

Dear Prof. Shao,

On behalf of my co-authors, I would like to thank you very much for your time handing our paper “acp-2015-26 (Size-resolved source apportionment of particulate matter in urban Beijing during haze and non-haze episodes)”. We also appreciate the comprehensive comments and valuable suggestions given by the reviewers for improving the quality of the manuscript. All of the comments have been considered to improve, revise, and correct the manuscript. Detailed responses to the comments are given below in blue font (the corrections in the revised manuscript were marked in red font). In addition, the language of this version was further polished by several native English speakers at the company of AJE (editorial certificate attached). If you have any additional questions about this paper, please do not hesitate to contact me.



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Manuscript title:
Size-resolved source apportionment of particulate matter in urban Beijing during haze and non-haze episodes

Authors:
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Sincerely yours,

Yuepeng, Pan

August 26, 2015

Response to Anonymous Referee #1:

Review of “Size-resolved source apportionment of particulate matter in urban Beijing during haze and non-haze episodes” by Tian and co-authors.

Tian et al. present size-resolved chemical composition of aerosol particles during haze and non-haze episodes in Beijing. In addition a source apportionment analysis is performed to the data. The authors present valuable data, however, the data treatment is unclear and need to be clarified before publication in ACP. The English grammar needs to be revised by a native.

Reply: Thank you for your suggestions and comments. As shown below, your suggestions and comments will strengthen the quality of our manuscript. We have revised the paper accordingly. In addition, the revised manuscript was edited by several native English speakers at AJE to improve the language.

P9411-L2: The references of Paatero and Tapper, (1994) or Paatero (1997) are probably more appropriate to refer to the PMF technique.

Paatero, P., Tapper, U., 1994. Positive matrix factorization: a nonnegative factor model with optimal utilization of error estimates of data values. *Environmetrics* 5, 111-126.

Paatero, P., 1997. Least square formulation of robust non-negative factor analysis. *Chemometrics and Intelligent Laboratory Systems* 3, 23-35.

Reply: Thank you for this advice. These references were added in the revised paper.

P9412 – L14: Change the section title to “PM mass concentrations and chemical composition”.

Reply: Done.

P9419–L23: Other runs of the PMF model did not result in additional sources or different sources between size fractions? The use of both size fractions is not

providing additional insights, since the same sources were identified in both fractions...

Reply: Thank you for your advice. In the original manuscript, the PMF model was applied separately to PM_{2.1}, PM_{2.1-9} and each size fraction (< 0.43, 0.43-0.65, 0.65-1.1, 1.1-2.1, 2.1-3.3, 3.3-4.7, 4.7-5.8, 5.8-9 and > 9 μm). In addition, only 52 samples were collected during each run. To improve the quality of the source apportionments, PMF analysis was performed in the revised version of the manuscript for the fine mode (the input data included the mass concentrations and chemical species of the 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 and 5.8-9 μm) (Contini et al., 2014). The new runs identified different sources for the fine and coarse fractions. For example, mineral dust and road dust were identified in the coarse fractions but only road dust was identified in the fine fractions. In addition, the contributions of most anthropogenic-related sources increased from non-haze to haze days. We believe that these new findings are more reasonable compared with the original findings. We have added more discussion regarding potential improvements of the PMF model results in the revised paper (See Sect. 4.3.1. and 4.3.2).

P9420 – L3: Rephrase this statement. This is confusing since this source is named as SIA.

Reply: Thank you for your advice. “SIA” was changed to “secondary inorganic aerosol (SIA)” in the revised version of the manuscript.

P 9420 – L8: As concentrations

Reply: Done.

P 9421 – L5: Add a reference about K⁺ as a tracer for biomass burning.

Reply: Done.

P 9421: It would be interesting to study the seasonality of the sources identified in both fractions. This exercise can offer additional insight into the results.

Reply: Thank you for your insightful suggestion. We have discussed the seasonality of the sources identified in the fine and coarse fractions in the revised manuscript. See Sect. 4.3.1.

P9424 – L23: All anthropogenic-related sources increased during haze days except industrial pollution. Do the authors have any explanation for this?

Reply: 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. Next, the PMF analysis was improved and performed separately for the fine and coarse fractions. The results showed that the absolute mass concentration of industrial pollution on haze days were higher than those on non-haze days.

P9422 – L14: How the results shown in Figure 7 are obtained? The PMF analysis was performed in the fine and coarse fractions, how is then extrapolated to 11 size fractions?

Reply: We apologize for any confusion. In the original manuscript, the PMF method was applied separately to PM_{2.1}, PM_{2.1-9} and each of the size fractions (< 0.43, 0.43-0.65, 0.65-1.1, 1.1-2.1, 2.1-3.3, 3.3-4.7, 4.7-5.8, 5.8-9 and > 9 μm); thus, the source apportionment results for the 11 size bins are shown in Figure 7. To further improve the accuracy of the results from the PMF model in the revised manuscript, the PMF analysis was performed for the fine (the input data included concentrations of mass and chemical species for particles in size fraction < 0.43, 0.43-0.65, 0.65-1.1, 1.1-2.1 μm) and coarse fractions (the input data included concentrations of mass and chemical species for particles in size fraction 2.1-3.3, 3.3-4.7, 4.7-5.8 and 5.8-9 μm) (Contini et al., 2014).

P9423 – L21: The section title “Back trajectory cluster analysis”

Reply: Done.

P9425 – L12: The different chemical constituents are included in the fine or coarse particles. This sentence needs to be rephrased.

Reply: Thank you for your advice. “In addition to the fine and coarse particles” was changed to “In addition to particle size distributions”.

Figures:

- Improve the readability of all figures.
- Y axis: units should be between brackets, e.g. ($\mu\text{g}/\text{m}^3$) instead of $\mu\text{g}/\text{m}^3$.

Reply: Thank you for your advice. We have redrawn all of the figures, and the units for the Y-axes were placed between brackets.

Figure 5: Include the % of species in this graph and include comments about this in the manuscript.

Reply: Done.

Figure 6: Apart of the relative contribution (%) of each source to each fraction, show the mass concentration of the sources in $\mu\text{g}/\text{m}^3$. It might be the case of a source increasing its contribution from haze to non-haze days but not its absolute mass concentration, for example.

Reply: Thanks you for your valuable suggestion. Your suggestions have been implemented in the final version of the manuscript.

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: Thank you for your advice. Haze is defined as a weather phenomenon that features a high concentration of fine particles that result in 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 were collected during haze days (marked in Fig. 2). Of the remaining 21 sets, 15 sets were collected during rain, snow or fog days and 6 were collected during dust days (visibility < 10 km, RH < 40%). These samples were excluded from the dataset when discussing the differences between haze and non-haze days. The table below shows how the samples collected over 52

weeks 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

In addition, we tried to use the mass concentrations of the particles as a standard for judging haze ($PM_{2.5}$ or $PM_{2.1} > 75 \mu g m^{-3}$) and non-haze days ($PM_{2.5}$ or $PM_{2.1} < 75 \mu g m^{-3}$). However, the results were similar to those obtained when using visibility and RH, except that the rain, snow, fog and dust days were divided into haze or non-haze days. Finally, we chose to use the visibility and RH standard in the revised version of the manuscript.

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

Reply: Thank you for pointing out this problem. However, according to the ACP guidelines, an asterisk (*) is not used to denote 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. In addition, we have also checked the manuscript and defined all other terms as needed.

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: Thank you for your advice. We have added background information about extreme haze events in the 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) (Guo et al., 2010; Wang et al., 2012). However, to simplify our calculations, the particle modes were divided directly by the cutting points in this study. The condensation mode particles were defined within the size range of 0.43-0.65 μm, and droplet-mode particles were defined within the size range of 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: Thank you for your advice. For the first samples obtained by the Andersen sampler, size-resolved particles were collected on 9 separate filters. The sampling period was extended to 48 h to collect fine particles (PM_{2.1}) on 4 filters because it was difficult to collect enough particles for chemical analysis. To obtain size-resolved information, we reduced the time resolution. In addition, we focused on the size distributions and associated chemical species because the size parameter is crucial for evaluating the effects of PM on human health, visibility, and regional radiative forcing and for determining the sources, formation mechanisms and conversion processes of the particles (Contini et al., 2014; Duarte et al., 2008; Liu et al., 2008; Pillai and Moorthy, 2001).

To highlight the size-resolved information, we added in depth discussions regarding the differences between the size fractions for chemical species and their variations from non-haze to haze days (in Sect. 4.1). In addition, chemical mass closure (in Sect. 4.2.2 and 4.2.3) and the PMF analysis (in Sect. 4.3.3) results for the particles in different size fractions were 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: Thank you for your advice. Background information describing the contributions of different sources to the chemical species (organic carbon and elemental carbon, water-soluble ions and heavy metals) in PM was added in the Introduction section. In addition, a literature review of the source apportionment of PM in Beijing was performed and was 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: Thank you for your advice. A map of the site location in Beijing was given in the supplementary material in 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 placed the quartz fiber filters and cellulose membranes in two 9-stage samplers that were used to simultaneously collect particles. The masses of the particles on each quartz filter were determined by weighing the filter before and after sampling to obtain the mass concentrations of the particles in different size fractions. For each quartz filter, a quarter of the filter was used to measure the concentrations of water-soluble inorganic ions and another quarter of the filter was used to determine the concentrations of OC and EC. For the cellulose membranes, a quarter of each filter was used to determine the trace element concentrations.

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

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

Reply: Three main types of source apportionment methods were used: emissions inventory, diffusion models and receptor models. Among these methods, receptor models have been widely used because they are not limited by the pollution discharge conditions, weather or terrain factors. The receptor models based on

chemical analysis can be divided into two types (Yin et al., 2015). The first type of model includes the chemical mass balance (CMB) method and requires source profiles. The second type of model includes the positive matrix factorization (PMF) method and does not require source profiles (Paatero and Tapper, 1994). Because it is difficult to build large and accurate source profiles, we used the PMF method for source apportionment in our study. However, we would like to create additional profiles in the future and compare results from PMF and CMB.

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 for “total suspended particulate” (TSP, mass of particles with aerodynamic diameters less than 100 μm).

Line 172: Authors need to clearly define “PM₉” and “PM_{2.1-9}”.

Reply: PM₉ was defined as particles with aerodynamic diameters less than 9 μm , and PM_{2.1-9} was defined as particles with aerodynamic diameters between 2.1 and 9 μm .

Line 175: Is it Chinese National Ambient Air Quality Standard (NAAQS)? What is the daily standard for PM_{2.5} and PM₁₀

Reply: In the new Chinese National Ambient Air Quality Standard (GB3095-2012), the daily standards (Grade I) for PM_{2.5} and PM₁₀ were 35 and 50 $\mu\text{g m}^{-3}$, 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) were defined as “fine mode”, and PM_{2.1-9} (particles with aerodynamic diameters between 2.1 and 9 μm) were defined as “coarse mode”.

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

Reply: of the concentrations of OC in the PM_{2.1} decreased between seasons in the following order: 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 the summer primarily resulted from the generation of more secondary organic carbon (SOC) through photochemistry. This result can be confirmed by the OC/EC ratios, which decreased between the seasons as follows: 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 result from the transport of dust over long distances and from local anthropogenic sources (construction dust and mechanical abrasion processes). The relatively high wind speed in the spring facilitates the ascent of road dust into the atmosphere and results in relatively high atmospheric dust concentrations in the coarse fraction (Liu et al., 2014).

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

Reply: The emissions from coal combustion for heating are higher during the winter than during the other seasons, especially for retail coal combustion in the surrounding areas, which is difficult to control (Wang et al., 2006). However, the meteorological conditions in winter are unfavorable for the diffusion of fine particles and precursors (SO₂, NO_x, VOCs), making secondary particle emissions 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, Mn, Cu and Mo all belonged to the third group. Here, we want to state that Cl^- and K^+ are good tracers for biomass burning and Mn and Cu are good tracers for industrial pollution. 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 mainly include volatile organic compounds (VOCs), which contain 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 because using the Andersen sampler was difficult. During this observation period, 12 sets of size-resolved PM samples were collected during non-haze days and 19 sets were collected during haze days (marked in Fig. 2), which can cover 36 days and 57 days, respectively. Overall, there were 132 non-haze days with an average visibility of more than 10 km and 133 haze days with an average visibility of less than 10 km and RH lower than 90% from March 2013 to February 2014. The Andersen samples represented the concentrations of particles during this study period well. The annual average concentration of fine particles was $67.3 \mu\text{g m}^{-3}$ based on the Andersen dataset in this study, which was very similar to the hourly

average PM_{2.5} data obtained throughout the year (70.9 μg m⁻³). However, we would like to perform continuous sampling work in the future.

Of the remaining 21 sets, 15 were collected during rain, snow or fog days and 6 were collected 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: $R_{H/N} = C_H / C_N$, where C_H is the concentration of the chemical species on haze days and C_N is the concentration of the 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: Thank you for your comment. In the revised paper, “These species had the highest toxicity” was changed to “Among these species, 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} values for Na⁺, K⁺ and Cl⁻ in the coarse fraction were observed during the summer, mainly due to their low concentrations on non-haze days and relatively high concentrations on haze days. Lower coarse particle concentrations occurred in the summer and were likely related to more precipitation in this season. High K⁺ and Cl⁻ concentrations in the coarse mode were mainly associated with biomass burning on haze days (Du et al., 2011). One of the samples that represents haze days during the summer was collected

between June 17 and 19. During this period, wheat straw burning in the surrounding areas would affect fine and coarse particle pollution in Beijing (Cheng et al., 2014; Wang et al., 2015b; Yan et al., 2015).

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 NH_4NO_3 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: Thank you for your advice. To investigate the effects of RH and temperature, the correlations between NO_3^- , SO_4^{2-} and NH_4^+ during different seasons were discussed in the revised version of the manuscript (in Sect. 3.4). However, based on the correlation results, we inferred the possible existence of NO_3^- , SO_4^{2-} and NH_4^+ rather than their formation. In addition, we calculated the ion balances for fine and coarse particles. These results were added in Sect. 3.4 and 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 mainly resulted from the meteorological conditions during the winter, which are unfavorable for the diffusion of precursors. In addition, more SO_2 was emitted from coal combustion during the winter heating period.

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

Reply: We apologize for this confusion. The results were shown in Table S3, which is indicated in the revised version of the 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 for the multiple linear regressions. This information was added in Sect. 4.4 in the revised manuscript and references are 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. The equations were labeled with numbers in the revised manuscript.

In this study, 93 variables were investigated; however, only 7 variables were selected because they were highly correlated (> 0.5) with visibility. The factors of RH, WS and Ca^{2+} are important for explaining visibility changes. High RH promotes the hygroscopic growth of particulate matter and the generation of secondary species, which reduce visibility. In addition, Ca^{2+} affects visibility because it is associated with dust, which strongly reduces visibility. In contrast, high wind speed is favorable for fine particle diffusion 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: Thank you for asking this question. After reviewing the literature

regarding particle pollution in Beijing, we found that the Ca^{2+} in the coarse particles could result from construction dust and long transportation dust (Liu et al., 2014). However, long transportation dust is not easy to control. Thus, we particularly stressed that construction dust must be controlled to improve visibility. Meanwhile, additional references were conducted in Beijing and are cited here.

Line 371 to 372. Where do those data come from (from March 2012 to February 2013)? More information is needed.

Reply: The data used to validate the equation (from March 2012 to February 2013) were obtained from previous studies. The appropriate references were 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: We used 15 km in the original paper mainly because discrete points primarily appeared in the scatter diagram for visibilities greater than 15 km. In the revised manuscript, 10 km was used because it is the cut-off point for hazy and clear days. In addition, a regression equation was developed to characterize the relationships between the visibility and chemical species concentrations when the visibility was less than 10 km.

Line 386: Change “contributions of OM to $\text{PM}_{2.1}$ were” to the “contribution of OM to $\text{PM}_{2.1}$ was”

Reply: Done.

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

Reply: We apologize that did not clearly describe this result. The contributions of these species in the coarse particles decreased as follows on both haze and

non-haze days: CM > OM > SNA. However, in the fine particles, 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 and the relative contributions 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 the less than 0.1 μm size fraction mainly result from primary emissions. Because we made a mistake in our original manuscript, the following sentence was deleted: “These fractions are related to the primary emissions of PM”.

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 variations of Q from PMF with a 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 of the size fractions. However, in the revised manuscript, 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 than fine mode?

Reply: Thank you for your question. In the revised manuscript, haze and non-haze days were reclassified based on visibility and RH together. Next, PMF analysis was improved and performed 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 and 5.8-9 μm) (Contini et al., 2014). The results showed that the contributions of coal combustion were higher in the fine mode than in the coarse mode.

Line 454: The authors need to mention that vehicles equipped with three-way catalysts are an important source of NH_3 , 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: Thank you 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, and PMF analysis was improved and performed separately for the fine and coarse fractions. The results showed that the contributions of industrial pollution were higher on haze days than 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 redrawn Figure 7 to improve its readability.

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

Reply: Done.

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

Reply: Done.

Response to Xia Ke's comments:

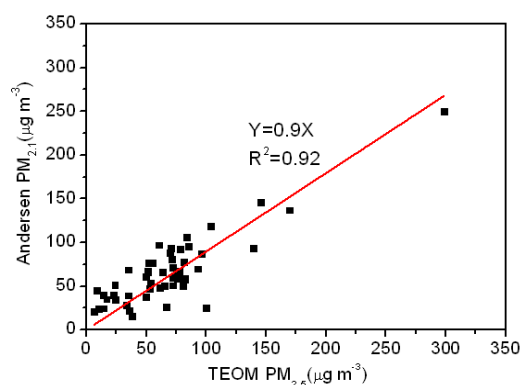
This study presents data of size-resolved aerosol chemical components in Beijing and analysis of their sources during the four seasons. While I see its scientific value, I also feel that there are some critical issues that need to be addressed.

1. The study used PM_{2.1} and PM_{2.1-9} data to represent fine and coarse parties, respectively, instead of the traditional PM_{2.5} and PM₁₀. How will this choice affect the final results? Uncertainty assessments can be easily done using known mass size distribution data.

Reply: Traditionally, fine and coarse particles are defined as particulate matter (PM) with aerodynamic diameters less than 2.5 μm (PM_{2.5}) and PM with aerodynamic diameters between 2.5 and 10 μm (PM_{2.5-10}), respectively. Thus, coarse particles are defined as PM_{2.5-10} rather than as PM₁₀.

However, no uniform definitions for fine and coarse particles are generally used for field observations, which depend on the cut off size of the individual sampler. In previous studies, for example (Matsumoto et al., 2012), PM samples with diameters less than 2.0 μm and between 2.0 and 10 μm were defined as fine and coarse particle, respectively. For the PM samples collected by MOUDI, PM_{1.8} was used to represent fine particles (Sun et al., 2013; Wang et al., 2012). Similarly, when Andersen samples were collected, fine particles were defined as PM_{2.1} (Li et al., 2011; Wang et al., 2011).

The definition of fine and coarse particles did not significantly affect the results in this study. For example, we compared the mass concentrations of PM_{2.1} obtained by weighing the filters before and after sampling with those obtained for PM_{2.5} by the commercial TEOM instrument. The figure below shows that the linear correlation between PM_{2.1} and PM_{2.5} was good ($R^2=0.92$, $P<0.05$) and that the concentrations of PM_{2.1} were only 10% lower than those of PM_{2.5}.



2. Why chose the weighing condition of RH as 10%? Cellulose filter or even quartz filter should be taken with static at such dry condition. Although the filters were eliminated static, the results of microbalance should not be stable during the multiple weighing processes. Thus, the uncertainty of aerosol mass should be addressed.

Reply: Thank you for your comment. We apologize for our mistake. The weighing condition of RH is $22 \pm 3\%$. Throughout the weighing process, static was eliminated from each quartz filter (AD1683, A&D company, limited, Japan). In the lab, the microbalance was stable and the balance time for each weighing process was less than 10 s.

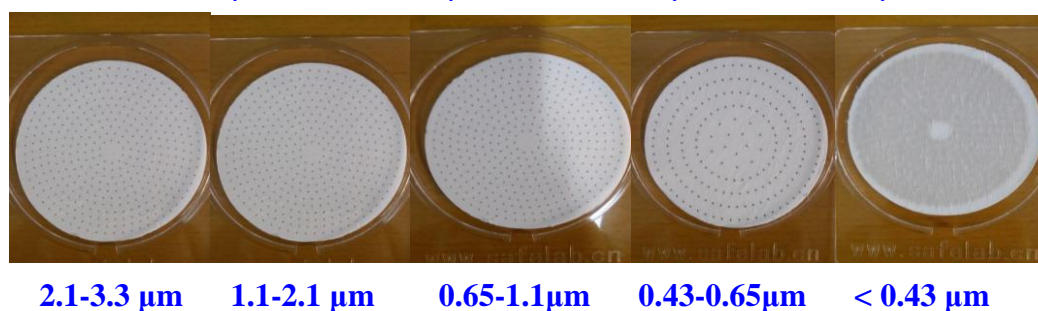
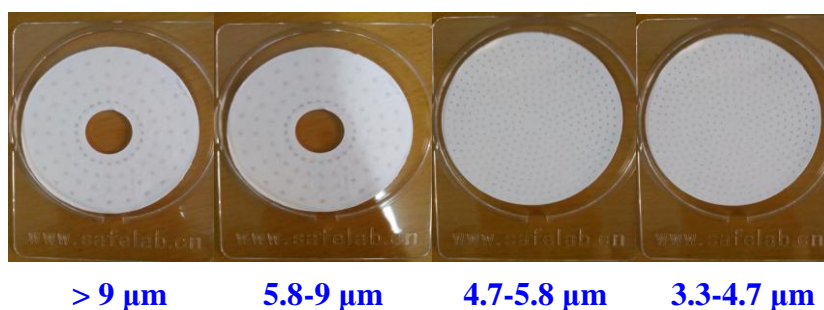
Similar equilibrium conditions have been widely used for weighing in previous studies. For example, the mass of particles on each quartz filter was determined after 48 h of equilibration in a glass chamber (25 ± 0.5 °C, $35 \pm 2\%$ RH) when the dust storm was studied in Xi'an based on Andersen samples (Wang et al., 2015a). During the observations in Nanjing, the glass fiber filters were baked at 500°C for 2 h to eliminate organic species and were conditioned in a desiccator for 24 h before weighing (Wang et al., 2003).

In addition, as shown in the above figure, the mass concentrations of $PM_{2.1}$ were comparable to those of $PM_{2.5}$, the latter of which is an alternative standard method recommended by the US EPA. These results indicated that the weighing method is sound and reasonable for further mass closure analysis.

3. QA/QC procedures of sampling process are missed in this manuscript, which are important for a scientific paper presenting the first-hand data. The Anderson sampler should be blocked during heavy pollution conditions, and then the collected samples were not evenly distributed. This phenomenon should affect the chemical analysis, especial for OC and EC (choice of spots).

Reply: Thank you for your suggestions. We have added detailed descriptions of the QA/QC procedures used in the revised manuscript.

To prevent particles from blocking the sampler during sampling, the samplers were cleaned using an ultrasonic bath for 30 min before each sampling. In addition, the sampling flow rates were calibrated before collecting each sample and were monitored using a flow meter throughout sampling. These control measures ensured that the Andersen sampler was not blocked during the sampling period. Even on heavily polluted days with $PM_{2.5}$ values of $169.8 \mu\text{g m}^{-3}$, the filter samples obtained using the Andersen sampler were evenly distributed and the spots were obvious, as shown below.



4. Meteorological parameters seemed to be collected, but was not shown in the manuscript. Aerosol should bound when $RH < 40\%$, and thus, samples under these conditions should be removed.

Reply: Thank you for your advice. In the revised manuscript, we added a

figure in the supplementary material to describe the meteorological parameters.

Additionally, we used visibility and RH to determine the haze/no-haze days as follows: sampling days with visibility < 10 km and RH < 90% were defined as haze days, while 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 were collected during haze days (marked in Fig. 2). Of the remaining 21 sets, 15 were collected during rain, snow or fog days and 6 were observed during dust days (visibility < 10 km, RH < 40%). These sets were excluded from the dataset when discussing the differences between haze and non-haze days. The table below shows how the samples obtained over 52 weeks 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

To determine whether the aerosols were bound or not when RH < 40%, we compared the mass concentrations of PM_{2.1} with those of PM_{2.5} measured using the commercial TEOM instrument under different RH conditions. A good linear correlation was observed between PM_{2.1} and PM_{2.5} under both RH > 40% (slope = 0.93, R² = 0.94, P < 0.05) and RH < 40% days (slope = 0.87, R² = 0.92, P < 0.05). After reviewing the literature, we observed that Andersen samplers can be used to collect dust storm samples when the RH is much lower than 40% (Wang et al., 2015a; Wang et al., 2014). In the revised paper, samples obtained when the RH was lower than 40% and when the visibility was less than 10 km were defined as dust samples and were removed from the haze days.

5. In general, results generated from PMF model could be questionable if less than 100 samples were used in the model.

Reply: In the revised manuscript, the PMF analysis was performed for the fine mode (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 and 5.8-9 μm) (Contini et al., 2014). The numbers of samples used in the model for both fine and coarse particles were 208.

Why the percentage contributions for chemical species estimated from the six source profiles are not shown in the manuscript? (Figure 5).

Reply: We have redrawn Figure 5 to include the percentage contributions in the revised version of the manuscript.

6. Figure 1 and Figure 3 are likely wrong due to the samplers with cut points of 0.43-9.0 μm .

Reply: Thank you for your comments. We have redrawn Figure 1 and Figure 3 in the revised version of the manuscript.

7. Discussions in section 4.4 seemed to be out of the scope of this manuscript.

Reply: The main aim of Sect. 4.4 was to investigate the effects of chemical species, especially in the different size fractions, and meteorological factors on visibility. This section may be useful for reconstructing the relationships between visibility and particulate matter source (Sect. 4.3.3). In the future, we will do more discussion regarding this topic. Thus, this section may help improve our understanding of haze formation mechanisms and help policy-makers design emission control strategies to reduce the effects of haze.

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