The authors performed simultaneous observations of size-resolved single-particle chemical composition and mixing state in both urban and rural areas of Beijing. The campaigns collected more than 4 million particles being at both sites. The authors have investigated the different sources, processing, and origination of atmospheric particles at both sites. Generally, the manuscript illustrates a substantial contribution to the scientific understanding of urban particulate pollution in China. Particularly, the clustering strategy used in this work can be very useful to illustrate different stages of atmospheric processing. The manuscript is concisely organized and well written. Therefore, the reviewer prefers publishing if the following concerns are addressed.

Major comments

1. The statement in the Introduction should be more clarified between Lines 47–53. The discrepancies should be described, and a conclusion or hypothesis should have been drawn from the literature review.

Ans: The following passage has been added to the text (lines 53–62):

"For example, the mass loading of $PM_{2.5}$ can rapidly increase to hundreds μg m$^{-3}$. Both Wang et al. (2016) and Cheng et al. (2016) suggested the secondary formation of sulfate from the oxidation of $NO_2$, 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 $PM_{2.5}$. According to Sun et al. (2014) and Zhai et al. (2016), regional transport plays 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. Also, could you be specific on this "limited attention to the atmospheric particulate processing?" It is very important because it provides the novelty of this work compared to previous studies.

Ans: We completely agree with the reviewer. The following statement has been added to the text to enhance the argument (lines 76−83):

"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 with 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."

3. Lines 101–112. The use of relative peak area to determine the aging of particles is interesting. As we know, matrix effect could alter the ion intensities of each ion. The authors should address the possible influential factor for bias.

Ans: We have added this part to the text (lines 128–131):

"Indeed, the matrix effect can affect ionic intensities between different particles during single-particle mass spectrometer analysis. However, the effect can be reduced by using the average mass spectra of particles within a similar size distribution and chemical composition."

4. Table 1. The reviewer strongly suggests a column of comment on the source of particle types at both sites, respectively.

Ans: We have added a comment on the sources of particles.

5. Section 3.2.5. Is it possible to use the polar plots at both sites to locate the potential source of Fe-rich particles?

Ans: Yes, according to the polar plots at both sites, the Fe-rich particles originated from the south of both sites which is the direction of Hebei Province, as we described in the text.

6. Please compare the -Sul particles at both sites because the sulfate-rich particles can be formed from the oxidation of SO2. Any difference in the origin of sulfate-rich particles at different sites? Please add additional analysis or comments to Discussion.

Ans: A detailed interpretation of sulfate-rich particles has been added in lines 433–446:

"$SO_2$ was strictly controlled in Beijing. However, the emission of $SO_2$ is still significant

in the nearby Hebei and Shandong provinces (Shi et al. 2019). The different control measures produced a low concentration area of $SO_2$ around Beijing. Sulfate-rich particle types such as EC-Sul, OC-Sul, K-Sul, and NaK-Sul usually arrived at the PKU site when the wind speed was high (> 3m $s^{-1}$). The wind directions, along with the transport of sulfate-rich particles, were east, southwest and south. In these directions, sulfate was either primarily emitted from coal burning for residential heating, power generation and industry, or secondary uptake on pre-existing particles (Zhang et al., 2015). Likewise, a portion of sulfate-rich particle arrived at the PG site when the wind speed was high. However, locally formed sulfate was also pronounced, especially for ECOC-Sul, K-Sul, and NaK-Sul. As discussed in Section 3, ECOC-Sul and NaK-Sul were mainly from coal burning for residential heating, and K-Sul was formed due to the uptake of secondary sulfate. Conclusively, the particulate characterization in rural areas around Beijing is significantly influenced by residential coal burning."

7. Section 3.4. according to the claim in the manuscript, both OC-PG and ECOC-PG are supposed to be local. However, the polar plot suggests that these particle types are from multiple directions, please explain.

Ans: Yes, OC_PG was mainly from the east, south, and west, and ECOC_PG came from the northeast, southeast, and east. They certainly came from multiple directions. However, the highest concentrations of these particles were at the centers of the polar plots, indicating that emissions of OC_PG and ECOC_PG were high in the Beijing region. Therefore, the following statement has been added to line 381:

"Also, the emissions of OC_PG and ECOC_PG are high in the region."

Minor and technical

Lines 385: "detailed" should be "details."

Ans: We have changed this part (line 412).

Line 421. "small" should be "limited."

Ans: We have changed this part (line 421).

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