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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 #3

General Comments:

This paper presents a model study upon the haze formation in Beijing, China. It concluded that high PM2.5 loading was the main cause of haze events in Beijing, and that water uptake by aerosols resulted in the frequent formation of haze in Beijing, particularly during summertime. In general, this paper is well organized except some technical defects. However, my major concern is that this paper did not provide new concept or scientiïňĄc ïňĄndings relevant to haze. Coupling RAMS-CMAQ with an aerosol optical scheme is not a new idea as a similar study from the same group has been published C13573

in another Journal (Atmospheric Environment, 2013, 72: 177-191). Moreover, It is well known in atmospheric physics that high levels of aerosol concentration will resulted in cases of low visibility, and hygroscopic growth of aerosols will enhance the light scattering capability, or mass-speciiňĄc light extinction efficiency of aerosol particles. The case of Beijing is interesting because the microphysical properties of aerosols could be different from those observed in US or Europe. Unfortunately, the authors stopped at a general description of the phenomena of haze formation and did not advance further into the details of aerosol chemistry and/or physics. Therefore, I suggest reject this paper from ACP because lack of scientiïňĄc merits.

Reply:

We would 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.

In general, besides the routine analysis of simulation results, this study also wants to provide some further study about the mechanism of haze formation and how to efficiently decrease the possibility of haze occurrence over NCP by using the modeling system. Due to the improper structure arrangement and descriptive approach of the paper, the main scientific view may not be well expressed. Therefore, we adjusted the paper structure and made substantial modification. Section 4.2, Section 4.3 and the fourth and fifth point of conclusion are modified, 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 two scientific views can be expressed more explicitly.

(1) The mechanism of haze formation in Beijing in winter is obviously different from that in summer. The mass concentration of PM2.5 is relatively higher, and the ratios of inorganic salts and carbonaceous components are generally balanced in winter. Therefore, the high mass concentration of PM2.5 and diverse aerosol components

should be the major reasons of the serious haze occurrence in winter. While the mass concentration of PM2.5 in summer was relatively lower, but the ratio of hygroscopic inorganic salts, including sulfate, nitrate and ammonium, increased and their mass concentrations were even higher than those in winter. With obviously higher relative humidity, it could still form more serious haze even the mass concentration of PM2.5 is lower than that in winter. The water uptake of hygroscopic components played a key role in it. This indicated that it is important to apply emission reduction measures based on the specific pollution and meteorological characteristics in different seasons. In this way, the possibility of haze occurrence can be effectively decreased.

(2) 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 μ g 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 this 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 paper and give us valuable com-

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ments. Thanks again.

Specific Comments:

1. Method Sec: Calculation of light extinction coefficient of aerosols is the key component of this task. In addition to citing references, it is worth a detailed description in this Sec, so that readers know what parameters were used in the model and thereby can make judgment.

Reply:

Thanks for this comment.

We are sorry we did not give a clear description of the modeling system. We have modified the related description of the modeling system in the manuscript. Please see the statement from line 132 to line 141.

2. Model evaluation: it was indicated that the model performed well as shown in the figures. However, there were indeed some cases where the model value was inconsistent with the observation. To perform a model validation, I suggest make the comparison in terms of statistics and refer to Eder and Yu (AE, 2006) and Appel et al. (AE, 2012).

Reply:

Thanks for this comment.

We have added some relevant content into the manuscript. Please see the statement in line 192-205. Here we just calculated the statistic parameters of PM2.5, O3, NO2, and visibility comparisons. For other variables, including the meteorological factors and aerosol components, the statistic samples are relatively small, so we did not provide the statistic parameters of their comparisons.

3. Sec 4.1: It was indicated that "the heavy mass burden of PM2.5 was mainly concentrated in four urban areas...". However, the urban hot spots were not shown in the figures. Actually, the pattern shown in those figures are more likely caused by a

regional pollution event.

Reply:

Thanks for this comment.

We have added the district areas of Beijing, Tianjin, Shijiazhuang, and Jinan in Fig. 1, and modified the description in section 4.1. We think that your advice is correct. The high mass burden of PM2.5 generally covered Beijing, Tianjin, the whole area of Hebei province and northwest part of Shandong province, not just the urban areas. Please see the modified statement in line 236-240.

4. Sec 4.1: It was indicated that "the distribution patterns of visibility broadly followed those of PM2.5...". Don't you think this is a result as expected and is determined by the calculation of visibility in model (i.e. EQ1)?

Reply:

Thanks for this comment.

Actually, except the mass concentration of aerosols, the calculation method used in this study also considers the influences of aerosol microphysical properties, including water uptake of soluble particles (by Kohler theory), internal mixing state (by Maxwell-Garnett mixing rule), and particle size distribution (the change of lognormal distribution parameters of three modes) as explained in line 132-141. Therefore, the mass burden of PM2.5 was not the only impact factor of visibility variation in our model. Even though the distribution patterns of visibility broadly followed those of PM2.5 mass burden, we can also see that the extinction was obviously enhanced by the higher relative humidity in July. Anyway, we are sorry that the description about the aerosol optical properties calculation is not clear, and hope the modified content is suitable.

5. Sec 4.2: Decline in pollution caused by the enhanced vertical convection is a classical case in PBL dynamics. I suggest move forward to investigate factors that were controlling the convection and, in turn, influencing air quality.

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Reply:

Thanks for this comment.

On second thoughts, we kindly think this part of analysis is not quite necessary for this study. The reasons are as follows. First one is that the vertical structure of modeled temperature was not evaluated by any observation data. Second one is that we would like to focus on the influence of aerosol characteristic (such as the particle components and microphysical properties) and related meteorological factors that can change the particle optical properties (such as the relative humidity which changes the water uptake of soluble particles). The impact mechanism of meteorological conditions on haze formation is not the main point of this study. Therefore, we kindly chose to delete this part of analysis, and added other content which is more suitable for the subject of this paper, including the discussion of microphysical properties of aerosols and haze occurrence threshold, etc.

6. Sec 4.3: The method for "contribution ratio" calculation is unclear. Are you turning off the formation of a specific compound in the model to investigate the corresponding effects? In that case, there could be some bias in the results. For instance, if you turn off the formation of ammonium sulfate then the ammonia will go to nitrate and change the partition and fate of N-containing species in the atmosphere.

Reply:

Thanks for this comment.

Actually, the input data of each specific aerosol component was just ignored when calculated the extinction coefficient by using the aerosol optical property module. The aerosol chemical and physical processes in the air quality modeling system was not modified or turned off. So the bias should not be existed in this method. Sorry we did not give a clear explanation. The statement in line 288-289 has been modified. Please check if it is OK.

7. Sec 4.3: Regarding the case study of size distribution, the mass fraction of accumulation mode was still âLij80% despite the increases in Aitken mode. Thus the changes in the cross section should be rather limited. I'm not convinced that the spike of "mass threshold" was due to increases of Aitken mode aerosols. Moreover, in terms of size distribution, I think that the cases of high coarse mode fraction also worth to be investigated further.

Reply:

Thank you very much for this comment.

This is an important advice and we think further analysis is necessary to make the problem clear. We calculated the contribution ratios of Aitken mode, accumulation mode, and coarse mode particles to the total extinction and added this information into Fig. 14 for investigating the detail impact mechanism. The related statement is added in line 348-368. As shown by the analysis, the particles of accumulation mode provided the most part of the extinction effect. A small variation of the extinction contribution ratio of accumulation mode could obviously change the mass threshold of haze occurrence. However, it seems that the spike of mass threshold appeared on July 29 was caused by the influence of the extinction contribution ratio of Aitken mode particles. Even though the mass concentration ratio of coarse particle could reach 10%-20%, their extinction was quite weak. Thus, the coarse particle may not play an important role unless its mass concentration is far higher than that of accumulation mode, such as the case of strong dust event. For the detail analysis, please check the contents in line 363-368.

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

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