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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Modeling analysis of the seasonal characteristics of haze formation in Beijing" *by* X. Han et al.

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Anonymous referee #1 This work analyzes the mechanism of haze formation in winter (February) and Summer (July) of 2011 using a regional air quality modeling system, and identiïňĄes that higher relative humidity (RH) is an important factor for the haze formation in summer. Considering the serious air pollution and deteriorated air quality in China, this kind of study is important and potentially useful for air pollution controls. This manuscript is in general well written although in some places there are wording issues. I have some general comments that the authors need to address before I recommend its publication in ACP. I suggest that the authors do more in-depth analysis of haze formation mechanisms and also evaluation of models results. Reply: We would

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like to thank the reviewer for his/her constructive comments for the improvement of our manuscript. Our responses to the comments from Reviewer 1 are included in the followings. Substantial modifications are made on the contents. They are Section 4.2, Section 4.3 and the fourth and fifth point of conclusion and some relevant contents are added. (please check the revised paper). We also modified Fig.1 and Fig. 14, and added Fig. 6, Fig. 8, and Fig. 13. In this way, we hope that the following scientific views can be expressed more explicitly. Even though the mass concentration of PM2.5 is closely inversely correlated with visibility, the influence effect is diversity when the mass concentration of PM2.5 locates in different intervals. The analysis of this study showed that the influence of PM2.5 mass burden variation on visibility is very weak when its value is relatively high (larger than 100 μg m-3). Only when the mass concentration of PM2.5 is cut down to a certain interval can its decrease make the visibility increase rapidly. Therefore, we suggest that it is more reasonably to set a haze occurrence threshold interval (the values of mass concentration of PM2.5 when the visibility reaches 10 km in different ambient conditions). Through sensitivity test, we estimated the possible values of haze occurrence threshold in Beijing in different seasons, and discussed the related impact factors. Detailed statement can refer to the relevant contents in the revised paper. By investigating the characteristics of haze occurrence threshold, one of the important viewpoints of this study is that the atmospheric haze and the atmospheric pollution should be distinguished. Only when the mass concentration of PM2.5 is cut down to a threshold interval can its decrease make the visibility increase rapidly, and the haze could be removed. Otherwise, if the mass concentration of PM2.5 fails to fall into the values of this threshold, the improvement of visibility would still be very weak when the emission reduction measures are taken. We sincerely hope that you can review the modified contents above and give us valuable comments. Thanks again.

(1) In addition to RH, are there other factors affecting the haze formation? Even with in one single month (e.g., February or July) there are huge changes in the visibility, while emissions should not change a lot. The authors brieïňĆy touch the stability and

ACPD
13, C13567–C13572, 2014

convection. I suggest the authors to do more qualitative analysis by calculating the atmospheric stability based on the temperature proïňĄles. I also suggest the authors to plot the surface precipitation as an indicator of wet scavenging of aerosols. Reply: Thanks for this comment. According to the valuable advice of you and other reviewers, substantial modifications are made on the contents. After careful consideration, we finally decide to delete the discussion of this part. The reasons are as follows. First, we could not obtain the observation data of vertical temperature profile right now. The evaluation of the model results is not available here. Second, we kindly think that the main point of this study should focus on investigating the influence mechanism of mass concentration and microphysical properties of aerosols on the haze formation in North China Plain by using the modeling system. To investigate the influence of meteorological field is not the main target of this study. Therefore, in the revised paper, this part is deleted and some more relevant contents are added (including the discussion of microphysical properties of aerosols and haze occurrence threshold, etc.). In addition, we accepted your suggestion, adding the content about verification and discussion of simulated precipitation. Please refer to line 182-188, line 200-204 in the revised paper.

(2) The authors mention that nitrate, sulfate and ammonium are the three major aerosol components in Beijing. How about the contribution of organic carbon (OC) to PM2.5? The model underpredicts OC mass concentration (Figure 8). Are there any aerosol mass spectrometry data available in Beijing in winter and summer to evaluate modeled aerosol components? How about the role of temperature in affecting the secondary formation of these aerosols between winter and summer? Reply: Thanks for this comment. According to the model simulation results and analysis in our study, the contribution of organic carbon aerosol to the mass concentration of PM2.5 was about 5%-15%. However, it is found that the model may underestimate about half of the organic carbon aerosol concentration by comparing with the observation data. According to the related researches, the uncertainty of organic components estimation should be a common problem in model simulations nowadays. Most models underestimate the mass concentration of organic carbon. On one hand, it is because the organic

ACPD
13, C13567–C13572, 2014

components and emission sources are too complex to recognize. On the other hand, it is difficult to add the simulation schemes of multiple organic components evolution processes, because it needs to modify almost all of the aerosol simulation modules, including the gas-phase chemistry solvers, aqueous-phase solvers, heterogeneous reaction schemes, scavenging schemes, etc. In general, the modeling system in this study can have accurate simulation on the seasonal variation features of organic carbon aerosol at least. However, it is also necessary to improve the accuracy of quantity estimation, which also is one major task of our current study. We regret that at present the observational data of mass spectrometry could not been collected by our group. Only daily observational data of carbonaceous aerosols which are measured by DRI Model 2001 Thermal/Optical Carbon Analyzer could be obtained. Thus, we cannot provide the evaluation with high time resolution. The CB05 mechanism was applied to treat the secondary organic aerosol (SOA) formation and its precursors in our modeling system. In the CB05, the SOA formation is modeled by forming semi-volatile products in volatile organic compound (VOC) reactions. The semi-volatile products are partitioned between the gas and aerosol phase according to the ambient conditions, such as temperature, relative humidity, vapor pressure, existing aerosol particles. If we just consider the impact of temperature, the higher temperature should favor the formation of SOA in summer. However, the higher SOA mass burden could also appear in winter because of the stronger emission of its precursors in North China Plain.

(3) The authors discuss the change of aerosol size distribution inïňĆuencing the threshold of haze occurrence. Are there aerosol size distribution data available for model evaluation? Reply: Thanks for this comment. We regret that the evaluation of modeled aerosol size distribution is not available because the observation data cannot be collected by our group. However, the air quality model CMAQ which was used by this study is one of the widely used regional atmospheric chemistry modeling systems in the world at present. The mechanism of particle coagulation and condensation growth in this modeling system should be reasonable. What's more, the analysis on the aerosol size distribution was just qualitative research (focusing on the decreasing or increas-

ACPD
13, C13567–C13572, 2014

ing trend). Therefore, we kindly think that the simulation results can be used by the analysis in this study.

SpeciïňĄc comments: 1. Page 30577. Lines 13-16. Why the mass burden of PM2.5 remain at high levels while SO2 emission has been reduced? Reply: Thanks for this comment. Even though the emission of SO2 decreases, the related studies showed that the emission loading of nitrate precursors NOx (Zhang et al., JGRïijŇ2007), primary aerosols from anthropogenic activities (including BC, primary organic aerosols, primary PM2.5 particles, etc.) (Lei et al., ACP, 2011) obviously increased in recent years. Therefore, the mass burden of PM2.5 should be remained at high level.

2. Page 30577. Line 25. Change "the said" to "these" 3. Page 30579. Line 2 Reply: Thanks for this comment. We have modified the statement.

3. Remove "efiňĄciently" Reply: Thanks for this comment. We have modified the statement.

4. Page 30580. Add brief description how the model treats SOA. Reply: Thanks for this comment. We added the description in line 106-111 in the revised paper. Please see if it is OK.

5. Page 30580. Line 24. Remove "efiňĄciently" Reply: Thanks for this comment. We have modified the statement.

6. Page 30582. Line 12. Remove "efiňĄciently" Reply: Thanks for this comment. We have modified the statement.

7. Page 30589. Lines 13-15. The authors conclude that the importance of nitrate from the transportation sector as the major sources of secondary particles in Beijing. How does this reconcile with the recent study of Zhang et al. (ACP, 13, 7053-7074) 2013 "Chemical characterizations: : :". Please add some discussions. Reply: Thanks for this comment. In this study, the conclusion that organic aerosol was higher in winter while inorganic aerosol (sulfate, nitrate, ammonium) was higher in summer in Beijing is

ACPD

13, C13567–C13572, 2014

consistent with that of Zhang et al. (2013). To investigate the contribution of transport sector to the air pollution in Beijing was not the main point of this study, so it did not discuss in detail. We deduced that the contribution of transport sector to the air pollution should be very important in Beijing because of the high mass burden of nitrate which converted from the precursors NOx (NOx is one of the main pollutants emitted from vehicle exhaust). In the study of Zhang et al. (2013), they used positive matrix factorization (PMF) to identify the possible sources of various aerosol components in PM2.5 samples in Beijing. There may be great differences on how to apportion the emission sources of PM2.5 particles between different source apportionment methods. The nitrate aerosol was identified as “secondary inorganic aerosol" in Zhang et al. (2013), not the pollutant from "traffic source". This shall be the main reason for the inconsistency between the relevant conclusions in this paper and those in Zhang et al., (2013).

Reference: Lei, Y., Zhang, Q., He, K. B. and Streets, D. G.: Primary anthropogenic aerosol emission trends for China, 1990–2005, Atmos. Chem. Phys., 11, 931-954, 2011. Zhang, Q., Streets, D. G., He, K., Wang, Y., Richter, A., Burrows, J. P., Uno, I., Jang, C., Chen, D., Yao, Z., and Lei Y.: NOx emission trends for China, 1995–2004: The view from the ground, J. Geophys. Res., 112, D22306, doi:10.1029/2007JD008684, 2007. Zhang, R., Jing, J., Tao, J. Hsu, S.-C., Wang, G., Cao, J. Lee, C. S., Zhu, L., Chen, Z., Zhao, Y., and Shen, Z.: Chemical characterization and source apportionment of PM2.5 in Beijing: seasonal perspective, Atmos. Chem. Phys., 13, 7053-7074, 2013.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 30575, 2013.