

Dear reviewer,

The authors are thankful for the reviewer's comments, suggestions, and time. We have prepared a point-by-point response highlighted in blue. We hope our efforts in revising the manuscript can improve it for the selection of ACP.

Anonymous Referee #3

This work did a nice job in measuring the chemical composition and mixing states of aerosols at an urban and a rural site in Beijing. More than 4 million particles were detected at each site, of which the chemically analyzed particles were grouped and analyzed to investigate the potential sources and atmospheric processing. The authors found that the urban particles were influenced significantly by rural processing and transport. The paper is generally well-written, and I recommend it for publication after some addressing the following comments.

Comments: 1. The authors listed several studies in paragraph 2 in Introduction, but didn't contain any conclusion. What does it mean by "discrepancies remain among these studies"? Authors need to be more specific about what discrepancies exist among the studies. Please revise/rephrase and be more specific.

Ans: We have added the following passage to illustrate the discrepancies and gaps in knowledge (lines 53–62):

“For example, the mass loading of PM<sub>2.5</sub> can rapidly increase to hundreds  $\mu\text{g m}^{-3}$ . Both Wang et al. (2016) and Cheng et al. (2016) suggested the secondary formation of sulfate from the

oxidation of NO<sub>2</sub>, while Guo et al. (2014) proposed a mechanism of particle formation and growth. Different from local secondary formation and accumulation, Li et al. (2015) proposed that particles via long-range transport cause the elevation of PM<sub>2.5</sub>. According to Sun et al. (2014) and Zhai et al. (2016), regional transport played an important role during heavy haze episodes. However, most studies have focused on the urban areas of Beijing, with limited attention paid to rural areas. To illustrate the sources, evolution, and transport of particles, the investigation of rural areas around Beijing is necessary.”

2. It should be noted that chemical bias of SPAMS might introduce uncertainties in representativeness of ambient particles and even in classification of chemically analyzed particles. I think there is a need to mention this caveat in your paper, and caution the readers that uncertainties may be expected for the results. This can be provided either in Introduction or Discussions.

Ans: We completely agree with the reviewer that the limitations of the instrumentation should be addressed (lines70–72).

“Due to the nature of laser desorption/ionization (LDI), the instrument is very sensitive to dust and other types of particles containing sodium and potassium, and this may cause bias in the particle matrix (Pratt and Prather, 2012).”

3. Please clarify the differences between this study and previous studies. The significance of this study is not well written in the paper.

Ans: Associated with what we mentioned in Lines 76–83, we have added the following paragraph:

“Organics, sulfate, nitrate, ammonium and other species have been found internally mixed in the atmospheric particles, and these particle types are mostly from the combustion of fuel or biomass. The abundance of secondary species can indicate the degree of aging during atmospheric processing. Particles are more secondary species with deeper processing. However, these studies lack the use of this data to provide a view of the dynamic particulate processing. Therefore, we used the relative abundance of secondary species to adequately illustrate the process of single particles at both sites, providing a tracing system on a regional scale.”

4. L150: The EC category has four types including EC-Nitrate (EC-Nit), EC-Sulfate (EC-Sul), and EC-Nit-Sul. Is it “four”, not “three”? Or there is another type?

Ans: Sorry, that was is a typo. We have changed it to “three.”

5. According to line 243, K-rich is one of branches of k-rich category. According to Figure 6a, K-rich means BB (BB in figure 6 caption, and K-rich in the figure) According to line 366, BB-related particle means K-rich category. The use of K-rich category, K-rich type, BB and BB-related particle is confusing. Please make the description more clear, specific and concrete.

Ans: Thank you very much for this suggestion. The K-rich category is from BB-related particles, and we will stick to this term. We have changed our description in the text, as well as the caption of Figure 6a.

6. As you mentioned in Line 253, the household BB is prohibited in urban Beijing,

which is inconsistent with figure 6c indicating that the highest number counts of K-rich were observed when wind speed was less than 2 m s<sup>-1</sup>, is that possible K-rich\_PKU can also from other sources?

Ans: This is a very interesting question.

The urban sampling site is around the 4<sup>th</sup> ring expressway of Beijing, 10 km from the 5<sup>th</sup> ring expressway. In these areas, there are still villages in which biofuels are used. Therefore, these K-rich\_PKU were also from biomass burning.

7. L295: “mass spectra of NaK category contained f Na<sup>+</sup>, K<sup>+</sup> . . .” Please correct.

Ans: We have made the correction (line 319).

## References

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