Anonymous referee #2

General Comments:

I appreciated very much the authors' efforts for this revision. However, I think further improvement is needed.

The authors argued that the formation of summer haze in the NCP area of China was a result of hygroscopic effects of aerosols for high RH in summertime. This argument is acceptable in principle. However, my major concern is upon the weak evidences for the argument. As shown in Table 2, the correlation coefficient between observation and simulated PM2.5 was 0.43 for summer case in Beijing, which means that the mechanisms considered in the model explained only ~20% of the variance in the observed PM2.5 concentration. Moreover, as shown in Figure 3, the model significantly underestimated summer RH at Miyun (Beijing) station. While the simulation of both key parameters is subject to high uncertainties, it's hard to draw a solid conclusion. I thought that the apparently fair R of 0.65 for visibility (Table 2) was a result of those extreme events with PM2.5 up to 0.2-0.3 mg/m3, where the impacts of uncertainty could be relatively small. Without a robust simulation and data analysis, the current results could not be extrapolated to cases of low PM2.5 loading as shown in Figure 15.

Reply:

Thank you very much for this comment.

The model performance could be different over divers regions because of several reasons, such as the applicability of parameterization, accuracy of underlying surface information, etc. Thus, we collected the hourly observation data of relative humidity from the CRAES measurements at Beijing to evaluate the model performance in Beijing. The comparison is shown in Fig. 10, and the discussion was added in Line 202-207. From the evaluation it can be seen that the model could provide more reasonable simulation of relative humidity in Beijing than those in Miyun and Tangshan, which kindly implies that the model performance

could be reliable in Beijing. For the simulation of $PM_{2.5}$, the comparison in Fig. 7 shows that the model may overestimate the mass concentration of $PM_{2.5}$ in the middle of July (from 14 to 19, July). This should be the main reason of the poor correlation coefficient 0.43 in Table 2, and the underestimation of the precipitation during the same period may lead to this deviation as analyzed in Line 192-194. However, the model generally performed well during the rest of July, especially when the peaks of $PM_{2.5}$ mass burden appeared in 10, 22, and 28 July. The simulation results are also in good agreement with the observations in February. In addition, the direct comparison with point measurements should be difficult because the model results are interpreted as the average of each grid cell (16×16 km). Therefore, we kindly think that the simulation results are generally reliable and can be used to analyze the hygroscopic effects of aerosols in Beijing.

Meanwhile, we admit that some uncertainties (such as the simulation of organic carbon) still exist in the modeling system, and these are the major tasks we will focus on in future work.

Specific comments:

1. Model evaluation: The authors claimed that "The modeled temperature, relative humidity, and wind speed were in good agreement with the observations at nearly all stations" (Line 173-174). However, there is indeed obvious inconsistency in the results shown in Figure 3. For example, the model underestimated RH for the second half of Feb at Tanggu and Miyun, and for almost the whole July at Miyun. In addition, overestimation of RH is significant for the summer case at Baotou station. The simulation of RH is particularly important to this study as the summer haze is attributed to the hygroscopic effects of aerosols. I'm not meaning that the model should be perfect. Nevertheless, the differences in the simulated and observed RH are so substantial in the Beijing-Tianjin area and thereby could have induced significant uncertainties in the conclusions. I'd like to suggest the authors make an in-depth analysis on the relevant uncertainty that will get their arguments more convincing than the current ones.

Reply:

We agree that the evaluation of model results need to be discussed in detail. Thanks for this comment. We have added some related discussion in Line 174-179. We also collected the hourly observation data of relative humidity in Beijing from the CRAES measurements, and used these data to evaluate the model performance. The Figure of comparison and related discussion was added in the manuscript (Fig. 10 and Line 202-207). It can be seen that the model could provide reasonable relative humidity in Beijing. Therefore, we kindly think that the simulation results are reliable and can be used for further analysis.

2. Model evaluation: The authors indicated that "A persistent underestimation of wind speed by the models was found at the Wutaishan and Taishan sites" (Line 174-175). This statement disagrees with the corresponding plots (Figure 4), which show underestimation for the case of Wutaishan but "overestimation" for Taishan. Thus, the relevant analysis and argument for the inconsistency in wind field need a substantial revision.

Reply:

Thanks for this comment. Actually, we rechecked the plots in Fig.4 and found that we kindly used the same plot for Taishan site as the one for Baotou site. We are very sorry for this rookie mistake. The plot has been corrected and we kindly think that the related analysis is suitable for this new version.

3. Line 205-206/Model evaluation: The large diurnal variation in NO2 was attributed to the "gas-phase chemical scheme in ISORROPIA". As the ISORROPIA is dealing with the thermodynamics of aerosols and their precursors, are you meaning that the diurnal variation of NO2 is dominated by heterogeneous reactions? This could be an interesting finding because the diurnal variation of NO2 was usually controlled by deposition flux and gas phase chemistry in urban areas.

Reply:

Thanks for this comment. Actually, we kindly think that the expression here is not correct. The related module which describes the reactions of NO_x should be the CB05. Sorry for this mistake, we have modified the sentence.

4. Sec4.1: The main argument here was that comparable degradation in visibility corresponded to less PM in summertime than in winter case because of high RH in summer. In this context, why not showed the correlation between mass- specific extinction efficiency (cross section) and the RH directly? A plot showing the extinction cross section vs. RH will depict the argument clearly.

Reply:

Thanks for this comment. Actually, we kindly think that the relationship between extinction cross section and RH cannot provide a better description about the feature of water uptake and the influence of particle size than that of total extinction coefficient. In our model, the wet radius of internal mixing particle (obtained by the water uptake scheme) is needed in two schemes: the calculations of extinction cross section and the cross area of wet particle. It means that the process of water uptake cannot be fully considered by just using extinction cross section because the extinction coefficient is obtained by multiplying extinction cross section and cross area of wet particle. On the other hand, aerosol particles have wide size range and we have to divide them into three major size distribution modes (Aitken mode, accumulation mode, and coarse mode) with different mode radii in the model. The specific extinction cross section can be used in only one mode with the same mode radius, not for all aerosol particles. Therefore, it is hard to describe the total aerosol extinction properties by just using extinction cross section because the extinction properties need to be separated and represented by different specific extinction cross sections in different modes, respectively.

However, we still think that this comment is valuable for us. In the future work, we will

consider how to describe the analysis with more effective way.

5. Sec 4.2: Line267-269 indicated that nitrate was the main particulate pollutant during winter. This was based on simulation I think. In fact, the observation (Figure 10) showed that OC was comparable with nitrate. Thus the total mass of organic matters (OM) would be larger than nitrate.

Reply:

Thanks for this comment. We agree that the discussion you mentioned in Line 267-269 should consider the total mass of organic matters, not just organic carbon. We have modified the expression in Line 262-266. Some related studies (Wang et al., 2014, Science China: Earth Science) also mentioned that the mass burden of organic matters was higher than nitrate in Beijing. However, the modeling system cannot provide a comprehensive simulation of all organic matters presently because the related physicochemical mechanisms are too complex to describe properly, and the investigation of organic matters is still insufficiency. This should be the general uncertainties in the Chemistry and Transport Models as well. Right now we are also working on improving the ability of organic matters simulation in our model.

6. Line 294-296 indicated that except nitrate the contribution of each aerosol component to the extinction "did not significantly change". However, as shown in Figure 12 the contribution of sulfate exhibited obvious diurnal variation. Moreover, sulfate behaved very much like a counterpart of nitrate and the total contribution of sulfate+nitrate was leveled off, particularly in summertime. As the mechanisms dominating sulfate and nitrate aerosols are very different, I wonder how the two species can compensate each other so closely. It seems to me that the total contribution of S+N could be controlled by something in the model.

Reply:

Thanks for this comment. Actually, we kindly meant that the contribution of each aerosol

component to the extinction did not significantly change when the mass concentration of $PM_{2.5}$ exceeded ~50 μg m^{-3} as described in Line 293-294.

For the counterpart relationship you mentioned about the extinction contributions of sulfate and nitrate, we also thought it is very interesting and conducted some deep analysis. The model uses the same refractive index and hyproscopicity factors for sulfate and nitrate aerosols. Thus, the mass burden should be the main impact factor of the extinction coefficient calculation. Please check the plots in Fig. R1. In plot (b), we can see that the variation of mass concentrations of sulfate and nitrate broadly coincided with each other, and they were obviously higher than those of black carbon and organic carbon. In plot (a), the mass burden percentages of black carbon and organic carbon were relatively small. The variation of these two aerosol components could be ignored (except the clean period, such as 7-9 July). In addition, the mass burden percentage of ammonium was generally constant. Therefore, the mass burden percentages of sulfate and nitrate were in fact the only two aerosol components with significant diurnal variation. When the mass burden percentages of nitrate increased, the mass burden percentages of sulfate should be decreased because other aerosol components were generally not change. We kindly think that this should be the main reason of the counterpart relationship between sulfate and nitrate extinction contributions.

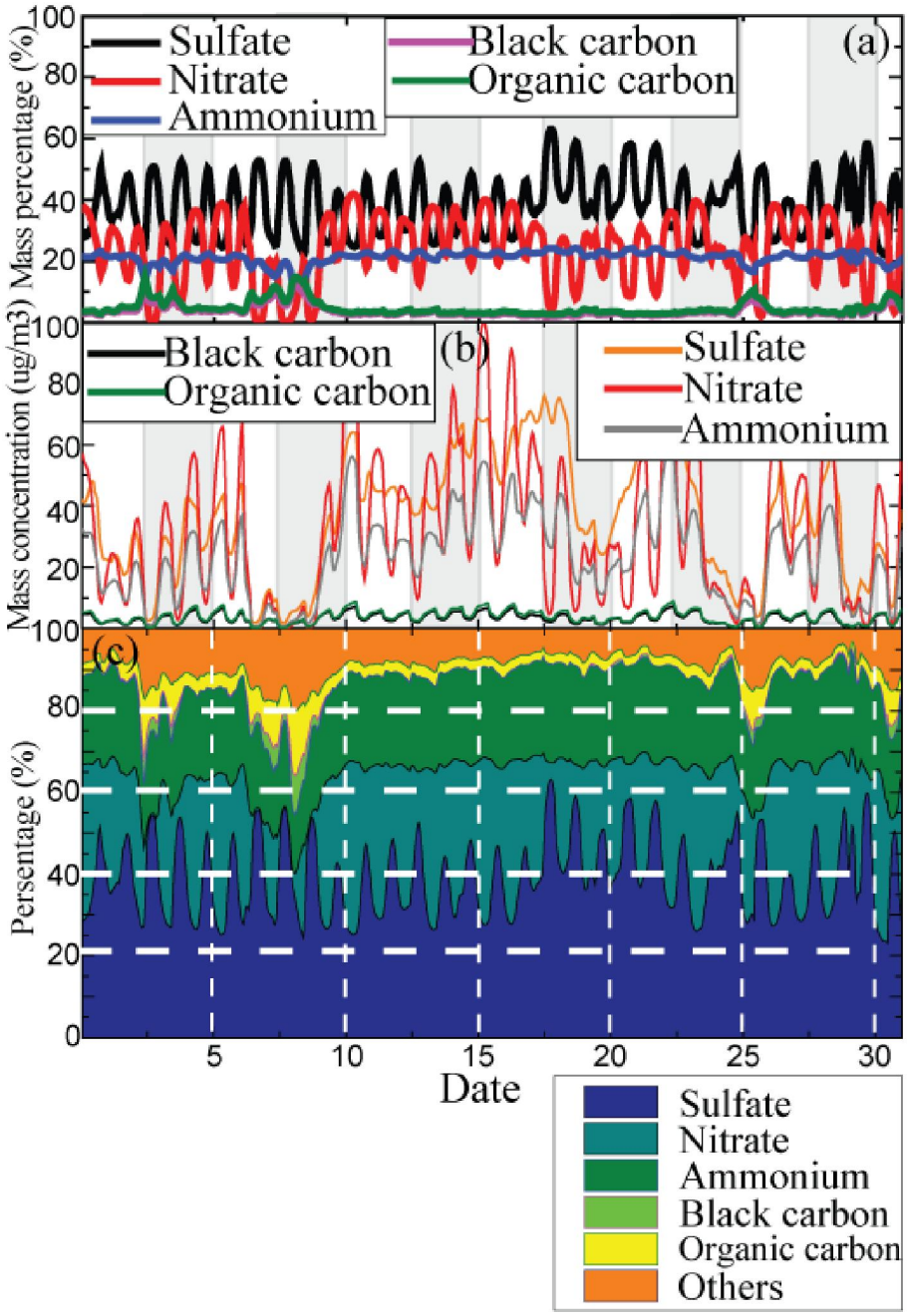

Fig. R1 The regional average values of mass ratios (%) of sulfate, nitrate and ammonium (a), mass concentrations of major aerosol components (μg m^{-3}; b), and extinction contribution ratios of major aerosol components (μg m^{-3}; c) in July over Beijing.

7. Sec 4.3: A nonlinear response in visibility to PM concentration was found from model simulation. I suggest make an in-depth analysis on the mechanisms for this result. This is particular for the drastic changes in visibility on July 25 (Figure 13), where the visibility increased from 20 to 70 km and decreased to ~40 km with only very minor changes in PM

within 12 hours.

Reply:

Thanks for this comment. The visibility was simply calculated by using the formula (2) in Line 133. Thus, the aerosol extinction coefficient should be the key factor influencing the visibility. We provided the time series of regional mean mass concentration of $PM_{2.5}$ and extinction coefficient (first layer of model) in Beijing from 23 to 25 July in Fig. R2. It can be seen that the variation features of extinction coefficient and mass concentration of $PM_{2.5}$ are quite similar with each other. During the period C, the variation ranges of these two variables were both very small. Therefore, we kindly think that the drastic changes in visibility during period C was caused by the inverse correlation between visibility and extinction coefficient. When the value of extinction coefficient becomes small, the value of visibility could change dramatically with a microvariation of extinction coefficient by applying formula (2). We added the relative expression in Line 315-321. Please check if it is OK.

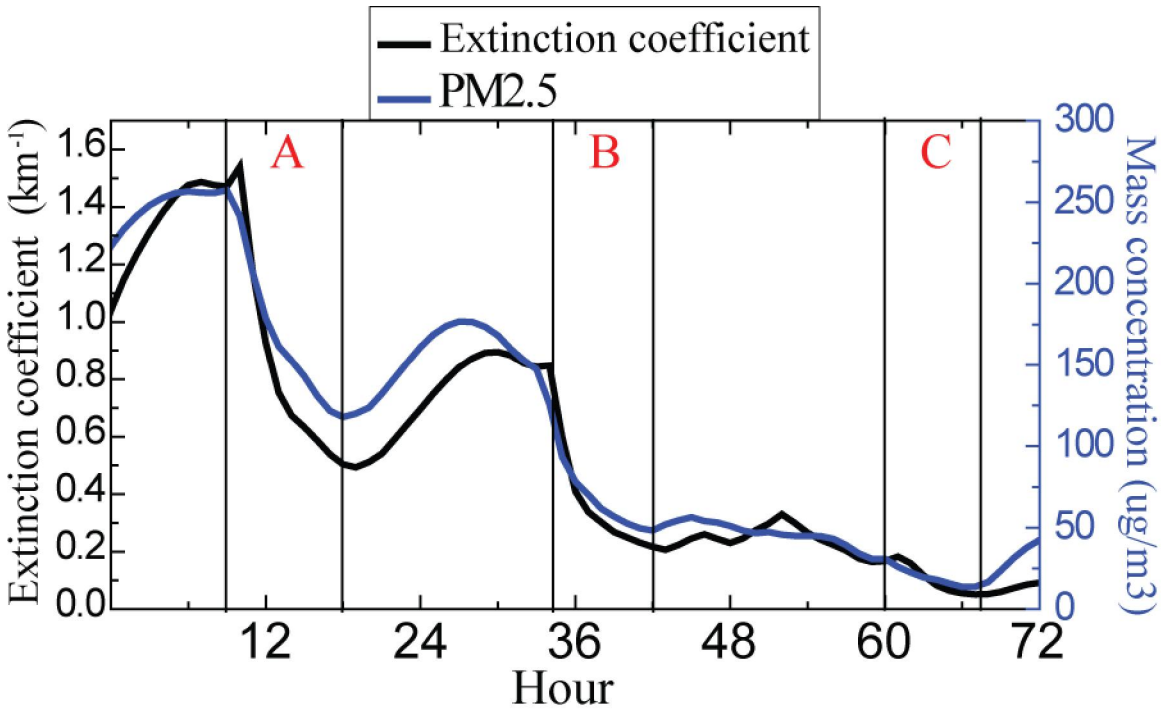

Fig. R2 The time series of regional average extinction coefficient (km^{-1}) and mass concentration of $PM_{2.5}$ from July 23 to 25 in Beijing.

8. Finally, the threshold of PM concentration for haze was described as a function of RH. The information is useful in air quality management. However, this information must be provided with cautions. Assumptions and thereby uncertainties should be addressed very clearly.

Reply:

Thanks for this comment. In our opinion, this is a preliminary study about how to properly quantify the impact factors of haze occurrence. The relationship between the relative humidity and threshold is just built by the results in Beijing. The ratio of each major aerosol component does not change dramatically in Beijing because the main emission sources of pollutions in this region are the anthropogenic activities, and the size distribution of particles should be mainly focused in accumulation mode. Therefore, we deduced that the relative humidity and mass burden are two major impact factors which should be considered and built the function. However, the function would be invalidation when the ratio of each major aerosol component or the size distribution of particles change, such as the aerosols over different regions or the dust storm happened. In summary, this function is just for the ordinary situation in Beijing. We have modified the expression in Line 463-467, please check if it is OK.

9. Furthermore, I compared the two plots in Figure 15 and I think they are same. Actually, according to the parameters in Table 6 and the equation 2, the RH will increase with M and the threshold of PM will be zero with RH of 97% in summer. This is certainly disagreeing with the reality. So PLEASE CAREFULLY check all the calculation and presentation .

Reply:

Thanks for this comment. For the similarity of the two plots in Figure 16, we kindly think that the main reason should be that the ratio of each major aerosol component does not

change dramatically in Beijing as we mentioned above.

We admit that the description about the function was not quite sufficient. Thanks for the suggestion. Actually, in this study we used the definition of haze from Wu et al. (2007): a day with atmospheric haze occurrence has a visibility below 10 km and the relative humidity of less than 90%. Therefore, all of our analysis and the sensitivity test were conducted while the relative humidity did not exceed 90%. The function would not be used when the relative humidity increased higher than 90%. We are sorry that the explanation about this point was not emphasized clearly, and the related expressions were modified (Line 47-48, Line 340-341, and Line 397-398).

Anonymous referee #1

The authors have improved their manuscript and addressed my comments in the revision. The authors also added a "Haze occurrence threshold" section to strength the science of this study.

There are many places where improvement of English writing is still necessary:

1. Line 25. Be specific "components are complicated". Do you mean components are not complicated in summer?

Reply:

Thanks for this comment. Actually, the phrase should be "components are complicated in winter". Sorry for the mistake. We have modified the expression.

2. Lines 37-38. define "contribution ratio" here.

Reply:

Thanks for this comment. It should be "extinction contribution ratio". We have modified the expression.

3. Abstract is too long. condense somehow.

Reply:

Thanks for this comment. We have modified the abstract and deleted about 200 words. Please see if it is OK.

4. Lines 138 and 139. What do you mean "under several Dp" and "40 Dp". Where and how do

you use these Dp.

Reply:

Thanks for this comment. The D_p represents geometric mean diameter of each aerosol mode which follows the lognormal size distribution. The value of D_p can be changed by the water uptake of hygroscopic aerosols in each time step. Thus, we calculated the aerosol optical properties under several possible size distributions with different D_p for building the fitting coefficients table. Then, the model can use the fitting coefficients table to calculate the aerosol optical properties with different aerosol size distributions. We have modified the relative sentences and tried to give more clear expression.

5. Line 187. Remove "kindly"

Reply:

Thanks for this comment. We have modified the expression.

6. Line 195. Change "evaluation of" to "evaluate"

Reply:

Thanks for this comment. We have modified the expression.

7. Line 205. Change "lager" to "Larger"

Reply:

Thanks for this comment. We have modified the expression.

8. Line 206. "gas-phase chemical scheme in ISORROPIA". Is ISORROPIA a gas-phase chemical scheme?

Reply:

Thanks for this comment. Yes, we also found this mistake. The gas-phase chemical scheme should be “CB05”. Sorry for the wrong expression.

9. Line 274. Is "biomass burning" a source of carbonaceous aerosol in February?

Reply:

Thanks for this comment. As we know, the straw (or other kinds of biomass) is used to heat in the countryside of North China Plain in winter. It is a very common phenomenon and the pollutants emitted from this source cannot be ignored in Northern China. Zhang et al. (ACP, 2014, 14, 2887–2903) also mentioned that the organic matters from coal combustion can be detected in winter. We have added this reference in the manuscript.

10. Table 4. Are these components added up to be 100%. if not, it is due to aerosol water. Then add the aerosol water fraction.

Reply:

Thanks for this comment. Yes, the sum should be 100%. The category “Others” includes the contribution of water.

11. Line 308, change "A period" to "period A". So do B,C period.

Reply:

Thanks for this comment. We have modified the expression.

12. Line 341. Change "threshold generally maintained a small value range" to "threshold generally maintained a small change"

Reply:

Thanks for this comment. We have modified the expression.

13. Line 396. Change "greatly" to "great" or "significant".

Reply:

Thanks for this comment. We have modified the expression.