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. Comments Line 22: Qualify this statement with "North China" after "haze episodes" because this statement is generally not true- many studies have focused on many different levels of hazes.
   Answer: We added "North China" after "haze episodes".

2. Line 22 : "freqent" is a typo.
   Answer: We revised this word to "frequent".

3. Line 72: Define "PM$_{2.5}$" at first usage.
   Answer: We added the definition of PM$_{2.5}$ (aerodynamic equivalent diameter $\leq$ 2.5 μm). Please see Line 71.

4. Line 80: Add "in China" after "episodes"
   Answer: We added them.

5. Line 99: How is a haze day defined with respect to time? How long do high concentrations or poor visibility have to last to be considered an episode? How different are the timescales for moderate versus heavy haze days?
   Answer: We added one statistic data for an example to show occurrence of haze episodes and the timescales (Figure S2).
   We statistically analyzed frequency of haze episodes in winter for nine cities. If one heavy haze episode persists more three days, the government will have the highest alert (red). Figure S2 shows that only two severe haze days occurred in wintertime. However, the light and moderate haze episodes are common and persistent longer (Figure S2).

6. Line 117: Define "BrC" at first usage.
   Answer: We added the full name of BrC. Please see Line 108.

7. Line 122: "inidividual" is a typo
   Answer: We revised this word.

8. Line 131: Were the same TEM grids used for all three analyses?
   Answer: 33 TEM grids were analyzed for TEM/EDX analysis. Three typical samples (one grid for each site) were chosen for AFM and NanoSIMS analysis because of the consistency of samples.

9. Line 142: Please provide elevations of S1 and S3.
   Answer: We added the elevations of S1 and S3.

10. Line 152: Remove "the" from between "is" and "downwind"

11. Line 157: Please provide more detail regarding the choice of 9-11.5 hour sampling period. Was this sampling repeated continuously? Or was it repeated daily only at the same time each day?

    Answer: We added the details about sampling periods (daytime: 7:30-19:00 and nighttime: 19:30-7:00 (next day)) and revised this sentence. Please see Line 156.

12. Line 164: Did the TEM grid sampling occur on the same sampling schedule as the bulk sampling?

    Answer: Yes. It should be noticed that different samples have different sampling duration. Individual particle samples must be collected in a short time. We added more information to explain it. Please see Table S2 which includes the details of samples.

13. Line 168: How were the 11 aerosol samples chosen? What time periods did the samples correspond to?

    Answer: We added the time periods and other information for samples at three sites in Table S2. The selected samples as much as possible represent the whole hazes.

14. Line 170: What was the order of the analysis for the three methods? How was destruction to particles from electron beams or vacuum minimized in the order of the analysis?

    Answer: The order of analysis is TEM, AFM, and NanoSIMS. Some particles (e.g., sulfate and nitrate) can be destroyed under the electron beams in TEM, but particles in other areas of the same sample still keep well. AFM doesn't destroy the samples. Finally, we used NanoSIMS to analyze the same samples. Because the TEM grids must install on the special plate in NanoSIMS, we cannot take them back anymore. We used the special TEM grids with letters which can help us to find locations. The method is the best way to integrate three different analyzed instruments for the same samples.

15. Line 202: Define EVD and ECD at first use.

    Answer: We added the definitions of EVD and ECD and added their formulas in supplementary material.

16. Line 211: What time periods to the MODIS images correspond to?

    Answer: We added the date. These two MODIS images were got on December 14 and 19, respectively.

17. Line 213-214: It is not clear what time periods averages correspond to? All periods above 75 μ g/m3?

    Answer: Yes, all the haze periods were above 75 μ g/m^3 here. Please see Figure S4.

18. Line 226: Point out that although the concentrations increased between haze and clear days, the fraction of PM$_{2.5}$ that is organic did not change that much. It appeared that the fraction of organics and inorganics remained fairly stable regardless of higher haze events.

    Answer: We revised this section and we added that "the fraction of OC to PM$_{2.5}$ remained fairly stable regardless of L&M haze and clear days". Please see Line 223.

19. Line 232: Was nanoSIMS performed on all TEM grids so that the carbon content of the particles could be confirmed this way? Obtaining carbon contribution from TEM grids using TEM/EDX is obviously very uncertain given the interference from the grids.

Answer: No, we could not do all the TEM grids. We just chose typical OM particles to confirm their chemical ions. We admitted TEM/EDX obtained uncertain carbon contribution, but it doesn't influence our classification based on all the elemental compositions of individual particles. The method is quite normal for individual aerosol analysis in TEM and SEM (e.g., Li et al., JGR, 2012; Moffet et al., ACP, 2010)

20. Line 237: The interference of the grid makes determining OM content of particles from TEM qualitative at best. How is this avoided with this analysis?

Answer: We used morphology and EDX data both to identify OM particles, and then we can account their number faction.

21. Line 240: Were the OM particle morphology characterized subjectively? Meaning, did a single user determine the type of each particle based on visual inspection, or was this somehow determined by a computer algorithm?

Answer: We made such a classification firstly based on visual inspection and then made their shape by a computer algorithm. I think the potential user can to identify the OM particles as this way.

22. Line 242: Can the authors provide some additional description of the "domelike" particles? What does this "domelike" structure imply?

Answer: Here we only define it based on their morphology. We suspect the domelike particles are organic gels. As we have known that organic gel is a type of material which is translucent. Indeed, we found similar particles from biomass burning and coal used in residential stoves in the laboratory experiments.

23. Line 243: Did any of these OM-type particles behave differently under the beam or vacuum?

Answer: In the TEM, these analyzed OM particles behaved stable. Obviously, they were non-volatile OM.

24. Line 245: For which site?

Answer: We made the three sampling sites together for analysis.

25. Line 261: It would be clearer if the equation for AR was moved up into the "Aspect Ratio" section.

Answer: We moved up the equation into the AR section.

26. Line 286: This information would fit better in the previous paragraph.

Answer: Yes, we moved it to previous paragraph and revised this sentence. Please see Line 301.

27. Line 283-292: According to line 166, the D50 for this sampler is 0.25 µ m. Was a collection

efficiency applied to the data to account for this? If not the size information should be considered qualitative at best (especially since the maximum bin is 4.5 μm). Some mention of this should be made in this section. Are the bin widths greater than the uncertainty in the size data? To assist with the interpretation of Figure 6, the data from all sites should have the same size bins and scale on the figures.

Answer: We didn't consider the sampling efficiency. We know that the sampler should have higher loss efficiency, so we used size bins to make possible comparisons. Otherwise, the particle number cannot be direct compared. We revised the size bins in figure 6.

28. Line 294: Parts (a) and (b) would be more easily compared if they had the same scale. Part (a) has a log-scale and (b) does not, so the size distributions are difficult to compare. Also, what is the significance of the OM-containing particle diagrams within part (a)? Does part (a) include all OM-containing particle types (1-6) while part (b) only include the subset 1-3? Which haze event does figure 7 correspond to? What does the bimodal peak around 0.8 um correspond to in part (b)?

Answer: Figure 7a represents the size distribution of individual particles in all L&M haze episodes during sampling period in NCP. Please noticed we covert the particle number N into dN/dlogDp in the y-axis, so the x-axis can be considered as log mode. Figure 7b only represents size of type 1-3 OM in one haze event. The y-axis is real number faction so x-axis should use the normal size. Figure 7b correspond to one haze event on December 14-15 (we added it in the Figure caption).

We checked all the data and found the peak is too low to give more strong information for particle sources.

29. Line 314: Change "coating" to "coated"
Answer: We deleted this sentence.

30. Line 316: Where in North America?
Answer: We deleted this description here.

31. Line 318-9: I am not sure how this conclusion follows from the previous comparisons?
Answer: We added some references (Li and Shao, 2010) and (Li et al., 2015) in this part. They found only a few type 1-3 OM particles in urban and remote mountain air in China. Based on the comparison, we conclude that the type 1-3 OM particles were not directly emitted by vehicular emissions in the NCP.

32. Line 327-328: State how OM 1-3 particles from coal-combustion from power plants and residential heating/cooking would differ that leads to this conclusion. What additional evidence?
Answer: In our previous studies, we studied aerosol particles associated with power plants, they didn't emit spherical OM. We revised the section and added other references for this conclusion.

33. Line 336: I have several questions/concerns from the data presentation and analysis in this paragraph. First, including and comparing C and O from the TEM/EDX analysis here is concerning given the interference from the grids. I am not sure that carbon data are very

meaningful in this context. I see the description of how Si-O-C line for haze determined from the supplemental, but some mention should also be included in the paper. Haze can correspond to very different particle composition and would not likely have a single Si-O-C ratio. Is corn combustion representative of biomass burning in the region during this time of year? Again, I am not convinced these data are meaningful given the C and O interference.

Answer: TEM/EDX only can obtain semi-quantitive 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.

We added some description in the paper about the Si-O-C line for haze.

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, the data points could not perfectly along with the lines. It should have one range like in Figure 8.

In the NCP, farmers harvest their corn in autumn and storage these corn stalks to burn in wintertime.

34. Line 345: The sphericity of OM 1-3 particles does not necessarily suggest that these emissions are from coal combustion. Many other studies have reported on spherical OM particles that originated from biomass burning. Shape alone does not necessarily correspond to emission type.

Answer: We agreed your comments. We revised the discussion about type 1-3 OM sources in the revised manuscript and deleted the description here between their shapes and sources. Please see Line 370.

35. Line 347: I suggest restating that the vehicular emissions at S1 led to higher contributions of soot particles because no mention of vehicular emissions at S1 has been made up to now. Instead, one might infer that the high contribution of soot particles at S1 could likely be from vehicular emissions in an urban area.

Answer: We deleted the descriptions about the sources of soot and fly ash particles here and focus on the OM particles in the revised manuscript.

36. Figure 6 shows fly-ash as part of two different types (OM-sulfate metal/fly ash and OM-fly ash) and how is that reconciled with the contributions shown in Figures S7?

Answer: Fly ash is a tracer of coal-fired power plant and heavy industrial, so it is an important kind of particle. Figure 2 shows only 19% of OM-sulfate particles were mixed with fly ash/metal. In Figure 2, we made OM-fly ash particles into a class and OM-sulfate mixed with fly ash into OM-sulfate group.

37. If the presence of fly ash is the evidence used for large stationary sources, than this designation should be made earlier (see comment for line 327).

Answer: Yes, we deleted the discussion about sources of fly ash and soot particles, but focus on the main sources of OM particles.

38. Line 356: The back trajectories for all sites look similar during haze events, so I am not convinced that aging can be determined separately for sites based on Figure 1.

Answer: We deleted the back trajectories because they only represent the air masses above 1500 m. We added more description about possible sources or location of the sampling sites.

39. Line 365: Define "coarse"

Answer: We revised this sentence. Please see Line 410-411.

40. Line 376: How would a secondary organic particle appear in the TEM analysis? One might argue that the mixed OM-sulfate or the coated particles are secondary in nature. I also disagree with the statement that not many inorganic aerosols were observed given that Table S1 states the inorganic fraction of $PM_{2.5}$ was actually higher than $TC/PM_{2.5}$ at all sites.

Answer: Thank you for your comments. Please notice that many OM-mixed particles more or less contain secondary inorganic species, which can be reflected in the classified names. The OM-coating particles cannot represent all the OM-sulfate particles. From the TEM observation, the OM-coating particle is much less in the samples collected in L&M hazes in winter than our previous study in summer or severe hazes in winter.

41. Line 381: Many hygroscopicity studies have demonstrated water associated with particles at RH values less than 60%.

Answer: Maybe it is true in some locations. We used the RH value for haze in North China from the reference (Zheng et al., 2015).

42. Line 386: This statement seems inconsistent with line 377 that states that SOA are common in heavy haze but only 31% in winter hazes. What type of hazes?

Answer: They did two researches in different haze levels and we added the haze type. Please see Line 438.

43. Line 394: Recall from Figure 1 that back trajectories suggest different transport on haze days. Are the authors trying to state that cooking and heating only from other regions are influencing the hazes? Can the authors reconcile and clarify this argument?

Answer: Your suggestion is very good, and we revised the Figure 1.

We did back trajectories at 1500 m for all sites in Figure 1 which could not represent ground pollutants' transportations.

44. Line 402: Some comment here on the bulk OC and EC data and comparisons to the single particle results would be useful. Is the relative abundance of soot particles on haze days consistent with higher EC measurements? Is this also true for OM-containing particles?

Answer: As the other reviewer's comments, we deleted the paragraph. The comparison can be done but we didn't focus on the EC in this study. Also, we want to mention that the particle classification cannot reflect the all the aerosol components. In the aged particles, many soot particles were internally mixed with OM and sulfate.

45. Line 410: The influence of direct emissions on haze stability has not been established in this

paper.

Answer: Thanks, we deleted this part.

46. Line 413: What does "70% aerosol particles" mean?

    Answer: We revised this sentence to "OM-containing aerosol particles". Please see Line 453.

47. Line 417: Transport must be taken into account when making the statements regarding differences in moderate and heavy hazes, as well as meteorological controls such as boundary layer depth, wind speed, etc. Heavier hazes could be associated with stagnant conditions when pollution builds up, but emissions could be the same.

    Answer: We agreed with your comments. The different haze levels must associate with meteorological data. Here we only focused on OM particle morphology.

48. Figure 1: What dates do the MODIS images correspond to?

    Answer: We added the specific date of the MODIS images.

49. Figure 6: As mentioned in the comments, the figure would be more easily compared if the bin widths and figure scales were the same.

    Answer: We revised this Figure's size bins.

50. Figure 7. Provide the significance of the OM-containing particle diagram within part (a); it can be interpreted a few different ways. Convert the x-scale in part (b) to log and use the same scale as part (a), same as with the y-axis.

    Answer: Thank you for your advice. These two figures have different purpose. Figure 7a shows size distribution of the analyzed particle number. Please notice we also convert the y-axis using dN/dLogDp, so the log mode can make clear size distribution in the limited particle number. In contrast, Figure 7b only show number fraction with particle size. If we use log mode, the peaks will not clear anymore. Therefore, we do not change the x-scale in Figure 8b

51. Figure 9: Does size here refer to EVD? How was sulfate core size measured?

    Answer: In Figure 9, particle size refers to EVD. We directly measure sulfate core size using the iTEM software (details in sections 2.2 and 2.4), and convert into EVD.

52. Table 1: It is not necessary to report so many digits for the average sphericity and AR values; only significant digits are necessary.

    Answer: We revised Table 1.