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Interactive comment on "Evaluating the degree of oxygenation of organic aerosol during foggy and hazy days in Hong Kong using high-resolution time-of-flight aerosol mass spectrometry (HR-ToF-AMS)" by Y. J. Li et al.

Anonymous Referee #1

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This paper presents an interesting contrast between foggy and hazy periods of various AMS variables indicating degree of OA oxidation or volatility. The results are likely to be of particular interest to the AMS community, but some of the findings are also of general interest to those investigating secondary organic aerosol formation. Although the data is interesting, in my view the author's way over interpret the data. The last line of the abstract and first line of the conclusions are strong statements despite no physical or chemical process actually being measured in this study.

The major premise of this paper is that the observations made at a specific time inform

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how the aerosol got to its observed chemical state. The authors implicitly assume that all chemical oxidation processes are so fast that this assumption is valid and that the air masses are completely uniform so that no composition variability is due to advection. For example, does particle pH or LWC at a specific time really mean these are the variables that explain the overall fine particle (i.e., bulk) chemical properties, such as SVOA, LVOA, etc? Over a single day, not even considering a particle's lifespan of say a week, it could be exposed to a wide range of pH and LWC. To illustrate, take sulfate, for example, since it's formation is much better known than OA. According to the AMS data presented in Table 1, sulfate concentrations are higher under hazy conditions. Because liquid water concentrations were lower during the hazy period (Table 1) does that mean the authors would conclude that a gas-phase oxidation route formed most sulfate observed during the hazy period? To really answer this question one may believe that some more complex analysis is needed, such as for example, a chemical transport model that accounts for the complex history that undoubtedly affects the overall sulfate mass? If one cannot definitively say the sulfate was mainly formed via gas phase oxidation during the hazy period, how can one make detailed predictions on OA partitioning pathways (even though no gases were measured), oxidation routes, etc based solely on particle composition?

More specific comments.

Some statements made in the background that are key to the analysis, which follows, are overly simplistic and should be clarified. This includes:

- 1) Pg 3538, L17-18. The statement that OOA is a surrogate of SOA is not strictly true. There are primary sources of oxygenated aerosol (e.g., organic acids, etc), both from mobile sources and biomass burning.
- 2) Pg 3538, L20-22. The link between oxygenation and hygroscopicity is not as strong as the authors imply. See, for example, Atmos. Chem. Phys., 13, 2735–2756, 2013 and references therein.

How was the quantitative accuracy of the AMS determined? How do uncertainties in key species affect predictions of pH, LWC, etc. Example, pg 3541 L17, how does an inaccurate CE not affect the analysis, be specific. pH is going to be very sensitive to SO4= and NH4+ concentrations. Was the same CE used to calculate both? Which leads to, what are the uncertainties in IS and pH given the typical uncertainties in AMS ion measurements? An uncertainty analysis is needed.

Pg 3541, L14, define what the measured to predicted NH4+ ratio is? That is, how is it predicted?

What is the uncertainty in RH when RH>95%. RH meters typically have issues with accuracy at high RH. Also, at RH greater than roughly 90% the LWC is highly sensitive to RH. At these RHs the LWC must be highly uncertain. This should be discussed and the uncertainty estimated and included in the analysis and figures.

Section 3.3 comparing LVOOA and SVOOA. It seems highly speculative to discuss partitioning mechanisms bases solely on aerosol composition and LWC data. I don't see any direct evidence to support these types of statements (pg 3548, L2 to 11), all one really can say is that the data are consistent with ... and note that it is really speculation.

Section 3.4, again I find it somewhat surprising that such definitive statements can be made on partitioning and oxidation processes from a correlation analysis, despite no gas phase measurements. Much of this just seems to be speculation.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 3533, 2013.

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