Direct observations of organic aerosols in common wintertime hazes in North China: insights into direct emissions from Chinese residential stoves

Chen et al.,

We appreciated the reviewers' comments which significantly improve quality of the manuscript. We carefully answer them one by one as below.

1. Selection of sampling sites needs to be introduced in more details, especially the importance and significance of the three sites. I understand that the Jinan city is regarded as the representative of uncontrolled coal combustion site, but in fact the urban area of this megacity might not rely on coal for domestic energy anymore and instead, the petrol and gas might be its major energy sources. This manuscript is designed to explain domestic coal combustion in the vast area of countryside, but no this type of sampling sites were selected.

   Answer: We added more information about sampling sites. We admit that air pollutants in Jinan city represented complicated sources such as industrial, vehicular, and residential emissions. If the reviewer noticed the finding of the whole paper, you can find aerosol particles collected in weak photochemical environment in winter. In other words, most of aerosols were primary particles instead of less secondary particles transformed from SO_2, NOx, and VOCs. Although the industrial and vehicular emission could be dominant, they mostly emitted trace gases (e.g., SO_2, NOx, and VOCs). Among these major sources in Jinan city, coal combustion in residential stoves only emitted large amounts of primary particles, in particular, they emitted abundant primary organic particles.

2. "1 Introduction" part: the introduction needs to be succinct and should be more concentrated on the organic aerosol particles and related hazes. The current text in the introduction part is too complicated and not well focused on the main aims.

   Answer: We revised introduction. Please notice that we deleted large part in second paragraph.

3. The classification of the six types of organic particles needs to be careful. For example, the type 4 particle (domelike) looks more like a mixture of possible organic and other materials such as ammonia or nitrates, and the type 5 particle (dispersed) may be the results of the shrinkage of organic-coated particle.

   Answer: In this study, we used EDX to examine chemical composition of individual particles. If the type 4 particle contain ammonia or nitrates (except organic nitrate), these particles should not be stable under the electron beam. We found that these particles were stable in the TEM. The type 5 particle is other important particle types which is different from organic-coated particles. The particle types have been well described in our latest paper in JGR-Atmos. which is under review.

   About the type 4 particle, we collect similar particles from coal combustion in stoves in laboratory. Based on our observations in previous studies and laboratory experiments, we can surely that the classification is no problem.

4. Line 30: "(Tai, S2)" might be "(Mt. Tai, S2),"

   Answer: We revised this to Mt. Tai

5. Line 33: I suggest to change “OM-coating” into “coating OM” for the type 6 particle.
   Answer: To make consistent with our previous study, we want keep the OM-coating particles. We used the particle name in one new paper under review in JGR.

6. Line 71: “the various air pollution levels” may be changed into “the various air quality levels”
   Answer: We changed the “pollution” to “quality”.

7. Line 74: the definition of “Haze as a weather phenomenon is defined by visibility ≤ 10 km and RH ≤ 95%” requires references.
   Answer: We revised this sentence and added reference for this definition. Please see Line 75.

8. Line 117 and also throughout the whole manuscript, acronyms and abbreviations must be explained at first occurrence. For example, the first appeared “BrC” should have a full word phrase.
   Answer: We added the full name of BrC: “brown carbon (BrC)”.

9. Line 129-133: the methods mentioned here are repetitive of the “2 Experimental Methods” part.
   Answer: We deleted the repetitive sentence and revised this part. Please see Line 120.

10. “2.1 Sampling sites and particle collection”: In Line 145, the authors mentioned that “aerosol particles collected at S1 mainly reflect local, ground-based urban and industrial emissions”. This means the S1 site can’t represent the potential uncontrolled coal combustion source?
    Answer: Please see our first reply (1).

11. Line 151: “During the winter monsoon season, S3 is the downwind of the Jing-Jin-Ji area … and Shandong province.” This looks not correct. From the map, S3 is located in the east of the JJJ area, and how can we regard it is the downwind of S1 and S2? Furthermore, S3 is not located upwind area, it may not appropriate to serve as a background (clean) site.
    Answer: Maybe the reviewer misunderstood our meaning. We change words to make more clearly here. We didn't address that S3 site is downwind S1 and S2. Here, three different sampling sites represent different environmental functions. In this study, S3 is treated as a polluted background site instead of the normal background site.

12. “2.3 NanoSIMS analysis” part: It is good to see that the NanoSIMS gives the ions $12C^-$ and $12C14N^-$ which could represent the organic matter in individual particles. However, to the study of this manuscript, how many of the individual particles were analyzed? Were all particles measured by NanoSIMS?
    Answer: The NanoSIMS is very complex and expensive instrument. It is not necessary to examine all the organic particles. The NanoSIMS only confirmed the organic findings from TEM. The routine procedures for this study will be shown as below.

    Firstly, we used the TEM to observe organic particles.

    Secondly, we selected some typical samples which contain large amounts of organic

particles for NanoSIMS experiments.

13. Line 198: “with 20, 25, and 13 individual particles analyzed by this method for each of the three sampling sites.” Which sites exactly these numbers correspond to?
    Answer: We added sites here. Please see Line 199.

14. Line 229-230: Soot may also be the C-dominated particles?
    Answer: We agreed and changed “C-dominated” to “OM-like” particles.

15. Line 254: Please check if it is “…ratio of width and height…” or ““…ratio of height and width…”.
    Answer: We revised this sentence to “ratio of length and width…” in Line 267.

16. Line 275-276: The category of “soot, mineral, metal, fly ash, and sulfate particles” is not same as that of the line 229-230?
    Answer: We corrected them and added the metal in Line 230.

17. Line 278: The OM-fly ash might be the overlapped particles during sampling and not necessarily the mixed particle in the air?
    Answer: The sampling duration was controlled to avoid overlap among different particles on the substrate. If the reviewer carefully looks at Figure 5c, you will find the connection between OM and fly ash which could not form during sampling process.

18. Line 314, “For example, Moffet et al. (2013) suggested : : :: : :Based on these comparisons, we conclude that those type 1-3 OM particles were not emitted by vehicular emissions in the NCP”. However, all these data for comparisons were from North America and Japan, which I don’t think can exclude the type 1-3 OM particles present from vehicular emissions in urban areas of Chinese Cities.
    Answer: Thank you very much. We added more explanation here including our previous studies in urban air and remote mountain air. Please see Line 351.

19. Line 325, the authors didn’t analyze the emissions from heavy industries or coal fired power plants, so I don’t think they can obtain the conclusion that “the type 1-3 OM particles were not emitted from heavy industries or coal-fired power plants” and that “they were from coal combustion or biomass burning for household heating and cooking in wintertime”. More evidence needs to be provided.
    Answer: Same to the above reply. We added more explanation here.

20. Line 413: Please check if the 1-3 OM occupy 70% of aerosol particles or 70% of the organic particles?
    Answer: We revised this to “70% OM-containing aerosol particles”. Please see Line 453.

21. “5. Conclusions and atmospheric implications” needs to be simplified, and what are major conclusions?

Answer: We revised and shortened the “conclusions and atmospheric implications” section.

22. “Acknowledgments”: There are some repetitive words between line 424 and line 425.
    Answer: We removed the extra words here.

23. Table 1: The decimal number should keep consistent.
    Answer: We unified the data format in Tables.

24. English of the text needs to be polished by a native English speaker.
    Answer: We invited a native English speaker (Dr. Peter Hyde) to polish the English writing.