Response to Anonymous Referee #2:

Review of "Size-resolved source apportionment of particulate matter in urban Beijing during haze and non-haze episodes" by S. L. Tian, Y. P. Pan, Y. S. Wang

The authors report one year of measurements of numerous chemical species in size-segregated particle samples in Beijing with the analysis results from a PMF model and back trajectory cluster. The results of this paper are quite interesting.

However, the main problem in this manuscript was the standard for how to judge haze and no-haze episodes. Authors use visibility (10km) as their standard, but in previous studies, scientists have used visibility and RH together to determine the haze/no-haze days (i.e. visibility < 10 km and RH < 90%) (Zhang et al., 2015). Since the Beijing government has already published its daily air quality data, I highly suggested the authors use Air Quality index (API) or $PM_{2.5}$ concentration as your standard. I also noticed the authors measured mass concentration for each 48 h sample. Authors could also use mass concentration as their standard. Overall, I would like to reconsider whether to accept or reject after receiving major revisions from authors based on my specific comments below:

Reply: Thanks for your advices. Haze is defined as a weather phenomenon featuring a high concentration of fine particles that leads to a visibility of less than 10 km at a relative humidity (RH) lower than 90% (Sun et al., 2006; Tan et al., 2009; Zhuang et al., 2014). Hence, in the revised manuscript, we use visibility and RH together to determine the haze/no-haze days: sampling days with visibility < 10 km and RH < 90% were defined as haze days, and sampling days with visibility > 10 km and RH < 90% were defined as non-haze days. During the observation period, 12 sets of size-resolved PM samples were collected during non-haze days and 19 sets during haze days (marked in Fig. 2). Of the remaining 21 sets, 15 sets were collected during rain, snow or fog days and 6 sets were observed during dust days (visibility < 10 km, RH < 40%). These samples were excluded from the dataset when we discuss the differences between haze and

non-haze days. The table below shows how 52 weeks samples were divided into different types, according to visibility and RH.

	Visibility	RH	Sample quantity
Haze	**<10 km**	**40%<RH<90%**	**19**
Non-haze	**>10 km**	**no rain, snow or fog**	**12**
Dust	**<10 km**	**RH<40%**	**6**
Other events		**rain, snow or fog**	**15**

We have also tried to use mass concentrations of particles as the standard to judge haze ($PM_{2.5}$ or $PM_{2.1}$ > 75 μg m^{-3}) and non-haze days ($PM_{2.5}$ or $PM_{2.1}$ < 75 μg m^{-3}). However, they shared similar results with that by visibility and RH standard with the exception that some rain, snow, fog or dust days were divided into haze or non-haze days. Finally we chose the visibility and RH standard in the revised version.

Line3: Please labeled the author with "*" to show who is corresponding author.

Reply: Thank you for your careful reminder. According to the guideline of ACP, asterisk (*) was not used to show who is the corresponding author. Instead, the corresponding authors are indicated by names and emails.

Line20: "SO_4^{2-}, NO_3^- and NH_4^+"need to be defined at their first mention in the manuscript. Authors have this problem with other chemical species as well. Please go through the manuscript and change all of them.

Reply: As suggested these species were defined at their first appearance. We have also checked through the manuscript and changed all other items that need to be defined.

Line38: Change "any mitigation strategy" to "future control strategies for air pollution"

Reply: Done.

Line39: Change "pattern" to "patterns" and “periods” to “episodes”

Reply: Done.

Line 49: Change “global climate” to “global climate change through its direct and indirect affects”

Reply: Done.

Line54: More background about extreme haze events needed such as time, PM concentration during the haze episode etc.

Reply: Thanks for your advice. We have added the background information about extreme haze events in the part of Introduction in the revised paper.

Line60: “$PM_{2.5}$” needs to be defined.

Reply: Done.

Line68: What is “droplet mode”? Author also mentioned the “condensation mode” in the following section which also needed a clear definition.

Reply: Typically, the mass distribution is dominated by three modes (or sub-modes): the condensation mode (~0.1- 0.5 μm), the droplet mode (~0.5- 2 μm) and the coarse mode (>2 μm) (Wang et al., 2012; Guo et al., 2010). To simplify the calculation, the particle modes were divided directly by the cut points in this study. The condensation mode particles were designated within the size range of 0.43-0.65 μm, and droplet-mode particles were within 0.65-2.1 μm.

Line83: Authors need to clearly highlight the difference between their research and Zhang et al. (2013). You measured almost the same chemical species and both use PMF, back trajectory cluster and chemical mass closure. The difference in the size-stages should be highlighted. Zhang et al. (2013) also did one year of measurements with higher time resolution (24h), the author’s work was “over short

periods" with shorter time resolution.

Reply: Thanks for your advice. For the Andersen sampler, size-resolved particles were collected on 9 separate filters. Among that, fine particles ($PM_{2.1}$) were collected on 4 filters, so the sampling period was extended to 48h, otherwise it is hard to collect enough particle loading for chemical analysis. To get the size-resolved information, we reduced the time resolution. We focused on the size distributions and associated chemical species because the size parameter is crucial for the evaluation of the effects of PM on human health, visibility, and regional radiative forcing, as well as the determination of the sources, formation mechanisms and conversion processes of the particles (Pillai and Moorthy, 2001; Duarte et al., 2008; Liu et al., 2008; Contini et al., 2014).

In order to highlight the size-resolved information, we have added in depth discussions about the differences between size fractions for chemical species and their variations from non-haze to haze days (in Sect. 4.1). Chemical mass closure (in Sect. 4.2.2 and 4.2.3) and PMF analysis (in Sect. 4.3.3) results for particles in different size fractions were also discussed in detail in the revised manuscript.

Line 86: The author discussed the contributions of different sources to the chemical species in PM in the following sections, so background information is needed in the introduction section.

Reply: Thanks for your advice. The background information of the contributions from different sources to the chemical species (organic carbon and elemental carbon, water-soluble ions and heavy metals) in PM was added in the part of Introduction. Meanwhile, literature reviews on source apportionment for PM in Beijing were also performed and added in the revised manuscript.

Line 90: Change "Positive matrix factorization (PMF)" to "Positive Matrix Factorization (PMF)"

Reply: Done.

Line 98-104: A map is needed to show the location of the site.

Reply: Thanks for your advice. A map of the site location in Beijing was given in the supplementary material as Fig. S1.

Line 106: Authors need to clearly describe how they use two 9-stage samplers. Did they put different filters in each sampler?

Reply: Yes, we put quartz fiber filters and the cellulose membranes in two 9-stage samplers, respectively, which were used to simultaneously collect particles. The mass of particles on each quartz filter was determined by weighing the filter before and after sampling to get mass concentrations of particles in different size fractions. For each quartz filter a quarter was used to measure the concentrations of water soluble inorganic ions and another quarter was used to determine concentrations of OC and EC. For the cellulose membranes, a quarter of each filter was used to determine the concentrations of trace elements.

Line 133: QA/QC should be briefly described in the manuscript and not just by simply citing 2 papers. How did the authors obtain meteorological data? A table or several plots needed in the supplement section.

Reply: QA/QC procedures of sampling process and chemical analysis were briefly described in the revised manuscript. We added a figure in supplementary materials to describe meteorological parameters.

Line143: Authors need to clearly illustrate why they use PMF model in their search.

Reply: The main source apportionment methods can be divided into three categories: emissions inventory, diffusion model and the receptor model, among which, the receptor models have been widely used because the methods are not limited by the pollution discharge conditions, weather and terrain factors. The receptor models based on chemical analysis can be divided into two categories (Yin et al., 2015): the first one, source profiles are needed, such as chemical mass balance (CMB) method; the other one, source profiles are not needed, such as

positive matrix factorization (PMF) method (Paatero and Tapper, 1994). As it is difficult to build huge and accurate source profiles, we take PMF method to do the source apportionment in our study. However, we would like to do further source profiles work in the future and compare the results from PMF and CMB as the subject of our next study.

Line 163: What is "a.g.l" ?

Reply: The definition of "a.g.l" is given in the revised paper, i.e., the abbreviation of "above ground level".

Line 172: What is "TSP"?

Reply: "TSP" is the abbreviation of "total suspended particulate" (TSP, mass of particles with aerodynamic diameters less than 100 μm).

Line 172: Authors need to clearly definite "PM_9 " and "$PM_{2.1\text{-}9}$ ".

Reply: Particles with aerodynamic diameter less than 9 μm were defined as PM_9, while that between 2.1 and 9 μm were defined as $PM_{2.1–9}$.

Line 175: Is it Chinese National Ambient Air Quality Standard (NAAQS)? What is the daily standard for $PM_{2.5}$ and PM_{10}

Reply: In the new Chinese National Ambient Air Quality Standard (GB3095-2012), the daily standard (Grade I) for $PM_{2.5}$ and PM_{10} were 35 and 50 μg m^{-3}, respectively.

Line 179 and 180: Authors also talked about "fine mode" and "coarse mode" in the following sections. Clear definition is needed.

Reply: $PM_{2.1}$ (particles with aerodynamic diameters less than 2.1 μm) represented "fine mode", and $PM_{2.1\text{-}9}$ (particles with aerodynamic diameters between 2.1 and 9 μm) represented "coarse mode".

Line 223 and 224: What is OC in spring and winter? What is OC/EC ratio in spring and winter?

Reply: Concentrations of OC in $PM_{2.1}$ followed the order of summer (20.2 μg m^{-3}) >spring (16.5 μg m^{-3}) > winter (16.2 μg m^{-3}) >autumn (13.4 μg m^{-3}). The high OC concentration in summer was primarily a result of the photochemistry generating more secondary organic carbon (SOC). This result can be confirmed by the OC/EC ratios, which exhibited a seasonal pattern of summer (16.7) >spring (12.7) > autumn (6.7) >winter (4.9).

Line 234: Where is re-suspended soil dust from (long transport from a sandstorm)?

Reply: Re-suspended soil dust may be from both long transport dust and local anthropogenic sources (construction dust and mechanical abrasion processes) dust. The relatively high wind speed in spring, facilitated the ascending of road dust into the atmosphere and hence resulted in the relatively high value of the species in coarse mode (Liu et al., 2014).

Line242: Please give more background information on why the emissions were complex in Beijing during the winter.

Reply: On the one hand, there are more emissions from coal combustion for heating during winter compared with other seasons, especially the retail coal combustion in surrounding areas, which is hard to control (Wang et al., 2006). On the other hand, meteorological conditions in winter are unfavorable for the diffusion of fine particles and precursors (SO_2, NOx, VOCs), this makes the secondary emissions of particles also more complex.

Line 255: Need to cite papers to support Cl^- and K^+ were from industrial pollution. Author also mentioned K^+ was from biomass burning in the following manuscript.

Reply: OC, Cl^-, K^+, Na^+, Na, K, V, Cr, Mn, Cu, As and Mo all belonged to the third group. Here we want to say Cl^- and K^+ are good biomass burning tracers and V, Cr, Mn and Cu are good industrial pollution tracers. Hence, the species

in the third group may represent mixed sources from biomass burning and industrial pollution.

Line 264: What are the precursors of SOC?

Reply: The precursors of SOC are mainly volatile organic compounds (VOCs), which contain both biological sources (such as monoterpene and sesquiterpene) and anthropogenic sources (such as aromatics) (Jacobson et al., 2000).

Line 274: Authors need to mention how many haze and no haze days occurred from March 2013 to February 2014 in total. Authors only sampled from Monday to Wednesday at each week. The authors need to defend how representative the samples are. Beijing can be influenced by sandstorms, especially in spring and fall. Are there any sandstorms that occurred during the observation period? How did authors deal with those samples during the sandstorm period?

Reply: The size–resolved particles were collected weekly as Andersen sampling is hard to be conducted. During this observation period, 12 sets of size-resolved PM samples were collected during non-haze days and 19 sets during haze days (marked in Fig. 2), which can cover 36 days and 57 days, respectively. There were 132 non-haze days with average visibility more than 10 km and 133 haze days with average visibility less than 10 km and RH lower than 90% from March 2013 to February 2014. The Andersen samples can well represent the concentrations of particles during this study period. Annual average concentration of fine particles was 67.3 μg m^{-3} based on the Andersen dataset in this study, which was very close to that from the hourly average data of $PM_{2.5}$ through the year (70.9 μg m^{-3}). However, we would like to do some continuous sampling work in the future.

Of the remaining 21 sets, 15 sets were collected during rain, snow or fog days and 6 sets were observed during dust days (visibility < 10 km, RH < 40%). These sets were excluded from the dataset when we discussed the differences between haze and non-haze days.

Line 282: Change “markedly” to “significantly”

Reply: Done.

Line 284: Authors need to give the equation to show how to calculate $R_{H/N}$

Reply: $R_{H/N}=C_H/C_N$, C_H--concentration of chemical species on haze days, C_N--concentration of chemical species on non-haze days.

Line 300: I do not know of any references that indicate any toxicity of Na^+ , K^+ and Cl^-, and perhaps other species listed. The toxicity of all species listed should be verified.

Reply: Thanks for the comments. In the revised paper, “These species had the highest toxicity” was changed to “Among these species, Mo, Pb, Cd and Tl had high toxicity”.

Line 305: The author needs to discuss the reasons for “the highest $R_{H/N}$ for Na^+, K^+ and Cl^- in the coarse fraction was observed in summer”

Reply: The highest $R_{H/N}$ for Na^+, K^+ and Cl^- in the coarse fraction was observed in summer mainly due to low concentrations on non-haze days and relatively high concentration on haze days. The lower concentrations of coarse particles occurred in summer were likely related to more precipitation in this season. High concentration of K^+ and Cl^- in coarse mode on haze days mainly associated with biomass burning (Du et al., 2011). One of the samples that represent haze days in summer was collected between June 17 and 19. During this period, the wheat straw burning in the surrounding areas would affect both fine and coarse particle pollution of Beijing (Wang et al., 2015; Yan et al., 2015; Cheng et al., 2014).

Line 306-315: In the discussion of “NO_3^-, SO_4^{2-} and NH_4^+,” especially in the discussion of correlation, the authors miss the important fact that the formation of

NH4NO3 is thermodynamically favored by high relative humidity and low temperatures (winter). NH_4NO_3 would dissociate to NH_3 and HNO_3 at high temperatures (summer).

Also, it will be interesting to calculate the ion balance to see if any variations of the ionic charge balance (deficiency of anions) in haze and no-haze days.

Reply: Thanks for the advice. To investigate the effect of RH and temperature, correlation of NO_3^-, SO_4^{2-} and NH_4^+ in different seasons were discussed in the revised version (in Sect. 4.1.1). Based on the correlation results, however, we can infer the possible existence form of NO_3^-, SO_4^{2-} and NH_4^+ rather than the formation process. Besides, we calculated ion balance for both fine and coarse particles on haze and non-haze days. The results were added in Sect. 4.1.1.

Line 337: What are the precursors and why are the concentrations of those precursors high?

Reply: The precursors are SO_2, NH_3 and NO_x. The high concentrations of these precursors were mainly due to meteorological conditions in winter, which are unfavorable for the diffusion of precursors. In addition, more SO_2 were emitted from coal combustion during winter heating period.

Line 345: Where did authors show the results? Any table or figure?

Reply: Sorry for the confusion. The results were shown in Table S3 and we have added the indication in the revised paper.

Line 340: Change “models” to “model”. What software did authors use to run multiple linear regression? If this model have been used in other research, please cite those papers. More information needed.

Reply: SPSS 16.0 was used to run multiple linear regressions. This information was added in Sect. 4.4 in the revised manuscript and references were cited here.

Line 352-353: Equation needs to be labeled with number (i.e. Line 152).

Why do the authors only include those 7 variables in this equation? How did authors drop the other variables?

From the coefficients in the equation, it looks like the RH, WS and Ca^{2+} dominated the visibility changes. More information and careful discussion are needed in this part.

Reply: Thank you for your advice. Equations were labeled with number in the revised manuscript.

In this study 93 variables were investigated first but only 7 variables were selected finally because they had high correlation coefficients (> 0.5) with visibility. The factors of RH, WS and Ca^{2+} are important in the explanation of visibility changes. High RH is conducive to particulate matter hygroscopic growth and generation of secondary species, in turn reduces the visibility. Besides, Ca^{2+} is crucial in affecting visibility because it associated with dust, which will strongly reduce the visibility. On the contrary, high wind speed is favorable for the diffusion of fine particles and can improve visibility.

Line364: Change "thereby" to "therefore"

Reply: Done.

Line 368: The research in Maenhaut's paper was not conducted in Beijing. Authors need more strong support to conclude that "Ca^{2+} in coarse particles, which was primarily from construction dust". What about the contribution of dust from long transportation?

Reply: Thanks for the question. After a review on the literatures about particle pollution in Beijing, we found that Ca^{2+} in the coarse particles might be from both construction dust and long transportation dust (Liu et al., 2014). However, long transportation dust is not easy to control. Thus, we particularly stressed here that construction dust must be controlled to improve visibility. Meanwhile, more references which were conducted in Beijing were cited here.

Line 371 to 372. Where do those data come from (from March 2012 to February

2013)? More information is needed.

Reply: Data used to validate the equation (from March 2012 to February 2013) were obtained from previous studies. The reference is added in the revised manuscript (Miao, 2014).

Line 376: Why did the authors choose 15km to do the analysis instead of 10km as they mentioned before?

Reply: In the original paper, we chose 15 km mainly because in the scatter diagram discrete points primarily appeared for visibilities greater than 15 km. In the revised manuscript, 10 km was chosen to do the analysis as it is the cut-off point of haze and clear days. And a regression equation was developed to characterize the relationship between the visibility and the chemical species concentrations when the visibility was less than 10 km.

Line 386: Change "contributions of OM to $PM_{2.1}$ were" to the "contribution of OM to $PM_{2.1}$ was"

Reply: Done.

Line 397: Was the order of CM > OM > SNA for both haze and no-haze days ? Why did this happened? More careful discussion is needed.

Reply: Sorry for the confusion. The contributions of these species in coarse particles followed the order of CM > OM > SNA on both haze and non-haze days. In fine particles, however, the order was OM>CM>SNA on non-haze days and OM>SNA>CM on haze days. In summary, the relatively contributions of OM and CM to the particle mass decreased from non-haze to haze days whereas that of SNA increased from non-haze to haze days.

Line 399-401: This conclusion is interesting. More explanations are needed, because most of ultra-fine particles were from the secondary chemistry formation instead of primary emissions.

Reply: Thank you for your advice. Particles in size fraction less than 0.1 μm are mainly from primary emissions. We made a mistake here and the sentence "These fractions are related to the primary emissions of PM" was deleted in the revised manuscript.

Line 427-429: Why did authors choose six sources instead of five or seven sources?

Reply: The optimal number of sources was selected by inspecting the variation of Q from PMF with varying number of sources (from 4 to 8) and by studying the physical meaningfulness of the calculated factors. In the original paper, six sources were identified for all the size fractions. In the revised manuscript, however, 6 and 7 sources were selected for fine particles and coarse particles, respectively.

Line 439: Authors need to explain why the contribution of coal combustion was higher in coarse mode that fine mode?

Reply: Thanks for your question. In the revised manuscript, haze and non-haze days were reclassified based on visibility and RH together, and then PMF analysis was improved and performed respectively for the fine (the input data included the mass concentrations and chemical species in particles with size bins of <0.43, 0.43-0.65, 0.65-1.1 and 1.1-2.1 μm) and coarse fractions (the input data included the mass concentrations and chemical species for particles in size fractions of 2.1-3.3, 3.3-4.7, 4.7-5.8, 5.8-9 and >9 μm) (Contini et al., 2014). The results showed that the contribution of coal combustion was higher in fine mode than that in coarse mode.

Line 454: The authors need to mention that vehicles equipped with three-way catalysts are an important source of NH3, which may also contribute to the SIA.

Reply: Thank you for your advice. This information was added in the revised manuscript.

Line 476: Why does the industrial pollution not contribute as much on haze days?

Reply: Thanks for your question. We agree with the reviewer that the results regarding industrial pollution in the original manuscript were unreasonable. In the revised manuscript, haze and non-haze days were reclassified based on visibility and RH together, and then PMF analysis was improved and performed respectively for the fine and coarse fractions. The results showed that the contribution of industrial pollution on haze days was also higher than that on non-haze days.

Line 513: Change "result" to "results"

Reply: Done.

Line518: Change "to" to "in order to"

Reply: Done.

Line539 to 540: NW, SE and NE need definition.

Reply: Done. Northwest (NW), southeast (SE) and northeast (NE).

Line545: Change "strong effect" to "strong impacts"

Reply: Done.

Line 593: This full equation does not need to be included in the conclusions.

Reply: The equation in the conclusions has been deleted.

Line 831: Great improvements need to be done for figure 7. First, some numbers (i.e. 16% and 15%) are overlapped with the bold dash line and cannot be read. Second, the numbers on the pie charts are hard to read.

Reply: We have redraw Figure 7 to improve the readability.

Line 797: Check the reference. "2013a" was not necessary.

Reply: Done.

Line 808: Check the reference. “2013b” was not necessary.

Reply: Done.

Reference:

Cheng, Y., Engling, G., He, K. b., Duan, F. k., Du, Z. y., Ma, Y. l., Liang, L. l., Lu, Z. f., Liu, J. m., Zheng, M., and Weber, R. J.: The characteristics of Beijing aerosol during two distinct episodes: Impacts of biomass burning and fireworks, Environ Pollut, 185, 149-157, 2014.

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