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Interactive comment on "Vehicular emissions in China in 2006 and 2010" *by* N. Chao et al.

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We would like to thank editor and referees for your comments and helpful suggestions. We revised our paper according to these comments and suggestions.

Response to the comments by referee 2

Major Concerns

Question 1:

Page 4907, Line 22-25: The author should precisely clarify whether the standards meant for light-duty vehicles or heavy diesel vehicles. If for light-duty gasoline vehicles, standards should be presented using Arabic numerals instead of Roman numerals, such as "Euro 3" instead of "Euro III". TWC has been a required after-treatment device

since the Euro 1 standard, which was adopted in China starting from 2000. For heavy-duty diesel vehicles in China, they in general didn't apply after-treatment deceives (e.g., SCR or DPF) to comply with the Euro III standards. The author should refer to some appropriate literatures.

Response 1: We are sorry to clarify this point unclearly. The TWC was just a required after-treatment device for light-duty gasoline vehicles, and we have modified on page 4907, Line 22-25 as follow:

To satisfy the Euro 3 emission standards for vehicles, newly registered light-duty gasoline vehicles must install three-way catalytic converters (TWCs).

Question 2: Page 4909 to page 4910: Please substantially streamline the literature review of the vehicular emission inventories. The author should summary the methodologies, implications and potential limitations from those references, rather than list them one by one in the manuscript.

Response 2: In the introduction, we first collected the references both at home and abroad, and then show them in time sequence. At last we summarized the methodologies and potential limitation of those references in Page 4910, Line 19 to Page 4911, and Line 4 in five aspects. We think this is one of the modes to write the literature review.

Question 3: Page 4910, Line 24: Please rewrite standards throughout the manuscript. For example, Euro 1 to Euro 4 is for light-duty vehicles, and Euro âĚă to Euro âĚč for heavy-duty diesel vehicles. In some megacities, such as Beijing, Shanghai and Guangzhou, emission standards implemented there were earlier than the rest of China. The authors need to address this point.

Response 3: We are sorry for our carelessness. The implementation time of Euro 4 emission standard Shanghai and Guangzhou had been reconsidered, and we had recalculated the emissions and modified in the manuscript.

Response 4: Page 4911, Line 9: Why choose two years 2006 and 2010 in this manuscript? I think those two years might be too close. The changes of vehicular emissions would be minor compared to the uncertainties in vehicular emissions. I suggest the authors should extent the study to a longer and continuous period, e. g. 2000-2010.

Response 4: One the one hand, the mission of our project (Grant No.: XDB05020000) is to develop the emission inventory of 2010, after this inventory was developed, we desired to verify the method used our study by comparing with previous study, so we choose one of the acknowledged results of INTEX-B, which was based on the year of 2006. On the other hand, we choose 2006 and 2010 in our study because they are the beginning and end of the 11th Five-Year Plan, respectively. Thanks for your advice for us to extent the study to a longer and continuous period. We can manage to do this in our future work.

Question 5: Page 4912, Lines 14 to 19: The dentition of vehicle classifications is not consistent with the statistical yearbooks in China. Please provide needed description of each vehicle classification and illustrate how to merge the census data into vehicle population by classification used in this study. It is very fundamental to final results. In addition, do the HDDVs only mean coaches and buses? If so, I suggest the authors should present as heavy-duty diesel buses or heavy-duty diesel passenger vehicles to avoid possible misunderstanding, since the HDDVs usually include buses and trucks. Similar to LDGVs.

Response 5: In the statistical yearbook, vehicles are divided into the mini-duty passenger vehicles, light-duty passenger vehicles, medium-duty passenger vehicles, heavy-duty passenger vehicles, mini-duty trucks, light-duty trucks, medium-duty trucks and heavy-duty trucks. In our study we merged the mini-duty and light-duty vehicles as light-duty vehicles and merged the medium-duty and heavy-duty vehicles as the heavy-duty vehicles. Both of the HDDVs and LDGVs in our study mean the passenger vehicles, and we had added the instruction in our manuscript.

Question 6: Page 4913, Line 23 to 4914, Line 2: Vehicle-use intensity indicated by VMT is fundamental to emission inventory. However, the authors assumed national-averaged VMT by vehicle type only based on one literature (i.e., Liu et al., 2008). The data fundamental is very weak and can bring substantial uncertainties in results. The authors should validate their VMT values and their variances, which are essential to simulate the uncertainties by using the Monte Carlo method. For example, I think the authors overrated the VMT values for light-duty vehicles and motorcycles meanwhile underestimated the VMT values for heavy-duty trucks, especially long-haul freight trucks. As a result, such great uncertainties could result in higher CO and NMHC emissions but lower NOX and PM2.5 emissions. VMT for each vehicle classification can vary by province. For example, Beijing adopted the restrictions on vehicle use for motorcycles, trucks and light-duty passenger vehicles. These restrictive policies can significantly influence the VMT for those vehicle classifications. The authors should consider the provincial differences in this study, considering the spatial resolution of emission inventory is emphasized in this study. In addition, the authors should take the deterioration of VMT with vehicle age into account. Namely the VMT for older vehicles (e.g., Euro0) should be lower compared to newer vehicles (e. g, Euro 3, Euro 4). Otherwise, the emission contribution of older vehicles will be overly estimated. I recommend the following paper to the authors for more information and insights. Huo, H., Zhang, Q., He, K., et al. Vehicle-use intensity in China: current status and futuretrend. Energy policy, 2012, 43, 6-16.

Response 6: Thanks for supplying the useful advice and literatures. During our study, we desired to obtain more information of VMT to decrease the uncertainties, but it is pity that few researches were conducted on it, so the VMT in each province is inaccessible. This is really a big problem. We can't agree more that the vehicle age may have influence on the VMT, but the corresponding information is very difficult to obtain. For these reasons, we used national-averaged VKM in our study. This may lead to the uncertainty in our result, but we think it is the best way at present. We will try our best to update our results as soon as we obtain more information of VMT.

Question 7: Page 4914, Section 2.1.3: The authors refer to several studies to determine the distance based emission factors by vehicle classification, fuel type and emission certification level. However, for each vehicle type, the authors relied on just one single study without needed description. Many of the cited references are too old and unsuitable, even much earlier than when the emission standards were implemented in China. That means those estimates cannot be supported or validated by local measurement data, which make the database with substantial uncertainty. I strongly suggest the authors should make a throughout critical review again, particularly to involve recent local measurement results. I wonder whether the emission factors are the same for various provinces. A lot of local features can influence emission factors even vehicles meet a same emission certification level, such as driving cycles, altitude, temperature, inspection and maintenance programs, and fuel quality. The author should clarify the impacts on emission factors of vehicle operating conditions other than emission standards. For example, there were always mismatches between the actual fuel quality and emission standards for many provinces (Zhang et al., 2010). Zhang, K., Hu, J., Gao, S., et al. Sulfur content of gasoline and diesel fuels in northern China. Energy Policy, 2010, 38(6), 2934-2940.

Response 7: Thanks for your suggestions very much. The old references cited in our study are used to obtain the emission factors of vehicle with old emission standards and the new references are used to obtain newly registered vehicles. During our work, we managed to investigate the emission factors in each province, but it is very difficult to obtain. In some previous studies, such as the INTEX-B and TRACE-P, which developed emission inventories on large scale, they also did not take the differences of each province into consideration. Therefore, we have tried our best to do this work. But we will not give up this work, and we will update this part as soon as more investigation information is accessible.

Question 8: Is evaporative HC emissions included for gasoline powered vehicles? The author should clarify. Since for tropical provinces, the evaporative emissions and cold

started missions are substantially different with those in cold provinces. Please reduce significant digits for BC and OC emission factors for all vehicle types. If you cannot achieve that accuracy, they would be meaningless. Is it appropriate to apply same mass ratios of BC or OC for various emission standards? For LDGVs, why the NOX EF for Euro 4 is higher than that for the Euro 3? Is this reduction statistically significant, since it is not consistent with the findings from most measurement results? The authors should carefully review the reference. Furthermore, I think the PM and OC emission factors for LDGVs in China should have been reduced in accord with over the past decade, especially when compare Euro 0 and post-Euro 0 LDGVs. Similar concerns on PM emission factors for other gasoline vehicle categories. The following paper might be helpful to the authors. May, A.A., Nguyen, N.T., Presto, A.A., et al. Gas and particle-phase primary emissions from in-use, on-road gasoline and diesel vehicles. Atmospheric Environment, 2014, 88, 247-260. For LDDVs, the authors referred to a Chinese project report released in 2005. However, considering that the LDDVs are limited in many places of China, I doubt the samples of LDDVs. In addition, theEuro 3 and 4 emission standard was not adopted in 2005. Therefore, I think the authors should consider other studies. Recent studies in Europe indicate that real-world NOX emission factors of diesel passenger cars have not been improved even through the Euro 5, which is one of the most important concerns for urban air quality. Chen, Y., Borken, J. Real-driving emissions from cars and light commercial vehicles–Results from 13 years remote sensing at Zurich/CH. Atmospheric Environment, 2014, 88, 157-164. Weiss, M., Bonnel, P., Hummel, R., et al. On-road emissions of light-duty vehiclesin Europe. Environmental Science & Technology, 45, 8575-8581. For HDVs, the drive cycles for urban coaches and long-distance is completely different. Therefore, urban diesel buses have significantly higher emission factors than long-distance buses with higher average speed. The authors should state how to aggregate two categories of heavy-duty diesel coaches in this study. In particular, the Euro IV emission standards have only urban public fleets like transit buses in Beijing, Shanghai and Guangzhou by 2010. Considering the low-speed driving conditions for urban buses,

the author's might under estimate the NOX emission factors for those Euro IV urban HDDVs. Besides, there are significantly reductions in VOC and CO emission factors of the Euro IV HDDVs compared to the Euro III. Which factors can result in reductions nearly 80%-90%, the authors provide needed information in the table footnote. Please refer to the following papers: Fu, M., Ge, Y., Wang, X., et al. NOx emissions from Euro IV busses with SCR systems associated with urban, suburban and freeway driving patterns. Science of the Total Environment, 2013, 452-453, 222-226. Wu, Y., Zhang, S.J., Li, M.L., et al. The challenge to NOx emission control for heavy-duty diesel vehicles in China. AtmosphericChemistry and Physics, 2012, 12, 9365-9779. For HDT, the trends in NOX in emission factors are awkward. Please explain why NOX emission factor for the Euro IHDT is much lower than those of the Euro II and Euro III. In addition, the Euro IV emission standard has not been adopted in China since 2013. Why the authors estimate the emission factors for the Euro IV HDTs? The following paper might be informative. Huo, H., Yao, Z., Zhang, Y., et al. On-board measurements of emissions from diesel trucks in five cities in China. Atmospheric Environment, 2012, 54, 159-167.

Response 8: Thanks for your advices and references. Firstly, the evaporation of gasoline is out of consideration in our study. The significant digits for BC and OC emission factors for all vehicle types have modified in our manuscript. As for choose of emission factors, although there are some references about it at present, we think it is very unsuitable to apply these separated results to a national scale inventory systemically. For example, we know that the vehicle speed in each condition may have great influence on the emissions, but the traffic flow in the whole China is difficult to obtain, even in separate province. Therefore, simplify the calculation appropriately is a better way to develop under the present condition. Although the simplicity may lead to larger uncertainties, we can calculate a relatively reliable emission inventory completely. By referring some previous inventory on large scale, such as the INTEX-B and TRACE-P, they also could not obtain the data in detail, so they took measures to simplify the calculation as well.

Question 9: Page 4915, Lines 5 to 11: The descriptive text is too wordy and not clear enough. Some equations are needed to illustrate the method. Why should the productions of secondary and tertiary industries be investigated? Were the values in 2006 and 2010 based on a same pricing level? I suggest the authors rewrite the method of spatial location.

Response 9: It is difficult to obtain the population of vehicle in the county level, so we analyzed the correlativity between the population of the vehicles in each province and the primary industry value, the secondary industry value, the tertiary industry value, the sum of primary and secondary industry value, the sum of primary and tertiary industry value, and sum of secondary and tertiary industry value. Then we found that there was good correlation between the population of the vehicles and the sum of secondary and tertiary industry value, we used the sum of secondary and tertiary industry value to allocate the emissions to each county. The values of both years were based on the sum of secondary and tertiary industry value in 2006 and 2010, respectively. We modified this paragraph as follow:

The city-level inventory should be calculated first based on the emissions in each province, by combining with the proportions of vehicles for each city from the China Statistical Yearbook (National Bureau of Statistics, 2006, 2010). Then, the sum of the secondary and tertiary industries values of each county both in 2006 and 2010, which was from the China County Statistical Yearbook, were used to allocate the city-level emissions to each county.(Investigeion general team of rural economic society in State Statistical Bureau, 2006, 2010)

Question 10: Page 4915, Lines 15 to 22: The units for all parameters are missing, which makes the method unclear. For example, is the turnover volume for freight trucks unit in ton*km or veh*km? The authors need to clarify.

Response 10: We are really sorry for the omission of the units of each parameter in Equation (3), so here we added the units as follow:

Where k represents the grade of the road; Fm represents the average traffic density in province m (people or ton); Tm represents the passenger and cargo turnover volume in province m (100 million people×km or ton×km); TLm,total represents the length of transport routes in province m (10 thousand kilometers); Fm,k represents the road traffic density of grade k road in province m (people or ton); and Pm,k represents the proportion of standard daily traffic flow of grade k road(%) (Ministry of Transport of the People's Republic of China, 2004).

Question 11: Page 4916, Line 6: Please provide more information of the SF, such as the way to determine the SF values. Are SF values same for various classes of roads? If the authors use the same SF value for different classes, the impacts of traffic patterns would be ignored. For example, the urban driving conditions are usually congested compared to inter-city driving conditions, which make the emission factors of major air pollutants higher in urban areas.

Response 11: SF represents the standard traffic density, and it is the average traffic density of all the provinces. Zheng et al. proposed a good way to allocate the vehicular emissions on the road, but the traffic flow needed in that method is inaccessible. To make this method more practicable, we use the passenger and cargo turnover volume and the length of transport routes from the China Statistical Yearbook to calculate the road traffic density to characterize the busyness of a road. So traffic density used in our study replacement for traffic flow in Zheng's study.SF is a standard value; Fm,k in Equation (4) means the road traffic density of the road of grade k in province m, and Fm,k can characteristic how busy the road of grade k is; and Am,k means the standard length commutation factor of the road of grade k in province m. Therefore, if the value of Am,k is large the emissions allocated to the road of grade k would be more, otherwise.

Question 12: Page 4918, Lines 7 to 9: Rewrite this sentence. It is not clear. From my understanding, two study years (i.e., 2006 and 2010) are too close. The authors could hardly conclude any statistically discussion the changes of estimated national vehicular

emissions if taking the uncertainty range into account.

Response 12: We are sorry for our poor presentation in our manuscript, and we had deleted this misleading sentence.

Question 13: Page 4917 Line 24 to Page 4918 Line 20: I suggest the authors compare the estimated results for the year 2010 with the Vehicle Emission Annual Report released by the Vehicle Emission Control Center of Ministry of Environment Protection in 2011 (VECC-MEP), since the output frameworks are similar.

Response 13: Thanks for your advice to compare the results for the year 2010 with the Vehicle Emission Annual Report released by the Vehicle Emission Control Center of Ministry of Environment Protection in 2011, and it is really more favorable to compare with our results for the similar output framework. The comparison is discussed in detailed in Response 16.

Question 14: Page 4921, Line 13 to 15: Euro 1 and Euro 2 LDGVs also adopted TWC to control gaseous exhaust pollutants as a principle after-treatment device. Is there any observation could support your result that NH3 vehicular emission intensity in Beijing is higher than other provinces? NH3 emissions from on-road vehicles are very lower compared the other sector. I don't think it is an issue of great significance unless urea-SCR systems are largely adopted for HDDVs.

Response 14: We think there is some misunderstanding caused by digit. We are really sorry for our poor presentation. NH3 emission from agriculture is 13570 Gg, which is much higher than vehicles.

Question 15: Page 4921, Line 16 to 22: Please remove this text from the result section. Furthermore, without link-based traffic flow or traffic demand data, the spatial allocation is only based on the infrastructure information and makes very limited improvement. Therefore, estimated results are still not the actual emissions.

Response 15: The spatial allocation is based on the road traffic density, and this pa-

rameter can describe the busy degree of the road traffic. The definitions of Fm are listed in detail and can be found in the instruction of each index in the Chapter 16 of the China Statistical Yearbook. Therefore, we think our spatial allocation method has made some improvement, and it really can allocate emissions to remote or sparely populated but busy regions, as shown in Fig. 6. For this we just deleted the last sentence "and the results have been demonstrated to approximate the actual road emissions" in Line 21-22.

Question 16: Page 4921, Line 25 to Page 4922, Line 24: The comparative analysis is crucial for inventory studies. However, the authors just listed the visible discrepancies with previous studies. I suggest the authors should carefully dig into the major factors resulting those differences and then review the key input data.

Response 16: Thanks for your advice to do some deep analysis, and we had improved this part as follow:

Vehicular emissions of previous studies, both domestic and international, are summarized and compared with the results of this study in Table 5. As seen from the table, China's total vehicular emissions for each pollutant in 2006 in our study were highly consistent with the inventory developed by Zhang et al., which was one of important achievements of the INTEX-B program executed by NASA and which has been widely acknowledged for its reliability. Specifically, the differences in CO and NOX emissions were within 10.7% and 10.9%, respectively; VOCs and OC emissions were within 4%; and PM2.5 and PM10 were within 1%. In contrast, there was a large difference in BC emissions, up to 30.7%, which may have been caused by the uncertainties in emission factors. Since few researches had exerted on the vehicular emissions of BC emission factors, our emission factors of BC were calculated according to the fraction of these three particulate matters in relation to the total PM10 (Streets et al., 2001), this may be lead to the difference with the result of INTEX-B. A glaring discrepancy has emerged between the results of our study and RAINS. This discrepancy may caused by the method. RAINS calculated the vehicular emissions based on fuel consumption

estimated by the RAINS-ASIA module, which used the fuel consumption in 1990 to predict the consumption in 2010. Fuel consumption based method was just used in early times to estimate the emission, and it inevitably led to uncertainties. The emissions in 2010 estimated in our study did not agree well with results calculated by Zhao et al. (except the NOX emission) either, because emission factors given in their paper were different from ours and there were some differences in vehicle classifications. For example, the unit of emission factor in Zhao's study is kg/tce, while in our study is kg/km. The Diesel and Gasoline car are divided separately in Zhao's study, but not in our study. Besides, the population of vehicles with each emission standards in our study is calculated by the newly registered vehicles, but no detail is described in Zhao' s study, and only the proportions of vehicle with each emission standards is shown in the supplement. Compared the estimated results for the year 2010 with the Vehicle Emission Annual Report released by the Vehicle Emission Control Center of Ministry of Environment Protection in 2011 (VECC-MEP), we found that our results were close to the VECC-MEPs', except the VOCs. However, RVs were included in our study, that is to say that emissions calculated in our study were smaller than the VECC-MEPs'. Since the source of the populations of vehicles and classifications were similar, these differences could be attributed to emissions factors and VKM. Vehicles emissions in BTH, YRD, and PRD regions had been calculated by some scholars and were also showed in table 5. Compared with these studies, differences could be found. These might have been caused by the sources of the VTM. In Zheng's study, the fuel ratio and VTM were from investigation and questionnaires. The VKM of trucks in our study was a little less than Zheng' s study, but the VKM of MC was larger than Zheng' s. Although the source of fuel ratio between Zheng's study and ours was different, there was little difference. It should be noted that emissions from RVs were significant but were neglected in Lang's and Huang's study, and the emission standards were out of consideration in Zheng' s study. Considering the old technology of vehicles registered earlier, emission factors of Euro 0 vehicles in our study were revised by referring previous studies, as discussed in Section 2.1.3. Therefore, big differences could be found

in CO, NOX, and VOCs emissions, compared with these three studies. Emissions on the city scale were also compared in this study. For example, emissions in Hangzhou in our study were close to Wang's results, whereas emissions in Guangzhou and Xi'an were not. NH3 emission was just involved in Zhao' study, but it was much lower than our result, and this mainly caused by the differences in emission factors. At present, few researches have been conducted in vehicular emission of NH3, and this may lead to the underestimation of total NH3 emissions, therefore, more researches should be done on it.

Question 17: Page 4925, Section 4.3: During the 11th Five Year Plan Period, anthropogenic NOX emissions from other sectors (e.g., power plants, cement industry, etc.) were also substantially increased due to the absences of the total emission reduction target. Based on the MEP' s estimate, estimated NOX emissions from on-road vehicles contributed approximately 26% of China's total anthropogenic NOX emissions, which was less than those from power plants. Only in the urban areas where power plants and industries are scare (e.g., Beijing) can the vehicular emissions dominate the source of NO2. Therefore, the observed trends in NO2 concentration cannot be just attributed to vehicular emissions.

Response 17: It is difficult to evaluate the rationality of the spatial distribution, so we had to use the satellite data, which covered a wide range of China, to discuss the results generally. We really found some differences between the satellite data and our results, for example, the vehicular emissions in Tianjin, south of Hebei, and north of Shandong were less significant in 2010 compared with satellite data. To understand these differences further, we collected the coal consumption, which is also one of the most important contributors of fossil energy of NOX emissions. The statistical data showed that coal consumption in these areas ranked within the top five of the whole China.

Question 18: Page 4925 Line 15 to Page 4926 Line 5: The author should quantitatively assess the benefits from implementation of tightened emission standards. Emission

control scenarios could be designed with various implementation date and certification level of emission standards taken into consideration. When it comes to the vehicular emission control in Beijing, control strategies and measures adopted were much more comprehensive than other regions. The author could not count the benefits merely on the role of stricter emission standards. The authors should improve their understanding of the vehicular emission regulations in China (Table 6). They for light-duty vehicles, heavy duty diesel vehicles, heavy-duty gasoline vehicles and motorcycles, not for gasoline and diesel!

Response 18: Thanks for your advice to design the emission control with various implementation data and certification level of emission certification level of emission standards, and these works are really of great significance. We can manage to do some researches by following these thoughts in our future work. The emphasis of this study is to calculate the vehicular emissions and describe characteristics of the emissions, so we did some preliminary analysis to find out how the oil quality and emission standards had impact on vehicular emissions. We hope that this preliminary analysis would support some theoretical foundation to the vehicular emissions reduction, and enlighten us on our further researches.

Question 19: Page 4926 Lines 13 to 23: There was very significant mismatch between the actual sulfur content and claimed fuel quality in China (Zheng et al., 2010). As a result, the authors overrated the benefit of emissions reductions from improved fuel quality. Zhang, K., Hu, J., Gao, S., et al. Sulfur content of gasoline and diesel fuels in northern China. Energy Policy, 2010, 38(6), 2934-2940.

Response 19: Thanks for supplying the literatures for us. Oil quality improvement indeed could reduce the pollutant emissions as discussed in this paper: Liu, H., He, K., He, D., Fu, L., Zhou, Y., Walsh, M., and Blumberg, K.: Analysis of the impacts of fuel sulfur on vehicle emissions in China, Fuel, 87, 3147-3154, 2008. So we did some preliminary analysis to find out how the oil quality and emission standards had impact on vehicular emissions. We hope that this preliminary analysis would support some

theoretical foundation to the vehicular emissions reduction in the future. More deep researches may conduct in our following work.

Question 20: Page 4927, Section 4.5: The uncertainties in vehicular emissions depend on the probability functions of key input data, such as emission factors, vehicle population by type, annual VMT. The authors should clarify the probability functions for those above input data, since the references cited here didn't provide detailed information. Otherwise, the uncertainty analysis becomes a kind of statistical play based on I also think it is strange and possibly misleading that the relative uncertainty range in 2010 is a bit wider than 2006. Usually, the relative uncertainty range would be narrowed with the improvement of vehicle emission control and data collection. I suggest the authors could read the following paper: Kioutsioukis, I., Tarantola, S., Saltelli, A., et al. Uncertainty and global sensitivity analysis of road transport emissions estimates. Atmospheric Environment, 2004, 38, 6609-6620.

Response 20: Thanks for supplying the literatures for us, and they really help us to understand the uncertainty analysis deeply. The uncertainty range in 2010 is a bit wider than 2006 because the population of RVs in 2010 is inaccessible in our study, so we just assumed that the population of RVs changed little from 2006 to 2010, and this might lead to uncertainties in 2010 inevitably. The probability functions is: f<x|μ,δ>=1/($\delta\sqrt{2\pi}$)(exp((-(lnx-μ)^2))/(2δ^2))/x

Minor Comments

Question 21: Page 4907, Line 5: The author should clearly note whether motor-cycles and rural vehicles included in the statistical data here.

Response 21: We are really sorry for the poor presentation in our manuscript, and we had added the note in the corresponding part. Motor-cycles and rural vehicles did not include in the statistical data.

Question 22: Page 4907, Lines 6-7: should be "within 12 years". Please present in an

appreciate way throughout the manuscript, such as Page 4908, Line 10.

Response 22: We are really sorry for the poor presentation in our manuscript, and we had modified in the corresponding part.

Question 23: Page 4907, Line 6-7: Does the NOx mean emissions or ambient concentration? The author should clarify this point. In addition, I strongly suggest that the authors should refer to more recent studies regarding the characteristics of vehicular emission in China.

Response 23: We are really sorry for the poor presentation in our manuscript, and here means emission. We had modified in the manuscript. In order to avoid duplication, we did not add some references here, because more studies were referred in the Page 4908, Line 6 - Page 4910, Line 18.

Question 24: Page 4907, Lines 12-15: This sentence read very awkward. Please rewrite it in a clear way and add some needed concrete data.

Response 24: We rewrote this sentence and added some data as follow:

The Beijing-Tianjin-Hebei region, the Yangtze River Delta, and the Pearl River Delta, which cover only 2.3%, 2.2%, and 1.9%, respectively, of the Chinese territory, generated about 10%, 19%, and 12%, respectively, of the total emissions in 2005, and became the most polluted regions.(Cai, H. and Xie, S.: Estimation of vehicular emission inventories in China from 1980 to 2005, Atmospheric Environment, 41, 8963-8979, 2007.)

Question 25: Page 4907, Line 17-27: In general, the carbon monoxide is a relatively stable pollutant category. Its contribution to fine particles ozone is considered minor compared to NOx and active species of hydrocarbons. In addition, the emissions of NH3 from vehicles are much lower compared to other process, such as agricultural activities. The authors should improve the understanding of pollutant emissionsand major reactions in the atmosphere, and avoid any wordy description.

Response 25: We are sorry for making such mistake for our carelessness, and deleted the carbon monoxide in this sentence. As for NH3 emissions from vehicle, few researches have exerted in China, however, some foreign scholars have revealed that NH3 emissions from gasoline-fueled vehicles have become an important source of pollution affecting urban air chemistry, and NH3 emissions changed in different driving cycles. More related literatures are listed in Response 38.

Question 26: Page 4907, Line 25-27: Please clarify the mass contribution of nitrates and sulfates in PM2.5 for China's megacities? The paper listed as below might be helpful to the authors. Yang, F., Tan, J., Zhao, Q., et al. Characteristics of PM2.5 speciation in the representative megacities and across China. Atmos. Chem. Phys., 2011, 11, 5207- 5219.

Response 26: Thanks for your advice for us to clarify the mass contribution of nitrates and sulfates in PM2.5 for China's megacities, and the paper you supplied for us is very useful and can make our introduction more comprehensive. We had added the contribution in Page 4907, Line 27 as follow:

At both urban and rural sites in the eastern region, the sum of sulfate, nitrate and ammonia typically constituted much higher fractions (40–57 %) of PM2.5 mass.

Question 27: Page 4907 Line 27 to Page 4908 Line 1: Please rewrite this sentence.

Response 27: We had rewritten this sentence as follow:

In January 2013, five times of heavy hazy pollution broke out in the Beijing-Tianjin-Hebei (BTH) region, and . . .

Question 28: Page 4908, Line 2: Why estimating vehicular emissions is convenient? Compare to which methods?

Response 28: We think that you misunderstand the mean of this sentence for our poor presentation, so we rewrite this sentence as follow:

Therefore, calculating vehicular emissions can help us to describe the characteristics of regional vehicular pollution in China clearly and . . .

Question 29: Page 4910, Line 20-21: Please provide some information about the change in vehicle stock and emission certification.

Response 29: We think that the change in vehicle stock is discussed in Page 4907, Line 5-9, and the emission certification is discussed in Section 4.4, so in order to avoid duplication, we did not discuss here.

Question 30: Page 4911, Line 1: should be "uncertainties because of the lack" Response 30: We had modified in the manuscript.

Question 31: Page 4913, Line 4: "emission standards" instead of "emissions standards"

Response 31: We had modified in the manuscript.

Question 32: Page 4913, Line 7: "complying with" instead of "with"

Response 32: We had modified in the manuscript.

Question 33: Page 4913, Line 10: delete "the total population and"

Response 33: We are sorry for our carelessness in our manuscript. We had rewritten this sentence as follow:

Therefore, the vehicle population by emission standards can be calculated by knowing the total population of the vehicles and the population of newly registered vehicles.

Question 34: Page 4913, Line 13: please rewrite the second half of that formula, which seems awkward, e. g., when j=Euro 0, meanwhile j=1 to 4

Response 34: Thanks for your advice, and we modified in the manuscript.

Question 35: Page 4913, Line 15: what is the vehicle "type", defined by vehicle classification or fuel type? Please clarify it.

Response 35: We are sorry for our poor presentation. The vehicle “type” here was defined by vehicle classification.

Question 36: Page 4913, Line 18-20: why the calculated population of Euro 0 vehicles can be negative? Some explanations are needed.

Response 36: These suppose was proposed before we calculated the population, because we were afraid that the error of statistical data might lead to this phenomenon. But in fact there was no negative data produced during our calculation, so we think that this suppose should delete now. We had deleted it in the manuscript.

Question 37: Page 4915, Line 21: Please clarify the traffic flow pattern for each road class.

Response 37: The traffic flow pattern for each road class had added in Page 4915, Line 23, as follow:

The roads are divided into four grades: express way, arterial road, sub arterial road and branches.

Question 38: Page 4917, Line 1 to 16: This is very simple and tradition cell gridding technology. The author may drop off this part.

Response 38: We think that we’d better not drop off this part to make this method of spatial distribution more complete.

Question 39: Page 4918, Line 5: please add some references to illustrate how the application of TWC influences the NH3 emissions from vehicles.

Response 39: We had added the following references to Page 4918, Line 5:

Durbin, T. D., Wilson, R. D., Norbeck, J. M., Miller, J. W., Huai, T., and Rhee, S. H.: Estimates of the emission rates of ammonia from light-duty vehicles using standard chassis dynamometer test cycles, Atmospheric Environment, 36, 1475-1482, 2002. Heeb, N. V., Forss, A.-M., Brühlmann, S., Lüscher, R., Saxer, C. J., and Hug, P.: Three-way

catalyst-induced formation of ammoniaâĂŤvelocity- and acceleration-dependent emission factors, Atmospheric Environment, 40, 5986-5997, 2006a. Heeb, N. V., Saxer, C. J., Forss, A.-M., and Brühlmann, S.: Correlation of hydrogen, ammonia and nitrogen monoxide (nitric oxide) emissions of gasoline-fueled Euro-3 passenger cars at transient driving, Atmospheric Environment, 40, 3750-3763, 2006b. Heeb, N. V., Saxer, C. J., Forss, A.-M., and Brühlmann, S.: Trends of NO-, NO2-, and NH3-emissions from gasoline-fueled Euro-3- to Euro-4-passenger cars, Atmospheric Environment, 42, 2543-2554, 2008. Kean, A. J., Littlejohn, D., Ban-Weiss, G. A., Harley, R. A., Kirchstetter, T. W., and Lunden, M. M.: Trends in on-road vehicle emissions of ammonia, Atmospheric Environment, 43, 1565-1570, 2009. Perrino, C., Catrambone, M., Bucchianico, A. D. M. D., and Allegrini, I.: Gaseous ammonia in the urban area of Rome, Italy and its relationship with traffic emissions, Atmospheric Environment, 36, 5385-5394, 2002.

Question 40: Page 4919, Line 3: It is not correct. For example, the Euro 4 emission standard was adopted in Guangzhou just before the Asian Game in 2010 (also in Shanghai). When it comes to the emission standards, the authors should note for which category, such as light-duty or heavy-duty diesel.

Response 40: We are sorry for our carelessness. The implementation time of Euro 4 emission standard Shanghai and Guangzhou had been reconsidered, and we had recalculated the emissions and modified in the manuscript.

Question 41: Page 4919, Line 11: “phasing out” instead of “eliminating”; “complying with” instead of “with”

Response 41: We had modified in the manuscript.

Question 42: Page 4919, Line 18: “fleet configuration” instead of “vehicle composition”

Response 42: We had modified in the manuscript.

Question 43: Page 4921 Line 11: Please define all the geographic terms, such as

Sichuan Basin, Urumqi regions?

Response 43: We had modified as follow:

The three provinces of northeastern China, the Sichuan Basin (the east of Sichuan Province and the west of Chongqing City), Wuhan and Urumqi, ...

Question 44: Page 4924, Line 1 to 3: Please add some necessary references.

Response 44: We had added the following references to Page 4924, Line 3:

Zhang, Q, Geng. G. N, Wang. S. W, et al., Satellite remote sensing of changes in NOx emissions over China: 1960-2010, Chin Sci Bull, 57, doi: 10:1007/ s11434-012-5015-4, 2012. Zhang, X. Y, Zhang, P, Zhang, Y, et al., The trends, temporal and spatial distribution and source of the NO2 in troposphere in China in recent 10 years, Science in China Press, 37, 1409-1416, 2007.

Question 45: Page 4924, Section 4.2: I suggest the authors should clarify the dentition of oil consumption. Gasoline, diesel or even including other petroleum products (e. g., kerosene)? Consumption for transportation sector total consumption?

Response 45: The gasoline and diesel were included in the oil consumption used in this section, and we added the corresponding introduction in section 4.2. Consumption of gasoline and diesel are accessible in the China Energy Statistical Yearbook compiled by the Department of Industry and Transport Statistics.

Question 46: Page 4923, Line 21 to Line 26: There are many other control measures adopted in Beijing and Shanghai in addition to tightening emission standards, to improve emission factors of air pollutants. Besides, the emissions standards adopted in Jiangsu were consistent with national requirements by 2010.

Response 46: Beijing and Shanghai took measures to control the vehicular emissions, for example, urea-SCR systems are largely adopted for buses, and speeded up the upgrade of vehicles without emission standards an so on. In our study, the upgrade of

vehicles without emission standards was taken into consideration, but the urea-SCR is out of consideration because there are few emission factors to refer at present. We are sorry to make a mistake that the emission standards adopted in Jiangsu were not consistent with national requirements, and we had modified it.

Question 47: Page 4925 Line 7-15: Please remove this paragraph. It just repeats the results presented upfront.

Response 47: We think that this paragraph serves as a connecting link between the preceding and the following part, so it is better to keep it. We just simplified it as follow:

Compared with 2006, the total fuel consumption of China in 2010 increased by 46.6% on average, and the total number of Euro 0 to I vehicles decreased by 6.5%, whereas the total number of other emission standard vehicles rose by 385.6%. However, a small increase was shown in vehicular emissions, as discussed in Section 3.1. Driven by curiosity, further work was conducted.

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