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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Development of a high temporal–spatial resolution vehicle emission inventory based on NRT traffic data and its impact on air pollution in Beijing – Part 2: Impact of vehicle emission on urban air quality" *by* J. J. He et al.

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1. My main concern is about the ammonium aerosol considered in this study. I do not see how NH3 is included the emission inventory. As we know, NH3 emissions from agriculture and other sectors cannot be ignored in East China, and the mass loading of ammonium should be a major contribution to PM2.5 in North China. On the other hand, the NH3 is an important factor that determines the formation of nitrate, which

is another major aerosol species in PM2.5, through the reaction below: NH3 (gas) + HNO3 (gas) NH4NO3 (solid). Therefore, if the emission of NH3 was not correctly considered in this study, the simulation about nitrate and ammonium would be deeply affected. This will lead to two significant uncertainties: One is the uncertainties in the simulation of the total mass burden of PM2.5 in model domain. Even in Beijing, the transport of ammonia from surrounding region is important as well. Another is the uncertainties of the nonlinearity in the processes of nitrate formation. Nitrate is a secondary aerosol component. The nonlinearity means if we cut down 50% the precursor NOx, the variation of secondary aerosol nitrate may not decrease 50% as well, and sometimes they can be enhanced (Burr and Zhang, 2011, APR). What's more, NOx is the major pollutant emitted from vehicle sources. Thus, the sensitivity tests in this study may provide unreasonable results because of the lack of description of ammonium. I suggest the authors conduct the simulation works with nearly compiled NH3 emissions from Song Yu (Beijing University), and the simulation results of nitrate should be provided at least, as it is the main secondary pollutant of vehicle sources.

Response: Source apportionment based on air quality numerical model includes source sensitivity simulations using the brute force method (also referred as zero-out method) or the decoupled direct method (DDM), air pollution tagged method, and the adjoint method, which was detailedly descripted in the response of question 1 of referee 1. In pervious study, the impact of Beijing local emission on air pollution is almost linear via source sensitivity analysis (An et al., 2007). Sensitivity analysis is suitable to investigate the contribution of vehicle emission in Beijing due to the limited change of emission in this study. NH3 emission is an important component in CUACE emission inventory (Fig. 1), and it is comparable with previous study (Zhao, 2007). The revised manuscript adds the analysis of the influence of vehicle emission on ammonium (NH4+) and nitrate (NO3-) (Table 5). The chemical components of PM2.5 represents the characteristics of emission source and complexity chemical processes of pollutant in atmosphere. Based on sensitivity test, the VECs of BC, OC and NI are large, while relative small for SF, and AM. The VECs of BC and OC in December are approxi-

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mately twice of that in July. Seasonal changes for the rates of BC and OC from vehicle emission in total emission are inapparent which indicates that it is not the reason for seasonal change of VECs. Beijing is controlled by southerly wind dominantly, which results in significant regional transport. And it causes small (large) VECs of BC and OC in summer (winter). Atmospheric chemical processes and dispersion conditions are also the reason for seasonal change of different components VECs. Using MM5-CMAQ model simulation, Cheng et al. (2013) investigated the VEC to the PM2.5 and found the VEC of BC was 32.3% and 30.7% in summer and winter respectively. Our results are comparable with Cheng et al. (2013) in winter, while show some difference in summer.

Reference: An, X., Zhu, T., Wang, Z., Li, C., and Wang, Y.: A modeling analysis of a heavy air pollution episode occurred in Beijing, Atmos. Chem. Phys., 7, 3103-3114, doi:10.5194/acp-7-3103-2007, 2007. Zhao, B.: The research of air pollution source emission for the north China. Chinese Academy of Meteorological Sciences, Master's Thesis, 2007.

Changes in manuscript:The details were supplied in section 2.2 and 3.2.

2. Section 2.1, more description about the model should be provided, especially the chemical part.

Response: More description of CUACE model has been provided in revised manuscript. Gaseous chemical is based on the Regional Acid Deposition Model (RADM) covering 66 gaseous species (Stockwell et al., 1990; Wang et al., 2015). Aerosol module includes mixing scheme, clear-sky processes, dry deposition, below-cloud scavenging, in-cloud processes. Seven aerosol species, i.e. sulfates, soil dust, black carbon, organic carbon, sea salts, nitrates, and ammonium salts are considered in aerosol chemical module. The details of sulphur chemistry, cloud chemistry, coagulation, nucleation, condensation etc. were depicted by Gong et al. (2003).

Reference: Gong, S. L., Barrie, L. A., Blanchet, J. P., von Salzen K., Lohmann, U.,

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Lesins, G., Spacek, L., Zhang, L. M., Girard, E., Lin, H., Leaitch, R., Leighton, H., Chylek, P., and Huang, P.: Canadian aerosol module: a size-segregated simulation of atmospheric aerosol processes for climate and air quality models 1. Model development, J. Geophys. Res., 108, 4007, doi:10.1029/2001JD002002, 2003. Stockwell, W. R., Middleton, P., Chang, J. S., and Tang, X.: The second generation regional acid deposition model chemical mechanism for regional quality modeling, J. Geophys. Res., 95 16343-16376, 1990. Wang, H., Xue, M., Zhang, X. Y., Liu, H. L., Zhou, C. H., Tan, S. C., Che, H. Z., Chen, B., and Li, T.: Mesoscale modeling study of the interactions between aerosols and PBL meteorology during a haze episode in Jing-Jin-Ji (China) and its nearby surrounding region-Part 1: Aerosol distributions and meteorological features, Atmos. Chem. Phys., 15, 3257-3275, doi:10.5194/acp-15-3257-2015, 2015.

Changes in manuscript: More description of CUACE model has been provided in section 2.1.

3. Page 19244, line 9, "ammonia"? If here means one aerosol species, I think it should be ammonium. And how does the model treat this aerosol?

Response: It has been modified in revised version. As mentioned above, aerosol module includes mixing scheme, clear-sky processes, dry deposition, below-cloud scavenging, in-cloud processes. The aerosol size spectrum was divided into a number of bins. Fig. 2 shows the flowchart of aerosol module. The details of sulphur chemistry, cloud chemistry, coagulation, nucleation, condensation etc. were depicted by Gong et al. (2003).

Reference: Gong, S. L., Barrie, L. A., Blanchet, J. P., von Salzen K., Lohmann, U., Lesins, G., Spacek, L., Zhang, L. M., Girard, E., Lin, H., Leaitch, R., Leighton, H., Chylek, P., and Huang, P.: Canadian aerosol module: a size-segregated simulation of atmospheric aerosol processes for climate and air quality models 1. Model development, J. Geophys. Res., 108, 4007, doi:10.1029/2001JD002002, 2003.

4. Page 19244, line 19, I cannot find the reference Li et al., 2013, should be 2014?

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Response: Thanks for your reminding. It has been corrected in revised revision.

5. Page 19248, line 10-24, why the authors used the evaluation results of previous studies? All of them were the results in 2008? The evaluation of simulation results in 2013 should be provided here.

Response: The performance of mesoscale meteorological model evaluated in recent years has been supplied in revised manuscript.

6. Page 19249, why not present the evaluation results in each observation stations? Why just presents the average results in Figure 2? More details can be seen if provided the evaluation results in each observation stations.

Response: Except for temporal trends, the evaluation of spatial distribution of NO2 and PM2.5 has been conducted in Fig. 3 (revised manuscript). It is found that CUACE can well reproduce temporal trends as well as spatial distribution characteristics of NO2 and PM2.5 concentrations. The performance statistics of hourly concentrations in July and December was supplied in supplement file.

7. Page 19254, line 16-17, switch off/on one emission sector would also change the back ground pollutant concentrations and chemical processes. This point is similar with "zero-out" method. And this is beneficial to capture the nonlinear relationship between precursors and secondary pollutants. Here the statement should be modified.

Response: Thanks for your reminding. It has been corrected in revised revision.

8. In Figure 2(d), I did not see the blue line. Does it coincide with the red one?

Response: Minor difference of PM2.5 concentration is observed between SIM1 and SIM2 due to little vehicle emission change (Table 3).

9. In abstract and at Page 9 Line 11, it is noted that the update emission HTSVE used in this study was presented in Jing et al. (2015). Actually, I cannot get any volumes and issues information of Jing et al. (2015) from the REFERENCE part (Page 19 Line

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12). The author should give the right citation of Jing et al. (2015).

Response: It has been corrected in revised revision.

10. The boundary conditions used in the simulations is from McKeen et al. (2002). I think this is inappropriate and may result in underestimates of the gas and aerosol concentration. The McKeen initial and boundary conditions are for the US. Western BC over the eastern Pacific Ocean and will be very low. I suggest rerunning the simulations using BC obtained from output from a global model.

Response: Thanks for referrer's advice. The accuracy of chemical boundary conditions is an important factor in regional air pollution numerical simulations. Even the boundary condition from global model has inherent errors. In this study, chemical boundary conditions were the default profile in WRF-Chem, which was widely used in recent studies (Gao et al., 2011; Zhang et al., 2015). On the other hand, the model is configured to have three nested domains to weaken the impact of boundary condition. Thirdly, the outer domain is large enough to reduce spurious boundary effects in the inner domain. Fourthly, the extra 10 day run (i.e. 21st June to 30th June, 21st November to 30th November) was conducted to reduce the effect of chemical initial and boundary conditions. Lastly, the major concern of this study is effect of vehicle emission on Beijing's air quality which mainly affected by the accuracy and the rate of vehicle emissions. Based on the reasons mentioned above, I think that the setting of boundary and initial conditions is reasonable thought these have some inherent uncertainties. And I also agree that the uncertainties of chemical boundary conditions need more deeply investigation, especially in regional transport research.

Reference: Gao, Y., Liu, X., Zhao, C., and Zhang M.: Emission controls versus meteorological conditions in determining aerosol concentrations in Beijing during the 2008 Olympic Games. Atmos. Chem. Phys., 11, 12437-12451, doi:10.5194/acp-11-12437-2011, 2011. Zhang, L., Jin, L. J., Zhao, T. L., Yin, Y., Zhu, B., Shan, Y. P., Guo, X. M., Tan, C. H., Gao, J. H., Wang, H. L.: Diurnal variation of surface ozone in mountainous

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areas: case study of Mt. Huang, East China. Sci. Total Environ., 538, 583-590, 2015.

11. This study only evaluates the site average concentration of NO2 and PM2.5. However, the vehicle emission and other emissions are different at different sites. In addition, only comparison of NO2 and PM2.5 are still limited. I think it is necessary to present the comparison of model results with observations at each site and add comparison of other gas and aerosol concentration (e.g., NO, O3, NO3-, SO42-, BC, OC).

Response: Thanks for your advice. The manuscript has been modified (Table 5).

12. At Page 11 Line 12, what is the reason for the low correlation of NO2? Is it related to the uncertainty of emissions or gas chemistry? The author should explain more for this.

Response: The uncertainty of emission inventory increases with the spatial resolution of numerical model, and it is one of the reasons for simulated bias because of high spatial resolution (3km). The rate of NOx emission from vehicle in total emission has a slightly seasonal change (Table 3). So the uncertainty of photochemical reaction which is more significant in summer (especially for NOx) might result in large bias compared to the performance of NO2 in winter.

Changes in manuscript: More explain are supplied in section 3.1.

13. In Table 2, please add the update emission HTSVE. The CUACE emission is very different with other studied especially for CO and NOX according to Table 2, what about HTSVE? The author should discuss about the resulting uncertainty in this study.

Response: Thanks for your advice. It has been modified in Table 2. The uncertainty has been discussed in revised manuscript.

Changes in manuscript: It was discussed in section 2.3.

14. The author should present the comparison of meteorological condition and the statistical analysis in supplement file.

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Response: The evaluations of meteorological conditions and air pollution are provided in supplement file with the form of statistical analysis.

15. At Page 14 Line 1, please explain more about Figure 7.

Response: low VEC presents in serious pollution, while high VEC presents in low pollution concentration level, especially for NO2. The absolute contribution of vehicle emission increases in severe pollution mostly because of adverse dispersion condition. However, pollutant regional transport is enhanced in severe pollution, which results in negatively correlation between VEC and pollution concentration level.

Changes in manuscript: More explain was listed in section 3.2.

16. Please discuss more about Figure 8 and 9 with distribution of vehicle emission and wind direction.

Response: The spatial distribution of RVEC are tremendously affected by vehicle emissions, as they are mostly consistent with the rate of vehicle emission in total emission (Fig. 4, revised manuscript). As pointed by Jing et al., (2015), the uncertainty of HTSVE is very small through multiple comparison with statistical data and real time observation. But the uncertainty of other sector emissions has a negative influence on the precision of RVEC, which need more improvement for accurate environmental management. Local circulation also determines the spatial distribution of RVEC. High PM2.5 emission from vehicle is found between north Fourth Ring Road and north Five Ring Road (See Part. 1, Fig. 9). Controlled by southwest wind, PM2.5 from vehicle is easily transferred out of the main urban areas, which results in low RVEC in July. However, the most of PM2.5 from vehicle stay in east main city controlled by northwest wind, which results in high RVEC in December.

Reference: Jing, B. Y., L. Wu, H. J. Mao, S. L. Gong, J. J. He, C. Zou, G. H. Song, and X. Y. Li: Development of a High Temporal-Spatial Resolution Vehicle Emission Inventory Based on NRT Traffic Data and Its Impact on Air Pollution in Beijing, Part

A: Development and evaluation of vehicle emission inventory, Atmos. Chem. Phys. Discuss., 15, 26711-26744, doi:10.5194/acpd-15-26711-2015, 2015.

Changes in manuscript: More explain has been listed in section 3.3.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/15/C9803/2015/acpd-15-C9803-2015-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., 15, 19239, 2015.

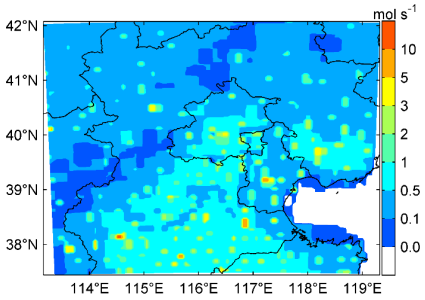

Fig. 1 Annual mean emissions of NH_3 in D02.

Fig. 1.

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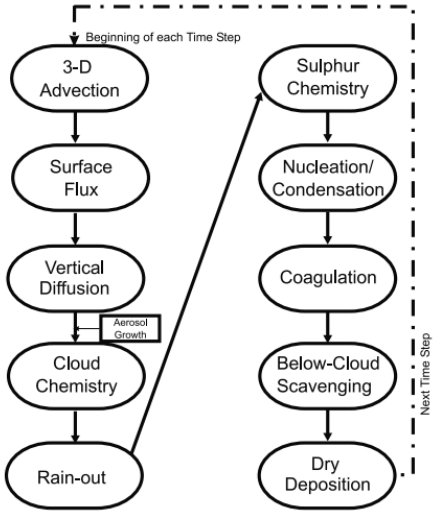

Fig. 2 The flowchart of aerosol module. (Gong et al., 2003)

Fig. 2.

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