

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 (AQI) 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:

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

Line20: “SO₄²⁻, NO₃⁻ and NH₄⁺” 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.

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

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

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

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

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

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

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.

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.

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

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

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

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.

Line 143: Authors need to clearly illustrate why they use PMF model in their search.

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

Line 172: What is "TSP" ?

Line 172: Authors need to clearly define "PM₉" and "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 PM₁₀

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

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

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

Line 242: Please give more background information on why the emissions were complex in Beijing during the winter.

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.

Line 264: What are the precursors of SOC?

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?

Line 282: Change “markedly” to “significantly”

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

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.

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”

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.

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

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

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.

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.

Line364: Change “thereby” to “therefore”

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?

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

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

Line 386: Change “contributions of OM to PM2.1 were” to the “contribution of OM to PM2.1 was”

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.

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.

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

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

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

Line 513: Change "result" to "results"

Line 518: Change "to" to "in order to"

Line 539 to 540: NW, SE and NE need definition.

Line 545: Change "strong effect" to "strong impacts"

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

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.

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

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

Reference:

Zhang, Q., Quan, J., Tie, X., Li, X., Liu, Q., Gao, Y., Zhao, D., 2015. Effects of meteorology and secondary particle formation on visibility during heavy haze events in Beijing, China. *Science of The Total Environment* 502, 578-584.

Zhang, R., Jing, J., Tao, J., Hsu, S.-C., Wang, G., Cao, J., Lee, C., Zhu, L., Chen, Z., Zhao, Y., 2013. Chemical characterization and source apportionment of PM 2.5 in Beijing: seasonal perspective. *Atmospheric Chemistry and Physics* 13, 7053-7074.