



(5) In Fig.6, the RBC ranges for clean and pollution periods are different, it is better to compare the variations on the same scale?

**Authors' reply:** In Figs.6c and 6d, the RBC ranges are set on the same scale.

(6) One general suggestion is that this dataset is unique as it measures only BC-particles in a highly polluted environment, the reviewer feels the discussion needs more comparisons with results in other locations or environments. As mentioned by the authors, such measurements were conducted in other sites in US, and Europe, even it is very scarce in China. This can help to show what is special or different or important of the findings observed in Beijing, and what are the implications of such findings to the atmospheric chemistry.

**Authors' reply:** Thanks for the suggestion. We in fact have included some comparisons with previous results including in Los Angeles, London, etc. during our discussion. Per the request, we have made some necessary minor changes in revising the manuscript. Please see the modified version for details.

### **Response to Reviewer #2**

BC also called as soot is an important aerosol from incomplete combustion of fossil fuels and biomass burning. Understanding the soot mixing state in polluted air of Beijing, it is quite important issue to evaluate their potential optical, hygroscopic, and human health. The authors used one SP-AMS to determine mixing state of soot particles collected in Beijing during the wintertime. They found coating/BC ratio was at 5.0, much smaller than highly aged soot in other places. Also, they studied coating chemical species and their possible formation mechanism. The scope of this study is suitable for ACP. However, the paper need to one substantial revision before it can be published. I list several concerns about the conclusions.

**Authors' reply:** We thank the reviewer for his valuable comments, and our point-to-point replies to the reviewer's comments are listed below.

(1) L28-29 deleted very (2) L30 only

**Authors' reply: Done**

(3) L33-34 how do the result indicate dominant contributions from primary emissions? You can say that these particles might source from local emissions instead of long-range transport particles. Am I right?

**Authors' reply:** We agree that this may not be appropriate, and we deleted this sentence.

(4) L35- 36, 38-40, seemly for me, the conclusion is contrast. One you mentioned primary emission. Other one you want to mention the secondary species.

**Authors' reply:** We agree with that the description is not very specific. In line with comment (3), we have modified the description. "Positive matrix factorization shows presence of significant primary fossil fuel and biomass burning organics."

(5) L41 at-during

**Authors' reply: done**

(6) L44-45, I don't think the conclusion is from your solid result. Most you speculate these results. (7) L44-47 the conclusion cover all the possible. I would ask the author revise it carefully. What is your conclusions during the sampling period. If these solid conclusions are not from this study, you need to remove it. Seemly, I like to see what you find on BC particles not for haze formation.

**Authors' reply:** The conclusions are based on our results, but are indeed only for BC-particles and two specific cases during the sampling period. We agree that over-interpretation should be avoided. Therefore, we have carefully revised this sentence. "However, for individual pollution events, sometimes primary species could also play a dominant role, as revealed by the compositions of BC-particles in two polluted episodes during the sampling period."

(8) L56 Morphology of BC might be altered greatly. These citations don't supply any morphology of BC particles. You need find others from electron microscopies.

**Authors' reply:** We have cited a couple of electron microscopic studies, including: Wang, Y., Liu, F., He, C., Bi, L., Cheng, T., Wang, Z., Zhang, H., Zhang, X., Shi, Z., and Li, W.: Fractal Dimensions and Mixing Structures of Soot Particles during Atmospheric Processing, *Environmental Science & Technology Letters*, 4, 487-493, 10.1021/acs.estlett.7b00418, 2017.

Li, W., Sun, J., Xu, L., Shi, Z., Riemer, N., Sun, Y., Fu, P., Zhang, J., Lin, Y., Wang, X., Shao, L., Chen, J., Zhang, X., Wang, Z., and Wang, W.: A conceptual framework for mixing structures in individual aerosol particles, *J. Geophys. Res. - Atmos.*, 121, 13,784-713,798, 10.1002/2016JD025252, 2016.

(9) L77, I don't agree with the claim. For example, Wu et al., 2017. Size distribution and source of black carbon aerosol in urban Beijing during winter haze episodes. *Atmos. Chem. Phys.* 17 (12), 7965-7975. The study seemly, give the online BC-containing particles in Beijing.

**Authors' reply:** Sorry, our claim is not clear, we meant to say that no chemical characterization of BC-containing particles only. It is now changed. And in fact, the work mentioned here was already cited as a previous work on BC-particles in Beijing.

(10) L161 Discussion, deleted s

**Authors' reply: done**

(11) L260-261, L284-285, L331-332 L347-348, all the parts discussed the aqueous reactions for nitrate and SOA formation during the nighttime. I take a look at the data from the study. It is too simply to get such conclusion. I might ask the authors cite more related references here. For example, Wu et al., 2018. *Environmental Science & Technology Letters* 5 (3), 160-166; Sun et al., 2018. *Journal of Geophysical Research: Atmospheres* 123 (2), 1234-1243. Kuang et al., 2016. *Geophysical Research Letters* 43 (16), 8744-8750.

**Authors' reply:** Thanks for the references provided. The aqueous-phase production of secondary species was a possible pathway in a qualitative manner. The references provided, which are also conducted in NCP, in fact strongly support our postulated aqueous-phase pathway therefore are very useful. They have now been cited along with our discussion in the main text. “Similarly, nitrate and sulfate formations driven by high RH in North China Plain have been proved previously (Kuang et al., 2016; Sun et al., 2018; Wu et al., 2018).”

(12) L278-279 I don't understand the sentence. Why was the large decrease of organics coating concentration?

**Authors' reply:** Since  $R_{BC}$  is the ratio of concentrations of total coating material to BC cores. There was a drop of  $R_{BC}$  at 4:00pm, however, since nitrate/BC, sulfate/BC and chloride/BC did not decrease around 4pm (there was even an increase of nitrate/BC), the decrease of  $R_{BC}$  must be caused by the decrease of organic/BC. And Fig.4f further shows that it is in fact the portions of BBOA and FFOA decreased since the OOA1/BC and OOA2/BC in fact increased. We have revised the sentence in the text. “In fact, the 4:00pm  $R_{BC}$  drop was mainly caused by the large decrease of Org/BC (as  $SO_4^{2-}/BC$ ,  $NO_3^-/BC$  and  $Cl^-/BC$  did not decrease at 4:00pm, Fig. 5d) - mainly the portions of fossil fuel and biomass burning OA (Fig. 4f). ”

(13) L292 at-during

**Authors' reply:** done

(14) L307 This can be expected for urban aerosols. I don't understand it. Why?

**Authors' reply:** We have expanded this sentence. “As BC-containing particle in urban Beijing were likely influenced by multiple local/regional primary sources, relative amount of secondarily formed coating species would be less than those of highly aged BC, therefore a lower  $R_{BC}$  is expected.”

(15) L328 of-at (16) L269 miss comma after ws (17) L317 at two polluted episodes

**Authors' reply:** done

(16) For section 3.5.2 Seemly, the authors found different coating species on soot particles. FE, the author found large SOA; SE the author proposed large POA instead of SOA. Do the authors answer how POA associated with BC? If these particles were emitted from sources, these mixing should occur in all the time, not just SE. Were there different sources in SE and FE? Seemly, the author didn't supply any wind and backtrajectories here. I would ask the authors carefully check the data. Make sure the differences in FE and SE are large. Here the authors only compared the organics. What about the sulfate and nitrate are in the coating of BC there. I am certainly struggling on the part.

**Authors' reply:** As requested, we have added the back trajectories, wind rose plots, as well as the vertical distributions of wind speeds/directions during these two episodes. It shows clearly that these episodes are very different, and therefore they would have

