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 for your comments.

The model performance could be different over diverse regions for 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 Chinese Research

Academy of Environmental Sciences (CRAES) measurements at Beijing to evaluate the model performance in Beijing. The comparison is shown in Fig. 10, and the discussion was added to Lines 201 – 206. From the evaluation it can be seen that the model could provide a more reasonable simulation of the relative humidity in Beijing than in Miyun and Tangshan, implying 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 is the main cause of the poor correlation coefficient (0.43) in Table 2: the underestimation of the precipitation during the same period may lead to this deviation as analyzed in Lines 192 – 194. However, the model generally performed well during the rest of July, especially when the peaks of $PM_{2.5}$ mass burden appeared on 10, 22, and 28 July. The simulation results were also in good agreement with the observations in February. In addition, the direct comparison with point measurements is difficult because the model results are interpreted as the average of each grid cell (16×16 km). Therefore, we think that the simulation results are generally reliable and can be used to analyze the hygroscopic effects of aerosols in Beijing.

Meanwhile, we accept that some major 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 have added a discussion in Line 174 – 178. 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. A comparison figure related discussion was added in the manuscript (Fig. 10 and Lines 201 – 206). The model could therefore provide reasonable relative humidity in Beijing.

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:

Upon rechecking the plots in Fig.4, we found that we had used the plot for Baotou site

instead of the Taishan site plot. This has been corrected.

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:

The expression here was not correct: the related module which describes the reactions of NO_x should be CB05. We have modified the sentence accordingly.

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. We think that the relationship between the extinction cross section and RH cannot provide a better description of water uptake or the influence of particle size than that given by the total extinction coefficient. In our model, the wet radius of the internal mixed particle (obtained by the water uptake scheme) is needed for the calculation of the extinction cross section, and the cross sectional area of the wet particle. This means that

the process of water uptake cannot be fully considered by just using the extinction cross section, because the extinction coefficient is itself obtained by multiplying the extinction cross section and the cross sectional 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), each with different mode radii, in the model. The specific extinction cross sections 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 the extinction cross section because the extinction properties need to be separated and represented by different specific extinction cross sections in the different modes.

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 Lines 267–269 should consider the total mass of organic matter, not just the organic carbon. We have modified the expression in Lines 260 – 264 accordingly. Some related studies (Wang et al., 2014, Science China: Earth Science) also mentioned that the mass burden of organic matter was higher than nitrate in Beijing. However, the modeling system cannot provide a comprehensive simulation of all organic matter presently because the related physicochemical

mechanisms are too complex to describe accurately. This adds to the general uncertainties in the chemistry and transport models as well. Right now we are also working on improving the ability of our model to simulate organic matter.

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 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 Lines 289 – 292.

For the counterpart relationship you mentioned the extinction contributions of sulfate and nitrate, we also thought it interesting and conducted some further analysis. The model uses the same refractive index and hygroscopicity factors for sulfate and nitrate aerosols. Thus, the mass burden should be the main impact factor for the extinction coefficient calculation. (Please see the plots in Fig. R1). In plot (b), the variation of the mass concentrations of sulfate and nitrate broadly coincided with each other, and 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 in clean periods, 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 correspondingly decreased because the other aerosol components generally did not change. We think that this is the main reason the counterpart relationship between sulfate and nitrate extinction contributions exists.

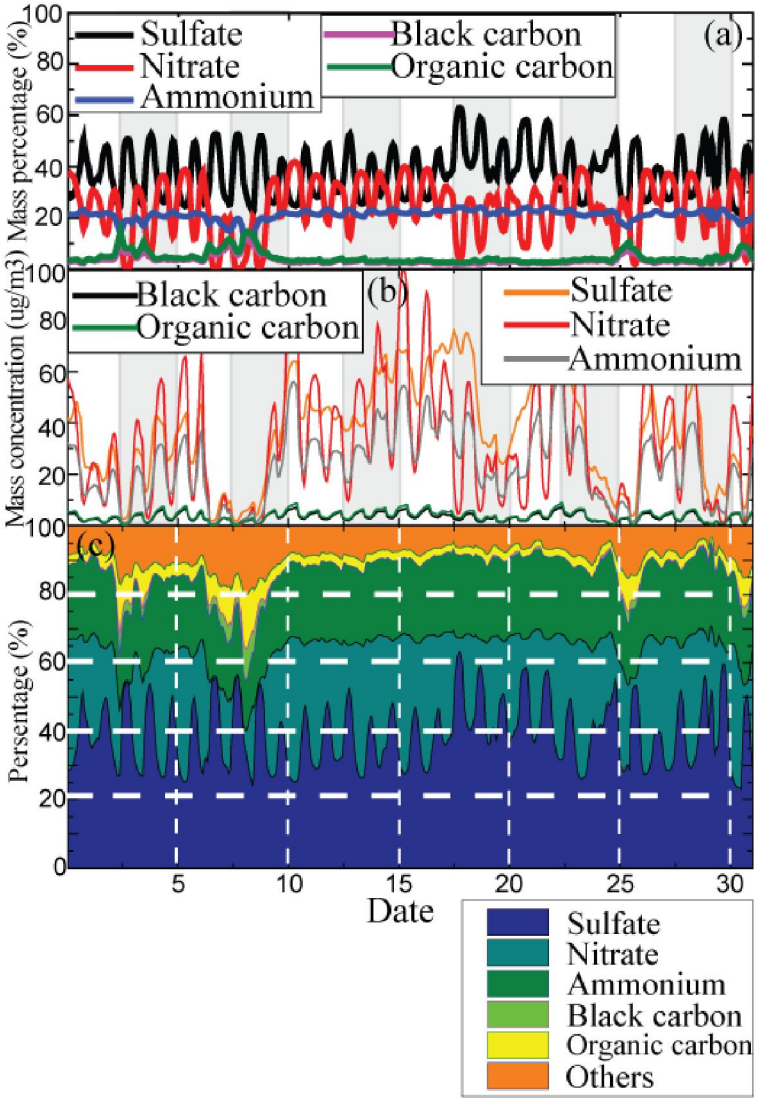

Fig. R1 The regional average values of the mass ratios (%) of sulfate, nitrate and ammonium (a), mass concentrations of major aerosol components (μg m^{-3}; b), and the extinction contribution ratios of the major aerosol components (μg m^{-3}; c) in July over Beijing.

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 Equation (2) in Line 132. The aerosol extinction coefficient is the key factor influencing the visibility. We provided the time series of the regional mean mass concentration of $PM_{2.5}$ and the extinction coefficient (first layer of model) in Beijing from 23–25 July in Fig. R2. The variation features of the extinction coefficient and the mass concentration of $PM_{2.5}$ were similar. During Period C, the variation ranges of these two variables were both very small. Therefore, we think that the drastic changes in visibility during period C were caused by inverse correlation between visibility and extinction coefficient. When the value of extinction coefficient became small, the visibility could change dramatically with a micro-variation of extinction coefficient, by Equation (2). We added the relative expression in Lines 312 – 317.

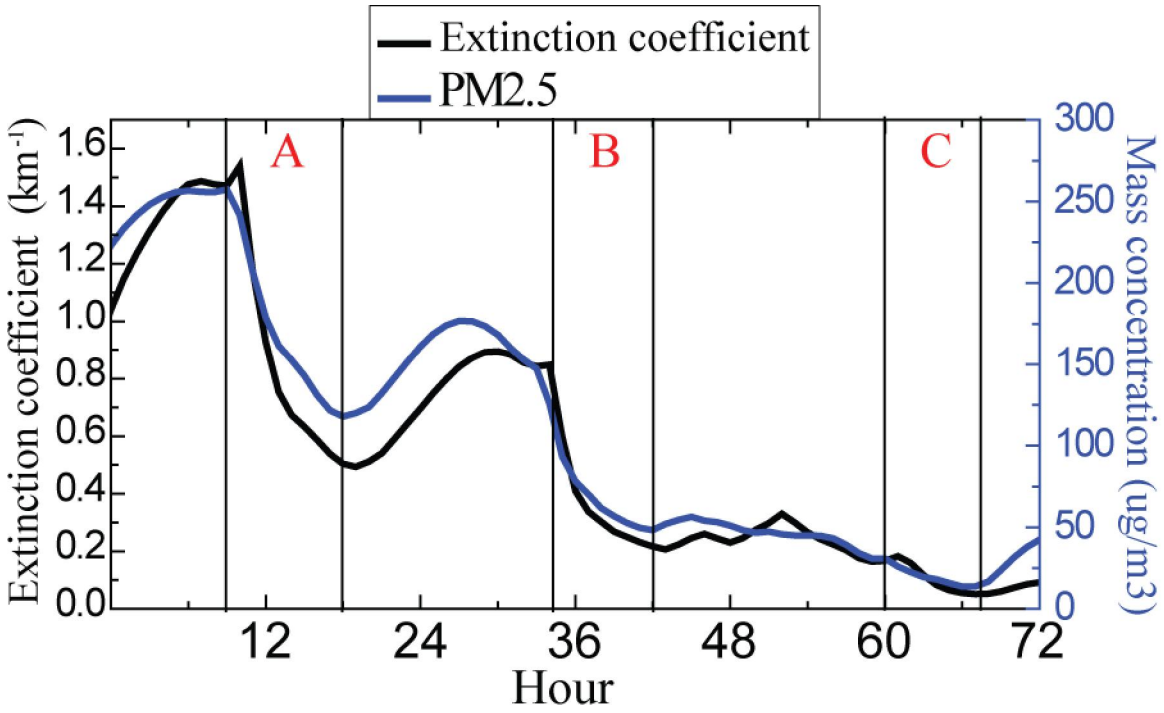

Fig. R2 The time series of the regional average extinction coefficient (km^{-1}) and mass concentration of $PM_{2.5}$ from July 23 to 25 in Beijing.

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 was built solely on the results for Beijing. The ratio of each major aerosol component did not change dramatically in Beijing because the main emission sources of pollutions in this region were anthropogenic activities, and the size distribution of the particles were mainly focused in the accumulation mode. Therefore, we deduced that the relative humidity and mass burden were the two major impact factors which should be

considered and built the function. However, the function is invalid when the ratio of each major aerosol component or the size distribution of particles changes, such as the aerosols over different regions, or when the dust storm happened. In summary, this function is just for the ordinary situation in Beijing. We have modified the expression in Lines 456 – 460

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 think that the main reason that they are similar is that the ratio of each major aerosol component did not change dramatically in Beijing as we mentioned above.

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 a relative humidity of less than 90%. Therefore, all of our analysis and the sensitivity tests were conducted for relative humidity not exceeding 90%. The function was not be used when the relative humidity was higher than 90%. The explanation of this condition has been reemphasized, and the related expressions were modified (Lines 47 – 48, Lines 336 – 337, and Lines 392).

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 your comments. Actually, the phrase should be "components are complicated in winter". We have modified the expression.

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

Reply:

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 condensed the abstract by about 200 words.

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

Reply:

The symbol D_p represents the geometric mean diameter of each aerosol mode which follows the lognormal size distribution. The value of D_p may change with 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 coefficient table. Then, the model used the fitting coefficient table to calculate the aerosol optical properties with different aerosol size distributions. We have modified the relative sentences and provided a clearer explanation.

5. Line 187. Remove "kindly"

Reply:

We have modified the expression.

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

Reply:

We have modified the expression.

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

Reply:

We have modified the expression.

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

Reply:

Yes, we also found this mistake. The gas-phase chemical scheme should be “CB05”. We have modified the expression.

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

Reply:

Thanks for this comment. Straw (and other kinds of biomass) is used for domestic heating in rural areas of the North China Plain in winter, and pollutants emitted from this source cannot be ignored in that area. Zhang et al. (ACP, 2014, 14, 2887–2903) also mentioned that the organic matter 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:

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:

We have modified the expression.

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

Reply:

We have modified the expression.

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

Reply:

We have modified the expression.