Response to reviewers

Title: Development of a real-time on-road emission (ROE v1.0) model for street-scale air quality modeling based on dynamic traffic big data (GMD-2019-74)

We are very thankful to both reviewers for their constructive criticisms and valuable comments, which are of great help in improving the quality of the manuscript. We have revised the manuscript accordingly and our detailed responses are shown below.

Reply to Reviewers:

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 PM_{10}, 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 PM_{10}, 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.*".

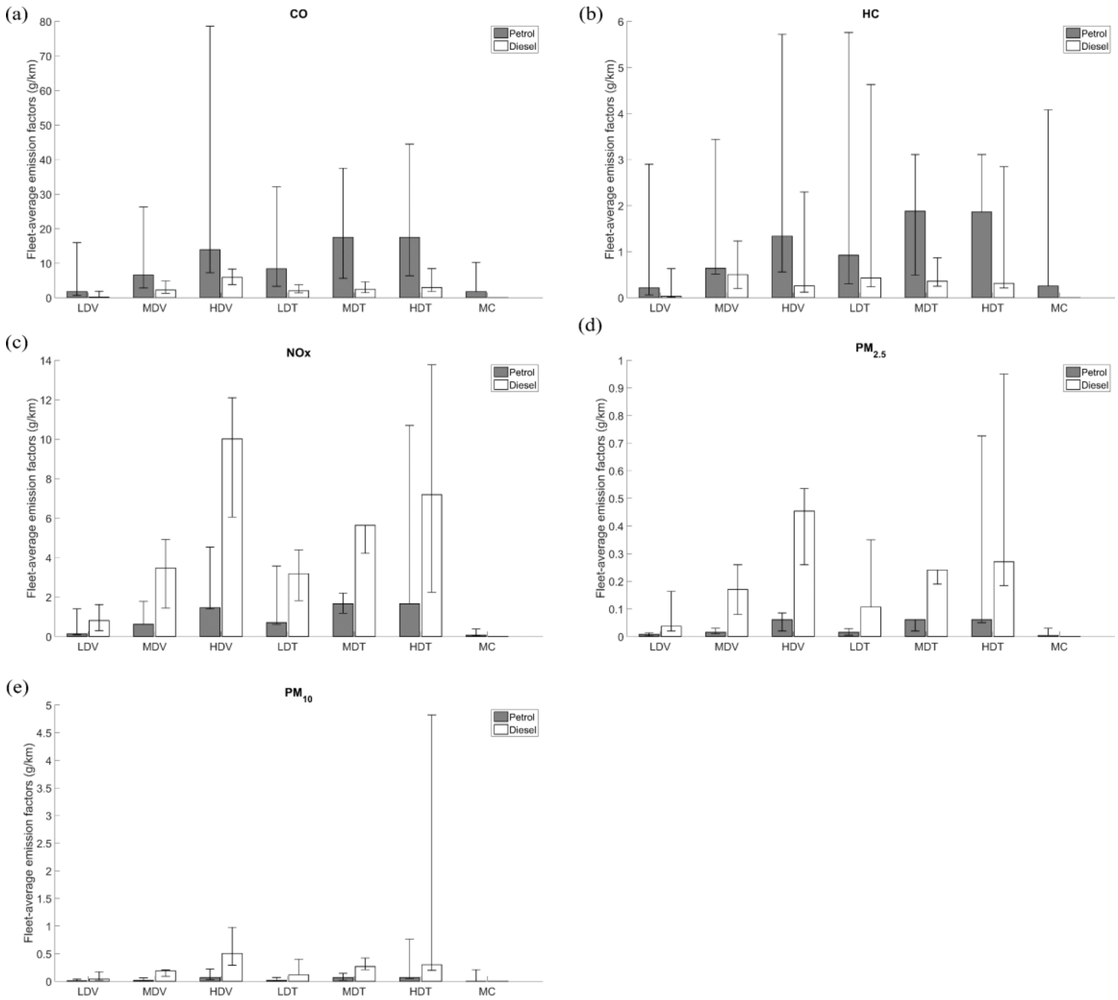

Figure S1. Fleet-average emission factors of (a) CO, (b) HC, (c) NO_x, (d) $PM_{2.5}$, and (e) PM_{10} (the value of the histogram represented the fleet-average emission factors in the study, the range represented the emission factors from other literatures).

2. How were the urban background concentrations be derived for the MUNICH model? What are the uncertainties/measurement errors associated with the measurements of NOx 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 NO_2 and O_3 were obtained from the Guangzhou environmental monitoring site network. NO_2 concentrations were measured with a chemiluminescence instrument (Model 42i, Thermo Scientific) and O_3 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 NO_2 and 1.0 ppbV (approximately 2.0 μg/m³) 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 NOx 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 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% < NMB < 50%, NME < 80% and R^2 > 0.3 for NO_2, and -15% < NMB < 15%, NME < 35%, and 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 NOx emissions estimated in this work higher than those from the other two? Why are the differences in gaseous emissions larger than those in PM2.5/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.

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 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 PM_{10} 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 PM_{10}. 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 NOx overpredictions of NOx 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 NOx titration (as the model tends to underpredict NOx 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 NO_2 and O_3 and analyze the results from the model sensitivity cases (Figure S2 to S5 in supplementary materials). We thought that NO_2 overprediction during the daytime was caused by the overestimation of background concentrations. Due to the only consideration of on-road emission and overprediction of NO_2, the VOCs-to-NO_x emission ratio was underestimated. Meanwhile, the simulated street network was in the VOC-sensitive regime. Thus, the O_3 concentrations were underpredicted during the daytime, especially on May 2nd. For the nighttime O_3 overprediction, both overestimated background concentrations and underestimation of NO titration were the reasons for higher predicted nighttime O_3. 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 NO_2 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, NO_2 concentrations were underestimated, while the O_3 were overestimated at night. However, the daytime model predictions were opposite to the nighttime prediction with NO_2 overprediction and O_3 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 concertation. 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 concertation 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.*".

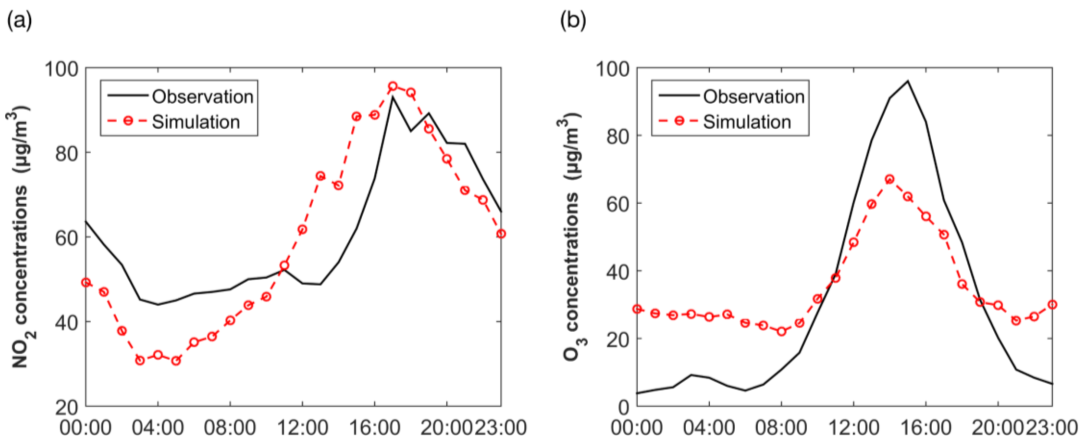

Figure S2. Diurnal variations of (a) NO_2 and (b) O_3 during the simulation period. (black solid line: observation; red dashed line: simulation).

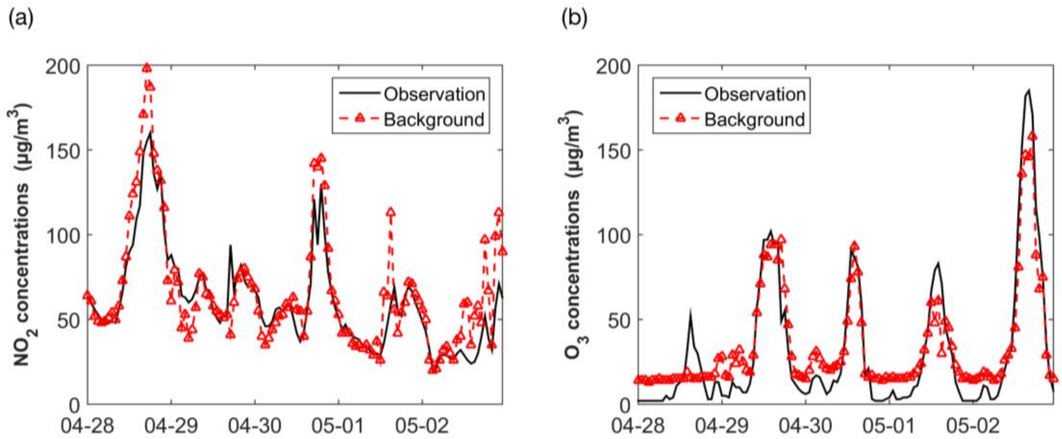

Figure S3. Time series of observational and background concentrations for (a) NO_2 and (b) O_3 during the simulation period (black solid line: observation; red dashed line: background).

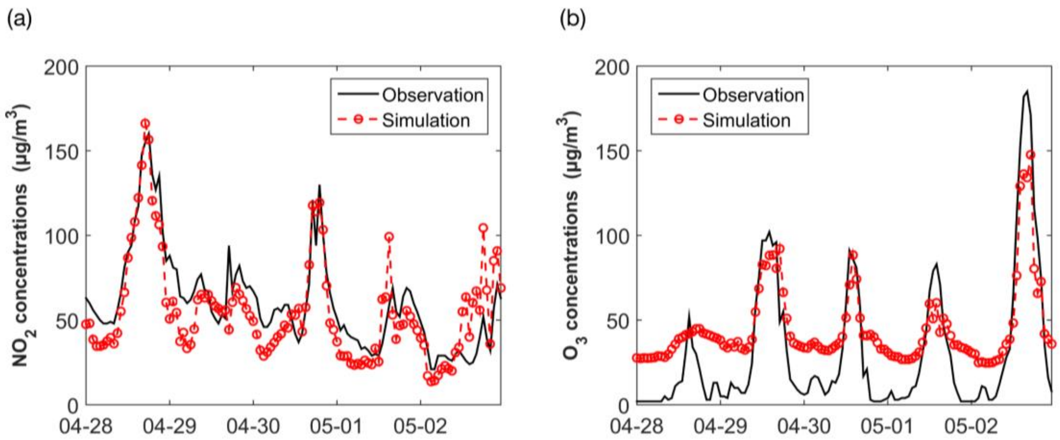

Figure S4. Time series of (a) NO_2 and (b) O_3 during the simulation period in no-emission sensitivity case. (black solid line: observation; red solid line: background).

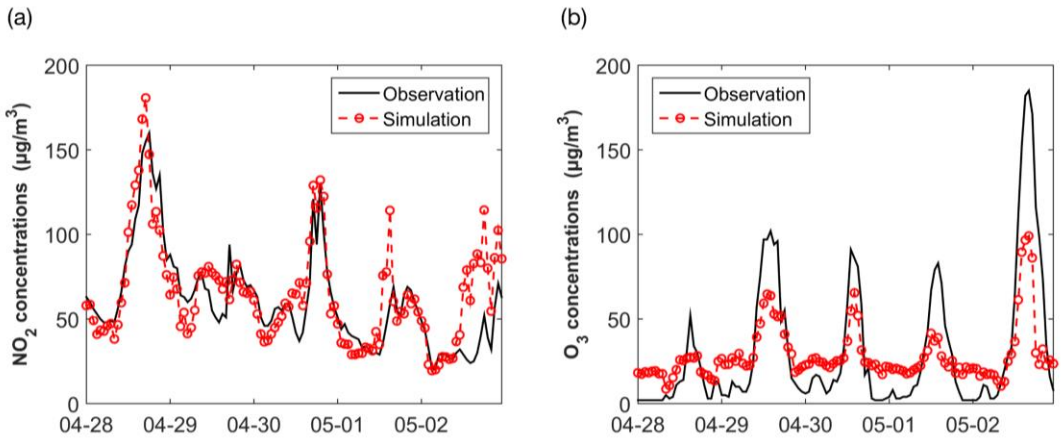

Figure S5. Time series of (a) NO_2 and (b) O_3 during the simulation period in double-background-NO case. (black solid line: observation; red solid line: background).

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 page1, 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, NOx, 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 though 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 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 PM_{10}, 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 PM_{10}, 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 significate 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 NOx 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 NO_2 and O_3 were obtained from the Guangzhou environmental monitoring site network. NO_2 concentrations were measured with a chemiluminescence instrument (Model 42i, Thermo Scientific) and O_3 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/m^3) for NO_2 and 1.0 ppbV (approximately 2.0 μg/m^3) for O_3. 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 NOx emissions estimated in this work higher than those from the other two? Why are the differences in gaseous emissions larger than those in PM2.5/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 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 PM_{10} 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 PM_{10}. 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 NOx overpredictions of NOx 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 NOx titration (as the model tends to underpredict NOx 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 NO_2 and O_3 and analyze the results from the model sensitivity cases (Figure S2 to S5 in supplementary materials). We thought that NO_2 overprediction during the daytime was caused by the overestimation of background concentrations. Due to the only consideration of on-road emission and overprediction of NO_2, the VOCs-to-NO_x

emission ratio was underestimated. Meanwhile, the simulated street network was in the VOC-sensitive regime. Thus, the O_3 concentrations were underpredicted during the daytime, especially on May 2nd. For the nighttime O_3 overprediction, both overestimated background concentrations and underestimation of NO titration were the reasons for higher predicted nighttime O_3. 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 NO_2 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, NO_2 concentrations were underestimated, while the O_3 were overestimated at night. However, the daytime model predictions were opposite to the nighttime prediction with NO_2 overprediction and O_3 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 concertation. 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 concertation 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.*".

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 NOx 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* 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% < NMB < 50%, NME < 80% and* $R^2 > 0.3$ *for* NO_2*, and -15% < NMB < 15%, NME < 35%, and* $R^2 > 0.4$ *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).* "

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.

Referee 2

1. p2 line 39: The authors mention "The real-time traffic data from the road network could be the most precise input data for on-road emission inventories and could significantly improve the spatial and temporal resolution of the inventories."

I believe this is the central point of the work presented. The targeted question is to know if "real" traffic data can help to improve the quality of modelled emissions and then the quality of modelled concentrations.

Of course the first step is to be able to use such data. The work presented shows it is technically possible. The second step is to show that it allows to get reasonable emissions and then reasonable concentrations. The manuscript provides some elements for this second step.

However what is missing in this work, from my point of view, is the demonstration of the interest of the proposed methodology in comparison to previously existing methods. It could have been relevant to compare a simulation with the emissions derived from the new methodology to a simulation with emissions derived from time-averaged data and applying hourly, daily and monthly factors as often applied within Top-Down approaches. Similarly the comparison to spatially-averaged data (at a chosen grid cell scale and per type of ways) would be of interest.

Reply:

Thank you for your helpful suggestion. We had known that this is important to compare our results with other existing emission inventories as many details as we can, no matter those inventories were establish by top-down or bottom-up method. However, due to the lack of data of the other inventories, some comparisons are unable to achieve in this study. Even so, we had tried out best to add more details about the comparison between our results and others. In section 4.1.2, we had compared the spatial distribution with other two inventories. In page 7, line 26-32, "*Moreover, the spatial distributions of these three emission inventories were compared in this study. Figure 10 shows the distribution of CO from the three different inventories. In urban areas, the results of both MEIC-2016 and PRD-2015 showed the urban areas as emission hotspots. However, the results from the ROE model were much lower for such areas. This may be due to the fact that the ROE model considers the traffic control policies, while the other two inventories do not. In suburban town centers, especially in the eastern and southern parts of Guangzhou, all three inventories showed the same results, namely that these areas were large contributors of on-road emissions. Notably, highways and arterial roads also contributed high emissions in all three inventories.*"

"*However, it should be noted that this comparison was only preliminary; the spatial resolutions of three inventories are inconsistent. Moreover, due to the lack of temporal information in the other two emission inventories, a comparison of the temporals difference could not be conducted. Future studies should focus on improving the accuracy of such comparisons.*" (In page 9, line 37-40)

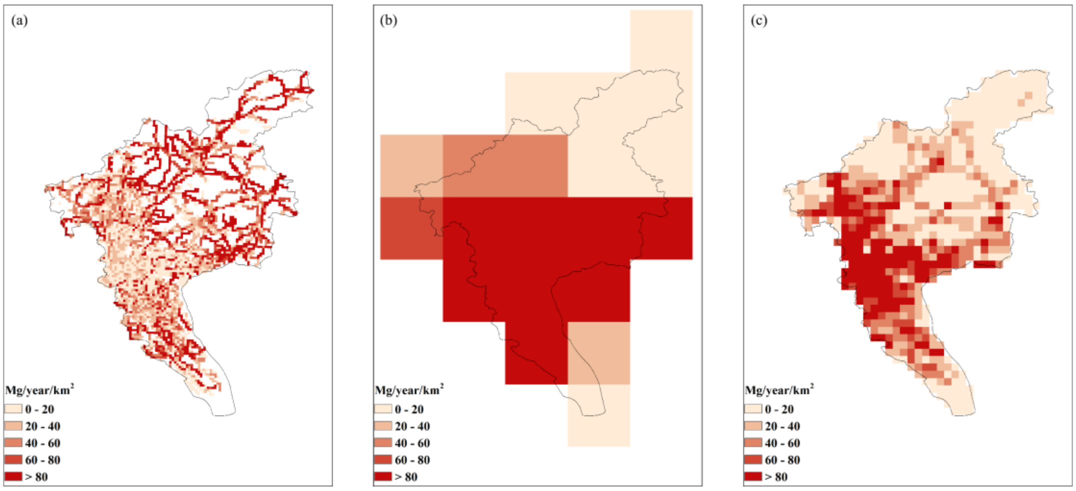

Figure 1. Spatial distribution of CO from (a) ROE model, (b) MEIC-2016 and (c) PRD-2015 in Guangzhou.

2. p3 line 4: Strictly speaking it is not the case for all air pollutants. This sentence could be rewritten avoiding this useless generalisation.

Reply:
Thanks for pointing out this. We had revised the sentence by "*Many studies have successfully applied the regional-scale CTMs to investigate the impact of on-road vehicles on the air quality in urban areas in the regional scale (~100 km).*" in page 3, line 4-5.

3. p3 line 26: This sentence should be rewritten to clarify what is available currently (it is always possible to develop a model to extend its functionalities).

Reply:
Thanks for your constructive comment. We had revised by adding a sentence "*The current version of the ROE model includes the crawler module for the from amap.com (also called the Gaode map) application (Figure 2), a widely adopted map application in China (additional details are provided in section 2.4.).*" to clarify the available data source in the current version of model in page 3, line 38-40.

4. p4 line 15 and section 2.2: More details on the emission factors building methodologies would be useful to appreciate their relevance in a near real-time / "instantaneous" framework. Does their temporal representativeness is fully consistent with the fine temporal description of the traffic data? If not, what are the expected impact on the results?

Reply:
Thanks for pointing out this. We had shown the emission factors and their correction factors in supplementary materials section S1. We had considered the traffic condition correction factors (Table S9 and Table S10) when we calculated the on-road emissions. The emission factors are different under different traffic conditions and the classification of these traffic conditions are based on the real-time traffic data. These correction factors were clarified in page 4, line 22-25, "*The*

correction factors involving environmental conditions (e.g., temperature, relative humidity, and altitude) and traffic conditions obtained from the technical guide were considered in the study. They are listed in Tables S4–S10 in the supplementary materials. These correction factors were applied to reduce the effects of uncertainties associated with the emission factors."

Table S9. Traffic condition correction factors of petrol vehicle

	Traffic Speed (km/h)				
	<20	20–30	30–40	40–80	>80
CO	1.69	1.26	0.79	0.39	0.62
HC	1.68	1.25	0.78	0.32	0.59
NO_x	1.38	1.13	0.90	0.96	0.96
$PM_{2.5}$, PM_{10}	1.68	1.25	0.78	0.32	0.59

Table S10. Traffic condition correction factors of diesel vehicle

	Emission Standard	Traffic Speed (km/h)				
		<20	20–30	30–40	40–80	>80
CO	Pre-China I to China III	1.43	1.14	0.89	0.84	0.61
	China IV, China V	1.29	1.10	0.93	0.70	0.61
HC	Pre-China I to China III	1.41	1.13	0.90	0.61	0.41
	China IV, China V	1.38	1.12	0.91	0.64	0.48
NO_x	Pre-China I to China III	1.31	1.08	0.93	0.74	0.66
	China IV, China V	1.39	1.12	0.91	0.60	0.28
$PM_{2.5}$, PM_{10}	Pre-China I to China III	1.22	1.08	0.93	0.71	0.49
	China IV, China V	1.36	1.12	0.91	0.65	0.48

5. p5 line 33: The table 2 only shows the global results without any analysis. I guess a comprehensive comparison of the three inventories is beyond the scope of the current paper, however some general considerations and analysis concerning the discrepancies between the three database appears mandatory for this manuscript.

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 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 PM_{10} 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 PM_{10}. 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."*

6. p5 line 35 and followings: The numbers provided in tables should not be recalled in the body text.

Reply:
Agreed. We had deleted it in section 4.1.1.

7. p7 line 14-15: From section 3 I understand the "boundary conditions" are considered to feed the MUNICH runs. It implies that other sources than on-road emissions are implicitly considered.

Reply:
Yes. The "boundary conditions" represented the "background concentrations" from outside the simulation street network. To make it understand more easily, we had changed the "boundary conditions" to "background concentrations" in page 6, line and line 21.

8. p8 line 35: One of the traditional aim of models is to be used for prospective (long term forecast) studies. Could the authors provide some hints on how their methodology could be extended too perform such study?

Reply:
This is a good point. In discussions and conclusions part, we had discussed the possibility of applying the street-level air quality model in forecasting the variation of pollutants. In page 10, line 24-28, "*Recently studies had shown that traffic forecasting models are effective within cities (Min et al., 2009; Cortez et al., 2012; Vlahogianni et al., 2014). These models allow one to obtain predicted traffic-based on-road emissions. Combined with the meteorological forecasting systems and regional air quality forecasting systems, which provide the meteorological and background*

concentration predictions, respectively, street-level air quality models could be used for street-level air quality forecasting as well.".

Response to editor

Editor

Thank you for your helpful advices about the code we uploaded. Our code now is available on GitHub (https://github.com/vnuni23/ROE) and Zenodo (https://doi.org/10.5281/zenodo.3264859). The users who interested in our ROE model could easily download the code from these two websites.

Additionally, it is our mistake that we did not upload any guide or documentation which could guide the users to use our model. We now have uploaded the user guide and some template files to the model package to make it easier for users to understand and use our model. Some plotting scripts are placed in the code repository as well. User guide for the ROE model are also shown in the supplement to the AC3 comment (https://www.geosci-model-dev-discuss.net/gmd-2019-74/gmd-2019-74-AC3-supplement.pdf).

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Development of a real-time on-road emission (ROE v1.0) model for street-scale air quality modeling based on dynamic traffic big data

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Abstract. Rapid urbanization in China has led to heavy traffic flows in street networks within cities, especially in eastern China, the economically developed region. This has increased the risk of exposure to vehicle-related pollutants. To evaluate the impact of vehicle emissions and provide an on-road emission inventory with higher spatial–temporal resolution for street-network air quality models, in this study, we developed the Real-time On-road Emission (ROE v1.0) model to calculate street-scale on-road hot emissions by using real-time big data for traffic provided by the Gaode map navigation application. This Python-based model obtains street-scale traffic data from the map application programming interface (API), which are open-access and updated every minute for each road segment. The results of application of the model to Guangzhou, one of the three major cities in China, showed on-road vehicle emissions of carbon monoxide (CO), nitrogen oxide (NO_x), hydrocarbons (HC), $PM_{2.5}$, and PM_{10} to be 35.22 × 10^4 Mg/yr, 12.05 × 10^4 Mg/yr, 4.10 × 10^4 Mg/yr, 0.49 × 10^4 Mg/yr, and 0.55 × 10^4 Mg/yr, respectively. The spatial distribution reveals that the emission hotspots are located in some highway-intensive areas and suburban town centers. Emission contribution shows that the dominant contributors are light-duty vehicles (LDVs) and heavy-duty vehicles (HDVs) in urban areas and LDVs and heavy-duty trucks (HDTs) in suburban areas, indicating that the traffic control policies regarding duty trucks in urban areas are effective. In this study, the Model of Urban Network of Intersecting Canyons and Highways (MUNICH) was applied to investigate the impact of traffic volume change on street-scale photochemistry in the urban areas by using the on-road emission results from the ROE model. The modeling results indicate that the daytime NO_x concentrations on national holidays are 26.5% and 9.1% lower than those on normal weekdays and normal weekends, respectively. Conversely, the national holiday O_3 concentrations exceed normal weekday and normal weekend amounts by 13.9% and 10.6%, respectively, owing to changes in the ratio of emission of VOCs and NO_x. Thus, not only the on-road emission, but other emissions should be controlled in order to improve the air quality in Guangzhou. More significantly, the newly developed ROE model may provide promising and effective methodologies for analyzing real-time street-level traffic emissions and high-resolution air quality assessment for more typical cities or urban districts.

1 Introduction

Rapid economic development and urbanization have led to the exponential growth in the number of vehicles in China has grown in recent years (National Bureau of Statistics of China, 2017). As one of the three major urban clusters, the Pearl River Delta (PRD) region, or its main city, Guangzhou, has experienced a significant increase in the number of vehicles. This increase has become the dominant contributor to carbon monoxide (CO), nitrogen oxide (NO_x), and hydrocarbon (HC) emissions (He et al., 2002; Zheng et al., 2009a), which in turn are causing more frequent and more severe public health problems in Chinese megacities (An et al., 2013). Previous studies have shown that on-road vehicle emissions can contribute approximately 22–52% of total CO, 37–47% of total NO_x, and 24–41% of total HC emissions detected in cities (Zhang et al., 2009; Zheng et al., 2009a, 2014; Li et al., 2017).

Reliable on-road emission inventories can be used as input data for the numerical air quality models which are applied to estimate the impact of on-road emissions on the urban air quality (Wang and Xie, 2009; He et al., 2016). For this purpose, the realistic on-road vehicle emission inventory should be developed as the pollutant source. The two main methodologies used in recent years to establish such an inventory are top-down and bottom-up technique.

Top-down methods, such as that used in the MOBILE model offered by the US Environmental Protection Agency (EPA) or other similar macroscale models, require the vehicle population, vehicle kilometers travelled (VKT), and mean vehicle speed of the entire city data to first calculate the total amount of vehicular emissions. Then, it allocates the emissions to each grid cell utilizing parameters such as the road density and road hierarchy (Saide et al., 2009; Jing et al., 2016; Liu et al., 2018). Many studies have adopted this method to develop city- or national-level vehicle emission inventories in China (Hao et al., 2000; Cai and Xie, 2007; Guo et al., 2007; Saide et al., 2009; Zheng et al., 2009a; Sun et al., 2016). However, the top-down inventories offer low-level spatial and temporal resolution because of the allocation method and input data. Generally, the spatial allocation of the top-down inventories is based on the road network. The higher the road density and length are, the greater is the amount of emission in the same grid. This allocation method simplifies the road emissions by assuming that every road of a specific road type (e.g., highway, arterial road, and local road) experiences the same traffic volume no matter where it is located. In addition, the emission factors used under the same traffic speed in the entire city also lead to inaccurate results for the inventory. Moreover, for some megacities (e.g., Guangzhou), there are traffic control policies in place in some specific urban areas that imply emissions should be different in these areas. Besides, the VKT data are usually given on the scale of years, which limits the temporal resolution of the inventory. For numerical modeling, the accuracy of the emission inventory may have a great impact on the simulation result because of the strong dependence of numerical models on the emission inventory (Jing et al., 2016). This scale of the emission inventory may not reflect the real emission conditions for the on-road vehicles within the city, which does not lead to effective evaluation of traffic-related impact on air pollution in a complex situation such as street-level traffic flow (Huo et al., 2009).

Consequently, several studies have established higher-resolution inventories using the bottom-up approach. The main difference in this method is that bottom-up inventories are based on information from the road segments. Therefore, spatial distribution is directly obtained from the input data and there is no need for spatial and temporal allocation. Among the input data, the traffic data are crucial for establishing the inventories and determining their accuracy. Some previous studies have used traffic simulation models to obtain the traffic speed or volume data within road networks (Pallavidino et al., 2014; Zhang et al., 2016; Chen et al., 2017; Ibarra-Espinosa et al., 2018). Based on the traffic model, the method could provide traffic data for each road from low-resolution average data. However, the results from such traffic models may not reflect the realistic situation quite well, which would reduce the accuracy of the inventories. Many other studies have used realistic traffic data, which are road-side or the on-board observational data obtained at certain road segments, to establish inventories and improve their accuracy (Huo et al., 2009; Wang et al., 2008, 2010; Wang and Xie, 2009; Yao et al., 2013). Although the observed traffic data are helpful for inventory establishment, their limitation is obvious in that large-scale observation for a whole city requires extensive human labor, financial and material resources, which are expensive and time consuming. Besides, such observation may not provide real-time traffic data, which reduces the temporal resolution for the inventories.

Recent developments in image identification technology and other observation detectors, are facilitating easy collection of real-time traffic data from road networks. The extensive implementation of closed circuit televisions and other detection subsystems in the cities helps in the implementation of intelligent transport systems (ITSs) in China (Wu et al., 2009), making it possible to attain real-time traffic data at city scale. Using the traffic data provided by ITSs, many previous studies have successfully developed inventories for different areas in China (Jing et al., 2016; Liu et al., 2018; Zhang et al., 2018). Such studies provided us a new direction for the establishment of bottom-up inventories. The real-time traffic data from the road network could be the most precise input data for on-road emission inventories and could significantly improve the spatial and temporal resolution of the inventories. However, there are still some difficulties in using the ITS data. In some cities, construction of the ITS is not complete yet or has not even been carried out. Moreover, the inconsistency of the data standards leads to an inefficient way of data utilization (Zhang, 2010). Furthermore, the low degree of the data sharing may be the biggest barrier to using traffic data obtained from the ITSs (Huang et al., 2017).

With the help of a high-resolution emission inventory, numerical models can assess the impact of on-road vehicle emissions on the air quality (Huo et al., 2009). The flow and air quality modeling in cities are commonly categorized into four groups by the length scales, i.e., street scale (~100 m), neighborhood scale (~1 km), city scale (~10 km), and regional scale (~100 km) (Britter and Hanna, 2003). A previous comprehensive literature review on this topic (Zhang et al., 2012) reports that regional-scale chemical transport models (CTMs) have been widely applied to investigate the chemistry and transport of air pollutants from their emission sources. Many studies have successfully applied regional-scale CTMs to investigate the impact of on-road vehicles on the air quality in urban areas in the regional scale (~100 km) (Che et al., 2011; Saikawa et al., 2011; He et al., 2016; Ke et al., 2017). In addition, some researchers have studied street-scale and neighborhood-scale pollutant dispersion and urban air quality by adopting computational fluid dynamics (CFD) models (Fernando et al., 2010; Kim et al., 2012; Kwak et al., 2013; Kwak and Baik, 2014; Park et al., 2015; Zhong et al., 2016; Hang et al., 2017). City-scale (~10 km) CFD modeling, however, usually requires consideration of billions of grids, because a city may include tens of thousands of buildings with high-resolution and complex street networks (Di Sabatino et al., 2008; Ashie and Kono, 2011). Thus, as city-scale CFD simulations are very expensive and time consuming, they are currently rare. Recently, some models have been developed and applied to investigate street-level air quality at the city scale (Davies et al., 2007; Righi et al., 2009; Zhang et al., 2016; Kim et al., 2018) by balancing the requirements of high resolution and low computational cost.

In this direction, the first purpose of this study was to find a new, open-access source of real-time and high-quality traffic data that could serve as the input for developing an on-road emission inventory with high spatial and temporal resolution for cities or urban districts. Guangzhou was selected as the target city for the initial application of this method not only because of the large number of vehicles in use there, but also because of its well-developed ITS which could obtain the traffic information from street networks (Xiong et al., 2010). A Python-based on-road emission model called the Real-time On-road Emission (ROE v1.0) model was developed in this study to utilize these traffic data and establish a bottom-up on-road emission inventory. A street-level chemistry transport model was then used to apply the emission results and study the impact of traffic volume variations on the air quality in the urban districts of Guangzhou.

2 Description of the ROE model

2.1 Model overview

The ROE model is intended to establish the street-level emission inventories using the emissions of on-road vehicle in the street segment of interest using a bottom-up approach. 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. Lastly, the ROE model outputs the results, that is, street-level air quality inventories.

2.2 Model structure

The ROE model was developed to calculate on-road vehicle emissions from real-time traffic data. The structure of the model is shown in Figure 1. The model, which has been implemented in Python 3, can be divided into four modules: crawler, preprocessing, emission calculation, and output modules.

(1) The most crucial part of the emission inventory involves obtaining the real-world traffic data. The crawler module is designed for "crawling" the real-time traffic data from the ITS, Internet, or any other data source if the code is updated to match the format of the data source. Moreover, the study area should be set in the module, and if it's needed (in case the coordinates differ), the coordinate transformation script should be activated. The current version of the ROE model includes the crawler module for the from *amap.com* (also called the Gaode map) application (Figure 2), a widely adopted map application in China (additional details are provided in section 2.4.). (2) The preprocessing module is used for fitting the time frequency between the data source and the air quality modeling system. Subsequently, the traffic volume data are also

calculated from the traffic speed data in this module if the traffic volume or vehicle fleet information is not available from the data source. Otherwise, the number of vehicles in each category can be used directly to the emission calculation. (3) The emission calculation module uses traffic information from the preprocessing module and information about vehicle fleets to calculate emissions for each street segment using the following equation:

$$E_{s,t} = \sum EF_{s,v} \times V_{v,t} \times L, \quad (1)$$

where $E_{s,t}$ is the emission of pollutant s at time t (g/h), $EF_{s,v}$ is the emission factor of pollutant s for vehicle category v (g/km), $V_{v,t}$ is the traffic volume of the vehicle (i.e., the number of vehicles, veh) category v at time t (veh/h), and L is the length of the street segment (km). The total emission in one specific area is given by the sum of emissions in every street segment within the area. (4) The output module sums up all the information given by the emission calculation module and can be modified to provide all the results produced during the calculation of the emissions. In addition, the model includes a tool that can modify the formats of the emissions, making it possible to provide the on-road emissions to other air quality models.

2.3 Emission factors

In this study, nation-wide vehicle emission factors mandated by the Ministry of Ecology and Environment (MEP) of the People's Republic of China were adopted to calculate the emissions from vehicles on road (MEP, 2014). They are listed in Table S1 and S2 in the supplementary materials. The emission factors of liquefied petroleum gas (LPG) vehicles were sourced from a previous study conducted in Guangzhou (Zhang et al., 2013). According to the MEP guide, vehicles are classified as one of the following: a light-duty vehicle (LDV), a middle-duty vehicle (MDV), a heavy-duty vehicle (HDV), a light-duty truck (LDT), a middle-duty truck (MDT), a heavy-duty truck (HDT), a motorcycle (MC), a taxi, or a bus. The fuel type is classified as petrol, diesel, or other (such as LPG or natural gas). The emission standard is classified as Pre-China I, China I, China II, China III, China IV, or China V. In addition, the evaporation of petrol was considered during the calculation of the emissions. HC evaporation was also considered as per the details provided in the MEP guidebook (Table S3).

The correction factors involving environmental conditions (e.g., temperature, relative humidity, and altitude) and traffic conditions obtained from the technical guide were considered in the study. They are listed in Tables S4–S10 in the supplementary materials. These correction factors were applied to reduce the effects of uncertainties associated with the emission factors.

To estimate the uncertainties of the emissions factors, the results of previous studies (Zheng et al., 2009a; Zhang et al., 2013, 2016; Tang et al., 2016; Wang et al., 2017) were summarized and compared with the emission factors obtained in this study. These results appear in Figure S1 of the supplementary materials.

In addition, the emission factors can be easily updated once the local emission factors data are available.

2.4 Traffic data of floating vehicle

In this study, the traffic speed data of each street segment were obtained from Gaode map. The Gaode map traffic data are quite extensive as it covers over 40 cities in China so far (with most of them being major cities of China). 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.

However, the map application cannot provide the traffic volume data directly. Many studies have shown that the traffic volume can be estimated using the average traffic speed based on the relationship between the traffic speed and the volume (Wang, 2003; Kuo and Tang, 2011; Xu et al., 2013; Yao et al., 2013; Hooper et al., 2014; Jing et al., 2016). Many speed–flow models exist for this purpose, and each of them has certain advantages and disadvantages. In this study, the Underwood volume calculation model (Underwood, 1961) was used to retrieve the information on traffic volume because of its history of successful application in China (Jing et al., 2016). The model is described by Eq. (2):

$$V = k_m u \ln \frac{u_f}{u}, \tag{2}$$

where V is the traffic volume at speed u (veh/h), k_m is the traffic density (veh/km), u is the traffic speed (km/h), and u_f is the free speed (km/h). In this study, k_m and u_f are given by fitting the model based on observation data obtained at the roadside and video identification data gained from different road types (Zheng et al., 2009a; Jing et al., 2016; Liu et al., 2018).

To calculate the traffic volume on national highways, another speed–flow model, which was previously applied in an observation-based study undertaken in China (Wang, 2003), was used. This model is described as follows:

When the speed limit is 120 km/h,

$$V = -0.611u^2 + 73.320u; \tag{3}$$

when the speed limit is 100 km/h,

$$V = -0.880u^2 + 88.000u; \tag{4}$$

when the speed limit is 80 km/h,

$$V = -1.250u^2 + 100.000u; \tag{5}$$

when the speed limit is 60 km/h,

$$V = -2.000u^2 + 120.000u; \tag{6}$$

where V is the traffic volume at speed u (veh/h), and u is the traffic speed (km/h).

Given Guangzhou's traffic control policies, the whole city is divided into two areas: urban area and suburban (Figure 3). Therefore, the traffic volume is also calculated accordingly (Figure 4). The main traffic control policies in urban areas are as follows: (1) No truck is allowed to enter the urban area during 7:00–9:00 (morning rush hours) and 18:00–20:00 (evening rush hours), (2) no middle- and heavy-duty truck is permitted to enter the urban area during 7:00–22:00, (3) no non-local truck can enter the urban area during 7:00–22:00, and (4) no motorcycle can enter the urban area, either.

2.5 Vehicle fleet information

In this study, the fleet information on each vehicle classification was sourced from the Statistical Yearbook of Guangzhou (Guangzhou Bureau of Statistics, 2017) (Figure 5(a)). The emission standards (Figure 5(b)) and fuel type data (Figure 6) for the vehicles were source from previous studies undertaken in Guangzhou (Zhang et al., 2013, 2015). Due to the lack of the street-level vehicle fleet information, this study used a uniform percentage of emission standard, fuel type and number of vehicles in each category for each segment. The number of each vehicle type was calculated based on the total traffic volume of each street segment and the vehicle fleet percentage. It should be noted that this information could be updated if the street-level fleet information becomes available in the future.

3 Description of the street-level air quality model

To evaluate the impact of on-road emissions on air quality at the street level in Guangzhou, an air quality model called the Model of Urban Network of Intersecting Canyons and Highways (MUNICH) was employed in this study with the on-road emission results from the ROE model. MUNICH is a street-network CTM that includes street-canyon and street-intersection components in the model (Kim et al., 2018).

In this study, the Weather Research and Forecasting (WRF) model (version 3.7.1) (Skamarock et al., 2008) was used to provide the meteorological data (wind profile, boundary layer height, and friction velocity) for the modeling. The WRF simulation was conducted with four nested domains at resolutions of 27 km, 9 km, 3 km, and 1 km (Figure 7a). The physical scheme is listed in Table 1.

In MUNICH, the CB05 chemical kinetic mechanism (Yarwood et al., 2005) was used to simulate the photochemical reactions at the street level in an urban street network. For the MUNICH run, the model was applied to simulate pollutant dispersion in Tianhe District, which serves as the Central Business District (CBD) of Guangzhou. The district is characterized by significant diurnal traffic variation compared with other districts in urban areas. 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.

The urban morphology data for the building height were obtained from the World Urban Database and Access Portal Tools (WUDAPT) dataset (Ching et al., 2018). The street data were sourced from the OpenStreetMap dataset (https://www.openstreetmap.org/). The street length data were calculated directly from the location of the start and end intersections of each street segment. Data on the street width were retrieved from the feature class of the road, and the width of each lane was assumed to be 3.5 m.

The simulation period of the study spanned from the April 28th, 2018 to the May 2nd, 2018, which included a Chinese national holiday between April 29th, 2018 to May 1st, 2018. There was a significate 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. Another 3-day simulation time was conducted before this period to spin up the model.

For modeling evaluation and background concentrations, the observational concentration data for NO_2 and O_3 were obtained from the Guangzhou environmental monitoring sites network. NO_2 concentrations were measured with a chemiluminescence instrument (Model 42i, Thermo Scientific) and O_3 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/m^3) for NO_2 and 1.0 ppbV (approximately 2.0 μg/m^3) for O_3. The total measurement uncertainty of these two instruments was estimated to be approximately 5% (Zhang et al., 2014).

Two monitoring sites, Tiyuxi (TYX) and YangJi (YJ) site were selected for this study (Figure 7c). 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. Due to the lack of NO observational data, the concentration ratio of NO_2 to NO was assumed as 4:1 in this study.

4 Application of the ROE model to Guangzhou

4.1 On-road emission inventory from the ROE model

4.1.1 Overview of the emission inventory

Using the high-resolution spatial and temporal traffic data from the map application, the emission inventory of on-road vehicles from the ROE model was established for this study. Table 2 shows the annual emissions from vehicles in Guangzhou city compared with two other gridded emission inventories in China: the MEIC model (http://meicmodel.org/) and a PRD region local emission inventory (Zheng et al., 2009b). 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.

Given the shorter total road length and traffic control policies in urban areas (Figure 3), the urban on-road emissions of CO, NO_x, HC, $PM_{2.5}$, and PM_{10} were comprised only 13.1%, 8.8%, 12.7%, 8.2%, and 9.1% of the total on-road emissions, respectively, suggesting that the suburban areas are the dominant contributor of on-road emissions in Guangzhou.

In general, the difference of $PM_{2.5}$ and PM_{10} 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 gaseous emissions, which led to the large difference for the gaseous emissions and the smaller differences for $PM_{2.5}$ and PM_{10}. 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.

As shown in Table 3, the emission contribution of local roads in urban areas is the highest component because of the total length of the local roads, which is 5.4 times and 4.8 times that of highways and arterial roads in urban areas, respectively. Although the total length of the highways is shorter, the traffic volume on the highway is much higher than that on the local roads (Figure 4), thus causing highest contribution of emissions from the suburban areas. Moreover, the emission contributions from urban and suburban areas differ on weekdays and weekends. In urban areas, the daily total weekday and weekend emissions are 129.94 Mg/d and 118.29 Mg/d of CO, 30.15 Mg/d and 27.71 Mg/d of NO_x, 14.74 Mg/d and 13.40 Mg/d of HC, 1.27 Mg/d and 1.16 Mg/d of $PM_{2.5}$, and 1.41 Mg/d and 1.29 Mg/d of PM_{10}, respectively. In suburban areas, the total weekday and weekend emissions are 873.97 Mg/d and 758.41 Mg/d of CO, 315.10 Mg/d and 267.91 Mg/d of NO_x, 102.46 Mg/d and 88.22 Mg/d of HC, 13.01 Mg/d and 10.98 Mg/d of $PM_{2.5}$, and 14.45 Mg/d and 12.19 Mg/d of PM_{10}, respectively. The total respective emissions of CO, NO_x, HC, $PM_{2.5}$, and PM_{10} on a weekday are 114.5%, 116.8%, 115.3%, 117.6%, and 117.7% of the values on a weekend, respectively.

4.1.2 Spatial distribution of emissions

Due to the vehicular activities, the spatial distribution of on-road emissions was consistent with the structure of the street network. For a better description of this spatial distribution, the emissions were mapped onto a 1-km-resolution fishnet and the total emissions of one grid cell were the sum of all on-road emissions from within the same grid cell. The spatial distribution of each pollutant is shown in Figure 9. Overall, the high-value grid cells were generally located along the highways. In suburban areas, high-value areas located away from the highways and arterial roads normally denoted suburban town centers that had more local roads and higher traffic volume density. In urban areas, the high-value areas were more closely related to the densities of the urban local roads. The emission hotspots were less prominent in urban areas than in suburban areas due to the strict traffic control policies in urban area. The spatial distribution indicated that the next effective control on-road emissions policy should pay more attention to the control of vehicles in suburban areas.

Moreover, the spatial distributions of these three emission inventories were compared in this study. Figure 10 shows the distribution of CO from the three different inventories. In urban areas, the results of both MEIC-2016 and PRD-2015 showed the urban areas as emission hotspots. However, the results from the ROE model were much lower for such areas. This may be due to the fact that the ROE model considers the traffic control policies, while the other two inventories do not. In suburban town centers, especially in the eastern and southern parts of Guangzhou, all three inventories showed the same results, namely that these areas were large contributors of on-road emissions. Notably, highways and arterial roads also contributed high emissions in all three inventories.

4.1.3 Emission contributions of vehicles by their classification

The emission contributions of different vehicle classifications in the urban and suburban areas are shown in Figure 8. As LDVs accounted for the largest number, their emission contribution comprised the dominant proportion of total emissions in urban areas for each pollutant. The contribution percentages of CO, HC, NO_x, $PM_{2.5}$, and PM_{10} were 80.9%, 84.1%, 26.4%, 38.3%, and 38.2%, respectively. The second largest contributor to on-road emissions was the HDVs, the relevant percentages being 5.8%, 2.9%, 30.3%, 35.2%, and 35.2% for CO, HC, NO_x, $PM_{2.5}$, and PM_{10}, respectively. As for the buses, except for the contribution of NO_x, which accounted for 20.5% of the total emissions mentioned above, the proportions of the other pollutants were less than 2% because of the use of LPG as the fuel. In the case of trucks, the total contribution of LDTs, MDTs, and HDTs were 10.3%, 9.3%, 21.2%, 23.3%, and 23.3% for CO, HC, NO_x, $PM_{2.5}$, and PM_{10}, respectively, considering the traffic control policies in the urban areas. The contribution of taxi was less than 1% because of the small number of taxis and their use of LPG. In suburban areas, the LDVs were the dominant contributor of CO and HC emissions because of its large amount. For

NO_x, $PM_{2.5}$, and PM_{10}, however, the HDT provided the largest contribution, at 36.5%, 43.2%, and 43.3%, respectively. Moreover, LDVs, HDVs, and buses were important contributors of NO_x, at 19.4%, 17.4%, and 10.8%, respectively. Regarding particulate matter, the respective percentages of emissions (for both $PM_{2.5}$ and PM_{10}) owing to LDVs, HDVs, and LDTs were 19.7%, 20.5%, and 9.0%, suggesting that these vehicles were also important sources of both $PM_{2.5}$ and PM_{10}.

4.2 Application of the ROE model's results to the street-level air quality model

4.2.1 Modeling performance in Guangzhou urban area

During the simulation period, the model results were evaluated for the TYX observation site located within the street network. The on-road emissions were provided by the ROE model, as discussed previously. Street segments to which high NO_x emission values were attributed were also responsible for high HC emissions because of the positive relationship between traffic volume and on-road emissions as shown in Figure 11.

The time series for the simulated NO_2 and O_3 concentrations within the street network were compared with the observed concentrations (Figure 12). As the results show, daytime NO_2 concentrations were overestimated while nighttime concentrations were underestimated during the simulation period. The O_3 concentrations, however, were underpredicted during daytime and overpredicted at nighttime. Serval modeling sensitivity cases were analyzed to identify what factors may have affected the model simulation. The sensitivity analysis results are provided in the supplementary materials section S3. 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.

Moreover, the performance statistics for NO_2 and O_3 are also shown in Table 4. Here, the statistical measures of the observation (OBS) mean, simulation (SIM) mean, mean bias (MB), normalized mean bias (NMB), normalized mean error (NME), mean relative bias (MRB), mean relative error (MRE), root mean squared error (RMSE), and the correlation coefficient (CORR) were used to validate the model. 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 $-40\% < NMB < 50\%$, $NME < 80\%$ and $R^2 > 0.3$ for NO_2, and $-15\% < NMB < 15\%$, $NME < 35\%$, and $R^2 > 0.4$ 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). Overall, the model showed good performance for the simulation and can be applied to the future studies to investigate the impact of on-road vehicles on air quality.

4.2.2 Impact of traffic volume variations on air quality

To investigate how traffic volume change affects air quality at the street level, a Chinese national holiday was chosen as the target simulation period for the modeling. Figure 13 shows the diurnal variation in the traffic volume during the national holiday, normal weekday, and normal weekend before and after the holiday in the simulation street network. On the normal weekday, two typical rush hour trends appeared during the 8:00–10:00 and 17:00–19:00 (although April 28th was a Saturday, it was a normal workday to compensate for the holiday). For the normal weekend and the national holiday, the peak in traffic volume was noted between 14:00 and 16:00 and no rush hour peak occurred on these days. At nighttime, not much difference was noted for the traffic volumes on the normal weekday, normal weekend, and national holiday, especially after midnight. However, the higher traffic volume between 21:00 and 23:00 on the 28th April at night may have been caused by people traveling out of the city before the national holiday (e.g., returning home across the city or traveling to other places).

Three sensitivity cases were carried out to study the impact of traffic volume change on the air quality in urban areas: (1) in the national holiday case, wherein the on-road emissions between April 29th and May 1st were regarded as the original emissions during the simulation period(this represents the base case), (2) in the normal weekday case, diurnal on-road emissions for three national holidays were replaced by the emissions of April 28th, and (3) in the normal weekend case, the national holiday period emissions were replaced by the diurnal on-road emissions of May 5th. The diurnal variations in NO_x and O_3 in the three cases are shown in Figure 14. During 0:00–5:00, because of similar traffic volume, there were no large differences in the NO_x and O_3 concentrations during this time. Due to the morning rush hour, the NO_x concentrations for the normal weekday case were much higher than those for the national holiday case in the morning. As shown in Table 5, the NO_x concentrations were 12.0–26.5% higher for the normal weekday case during this time. For in the normal weekend case too, the NO_x concentrations simultaneously increased by 9.1% compared to those on the national holiday in the morning. This increase was caused by people traveling for normal weekend engagements. In the afternoon, however, the difference between the NO_x concentrations was less than 10% due to the rising traffic volume on the national holiday. During the evening rush hour, although the traffic volume on the normal weekday was 1.3 times that on the national holiday, the maximum difference between the NO_x concentrations was only 7.3%. This shows that the variations in NO_x concentrations were more affected to a greater extent by the background concentrations (i.e., boundary conditions) in the evening.

Compared with the national holiday case, the O_3 concentrations were much lower in the normal weekday case. In the afternoon, as shown in Table 6, when photochemical reactions are more prevalent, the national holiday O_3 concentrations exceeded those on normal weekdays and weekends by 13.9% and 10.6%, respectively. This occurs because the simulation street network in the urban area is in the VOC-sensitive regime (Ye et al., 2016). The O_3 concentrations were positively correlated with the VOC emissions. As the NO_x emissions was higher than the VOC emissions, the reduction in the NO_x emissions was also much higher than in the VOC emissions when the number of vehicles decreased on the national holiday. The larger NO_x emission reduction led to a higher VOCs-to-NO_x emission ratio, which resulted in a higher O_3 concentration during the national holiday (Sanford and He, 2002).

5 Discussion and conclusions

Using real-world traffic information, the Real-time On-road Emission (ROE v1.0) model can provide real-time and high-resolution emission inventories for regional or street-level air quality models in China. The results show that the ROE model can simulate the emissions of CO, NO_x, HC, $PM_{2.5}$, PM_{10} and any other pollutant provided the relevant emission factors are included in the model. (This aspect will be updated in subsequent releases.) As it uses the bottom-up method, the ROE model facilitates the calculation of the emissions in each street segment.

In this study, the traffic information of Guangzhou was obtained from the Gaode map, the data for which are collected from map users while they are driving. The geographic and speed information were sourced from the map users' GPS devices and can be used through the map API. Using the ROE model and fully considering the traffic control policies of Guangzhou city, the annual total on-road emissions of CO, NO_x, HC, $PM_{2.5}$, and PM_{10} were modeled to be 35.22×10^4 Mg/yr, 12.05×10^4 Mg/yr, 4.10×10^4 Mg/yr, 0.49×10^4 Mg/yr, and 0.55×10^4 Mg/yr, respectively. Spatial distribution analysis showed that hotspots of on-road emissions were situated along the highways and suburban town centers. The comparison of spatial distribution between the ROE model's results and those of two other inventories showed that the ROE model provided had lower urban emissions as it the considered the traffic control polices. However, it should be noted that this comparison was only preliminary. The spatial resolutions of three inventories are inconsistent in this study. Moreover, due to the lack of temporal information about the other two emission inventories, a comparison of the temporal difference could not be conducted. Future studies should focus on improving the accuracy of such comparisons.

Owing to the number of vehicles and their respective distributions, LDVs constituted the dominant source of on-road emissions in Guangzhou. In suburban areas, however, the HDTs was the most important contributor of NO_x, $PM_{2.5}$, and PM_{10}. Daily emissions of CO, NO_x, HC, $PM_{2.5}$, and PM_{10} on a weekday were found to be 14.5%, 16.8%, 15.3%, 17.6, and 17.7%

higher than the daily emissions on a weekend, respectively. However, 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. Given the high spatial and temporal resolutions of the emission inventory of the ROE model, three sensitivity cases were analyzed to study the effect of vehicular on-road emissions on urban street-level air quality. On a national holiday, NO_x concentrations were 12.0–26.5% less than those on a normal weekday as no morning rush hours occurred on holidays. Moreover, compared with the normal weekend, the NO_x concentrations on a national holiday also show a decrease of 9.1% in the peak value in the morning. However, the reduction of the NO_x concentrations in the afternoon was smaller than that in the morning, suggesting that NO_x concentrations transportation from the surrounding boundaries is the main reason for the variation in the afternoon NO_x concentrations. In addition, as the simulation street network lies in the VOC-sensitivity regime, the lower traffic on a national holiday and a normal weekend caused the NO_x and VOC emissions to be lower than those on a normal weekday. However, the reductions in NO_x were higher than the decrease in VOC emissions, which led to a higher VOCs-to-NO_x emission ratio and O_3 concentrations on holidays and normal weekends. 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.

In general, the ROE model could provide a high-resolution on-road emission inventory when the real-time traffic information and emission factors were fed into the model. 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.

Recently studies had shown that traffic forecasting models are effective within cities (Min et al., 2009; Cortez et al., 2012; Vlahogianni et al., 2014). These models allow one to obtain predicted traffic-based on-road emissions. Combined with the meteorological forecasting systems and regional air quality forecasting systems, which provide the meteorological and background concentration predictions, respectively, street-level air quality models could be used for street-level air quality forecasting as well.

In summary, the newly developed ROE model was confirmed to be effective for analyzing real-time city-scale traffic emissions and performing high-resolution air quality assessments in the street networks of Guangzhou city. The methodologies presented in this work can be further extended to more typical cities or urban districts in China or other countries.

Author contribution. Luolin Wu and Xuemei Wang designed the experiments. Luolin Wu developed the model code and performed the simulations. Ming Chang organized and visualized the data. Jinpu Zhang collected and organized the observational data. Luolin Wu and Jian Hang prepared the manuscript with contributions from all co-authors.

Code availability. The python source code of the ROE v1.0 model and examples are available on Github (https://github.com/vnuni23/ROE) and Zenodo (https://doi.org/10.5281/zenodo.3264859). More information and help are also available by contacting the authors.

Competing interests. The authors declare that they have no conflict of interest.

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Table 1. Physical parameterization configurations for WRF v3.7.1 model

Physical parameterizations	
Microphysics Scheme	Morrison (2 moments) (Morrison et al., 2009)
Land-surface Scheme	Pleim-Xiu (Xiu and Pleim, 2001)
Cumulus Scheme	Kain-Fristsch (Kain, 2004)
Longwave Radiation Scheme	Rapid Radiative Transfer Model (RRTM) (Mlawer et al., 1997)
Shortwave Radiation Scheme	Dudhia (Dudhia, 1989)
Boundary-layer Scheme	Asymmetric Convective Model version 2 (ACM2) (Pleim, 2007)
Urban Surface Scheme	Urban Canopy Model (UCM) (Chen et al., 2011)

Table 2. Annual on-road emissions in Guangzhou (unit: 10^4 Mg/yr)

		CO	NO_x	HC	$PM_{2.5}$	PM_{10}
	Urban	4.61	1.07	0.52	0.04	0.05
This study	Suburban	30.61	10.98	3.58	0.45	0.50
	Total	35.22	12.05	4.10	0.49	0.55
MEIC-2016	(Gridded)	43.56	8.45	9.26	0.46	0.47
PRD-2015	(Gridded)	28.89	6.99	4.65	0.52	0.52

Table 3. Daily emissions on different road type in urban and suburban area (unit: Mg/day)

		Road type	Length(km)	CO	NO_x	HC	$PM_{2.5}$	PM_{10}
weekday	urban	highway	301.87	9.71	3.15	1.02	0.11	0.12
		artery	337.19	17.24	4.95	1.88	0.19	0.21
		local	1629.92	102.99	22.05	11.84	0.97	1.08
	suburban	highway	2316.73	417.49	168.29	45.51	6.50	7.22
		artery	747.63	61.12	26.54	7.24	1.11	1.23
		local	8867.69	395.36	120.27	49.71	5.40	6.00
weekend	urban	highway	301.87	7.47	2.34	0.79	0.08	0.09
		artery	337.19	13.20	4.23	1.40	0.15	0.17
		local	1629.92	97.62	21.14	11.21	0.93	1.03
	suburban	highway	2316.73	428.30	156.78	47.14	6.07	6.74

	artery	747.63	59.20	26.56	6.99	1.10	1.22
	local	8867.69	270.91	84.57	34.09	3.81	4.23

Table 4 The performance statistics for NO_2 and O_3 in modeling (unit: μg/m^3)

	Mean								
	OBS[a]	SIM[b]	MB[c]	NMB[d]	NME[e]	MRB[f]	MRE[g]	RMSE[h]	CORR[i]
NO_2	30.8	35.4	4.7	15.2%	68.8%	3.0%	3.2%	25.7	0.90
O_3	60.9	59.3	-1.6	-2.7%	24.3%	<0.1%	0.3%	18.7	0.80

[a] OBS (Observation). [b] SIM (Simulation). [c] MB (Mean Bias). [d] NMB (Normalized Mean Bias). [e] NME (Normalized Mean Error). [f] MRB (Mean Relative Bias). [g] MRE (Mean Relative Error). [h] RMSE (Root Mean Squared Error). [i] CORR (correlation coefficient).

Table 5. Daytime percentage difference of NO_x compared to National holiday case

time	6:00	7:00	8:00	9:00	10:00	11:00	12:00	13:00	14:00	15:00	16:00	17:00	18:00	19:00	20:00
normal weekday	12.7	21.7	16.8	26.5	14.7	12.0	4.9	0.6	8.6	2.2	0.7	0.2	7.3	5.9	7.1
normal weekend	-4.4	0.1	9.1	6.7	0.2	7.0	1.2	2.6	6.2	0.8	-0.6	-0.9	2.1	-5.7	4.9

Table 6. Daytime percentage difference of O_3 compared to National holiday case

time	6:00	7:00	8:00	9:00	10:00	11:00	12:00	13:00	14:00	15:00	16:00	17:00	18:00	19:00	20:00
normal weekday	-4.5	-15.7	-52.8	-48.9	-37.5	-25.9	-15.6	0.2	-7.9	-13.9	-7.4	-11.1	-46.3	-38.4	-32.3
normal weekend	2.9	6.3	-2.6	-4.9	-15.0	0.5	-4.0	-1.6	-5.7	-10.6	-0.4	12.4	-15.3	-0.1	3.7

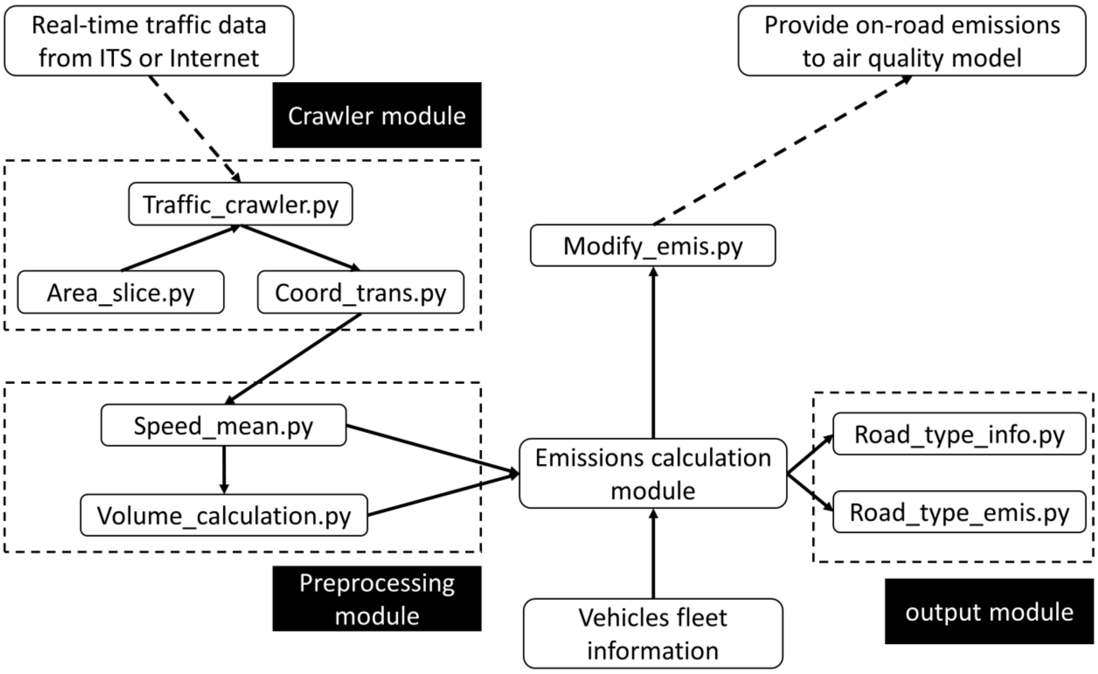

Figure 1. The structure of ROE model.

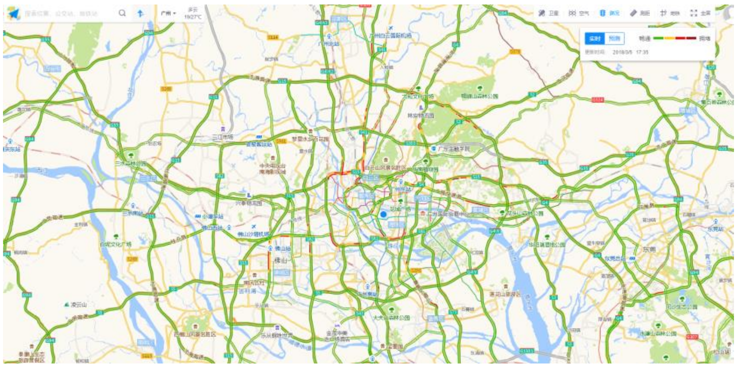

Figure 2. Traffic information from Gaode map.

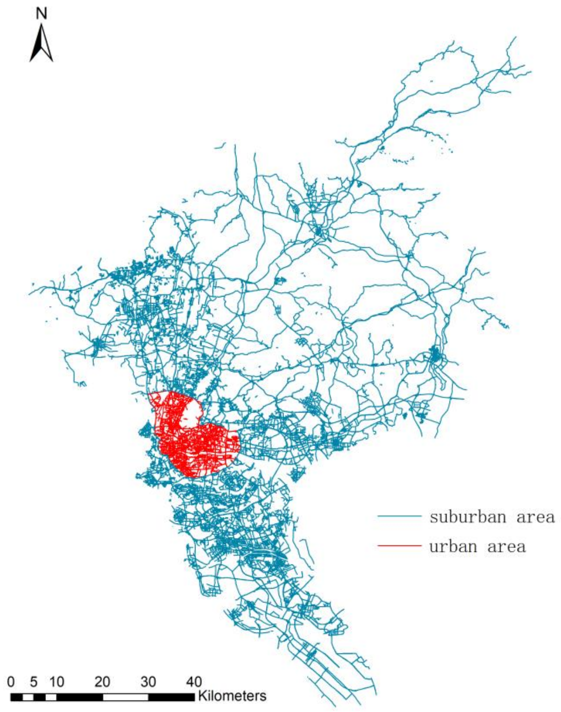

Figure 3. Traffic control area.

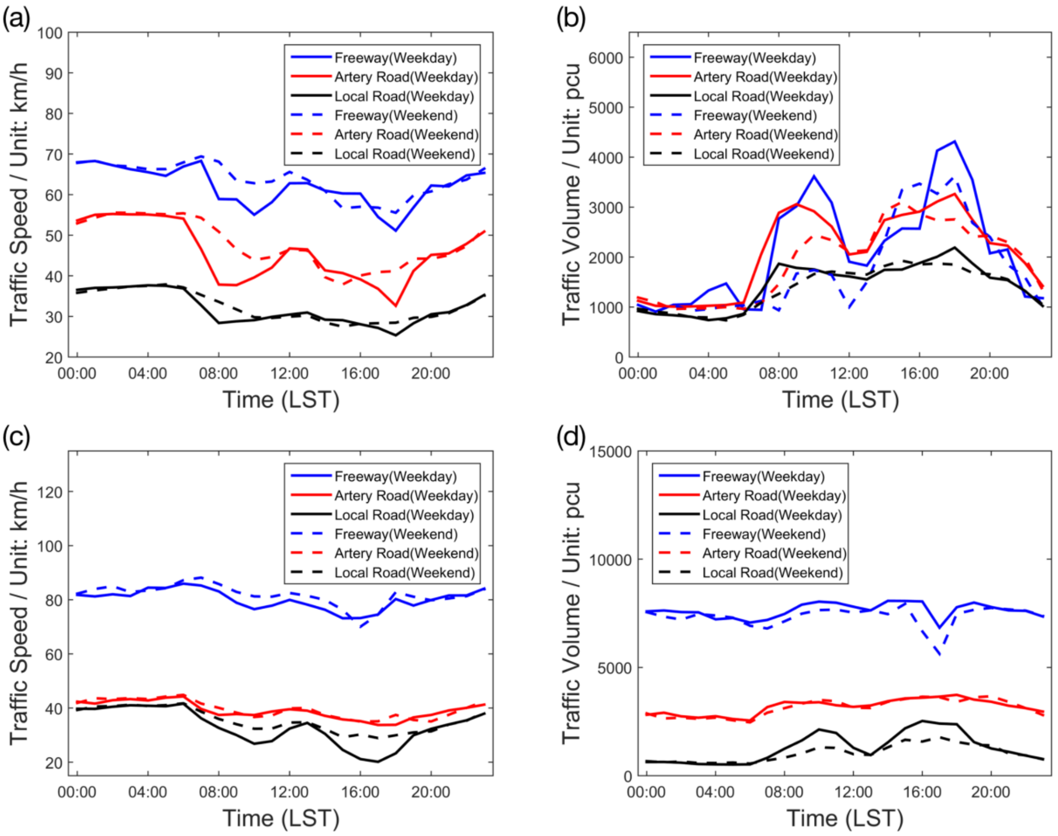

Figure 4. Diurnal variation of average traffic speed and traffic volume in (a, b) urban area and (c, d) suburban area during weekday and weekend.

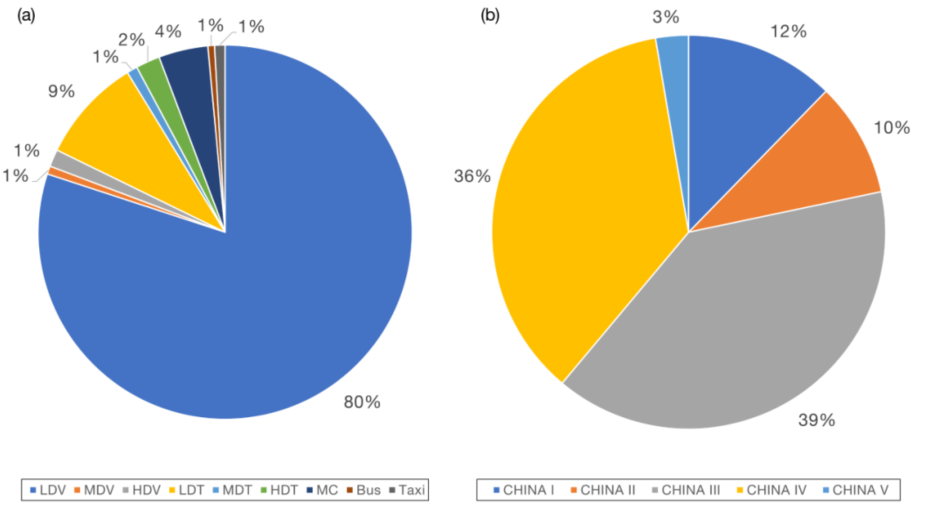

Figure 5. The percentage of (a) vehicle classification and (b) emission standard.

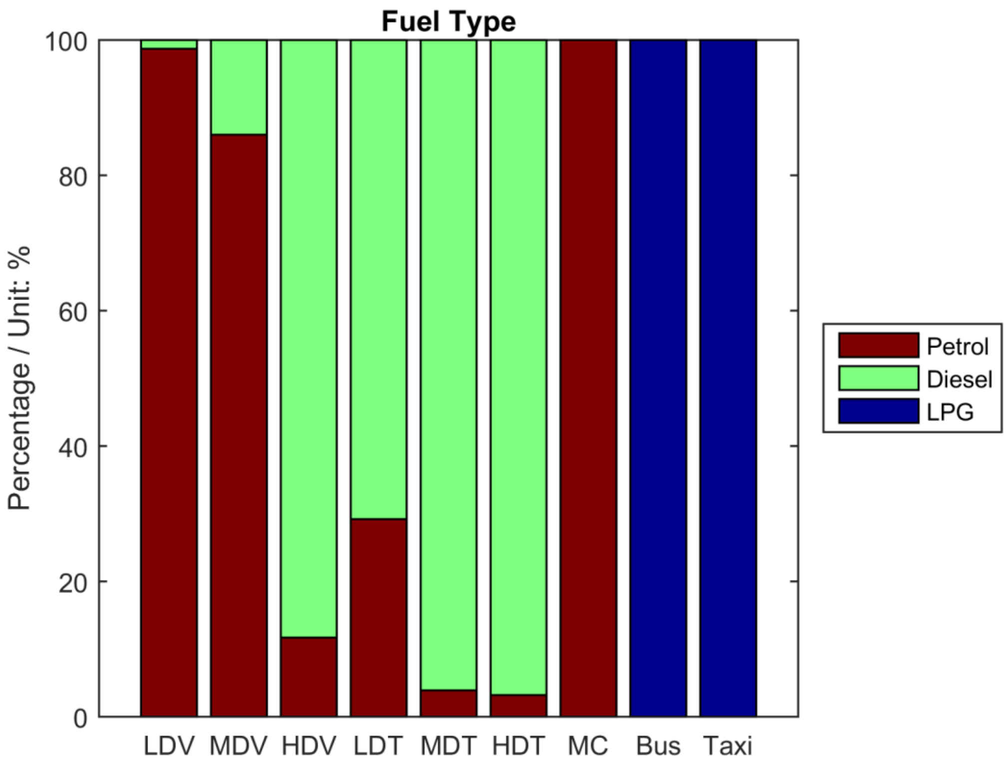

Figure 6. Fuel type percentage of each vehicle classification.

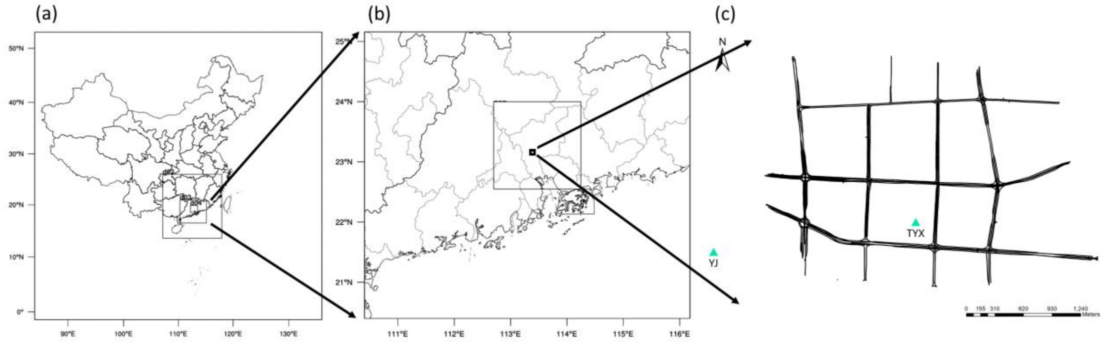

Figure 7. Simulation domain from regional scale to street-level scale: (a) four-nested simulation for WRF; (b) domains 3 and 4 covering the Pearl River Delta Region and Guangzhou city, the innermost box corresponds to the Tianhe District; (c) 31 street segments and two observation sites (triangle) within the MUNICH study domain.

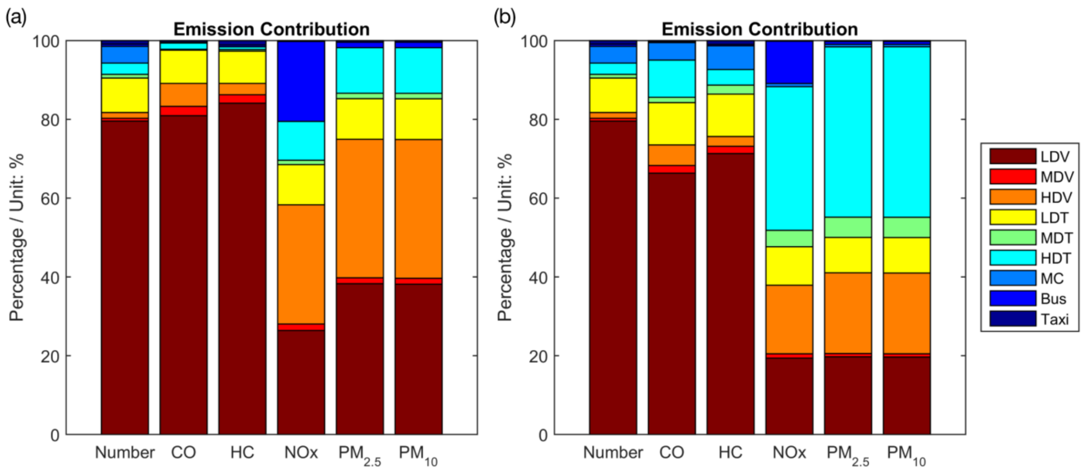

Figure 8. Emission contribution of each vehicle classification in (a) urban area and (b) suburban area.

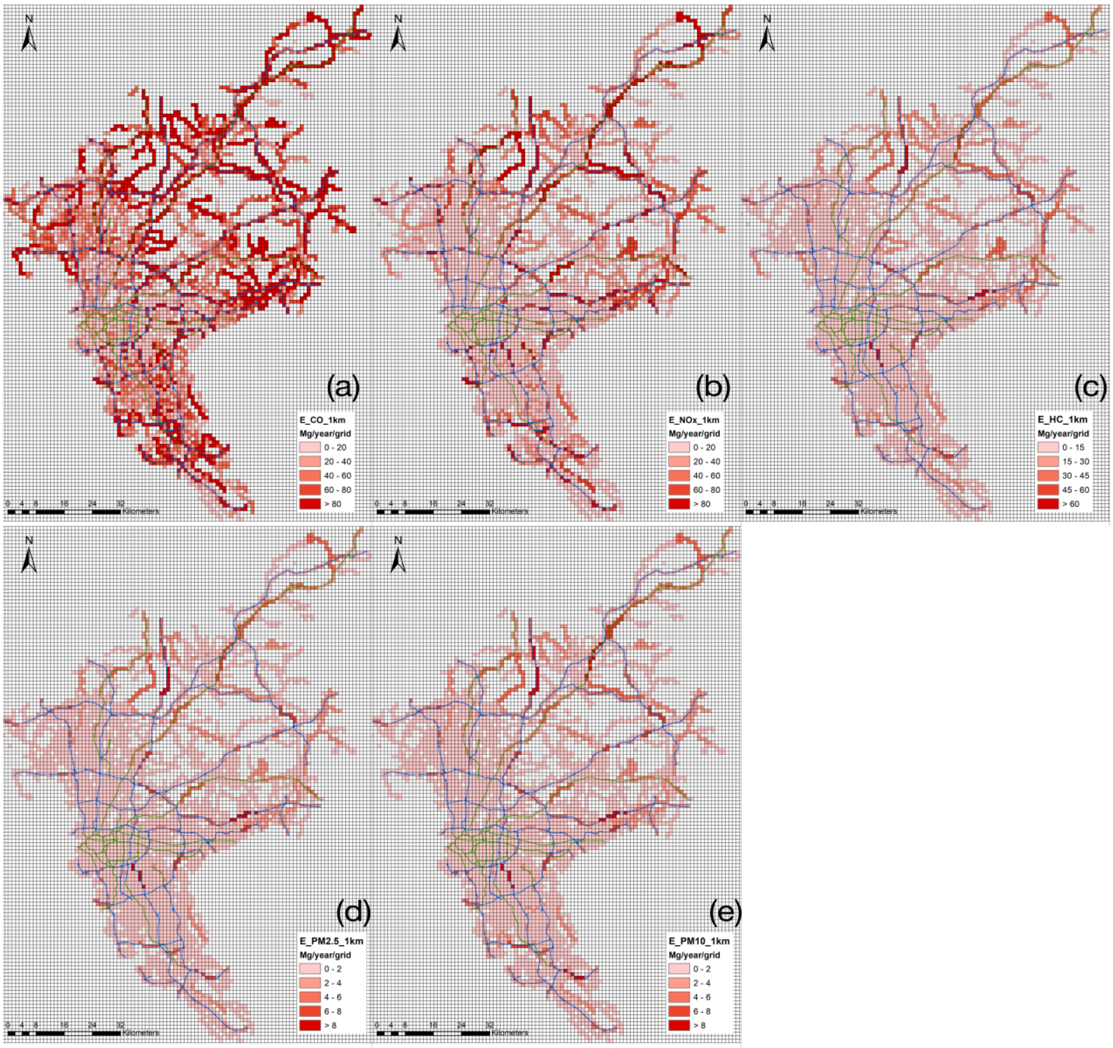

Figure 9. Spatial distribution of (a) CO, (b) NO_x, (c) HC, (d) $PM_{2.5}$, and (e) PM_{10} from the on-road emissions in Guangzhou (blue lines: highways; green lines: arterial roads; local roads are not shown).

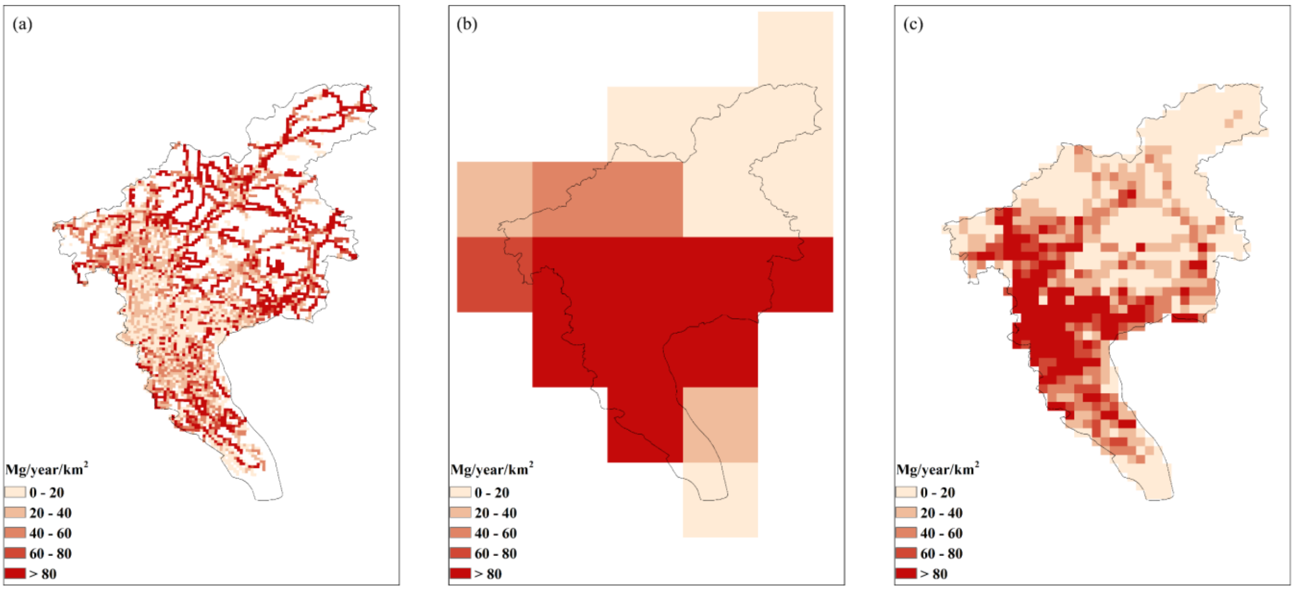

Figure 10. Spatial distribution of CO from (a) ROE model, (b) MEIC-2016 and (c) PRD-2015 in Guangzhou.

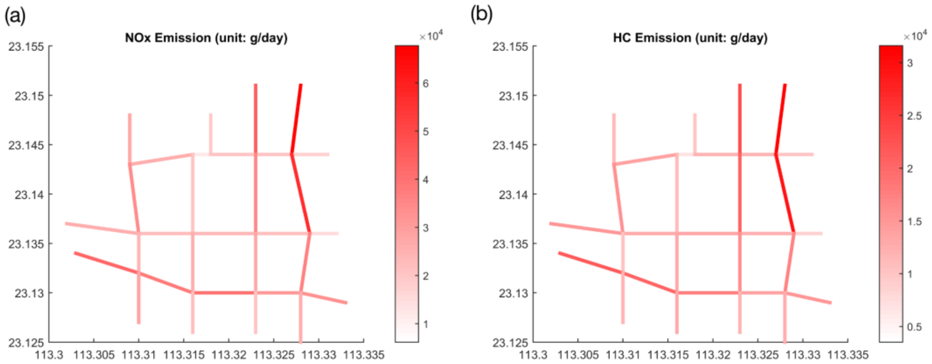

Figure 11. The spatial distribution of weekday (a) NO_x and (b) HC emission in the simulated street network.

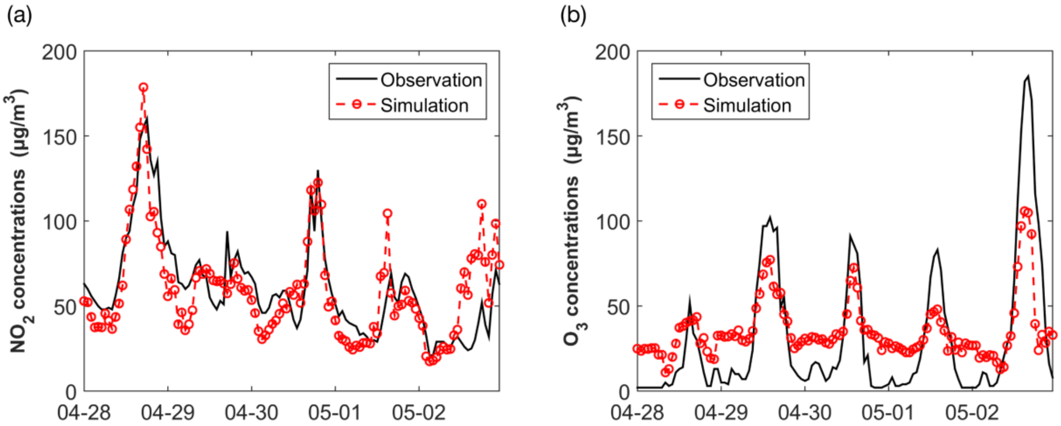

Figure 12. Time series of (a) NO_2 and (b) O_3 during the simulation period. (black solid line: observation; red dashed line: simulation).

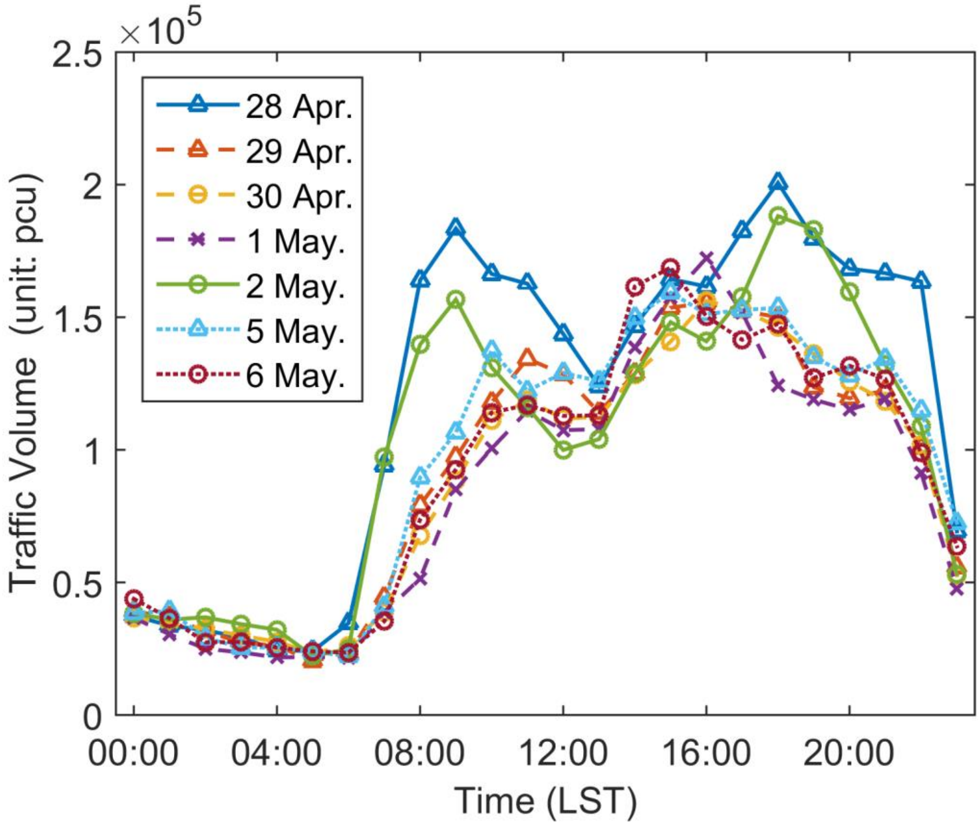

Figure 13. The diurnal variation of the total traffic volume in the simulation street network (solid line: normal weekday; dashed line: national holiday; dotted line: normal weekend).

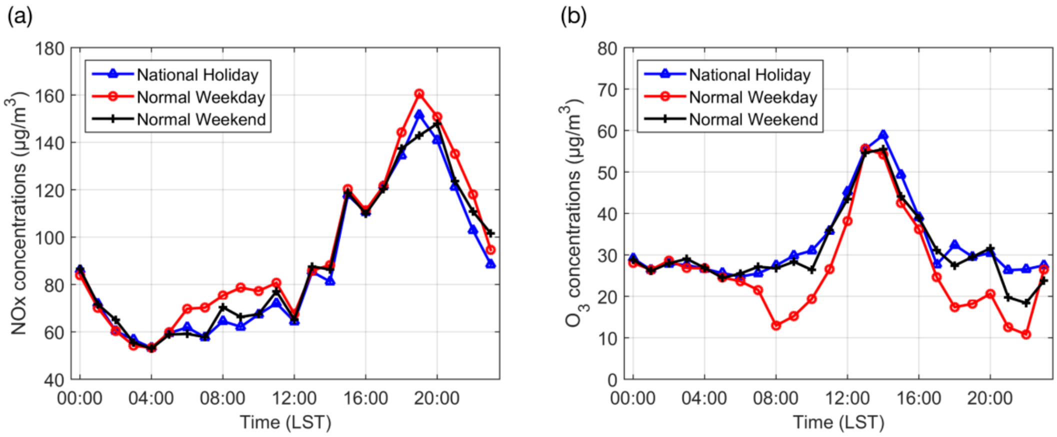

Figure 14. The (a) NO_x and (b) O_3 diurnal variation of different sensitivity cases in the simulation street network