Manuscript No.: acp-2014-309

Title: Exploring the severe winter haze in Beijing

We would like to thank the anonymous referee for his/her valuable and constructive comments/suggestions on our manuscript. We have revised the manuscript accordingly and please find our point-to-point responses below.

Comments in RC C5497 by Anonymous Referee #2:

The paper by Zheng et al. focuses the haze episodes occurred during the winter of 2012–2013. The author utilized hourly chemical composition data of PM2.5, model simulations, and meteorological data to characterize the processes involved in the development of these events. Their manuscript contained the following major points: 1. They determined on the basis of modeling that the severe winter haze was shown to result from stable synoptic meteorological conditions over a large part of northeastern China and not from a change in emissions. 2. The build-up of secondary species was the major driving force behind these polluted periods. 3. The contribution of organic matter decreased with increasing pollution level while sulfate and nitrate contributions increased. 4. There is a weakening of the photochemical activity due to the dimming effect of high loading of aerosol particles. 5. Regional transport of pollutants played an important role during these severe pollution events. The paper was generally well written, and I recommend that this paper can be considered for publication after the following issues are adequately addressed.

Major issues:

1. My main concern with this paper is that it would benefit if the paper can be more quantitative as a whole. There are many places when they author stated a conclusion, but did not back it up sufficiently with a number. For example in the model discussion, the paper stated that the change of emissions added "+- 10 ug m-3" (Ln 9). It would be helpful if the readers were presented with the initial average PM concentrations and the differing resultant concentrations. Another example in section 6.1, the only direct value presented was 2.77 MJm^{-2}, and there were no other values to allow the reader to understand how low this value is compared to the rest of the observational period.

Response:

We have revised the manuscript as suggested. For example, we have modified Paragraph 3 in Section 4 into:

"As expected, the influence of emission difference was negligible (Fig. 3a and 3c). For the whole simulation domain of the North China Plain (NCP), both simulation with Jan. 2012 meteorology (Scenario ***c***) and Jan. 2013 meteorology (Scenario ***a***) resulted in similar $PM_{2.5}$ concentration ranges (~50 to ~500 μg/m³) and spatial distributions. Difference of $PM_{2.5}$ concentration at any site was within ± 10 μg/m³ (Fig. 3e). Simulation results of Scenario ***a*** and ***c*** were not only similar in average concentration levels, but also in temporal variations. For example, in Beijing, simulated hourly $PM_{2.5}$ concentration results under this two scenarios presented not only similar concentration (being 279.1 ± 170.2 μg/m³ and 278.8 ± 168.9μg/m³, respectively) but also excellent correlation with R^2 reaching 0.97."

And also relative statement in Section 6.1 into:

"Take Beijing for example, during haze episodes, the amount of solar radiation reaching the ground was significantly lower (e.g., down to 2.77 MJ/m²/day, 13 Jan.) than clean days (averaging 9.36 ± 0.60 MJ/m²/day for all the six clean days), rendering high photochemical activity impossible."

2. How well did the model reproduce the observations? It would be nice if one could see a figure displaying the accuracy of the plot for a reader to have confidence in the conclusion.

Response:

The original WRF-CMAQ model cannot reproduce the highest observed concentrations. Therefore, a revised model was developed which included additional heterogeneous reactions (Wang et al., 2012). The simulations based on the revised model agree well with the heavily polluted periods, not only for the magnitude and temporal variation of $PM_{2.5}$ (with normalized mean biases (NMB) being 0.4 %), but also for the chemical composition (Figure R1). We have revised the manuscript by including a brief summary of the model performance:

"Since the original WRF-CMAQ model cannot reproduce the observed concentrations under heavily polluted conditions (B. Zheng et al. 2014), a revised WRF-CMAQ

system with enhanced heterogeneous reactions (Wang et al., 2012) was adopted to improve the model performance. The revised model could effectively capture the measured concentrations of total $PM_{2.5}$ (with normalized mean biases (NMB) being 0.4 %) and its different chemical compositions for both clean and heavily polluted haze days (B. Zheng et al., 2014). Details of the model configuration, modifications, and validation are described in B. Zheng et al. (2014)."

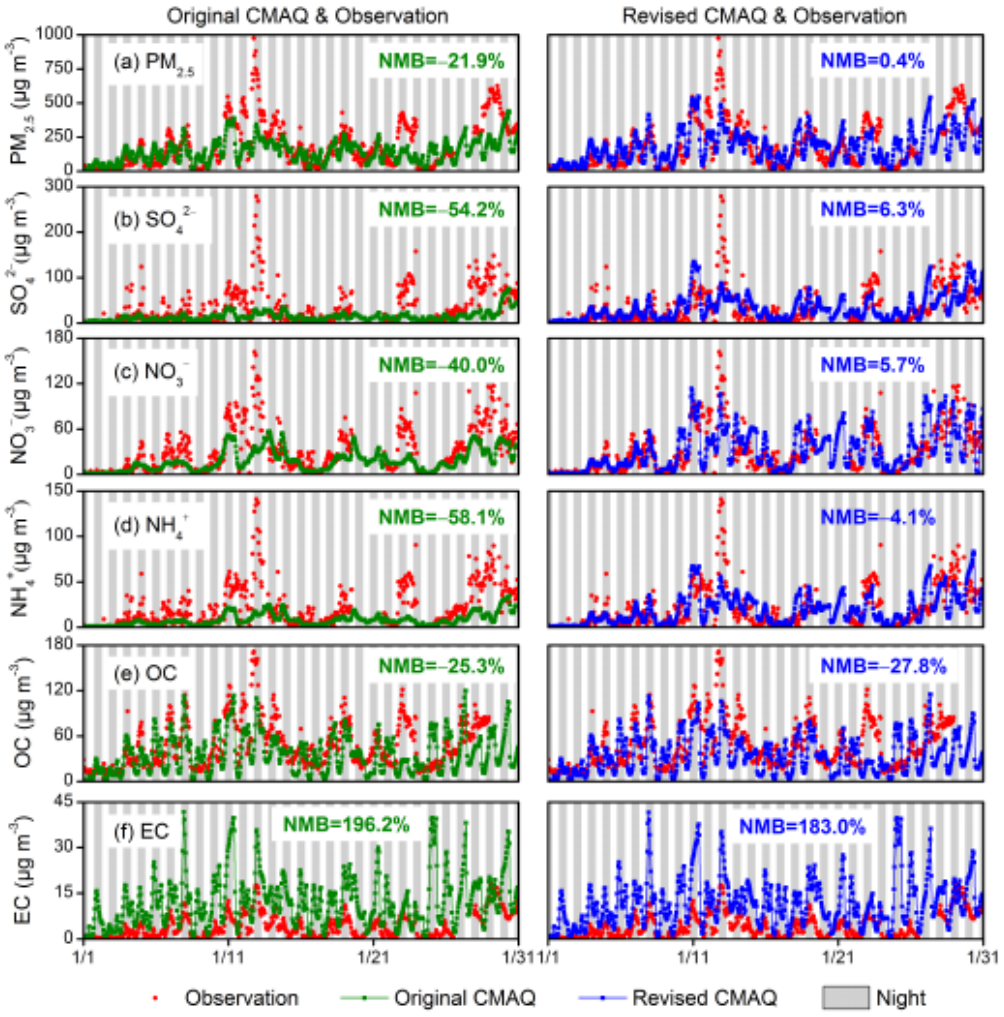

Figure R1. Observed and simulated hourly aerosol compositions from the original and revised CMAQ at the THU site: (a) $PM_{2.5}$; (b) SO_4^{2-}; (c) NO_3^-; (d) NH_4^+; (e) OC; (f) EC. NMB referred to normalized mean biases. (***Source: Fig. 3 in B. Zheng et al., 2014)***.

3. The use of quotation marks was awkwardly used throughout the paper. I think the paper would read better if they were removed.

Response:

Most quotation marks were now removed as suggested, except those indicating the quotation of the original text from other documents.

4. Pg 17919 ln 1-10: I did not follow the thought process in which the authors used the OC/EC ratio to determine the SOA production or how it connected with the boundary layer. The rational and assumption need to be better explained.

Response:

Thanks for pointing this out. The ratio to EC is used for two purposes: (1) one is to estimate the SOA production following the methods of Lim and Turpin (2002) and Lin et al. (2009); and (2) the other purpose is to account for the influence of different dilution/mixing conditions in the boundary layer. For the latter, the basic assumption is that EC comes only from primary emissions, thus, under certain emission rates, the change of EC concentration should be merely subject to atmospheric physical processes, such as the dilution/mixing effect, etc. We have included a paragraph to clarify this issue (See Paragraph 2 in Section 6):

"To evaluate the role of chemical productions, we analyzed the EC-scaled concentrations for individual compounds. The purpose of using EC-scaled concentration is to eliminate the influence of different dilution/mixing conditions on the variation of observed pollutant concentrations. The observed variations of pollutant concentrations are not only controlled by the chemical reactions but also subject to the influence of boundary layer developments. For the same emission rate and chemical production rate, different mixing condition will result in different pollutant levels. It is thus highly uncertain to conclude a stronger/weaker chemical production based on purely the concentrations data without considering the boundary layer effect. Since EC is an aerosol species coming from only primary emission and quite inertial to chemical reactions, its variations well reflect the influence of atmospheric physical processes (dilution/mixing effect). The ratio of other species to EC will to a large extent eliminate the variations due to mixing/dilution and better represent the contribution from chemical reactions."

Minor comments:

Pg 17917 ln 4: The use of "embrace" in this way personifies the weather system, which is not typically used in scientific writing.

Response:

The expression has been changed from "to embrace it" into "to be scavenged" as suggested.

Reference

Wang, K., Zhang, Y., Nenes, A., and Fountoukis, C.: Implementation of dust emission and chemistry into the Community Multiscale Air Quality modeling system and initial application to an Asian dust storm episode, Atmos. Chem. Phys., 12, 10209-10237, 2012.

Zheng, B., Zhang, Q., Zhang, Y., He, K. B., Wang, K., Zheng, G. J., Duan, F. K., Ma, Y. L., and Kimoto, T.: Heterogeneous chemistry: a mechanism missing in current models to explain secondary inorganic aerosol formation during the January 2013 haze episode in North China, Atmos. Chem. Phys. Discuss., 14, 16731–16776, doi:10.5194/acpd-14-16731-2014, 2014.