

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. Although the observations of individual particles probed by TEM can provide certain information to relate to the sources, the conclusions regarding the exact sources should be carefully evaluated. It could be expected that the sources during such haze events are complicate. To exclude the other sources and limit it to residential stove emission, additional information is needed to support such conclusion. For example, as discussed in the manuscript, there are two particle types, OM-fly ash and OM sulfate(K)-fly ash as shown in Figure 5, what are the possible sources of these fly ash containing particles, what are the composition of these fly ash? Is it possible that these are from coal-fired plants or industrial emissions? In Line 142, as indicated by the authors that S1 can be influenced by the industrial emissions. The statements regarding the sources in Abstract and Conclusion section should be reworded if no further supporting evidences is provided to constrain the sources.

Answer: We really appreciated your comments. Spherical fly ash typically contains Si and O which indicates coal-fired power plant. In the revised manuscript, we largely revised section 4.1 (Sources of OM-containing particles) and added more references. We also slightly revised abstract and conclusion.

2. The Introduction and Conclusion sections should be revised. In the Introduction section, there is a long discussion between the severe haze and L&M haze events and their differences, but later there is no comparison or further discussion in the main text. This part should be shortened. It may be beneficial to reader or to the context to focus more on single particle analysis or source characterization. The Conclusion and Atmospheric Implication section can be more concise and focus more on the findings from these observations. For example, the discussion of BrC and the implications, for such discussion should be limited to a certain extend unless data are provided showing the OM particles are BrC.

Answer: We revised and shortened the discussion between the severe haze and L&M haze events. We deleted most parts suggested by the reviewer.

3. The classification of the particle types is not very clear and straightforward in the current form. It is suggested to add a figure or table to describe how the particles are grouped.

Answer: The classification of particle types were based on their chemical composition and morphology in the TEM/EDX, and then calculated their shapes in the computer algorithm. We added Figure 2 to describe the flow chart of particle classifications.

4. Some definitions are not consistent. For example, the "OM-coating (type 6)" in L33, "C-dominated" in L230, "OM-containing particles" in L237, "OM coating" in L269, these terms are confusing and not consistent throughout the text.

Answer: We revised the name of "C-dominated" to "OM-like". TEM could not exactly determine OM particles, so called OM-like particles before we showed the NanoSIMS result in the context.

Because OM particles were internally mixed particles, we used OM-containing particles to represent all the OM-related particles. OM-coating represents one OM mixing structure in the internally mixed particles. The individual aerosol particles classifications were shown in Figure 2.

5. In Figure 3 (e), the type 5 dispersed-OM is very similar to the particle in Figure 5 (d) which was bleached by the beam, why it is classified as OM particle which seems to have S and K?

Answer: We revised Figure 2 to show the classification here. Figure 5d shows the OM-mixed containing S and K belong to the classifying rules in this study.

6. In Figure 3, for the type 6, what is the chemical composition of cores?

Answer: The cores in type 6 are secondary inorganic components such as sulfate and nitrate. Please see the Figure 5f, the core is sulfate.

7. Should type 6 belong to OM internally mixed particles as showing in Figure 5?

Answer: Yes, It belongs to OM internally mixed particles as shown in Figure 5. We revised the Figure 2 to show how we classified the particles.

8. L31, use “morphologies”?

Answer: We revised this word.

9. L37, what does “cooling, polluted plumes” mean?

Answer: We revised this sentence to: “formed in cooling process after polluted plumes emitted from...”. Please see Line 38.

10. L38, what kind of detector is used for EDX analysis? Please justify the use of “Si-O-C ratio” to estimate to contribution of coal combustion.

Answer: The EDX is from oxford instruments. TEM/EDX only can obtain semi-quantitative data for elements. Therefore, we could not make any significant conclusion from the each element in individual particles. However, it is significant to make comparisons of Si-O-C in many OM particles detected under the same TEM/EDX, which can avoid some impacts from the substrate or instrument. The same method has been used in Li et al., JGR, 2012 and Posfai et al., JGR, 2004 in the reference list.

11. L44, “aerosols” means “particles and gases”

Answer: We revised “aerosols” to “particles”.

12. L46, use “to” instead of “into”?

Answer: We revised this word.

13. L58, the sentence should be revised.

Answer: We revised this sentence. Please see Line 59.

14. L61, “Poschl” should be “Pöschl”

Answer: We revised this name.

15. L70-71, It is suggested to use “Ministry of Environmental Protection of People’s Republic of China”. I guess the Ministry of environmental protection is not monitoring the PM2.5 by itself. Please revise the statement.
Answer: We revised this to “Ministry of Environmental Protection of People’s Republic of China”.
16. L73, please use the right document citation, what is “HJ 663-2012”?
Answer: We revised this citation.
17. L75-76, use “associated with different levels of PM2.5 concentrations and RH”?
Answer: We revised this sentence to “haze levels normally are associated with different levels of PM2.5 concentrations and RH” in Line 77.
18. L93, delete “been”
Answer: We revised this sentence to “physicochemical properties...have been well understood”. Please see Line 83.
19. L94 delete “their”
Answer: Please see the answer for comment 18. We revised this sentence.
20. L98 delete “different”
Answer: We deleted this “different” in line 90.
21. section 2.1, more details should be provided regarding the sampling procedure and sample sites. What was the sample height at each site, what about the sampling time and duration?
Answer: Thanks, we provide it in supplement as Table S2.
22. L155, use “represent” instead of “display”
Answer: We revised “display” to “represent”.
23. L163, the impactors are used to collect particles not aerosols
Answer: We changed “aerosols” to “particles” here.
24. L168, one sample for each day was analyzed? What is the sampling duration for each grid?
Answer: Yes, We added the information about analyzed samples in Table S2.
25. L174, use “acquired”?
Answer: We revised the word to “acquired”.
26. L183, what is brand and model for NanoSIMS?
Answer: We added the brand and model “NanoSIMS 50L, CAMECA Instruments, Geneviers, France”. Please see Line 185.

27. L218-221, it is not clear what the authors try to discuss. Please revise.
Answer: Here, we deleted descriptions about back trajectories in this manuscript.
28. L253, please revise how the reference is cited.
Answer: We revised it in line 262.
29. L301-302, The sentence is not clear.
Answer: We revised this sentence like this: "This result suggests that the type 1-3 OM sources were similar in the same haze layer over the NCP". Please see Line 333.
30. L318, there is no sufficient evidence to support this conclusion.
Answer: We added more explanation here including our previous studies in urban air and remote mountain air. Please see Line 351.
31. L326-331, this section should be carefully revised as discussed in the General Comments.
Answer: We revised this section and added other references for this conclusion.
32. L333, what is the EDX detector; detector background may contribute to Si signal?
Answer: No, the background from EDX detector has been calibrated to remove the possible influence. As we used EDX to detect the background film, we could not detect the Si signal. It should not be worried about that.
33. L339, as it is shown in Figure 8, the ratios are not following the lines, the data points are sort of deviating from the lines. Please discuss in more details.
Answer: Thank you for your comments. As you know, we obtained the data from internally mixed particles. Elements from OM in the EDX data should not be like pure OM or influenced by other aerosol components mixed in individual particles. Therefore, it should have one range like in Figure 8.
34. L363-365, This is not very clear. Considering the scattering of the data points, is it significantly different among these three cases? It would be more straightforward if coating thickness is calculated and compared among these sites with statistical test.
Answer: As our result, it is not significantly different among these three sites. We added average coating thickness in Figure 9.
35. L368, the statement is not convincing if only consider the number fractions of type 6 OM particles.
Answer: Yes, we revised this sentence. Please see Line 414.
36. L393, "in other words"?
Answer: We deleted this sentence here.
37. Figure 1, are the two black lines for each site indicating the range of the backward trajectories?
Answer: As other reviewer requests, we deleted backward trajectories in the Figure 1.

38. Figure 6, it is not easy to distinguish the green colors for the OM-soot and OM-soot sulfate.

Answer: We revised the color in Figure 6.

39. Figure 9, “OM coating”, do you mean “OM-coating particles”?

Answer: Yes, We revised this. Please see the description for this Figure in Line 419.