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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Direct observations of organic aerosols in common wintertime hazes in North China: insights into their size, shape, mixing state, and source" *by* S. R. Chen et al.

Anonymous Referee #3

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Review of "Direct observations of organic aerosols in common wintertime hazes in North China: insights into their size, shape, mixing state, and source" by Chen et al. This is a well-written paper that presents single particle analyses of samples obtained during moderate haze events in North China during winter of 2014 at an urban, island, and mountain site. The analyses focused on the composition, size, morphology, and mixing state of organic particles (OM). Single particle analysis is critical to understanding the properties of OM in the atmosphere and for assessing modeling efforts to estimate their contribution to visibility degradation and climate change. This paper furthers our knowledge of OM morphology and is a useful contribution to the literature regarding OM properties, especially during moderate hazes. I recommend publication

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after major revisions that address the concerns and comments listed below.

A main concern is the use of carbon data obtained from EDX on TEM grids to comment on source emissions (coal, haze, corn).The contribution of carbon and oxygen from the TEM grids makes the C and O EDX data semiquantitative at best, yet the authors are using them to determine source profiles and to derive conclusions without a mention of the interference other than line 232. Also, the main motivation for the paper is the contribution of these particles to haze, yet no mention of optical properties of these particles was included. Can the authors extend their results/discussion to the impact of their results on our knowledge of OM optical properties? They have data on particle shape, for example, and could comment on the assumptions used to model optical properties of OM particles (typically assumed to be spherical). In addition, comparisons to bulk data would be helpful to understand whether OC/EC ratios and inorganic/OM fractions are reflected in the particle type/morphology. Finally, several arguments and conclusions provided in the paper would benefit from clarification and additional evidence; details are in the comments below.

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.

Line 22 : “freqent” is a typo.

Line 72: Define “PM2.5” at first usage.

Line 80: Add “in China” after “episodes”

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?

Line 117: Define “BrC” at first usage.

Line 122: “inidividual” is a typo.

Line 131: Were the same TEM grids used for all three analyses?

Line 142: Please provide elevations of S1 and S3.

Line 152: Remove “the” from between “is” and “downwind”

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?

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

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

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?

Line 202: Define EVD and ECD at first use.

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

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

Line 226: Point out that although the concentrations increased between haze and clear days, the fraction of PM2.5 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.

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.

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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?

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?

Line 242: Can the authors provide some additional description of the “domelike” particles? What does this “domelike” structure imply?

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

Line 245: For which site?

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

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

Line 283-292: According to line 166, the D50 for this sampler is 0.25 μm. 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.

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)?

Line 314: Change “coating” to “coated”

Line 316: Where in North America?

Line 318-9: I am not sure how this conclusion follows from the previous comparisons?

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?

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.

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.

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. 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? 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).

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.

Line 365: Define “coarse”

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 PM2.5 was actually higher than TC/PM2.5 at all sites.

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

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?

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?

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?

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

Line 413: What does “70% aerosol particles” mean?

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.

Figures/Tables: Figure 1: What dates do the MODIS images correspond to?

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

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.

Figure 9: Does size here refer to EVD? How was sulfate core size measured? Table 1: It is not necessary to report so many digits for the average sphericity and AR values; only significant digits are necessary.

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