

Interactive comment on “Secondary aerosol formation promotes water uptake by organic-rich wildfire haze particles in Equatorial Asia” by Jing Chen et al.

Anonymous Referee #2

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This manuscript describes an aerosol dataset collected in Singapore during a period in October, 2015 when the region was impacted by high concentrations of smoke particles. Though I have some concerns with the analysis, I support eventual publication in ACP in part because the region and this particle source are understudied. But major revisions would be required first. In addition to the concerns identified below, the writing would need to be improved prior to publication. The mistakes are too numerous to identify in this review.

A general concern I have with the manuscript is that it lacks a description of the meteorological and chemical setting that readers would need to understand the data. There

C1

is a brief mention of the location of the sampling site relative to a road and a petrochemical complex, but no discussion about whether and when those sources would be upwind. There is also no discussion of the typical transport time of the smoke prior to arrival at the sampling site. And to put the smoke-impacted measurements into context, there needs to be some data and discussion of the typical concentration and composition at the site.

Line 20 (and later): There is no discussion about the disconnect between the single size at which g was measured (100 nm) and the larger particles that dominate the composition measured by the ACSM. There is a mention that the number size distribution is dominated by particles in the 50 – 200 nm size range, but the number distribution is irrelevant. It seems likely that the diurnal variations in g would not be as important for particles near the mass median size of the distribution. So then the composition should be comparatively constant and the variation between it and g introduces scatter to the inferred hygroscopicity parameters. It also seems likely that much of the sulfate will be in >100 nm particles if it forms in cloud. So then it is questionable how much direct impact it has on g .

I am unfamiliar with the characteristics of PBOA. Is it reliably separated from BBOA? Could variation in fuel type or burning characteristics cause shifts in attribution between the two types? And what is known about the solubility of peat burning primary particles? I ask because it could influence the fraction of soluble species in solution at the 85% RH in the HTDMA. These points should be added to the manuscript and not simply provided in a response.

Page 8, line 13: The OPS does not measure aerodynamic size.

Page 9, Line 3: The only g distribution provided is the study average shown in Figure 1. The separation of particles into the three g categories implies that the distributions were generally multimodal. I don't doubt that, but it should be shown. Perhaps a few example distributions could be included in a supplemental document.

C2

Page 9, line 14: I think “More” would be a better word here. Highly to me implies much higher g than 1.27. Especially when those with $g < 1.15$ are “Nearly non-hygroscopic”; that’s a large change in type over a small change in g .

Page 10, line 9: Those are not salts

Page 10, line 12: Is sea salt expected to be important at the site? Unless operated at a high temperature the ACSM would not see it.

Page 10, line 21: The second sentence in this paragraph needs to be rewritten.

Page 11, line 15: I can’t understand why the authors chose to assume that both BBOA and PBOA have k appas of 0.0. Even if they are low, why not use best estimates of the values instead of just arbitrarily setting them to 0. Because they are not actually completely non-hygroscopic, the result will be an erroneous sensitivity of k appa_OOA to the organic type fractions. This needs to be changed in the revision.

Page 12, line 21: I agree that most of wildfire haze mass will be submicron, but that can’t be asserted based on the number concentration as the authors do. At a minimum the supermicron volume fraction should be used for this conclusion.

Page 12, lines 25 and 26: $dN/d\log D_p$ is not the same as number concentration.

Page 13, line 1: The authors note that traffic emissions may be important but take no steps to correct for the impact in their analysis. Should periods when winds are from the road and when traffic is heavy be excluded? The novelty of this dataset is obviously the wildfire haze and anything local is an interference.

Page 13, line 22: Add non-refractory in front of each instance of chloride.

Page 14, line 2: Related to the above comment about the assumption that only the OOA contributed to hygroscopicity, the fraction of the organic mass that was water soluble was quite high (average 64%, max 80%). How frequently was this higher than the fraction of organics categorized as OOA?

C3

Page 14, line 7: Is it surprising that there is much less K than SO₄?

Page 14, line 26: Somewhere in the paper there needs to be a conceptual explanation of what is responsible for the observed mix of HOA, PBOA, BBOA, and OOA. Do the authors believe that closer to the source measurements would show mostly PBOA and BBOA and that aging is responsible for the conversion to OOA? (the transport timescale issue mentioned above is relevant here). Is the daily variation thought to be mostly due to photochemical processing or could it also be daily variation in fire characteristics? And is it thought that the changes are due to processing of the existing particles or production of SOA from gas phase emissions from the fires? The one example size distribution time series presented does not seem to indicate particles are growing, but rather the size is about constant and only the concentration changes. Do the time series on other days look like that as well? And if so, what is responsible?

Page 17, line 15: NO₃ was a small contributor to the overall mass. It seems pointless to note that variations in that small contribution had little effect on k appa.

Figure 3: I would like to be able to see whether organic mass and sulfate mass are correlated. But with the use of the same y-scale for both in (a) that is not possible.

Figure 6: Specify date

Figure 9: Clarify what “averaged diurnal” means.

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C4