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Interactive comment on "Simultaneous Measurement of Urban and Rural Particles in Beijing, Part II: Case Studies of Haze Events and Regional Transport" *by* Yang Chen et al.

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Dear reviewer,

We are grateful for your valuable comments that have helped us to improve our manuscript. We went through your comments and prepared a point-to-point response below. Correspondingly, the changes are also highlighted in the manuscript. All our responses are marked in blue or with "Ans". Again, we appreciate your time and comments.

Online single-particle chemical composition analysis was used as a tracer system to

investigate the impact of heating activities and the formation of haze events in two parallel field studies at both urban and rural sites in Beijing. This manuscript focuses on case studies. One of the key points of this manuscript is that there is a pattern of transportation and accumulation of particles in both the urban and rural areas. The input of regional particles was a consequence of weakening atmospheric circulations, resulting in the stagnation of the air which provided favorable conditions for the accumulation of pollutants, ultimately leading to severe haze events. In the rural area, the heavy haze was mainly controlled by air stagnation and local emissions, but regional transport was also observed before the event.

This work represents a potentially substantial contribution to understanding the heavy haze formation in Beijing. However, I do have several concerns mostly related to this point. I will support the publication of this manuscript if the authors can properly address my following comments. The hypothesis of regional transport can trigger a high pollution event is interesting. The evidence provided in this manuscript, however, can only suggest that there is some possibility of this happening at best.

The evolution of particle compositions and fractions were not consistent or repeatable in the high PM events. The transition of particle fraction was not there for most of the cases. At least they are not obvious from the time series. The higher wind speed was used as another evidence for regional transport, which is far from robust. The authors did not discuss other possible causes, such as boundary layer height. The argument of why higher PM causes stagnation of atmosphere is also lacking. I wonder if the authors can provide some reasoning from the meteorological perspective. In order to support the current conclusion in the manuscript, the authors need to better illustrate those points with more than time series and address the reproducibility in all the high PM events. Otherwise, the authors can tighten their language and only provide this as one of the probable theories.

Ans:

We are very grateful for your detailed comments. This study is based on two field measurements, and we separated our presentation into two parts. Part I illustrates the particle types, single-particle chemical composition, mixing state, and sources (Chen et al., 2020); and Part II discusses pollution events and interactions between PG and PKU.

In Part I, we investigate mass spectra of major particle types. Particle types were more aged with higher relative abundances of secondary species. Hence, we determined the aged particles from the two datasets from both PKU and PG. Considering the particles can undergo aging locally or regionally, we also studied the evolution of these particles under variable meteorological conditions. The locally aged particles arrived at sampling sites with no unique wind directions, at low wind speed (commonly < 2 m sâĂŠ1); on the other hand, regionally aged particles responded to unique wind directions. Hence, at each site, particle types can be categorized as "local" or "regional" depending on their chemical composition, hourly number counts, and wind-direction responses. The meteorological perspective on pollution events has been fully described in Part I.

In Part II, the time series of aged particles along with high wind speed has been used as indicators of regional transport. It is regrettable that we do not emphasize the time series were from the aged particles. A particle transportation event is defined if the concentrations of aged particles rose at high wind speed. For the better presentation of this part, we have added the necessary descriptions of particle evolution, transport, and meteorology-dependent properties (lines 125-144).

"We observed five particle categories at both sites: elemental carbon (EC), organic carbon (OC), internal-mixed EC and OC (ECOC), potassium-rich (K-rich), and metals. According to their different stages of atmospheric processing, the five categories can be divided into up to 20 particle types, as shown in Table 1. Particles with relative peak areas of sulfate and nitrate greater than 0.1 were marked with nitrate (-Nit) or sulfate (-Sul), respectively, or both (-Nit-Sul). Particle types were more aged with higher relative abundances of secondary species. Besides, the suffixes "_PKU" and "_PG" are used

when the same particles appear. The typical single-particle mass spectra of all particle types are available in Supportive Information and (Chen et al., 2020).

After resolving the sources, the origins of particle types were ascertained at both sites. At PKU, the following particle types were local: EC-Nit, EC-Nit-Sul, ECOC-Nit-Sul, Ca-rich, and ECOC-Nit. These particles arrived at PKU with no unique wind directions, at low wind speed (commonly < 2 m sâĂŠ1) and with clear diurnal patterns. On the contrary, OC-Nit, OC-Sul, NaK-Nit, and NaK-Nit-Sul responded to unique wind directions, implying that these particle types were regionally transported. At PG, all particle types showed patterns that were both local and regional. For example, OC, ECOC, OC-Nit-Sul, and ECOC-Nit-Sul came from the local area, northeast, and southwest. Universal patterns can be used to determine the mechanisms of pollution event formation when combined with unique cases."

We also discussed the shift of PBL and provided other evidence to support our hypothesis (lines 252-263). "In the most recent study of aerosol-radiation feedback deterioration in Beijing during wintertime, Wu et al. (2019) proposed that the increase of near-surface PM2.5 from 10 to 200 μg mâĂŠ3 can result in a decrease of the planetary boundary layer (PBL) from 1,500 m to 400 m, consequently contributing to PM2.5 concentration by 20%. However, a 20% difference cannot explain that PM2.5 concentration increased from 100 μg mâĂŠ3 to 300 μg mâĂŠ3. Moreover, when PM2.5 exceeded 200 μg mâĂŠ3, the height of the PBL remained at 400âĂŠ500 m, and air stagnation occurred with weak horizontal wind and inactive advection. Zhong et al. (2017) observed that weak temperature inversion occurred almost at the same period, and near-surface RH increased after southerly transport, along with decreased vertical wind speed and increased RH during winter. Air stagnation was also observed in this study (Figure 2). Therefore, based on the evidence of chemical evolution, the southerly transport of PM strongly connected to pollution events. "

Transported aerosols weaken near-surface radiation, causing the shifting of the PBL with temperature inversion and increasing RH. The consequences are favorable for ac-

cumulation and secondary formation, which were both observed in this study. Therefore, the claim that the regional transport of pollutants is highly linked to pollution events is feasible.

Minor Comments:

1. Line 110: Adding the two sites on this map would be helpful.

Ans: A map of the sampling site has been added in line 101.

2. Line 138: Another Table 1? And it is unclear to me what these correlations are.

Ans: We are sorry, it was a typo, and we have fixed it (line 167).

3. Line 273-274: “These results are consistent with the analysis of particle categories.” Please expand and support this argument.

Ans: Yes, the argument is incomplete. We have added the following evidence to support our statement (lines 324âĂŠ327): “As shown in Figure 3, when transport occurred on November 4th, 19th, and 26th, regional particle types such as K-Nit-Sul, Nak-Nit-Sul, ECOC-Nit-Sul, and OC-Nit-Sul increased due to transport from the east (Part I). ”

Editorial Comments: It seems that the manuscript has many typos and I am only listing the ones I caught. Please proofread intensively before considering resubmission.

Ans:

Thank you for the reminder. A full proofread has been conducted by a native English-speaking scientist.

1. Line 22-33: I strongly recommend NOT to use abbreviations for particles types like EC-Nit, EC-Nit-Sul, ECOC-Nit-Sul, Nak-Nit, and OC-SulECOC-Nit in the abstract. PKU and PG have not been introduced either. Please fix. Ans: We have added the necessary information on those terms (Lines 96–99).

2. Line 56-57: “Sun et al. (2014); Sun et al.(2013a)” Format of citation needs correction. Ans: The citation format has been updated to “Sun et al. (2013) and (2014)”.

3. Line 113: “Particle types, their ratios at both sites,” Do you mean fractions instead of ratios? Ans: Yes, we mean their number fractions in the dataset. We have changed all terms to “fractions.”

4. Line 175: “control emissions from household emissions” fix typo. Ans: This has been changed to (lines 205-206): “Conclusively, the control of emissions from household heating is also key to improving the air quality in Beijing.”

5. Line 292: “such provinces as Hebei, Henan,” Check grammar Ans:

This has been changed to (lines 344): “. . .air pollutants in Hebei, Henan, and Shandong provinces are transported to Beijing (Shi et al., 2019; Du et al., 2019).”

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Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2019-1118/acp-2019-1118-AC1-supplement.pdf

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