE model only considers Pre-CHINA I to CHINA V currently. Thus, the model will be updated in the near future to include the CHINA VI emission standards.”* Moreover, another limitation is that in page 10, line 1-3, *“due to the lack of street-level vehicle fleet information, this study applied a city-level average uniform percentage for every street segment. This may increase the uncertainty of the inventory, but this aspect could be improved upon provided additional data become available in the future.”*

In page 3, line 17-18, the ITS work of Xiong et al. (2010) over Guangzhou aimed to establish a monitoring system which could obtain the traffic conditions automatically by using the traffic cameras, while the ROE model aimed to establish the high-resolution on-road emission inventories by using the traffic data from ITSs. The ROE model could be applied in other cities if the traffic data are available. If the official data from on-road cameras cannot be obtained, the ROE users can still able to use the data from amap.com (also called Gaode map), which is the same data source as this study used. The current version of crawler module of ROE was designed for obtaining the data from Gaode map. In page 4, line 31-32, *“The Gaode map traffic data are quite extensive as it covered over 40 cities in China so far (with most of them being China’s major cities).”* This could help the users apply the ROE model in other cities of China. We also had added some detail information about the data source in section 2.4.

The most innovative features and uniqueness of this work is that we used the real-time traffic data to establish the high-resolution emission inventories by bottom-up method, which could obtain the hourly or minutely on-road from some certain street segment. Besides, in page 4, line 32-37, *“Based on the GPS and mobile network information, details on vehicle speed and location are collected from the map user’s devices while using the map navigation on the road. This aspect saves a considerable amount of human labor and material resources with regard to traffic condition observations. These data are updated in real time and can be used through an open-access application programming interface (API), which remove the barrier of obtaining data. As the data can be updated in real time, the emission data can also be refreshed in real time.”*

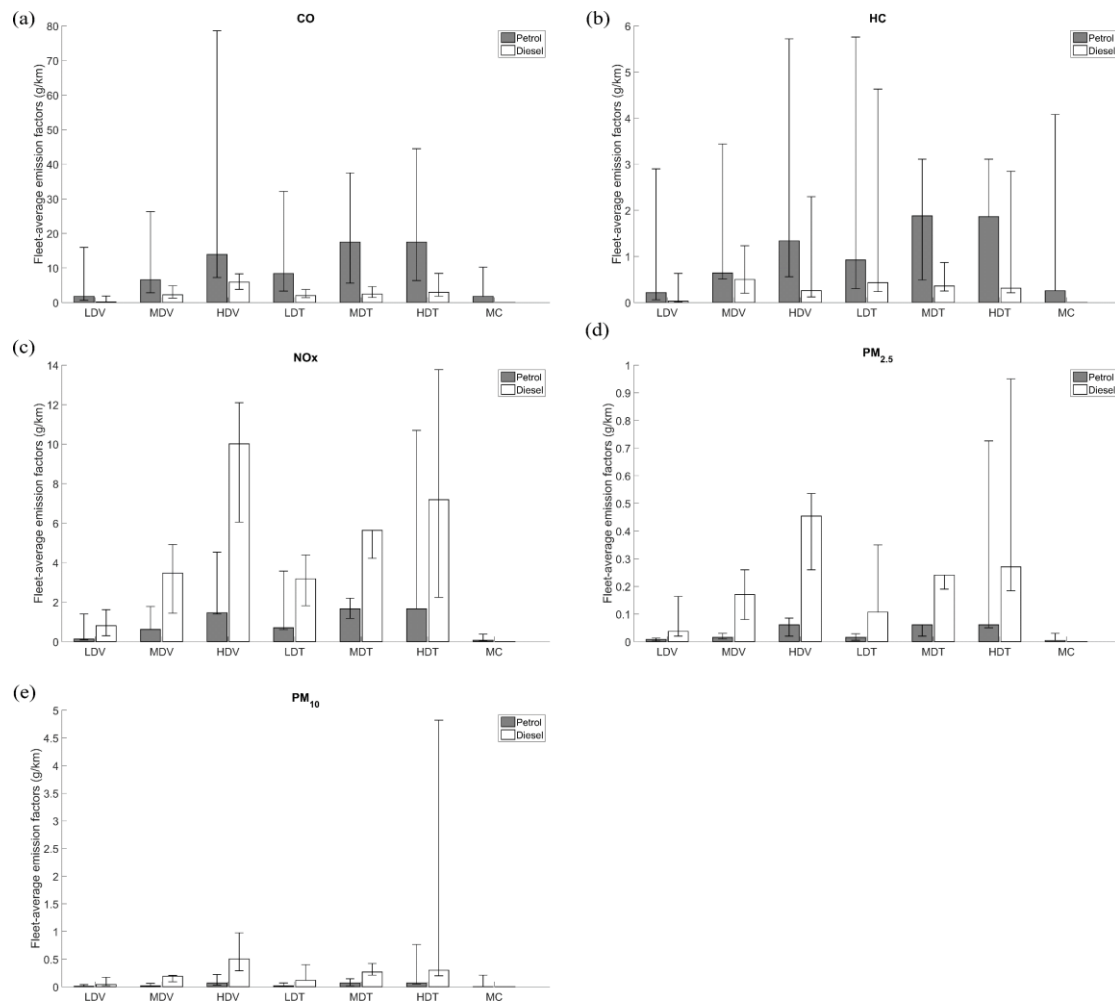


Figure S1. Fleet-average emission factors of (a) CO, (b) HC, (c) NO_x, (d) PM_{2.5}, and (e) PM₁₀ (the value of the histogram represented the fleet-average emission factors in the study, the range represented the emission factors from other literatures).

2. How were the urban background concentrations be derived for the MUNICH model? What are the uncertainties/measurement errors associated with the measurements of NO_x and O₃ concentrations?

Reply:

Thanks for your constructive comments. In page 6, line 18-21, “The observational data from TYX were used for modeling evaluation because TYX locates inside the simulation area which could be compared with the model results. In addition, YJ is located near but not within the simulation street network. The observational data from YJ could be used as the background concentration data for the modeling.”.

In page 6, line 12-17, “For modeling evaluation and background concentrations, the observational concentration data for NO₂ and O₃ were obtained from the Guangzhou environmental monitoring site network. NO₂ concentrations were measured with a chemiluminescence instrument (Model 42i, Thermo Scientific) and O₃ was measured by a UV photometric analyzer (Model 49i, Thermo

Scientific). The minimum detection limit (3S/N) of the analyzer was 0.4 ppbV (approximately 0.8 $\mu\text{g}/\text{m}^3$) for NO_2 and 1.0 ppbV (approximately 2.0 $\mu\text{g}/\text{m}^3$) for O_3 . The total measurement uncertainty of these two instruments was estimated to be approximately 5% (Zhang et al., 2014).”

3. What are the model evaluation criteria (e.g., threshold values for the statistical metrics) used to judge the model performance? How are those statistics compared with other model evaluation for simulated NO_x and O_3 concentrations reported in the literature?

Reply:

Thank you for providing these important points. We had added the recommended values from Ministry of Ecology and Environment of the People’s Republic of China technical guide in section 4.2.1. In page 8, line 23-29, “*The NMB, NME, and CORR values of NO_2 and O_3 in this study were within the recommended ranges in the MEP Technical Guide for Air Quality Model Selection (MEP, 2012). These recommended values were $-40\% < \text{NMB} < 50\%$, $\text{NME} < 80\%$ and $\text{R}^2 > 0.3$ for NO_2 , and $-15\% < \text{NMB} < 15\%$, $\text{NME} < 35\%$, and $\text{R}^2 > 0.4$ for O_3 . Additionally, the values obtained in this study fell within the range of those obtained by other modeling studies in Guangzhou; the NMB, NME and RMSE values for simulated urban NO_2 in Guangzhou were -27.5% to -6% , 29.2% to 53.0% and 16 to 37.3 , respectively, and the corresponding values for O_3 were and -21.2% to 20.0% , 38.2% to 98% , 9.4 to 40.1 (Che et al., 2011; Fan et al., 2015; Wang et al., 2016). ”*

4. More in-depth discussions of emission modeling results are needed, e.g., the discussion for Table 2 that compares the three emission datasets. Why are NO_x emissions estimated in this work higher than those from the other two? Why are the differences in gaseous emissions larger than those in $\text{PM}_{2.5}/\text{PM}_{10}$ emissions among the three inventories? Also, it would be useful to provide a brief description on the basis of MEIC-2016 and PRD-2015 inventories, which may help understand the differences across the three inventories.

Reply:

Thank you for your helpful suggestion. We had added some details about the MEIC and PRD inventories. In page 6, line 29-34, “*These two emission inventories used the top-down method to establish on-road emission inventories. Unlike the bottom-up method used in this study, these two inventories first calculated the total emissions based on the VKT data of vehicle categories. In the MEIC inventory, the total number of vehicles was obtained from the relationship between total vehicle ownership and economic development (Zheng et al., 2014), while the PRD inventory acquired information on the number of vehicles from the city-level statistics Yearbook. Then, the spatial distribution of these two inventories was established based on the road network density.*”

According to the uncertainty analysis of emission factors, the uncertainty of $\text{PM}_{2.5}$ and PM_{10} is much smaller than the gaseous emissions, leading the large difference of gaseous emissions.

As for NO_x emissions, we thought that the higher NO_x estimate could be due to our updated LPG bus emission factor based on the local study (Zhang et al., 2013). The NO_x emission factor of an LPG-fueled bus is 1.7 times that of a diesel-fueled bus. This maybe one of the reasons leading the

higher NO_x estimate. From figure 9, the results showed that the NO_x emission distribution of bus in urban and suburban area was 20.5% and 10.8%.

We had added this content in page 6, line 38 to page 7, line 4, *“the difference of PM_{2.5} and PM₁₀ amount was smaller than other gaseous emissions among different inventories. This was because that the uncertainty of particulate matter emission factors was lower than the corresponding values of the other emissions, which led to the large difference for the gaseous emissions and the smaller differences for PM_{2.5} and PM₁₀. For NO_x emissions, however, this study showed a higher NO_x estimate than that in the other two inventories. One of the reasons for the higher NO_x estimate may be the application of the updated LPG bus emission factors in this study. Based on a previous local emission factor study, the NO_x emission factor of an LPG-fueled bus is 1.7 times that of a diesel-fueled bus in Guangzhou (Zhang et al., 2013). The results in Figure 8 show that the NO_x emissions distribution attributable to buses in urban and suburban areas were 20.5% and 10.8% of the total NO_x, respectively, showing that the LPG-fueled buses may be responsible for higher NO_x estimates in this study compared to those in the other two inventories.”*

5. More in-depth discussions of air quality modeling results are needed, e.g., discussion for Figure 11, why does the model give larger NO_x overpredictions of NO_x and O₃ underpredictions on May 2? It looks that the model tends to overpredict O₃ mixing ratios at night, could you please explain the likely causes for this overprediction? Does this error come from the overestimated urban background O₃, or underpredicted NO_x titration (as the model tends to underpredict NO_x mixing ratios at night) or both? Is it possible to set up some sensitivity simulations to verify/pin-point your speculated causes for the model bias?

Reply:

Thanks for bringing up this meaningful question. Several sensitivity cases had been further carried out to figure out what affected the simulation results in supplementary materials section S3. We had compared the observational and background concentrations of NO₂ and O₃ and analyze the results from the model sensitivity cases (Figure S2 to S5 in supplementary materials). We thought that NO₂ overprediction during the daytime was caused by the overestimation of background concentrations. Due to the only consideration of on-road emission and overprediction of NO₂, the VOCs-to-NO_x emission ratio was underestimated. Meanwhile, the simulated street network was in the VOC-sensitive regime. Thus, the O₃ concentrations were underpredicted during the daytime, especially on May 2nd. For the nighttime O₃ overprediction, both overestimated background concentrations and underestimation of NO titration were the reasons for higher predicted nighttime O₃. As shown by the sensitivity case results shown (Figure S2 to S5 in supplementary materials), the underestimated NO titration had also led to a lower simulated NO₂ concentrations at night.

In supplementary materials section S3, *“In order to figure out what affected the simulation results, some modeling sensitivity cases had been carried out in this study. As the diurnal variations shown in Figure S2, NO₂ concentrations were underestimated, while the O₃ were overestimated at night. However, the daytime model predictions were opposite to the nighttime prediction with NO₂ overprediction and O₃ underprediction. Combined with the observation and background*

concentrations data in Figure S3, the daytime NO_2 and nighttime O_3 overprediction maybe be caused by the overestimation of background concentrations.

To evaluate the impact of background concentrations on the simulation results, the no-emission sensitivity case was carried out for enhancing the background effect. This case was run without any emissions and results were shown in the Figure S4. For NO_2 , daytime concentrations were still overestimated compared with the observational data, especially in May 2nd, with maximum 23.7% daytime overestimation during the simulation. Meanwhile, nighttime O_3 were also overpredicted in this case, showing the nighttime O_3 overestimation was mostly due to the overestimation of background O_3 concentration. In contrast, since only vehicle emissions were considered, the underestimation of daytime O_3 should relate to the lack of other sections of emission in the simulation street network.

Besides, another case was carried out to evaluate the nighttime NO_x titration. Since NO_2 were underestimated at night but overestimated during daytime, NO_2 titration was not underpredicted and probably overpredicted at night. The overestimation of nighttime should be due to the underestimation of NO concentration. Thus, the double-background- NO case was carried out with double background NO concentrations to evaluate the NO titration. As the background NO concentration increased, nighttime NO_2 had increased which offset the underestimation concentration in base case. Meanwhile, O_3 concentrations had decreased due to the enhancement of NO titration. These results had shown that on the one hand, the overestimation of background O_3 concentration could lead to the O_3 overprediction at night. On the other hand, the underestimated NO titration was also a reason for the nighttime overprediction of O_3 concentrations.”

We had summarized up the results from those sensitivity case. The conclusions were shown in page 8, line 16-19, “Generally, the overestimated background concentrations of NO_2 and O_3 caused the reason for the overprediction of daytime NO_2 and nighttime O_3 concentrations, respectively. Also, the underestimated NO titration was the other main reason for overprediction of O_3 and underprediction of NO_2 concentrations at night. Due to the only consideration of on-road emission in the simulation street network, daytime O_3 concentrations were underpredicted in the results.”.

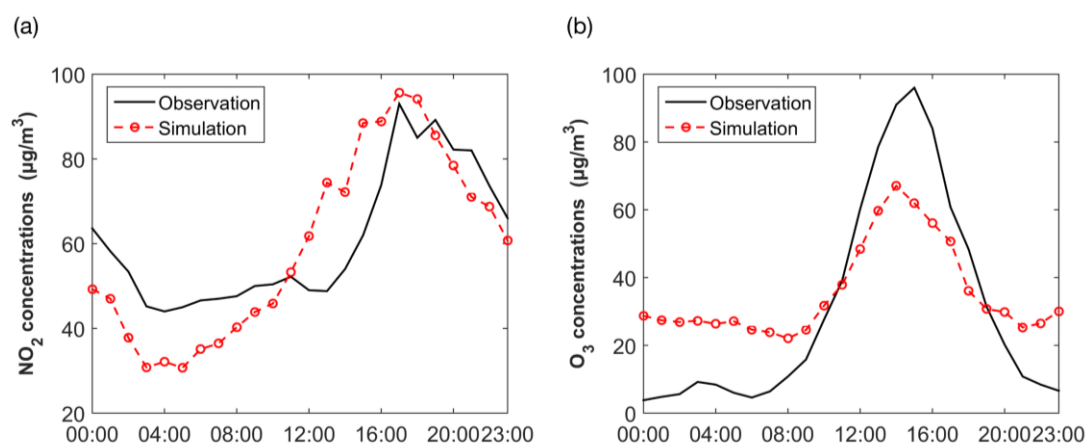


Figure S2. Diurnal variations of (a) NO_2 and (b) O_3 during the simulation period. (black solid line: observation; red dashed line: simulation).

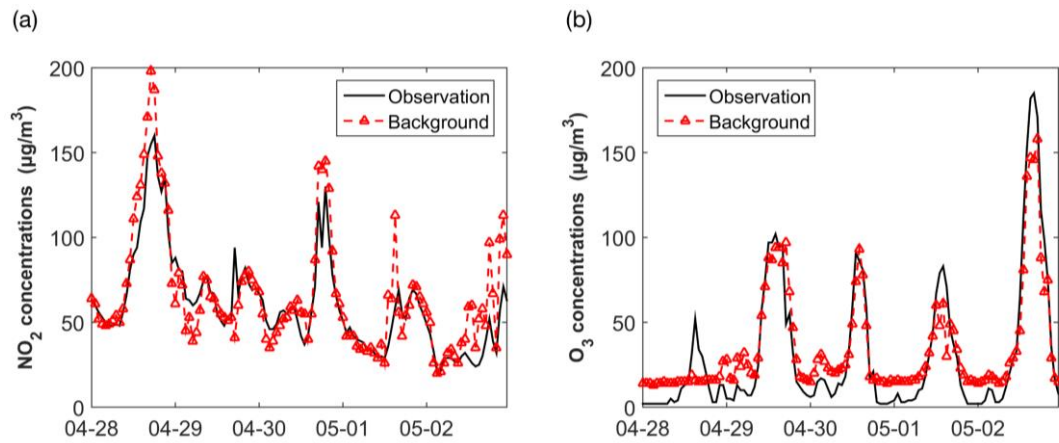


Figure S3. Time series of observational and background concentrations for (a) NO₂ and (b) O₃ during the simulation period (black solid line: observation; red dashed line: background).

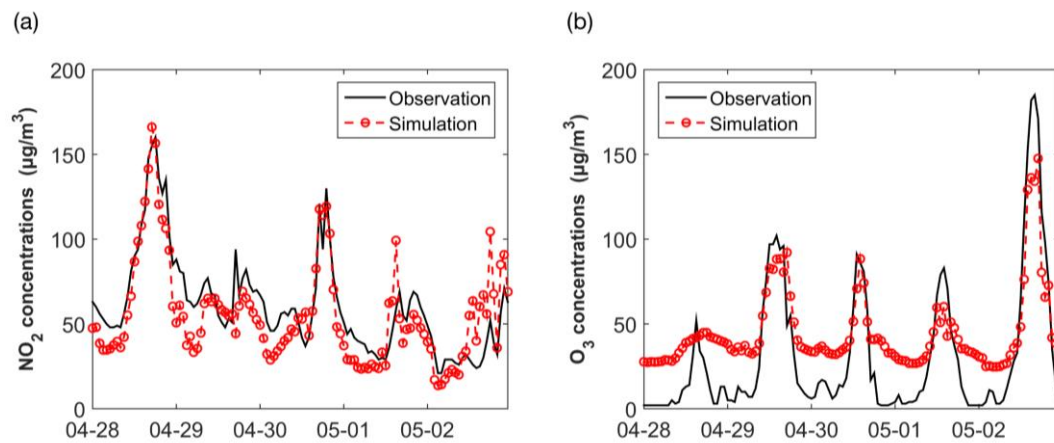


Figure S4. Time series of (a) NO₂ and (b) O₃ during the simulation period in no-emission sensitivity case. (black solid line: observation; red solid line: background).

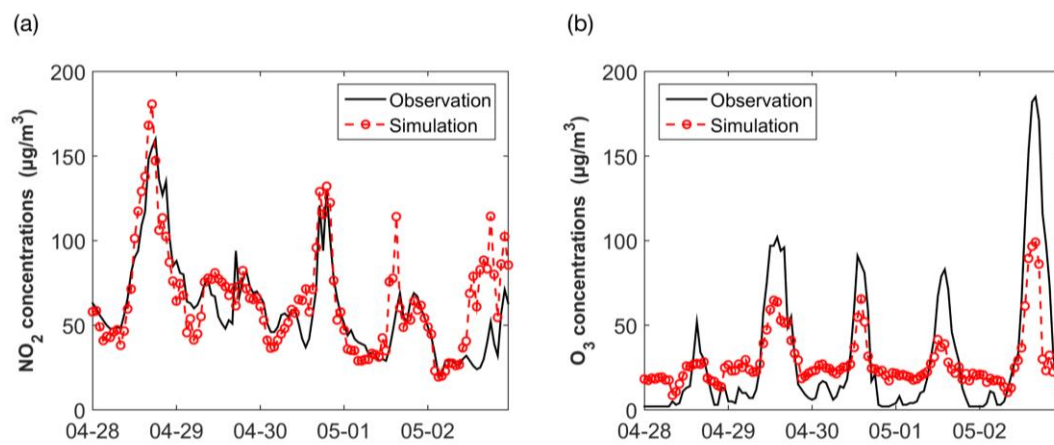


Figure S5. Time series of (a) NO₂ and (b) O₃ during the simulation period in double-background-NO case. (black solid line: observation; red solid line: background).

6. In the conclusion section, it would be useful to discuss the limitations of this work and future areas of improvement for both ROE and the MUNICH modeling work.

Reply:

Thank you for the reminder. The limitations of the ROE modeling work are listed as below:

In page 10, line 1-3, *“due to the lack of street-level vehicle fleet information, this study applied a city-level average uniform percentage for every street segment. This may increase the uncertainty of the inventory, but this aspect could be improved upon provided additional data become available in the future.”*

In page 10, line 21-23, *“In addition, China is promoting the CHINA VI emission standards for on-road vehicles. The ROE model only considers Pre-CHINA I to CHINA V currently. Thus, the model will be updated in the near future to include the CHINA VI emission standards.”*

As for MUNICH modeling work, in page 10, line 13-18, *“In this study, only 31 main street segments were selected to study the impact of a holiday on air quality in a certain urban area of Guangzhou. Additional investigations are required to understand the variations in street-level air quality in urban or suburban area of a megacity. The results of the ROE model showed that the suburban town centers of Guangzhou served as emission hotspots. These areas had relatively higher emissions than the other suburban areas and less stringent control policies than the urban area, which suffers from more serious air quality problems.”*

7. The paper contains some grammatical errors, typos, and undefined acronyms. Additional references should be cited in several places. It would benefit from an editorial review by an English-speaker.

Reply:

We had carefully rechecked the language problem and added more references for making the paper clearer.

Response to specific comments

1. Page 1, line 16, does “Mg/a” mean “Mg/year”? If so, I would suggest to use “Mg/yr”. A similar question for “Mg/a” in page 5, line 36.

Reply:

Agreed. We had revised it in page 1, line 16 and page 9 line 34.

2. Page 1, line 32, replace “has seen” by “has experienced”.

Reply:

Agreed. We had revised it in page 1, line 32.

3. Page 1, lines 33-34, “Zheng et al., 2009b” should be cited after “Zheng et al., 2009a”

Reply:

Agreed. We had revised it in page 1, line 33-34.

4. Page 1, lines 35-36, are those percentages concentrations or emissions of CO, NO_x, and HC? Please clarify.

Reply:

These percentages were the emissions of CO, NO_x and HC. We had revised it in page 1, line 35-36.

5. Page 1, line 37, “Numerical emission modeling” (rather than “Numerical air quality modeling”) is “an effective method to estimate on-road vehicle emissions”, please correct this.

Reply:

In here, what we want to express is that the on-road emission inventory can be used as input data for the numerical air quality models which were applied to estimate the impact of on-road emissions on the air quality. We thought it’s more suitable to use “numerical air quality modeling” here instead of “Numerical emission modeling”. To avoid the misunderstanding, we had rewritten the sentence by “*Reliable on-road emission inventories can be used as input data for the numerical air quality models which were applied to estimate the impact of on-road emissions on the urban air quality.*” in page 1, line 38-39.

6. Page 2, line 13, replace “leads” by “lead”

Reply:

Agreed. We had revised it in page2, line 13

7. Page 2, line 18, replace “heavy reliance” by “strong dependence”

Reply:

Agreed. We had revised it in page2, line 17.

8. Page 2, line 29, replace “observation” by “observational”

Reply:

Agreed. We had revised it in page2, line 29.

9. Page 3, lines 16-20, how is the ROE developed in this work different from that ITS work of Xiong et al., 2010 over Guang Zhou and also by other people over China? What are the innovative features and uniqueness of this work in the context of existing work?

Reply:

Thanks for bringing up this meaningful question. The ITS work of Xiong et al. (2010) over Guangzhou aimed to establish a monitoring system which could obtain the traffic conditions automatically by using the traffic cameras, while the ROE model aimed to establish the high-resolution on-road emission inventories by using the traffic data from ITSs. The ROE model could be applied in other cities if the traffic data are available. If the official data from on-road cameras cannot be obtained, the ROE users can still be able to use the data from amap.com (also called Gaode map), which is the same data source as this study used. The current version of crawler module of ROE was designed for obtaining the data from Gaode map. In page 4, line 31-32, "*The Gaode map traffic data are quite extensive as it covered over 40 cities in China so far (with most of them being China's major cities).*" This could help the users apply the ROE model in other cities of China. We also had added some detail information about the data source in section 2.4.

The most innovative features and uniqueness of this work is that we used the real-time traffic data to establish the high-resolution emission inventories by bottom-up method, which could obtain the hourly or minutely on-road from some certain street segment. Besides, in page 4, line 32-37, "*Based on the GPS and mobile network information, details on vehicle speed and location are collected from the map user's devices while using the map navigation on the road. This aspect saves a considerable amount of human labor and material resources with regard to traffic condition observations. These data are updated in real time and can be used through an open-access application programming interface (API), which remove the barrier of obtaining data. As the data can be updated in real time, the emission data can also be refreshed in real time.*"

10. Page 4, please discuss uncertainties associated with emission factors.

Reply:

Thanks for pointing out this. We had discussed the uncertainties of emission factors in the supplementary materials section S2. As shown in Figure S1, the uncertainty range of LDV was the largest for CO, HC and NO_x. Besides, the HDT has the largest uncertainty for PM_{2.5} and PM₁₀, whether it was petrol or diesel. However, more comprehensive emissions factor measurements should be done to analyze the uncertainty and improve the accuracy of the emission factors in the future.

In supplementary materials section S2, "*In order to estimate the uncertainty of emission factors, some results from previous studies had been collected and compared with the emission factors which were applied in this study. As shown in Figure S1, the uncertainties for each pollutant of petrol vehicles were much larger than the diesel vehicles. Overall, the uncertainties of LDVs for CO, HC, and NO_x were largest than other vehicle categories, whether it was petrol- or diesel-fueled. There were maximum 8.9 and 9.8 times higher for CO emission factors, 13.5 and 21.9 for HC, and 10.5 and 2.0 times for NO_x than that of the emission factors applied in this study. And for PM_{2.5} and PM₁₀, the HDTs had the largest uncertainty range, which were maximum 11.9 and 11.3 times higher for petrol HDT, and 3.5 and 16.1 times for higher diesel HDT, compared with the emission factors used in this study, respectively. However, it should be indicated more comprehensive emissions factor measurements should be done to analyze the uncertainty and improve the accuracy of the emission factors in the future*"

11. Page 4, lines 21-27, please explain why the Underwood volume calculation model was selected. This method was developed about 60-year ago, is it still better than more recent methods?

Reply:

In the work of Jing et al. (2016), they tested several speed-flow models and found the Underwood model had best goodness of fit among these models in China megacity. Thus, we applied this speed-flow model in our work. In page 4, line 41-43, we had clarified that *“In this study, the Underwood volume calculation model was used to retrieve the information on volume because of its history of successful application in China (Jing et al., 2016).”*

12. Page 4, line 41, replace “includes” by “include”

Reply:

Agreed. We had rewritten the sentence by *“The main traffic control policies in urban areas are as follows:”* in page 5, line 17-18.

13. Page 5, line 1, “7:00-22:00” covers not only daytime but also nighttime, please clarify.

Reply:

Agreed. We had revised it in page 5, line 20.

14. Page 5, after Section 2.4, it would be useful to discuss any limitation and uncertainties associated with the ROE model. Page 2, lines 41-44 indicated some issues with the ITS methods, are those issues applicable to the ROE developed for Guangzhou area? Also, what specific traffic information and emission factors will be needed if one applies the approaches/modules used in the ROE model to estimate real-time traffic emissions in other cities?

Reply:

The limitations of the ROE model had discussed in the section 5 Discussion and conclusions part. In page 10, line 20-23, *“It is worth noting that the ROE model is highly dependent on the ITS traffic data. For economically underdeveloped cities, this aspect may pose a barrier against the use of the ROE model. In addition, China is promoting the CHINA VI emission standards for on-road vehicles. The ROE model only considers Pre-CHINA I to CHINA V currently. Thus, the model will be updated in the near future to include the CHINA VI emission standards.”* Moreover, another limitation is that in page 10, line 1-3, *“due to the lack of street-level vehicle fleet information, this study applied a city-level average uniform percentage for every street segment. This may increase the uncertainty of the inventory, but this aspect could be improved upon provided additional data become available in the future.”*

The ROE model applied the bottom-up method to establish the on-road emission inventory. To apply the ROE model in other cities, in the best case, as it introduced in section 2.1, *“First, the ROE model collects the real-time traffic information to obtain the traffic volume for each street segment from*

the ITS. Then, according to the vehicle fleet information, the ROE model calculates the number of vehicles for each vehicle category on each street segment (if available, these data could be obtained from the ITS and need not be calculated by model). Thereafter, the ROE model calculates the emissions for street segments based on the vehicle fleet information, traffic conditions, and environmental conditions.”, the street-level traffic volume for each vehicle category (e.g., the number of each vehicle type, the percentage of emission standard and fuel type for each vehicle type), traffic speed of street-segments and street information (e.g., street length, street width) are needed in the model.

As shown in section 2.4, If the traffic volume data are unavailable, users could refer to the method of this study for obtaining the traffic speed data from the Gaode map and traffic volume data by the speed-flow model. The Gaode map traffic data are quite extensive as it covered over 40 cities in China so far (with most of them being China’s major cities). Then, users could use the city-level average uniform percentage of emission standard and fuel type to obtain the volume of each vehicle category.

For the emission factors, as shown in section 2.3, users could apply the recommended value from the MEP guidebook, which are also listed in the supplementary materials. Those emission factors are national-wide and can be used in other city of China. User could also update the emission factors once they have their own data.

This would ensure the normal use of the ROE model in different cities.

15. Page 5, line 12, which version of WRF was used?

Reply:

The WRF version we used in this study was 3.7.1. We had revised it in page5, line 34.

16. Page 5, lines 18-19, why were only 31 main street segments selected?

Reply:

These 31 main street segments all locate in the Center Business District (CBD) which has significant diurnal traffic variation compared with other district in urban area. The reason that we selected these 31 main street segments was clarified in page 5, line 41 to page 6, line 3, *“The simulation area comprised 31 main street segments selected to simulate the variation in pollutant concentrations, because continuous traffic data existed for these street segments during the simulation period which were representative within the street network.”*

17. Page 5, line 20, please spell out “WUDAPT”.

Reply:

We had revised it in page 6, line 4

18. Page 5, line 24, why was “the 28th April 2018 to the 2nd May 2018” selected? This needs to be explained up front, not in a section later.

Reply:

Agreed. We had revised it in page 6, line 8-11, *“The simulation period of the study spanned from the April 28th, 2018 to the May 2nd, 2018. There was a significant traffic volume change between holidays and non-holidays. This simulation period covered holidays and non-holidays, which was helpful to investigate the impact of traffic volume variations on air quality.”*

19. Page 5, lines 26-28, are “boundary conditions” the same as the urban background concentrations needed for MUNICH model simulations? How are the measured NO_x and O₃ concentrations used to derive the boundary conditions? What are the uncertainties/measurement errors associated with those measurements?

Reply:

The boundary conditions were the same as background concentrations which was needed for MUNICH model. We had changed the “boundary conditions” to “background concentrations” for easier understanding in page 6, line 12 and line 21.

The background concentrations were from a monitoring site (YangJi site, YJ) outside but near the simulation street network. The observational data from this site could provide the background concentrations for MUNICH model. We had clarified it in page 6, line 20-21, *“In addition, YJ is located near but not within the simulation street network. The observational data from YJ could be used as the background concentration data for the modeling.”*

The uncertainties of the measurements were declared in page 6, line 12-17, *“For modeling evaluation and background concentrations, the observational concentration data for NO₂ and O₃ were obtained from the Guangzhou environmental monitoring site network. NO₂ concentrations were measured with a chemiluminescence instrument (Model 42i, Thermo Scientific) and O₃ was measured by a UV photometric analyzer (Model 49i, Thermo Scientific). The minimum detection limit (3S/N) of the analyzer was 0.4 ppbV (approximately 0.8 μg/m³) for NO₂ and 1.0 ppbV (approximately 2.0 μg/m³) for O₃. The total measurement uncertainty of these two instruments was estimated to be approximately 5% (Zhang et al., 2014).”*

20. Page 5, lines 33-37 and page 6, lines 1-2. Please add some discussions on the comparison of the three emission datasets in Table 2, e.g., why are the NO_x emissions estimated in this work higher than those from the other two? Why are the differences in gaseous emissions larger than those in PM_{2.5}/PM₁₀ emissions among the three inventories? Also, it would be useful to provide a brief description on the basis of MEIC- 2016 and PRD-2015 inventories which may help understand the differences across the three inventories. Were MEIC-2016 and PRD-2015 based on the top-down or bottom up approaches? Can those differences be related to the limitations associated with the emission modeling methods discussed in page 2?

Reply:

Thank you for your helpful suggestion. We had added some details about the MEIC and PRD inventories. In page 6, line 29-34, *“These two emission inventories used the top-down method to*

establish on-road emission inventories. Unlike the bottom-up method used in this study, these two inventories first calculated the total emissions based on the VKT data of vehicle categories. In the MEIC inventory, the total number of vehicles was obtained from the relationship between total vehicle ownership and economic development (Zheng et al., 2014), while the PRD inventory acquired information on the number of vehicles from the city-level statistics Yearbook. Then, the spatial distribution of these two inventories was established based on the road network density.”

According to the uncertainty analysis of emission factors, the uncertainty of PM_{2.5} and PM₁₀ is much smaller than the gaseous emissions, leading the large difference of gaseous emissions.

As for NO_x emissions, we thought that the higher NO_x estimate could be due to our updated LPG bus emission factor based on the local study (Zhang et al., 2013). The NO_x emission factor of an LPG-fueled bus is 1.7 times that of a diesel-fueled bus. This maybe one of the reasons leading the higher NO_x estimate. From figure 9, the results showed that the NO_x emission distribution of bus in urban and suburban area was 20.5% and 10.8%.

We had added this content in page 6, line 38 to page 7, line 4, *“the difference of PM_{2.5} and PM₁₀ amount was smaller than other gaseous emissions among different inventories. This was because that the uncertainty of particulate matter emission factors was lower than the corresponding values of the other emissions, which led to the large difference for the gaseous emissions and the smaller differences for PM_{2.5} and PM₁₀. For NO_x emissions, however, this study showed a higher NO_x estimate than that in the other two inventories. One of the reasons for the higher NO_x estimate may be the application of the updated LPG bus emission factors in this study. Based on a previous local emission factor study, the NO_x emission factor of an LPG-fueled bus is 1.7 times that of a diesel-fueled bus in Guangzhou (Zhang et al., 2013). The results in Figure 8 show that the NO_x emissions distribution attributable to buses in urban and suburban areas were 20.5% and 10.8% of the total NO_x, respectively, showing that the LPG-fueled buses may be responsible for higher NO_x estimates in this study compared to those in the other two inventories.”*

21. Page 7, lines 7-8, discussion for Figure 11, could you explain why the model gives larger NO_x overpredictions of NO_x and O₃ underpredictions on May 2? It looks that the model tends to overpredict O₃ mixing ratios at night, could you please explain the likely causes for this overprediction? Does this error come from the overestimated urban background O₃, or underpredicted NO_x titration (as the model tends to underpredict NO_x mixing ratios at night) or both? Is it possible to set up some sensitivity simulations to verify/pin-point your speculated causes for the model bias?

Reply:

Thanks for bringing up this meaningful question. Several sensitivity cases had been further carried out to figure out what affected the simulation results in supplementary materials section S3. We had compared the observational and background concentrations of NO₂ and O₃ and analyze the results from the model sensitivity cases (Figure S2 to S5 in supplementary materials). We thought that NO₂ overprediction during the daytime was caused by the overestimation of background concentrations. Due to the only consideration of on-road emission and overprediction of NO₂, the VOCs-to-NO_x

emission ratio was underestimated. Meanwhile, the simulated street network was in the VOC-sensitive regime. Thus, the O₃ concentrations were underpredicted during the daytime, especially on May 2nd. For the nighttime O₃ overprediction, both overestimated background concentrations and underestimation of NO titration were the reasons for higher predicted nighttime O₃. As shown by the sensitivity case results shown (Figure S2 to S5 in supplementary materials), the underestimated NO titration had also led to a lower simulated NO₂ concentrations at night.

In supplementary materials section S3, *“In order to figure out what affected the simulation results, some modeling sensitivity cases had been carried out in this study. As the diurnal variations shown in Figure S2, NO₂ concentrations were underestimated, while the O₃ were overestimated at night. However, the daytime model predictions were opposite to the nighttime prediction with NO₂ overprediction and O₃ underprediction. Combined with the observation and background concentrations data in Figure S3, the daytime NO₂ and nighttime O₃ overprediction maybe be caused by the overestimation of background concentrations.*

To evaluate the impact of background concentrations on the simulation results, the no-emission sensitivity case was carried out for enhancing the background effect. This case was run without any emissions and results were shown in the Figure S4. For NO₂, daytime concentrations were still overestimated compared with the observational data, especially in May 2nd, with maximum 23.7% daytime overestimation during the simulation. Meanwhile, nighttime O₃ were also overpredicted in this case, showing the nighttime O₃ overestimation was mostly due to the overestimation of background O₃ concentration. In contrast, since only vehicle emissions were considered, the underestimation of daytime O₃ should relate to the lack of other sections of emission in the simulation street network.

Besides, another case was carried out to evaluate the nighttime NO_x titration. Since NO₂ were underestimated at night but overestimated during daytime, NO₂ titration was not underpredicted and probably overpredicted at night. The overestimation of nighttime should be due to the underestimation of NO concentration. Thus, the double-background-NO case was carried out with double background NO concentrations to evaluate the NO titration. As the background NO concentration increased, nighttime NO₂ had increased which offset the underestimation concentration in base case. Meanwhile, O₃ concentrations had decreased due to the enhancement of NO titration. These results had shown that on the one hand, the overestimation of background O₃ concentration could lead to the O₃ overprediction at night. On the other hand, the underestimated NO titration was also a reason for the nighttime overprediction of O₃ concentrations.”

We had summarized up the results from those sensitivity case. The conclusions were shown in page 8, line 16-19, *“Generally, the overestimated background concentrations of NO₂ and O₃ caused the reason for the overprediction of daytime NO₂ and nighttime O₃ concentrations, respectively. Also, the underestimated NO titration was the other main reason for overprediction of O₃ and underprediction of NO₂ concentrations at night. Due to the only consideration of on-road emission in the simulation street network, daytime O₃ concentrations were underpredicted in the results.”*

22. Page 7, lines 7-16, a reference is needed for those selected metrics. What are the criteria used to judge the model performance to be good? How are those statistics compared with other model evaluation for simulated NO_x and O₃ concentrations reported in the literature?

Reply:

Thank you for providing these important points. We had added the recommended values from Ministry of Ecology and Environment of the People's Republic of China technical guide in section 4.2.1. In page 8, line 23-29, "*The NMB, NME, and CORR values of NO₂ and O₃ in this study were within the recommended ranges in the MEP Technical Guide for Air Quality Model Selection (MEP, 2012). These recommended values were -40% < NMB < 50%, NME < 80% and R² > 0.3 for NO₂, and -15% < NMB < 15%, NME < 35%, and R² > 0.4 for O₃. Additionally, the values obtained in this study fell within the range of those obtained by other modeling studies in Guangzhou; the NMB, NME and RMSE values for simulated urban NO₂ in Guangzhou were -27.5% to -6%, 29.2% to 53.0% and 16 to 37.3, respectively, and the corresponding values for O₃ were and -21.2% to 20.0%, 38.2% to 98%, 9.4 to 40.1 (Che et al., 2011; Fan et al., 2015; Wang et al., 2016).*"

23. Page 7, line 11, "the model overestimated values", do "values" mean "observations"? Please replace "a MB" by "an MB".

Reply:

We had rewritten this paragraph deleted this sentence.

24. Page 7, line 12, please remove "respective"

Reply:

Agreed. We had rewritten this paragraph deleted this sentence.

25. Page 7, line 13, please add "respectively" after "0.90", replace "values" by "observations", replace "a MB" by "an MB".

Reply:

We had rewritten this paragraph deleted this sentence.

26. Page 7, line 36, replace "As Table 5 shows" by "As shown in Table 5"

Reply:

Agreed. We had revised it in page 9, line 8.

27. Page 8, line 12, replace "the emission" by "the emissions"

Reply:

Agreed. We had revised it in page 9, line 27.

28. Page 8, line 42, replace "observation" by "observational"

Reply:

Agreed. We had revised it in page 10, line 35.

29. Page 8, it would be useful to discuss the limitations of this work and future areas of improvement for both ROE and the MUNICH modeling work.

Reply:

Thank you for the reminder. The limitations of the ROE modeling work are listed as below:

In page 10, line 1-3, *“due to the lack of street-level vehicle fleet information, this study applied a city-level average uniform percentage for every street segment. This may increase the uncertainty of the inventory, but this aspect could be improved upon provided additional data become available in the future.”*

In page 10, line 21-23, *“In addition, China is promoting the CHINA VI emission standards for on-road vehicles. The ROE model only considers Pre-CHINA I to CHINA V currently. Thus, the model will be updated in the near future to include the CHINA VI emission standards.”*.

As for MUNICH modeling work, in page 10, line 13-18, *“In this study, only 31 main street segments were selected to study the impact of a holiday on air quality in a certain urban area of Guangzhou. Additional investigations are required to understand the variations in street-level air quality in urban or suburban area of a megacity. The results of the ROE model showed that the suburban town centers of Guangzhou served as emission hotspots. These areas had relatively higher emissions than the other suburban areas and less stringent control policies than the urban area, which suffers from more serious air quality problems.”*

30. Page 12, Table 1, please provide the full name of acronyms such as RRTM, ACM2, UCM in the footnote and references for each module. Please also indicate the version of WRF used in the table title.

Reply:

Agreed. We had revised it page 15, Table 1.

31. Page 13, Table 4, “RESE” should be “RMSE”. Please add a footnote to define all acronyms such as OBS, SIM, etc. to make the table self-explainable.

Reply:

Agreed. We had revised it in page 16, Table 4.

32. Page 13, Tables 5-6, it is not necessary to include “%” in all numbers in those tables. Suggest to delete “%” from all numbers in the tables and add “percentage” before “differences” in the title of the tables.

Reply:

Agreed. We had revised it in page 16, Table 5-6.

Reference

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