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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Mixing state of oxalic acid containing particles in the rural area of Pearl River Delta, China: implication for seasonal formation mechanism of Secondary Organic Aerosol (SOA)" *by* Chunlei Cheng et al.

Anonymous Referee #2

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The authors present a comprehensive analysis of the mixing state of oxalic acid in the Pearl River Delta area of China using a single particle aerosol mass spectrometer (SPAMS). The topic of the paper is important with regard to understanding formation pathways of secondary organic aerosol (SOA). The main findings are the following: oxalic acid containing particles accounted for <3% of total particles; in summer heavy-metals containing particles were the largest group containing oxalic acid while in winter it was the biomass burning group; the majority of oxalic acid particles were internally mixed with sulfate and nitrate; the fraction of oxalic acid particles containing ammonium increases significantly in winter versus summer. A couple of interesting speculations

are made about acid-catalyzed reactions and Fenton like reactions.

The paper is written well. The title and abstract are mostly reflective of the contents of the paper. The results are quite interesting and I support publication after each of my comments below are adequately addressed:

Specific Comments: While the title of the paper and parts of the manuscript make reference to comparison of summer and winter and also mention the word 'seasonal', it is important to consider that the authors are only looking at two short-term periods spanning ∼2 weeks. I suggest a relaxation of words in parts of the paper that make it seem as though full seasonal coverage was obtained. At the minimum, 'seasonal' needs to be revised in my view for the title.

Line 189: These percentages are quite interesting to me and they seem low based on how ubiquitous the literature suggests oxalic acid is in particles. Can the authors comment more about how these percentages compare to previous reports?

Line 189-193: While the authors refer readers to the Supplement, it would be useful here to at least provide a little more detail as to how the categorization was done assuming that not all readers will go to the Supplement. The categories are very important for the results, so some more discussion is warranted here as to how this was done.

Line 289-291: Could the lower mixing height in the winter have contributed in some way to this finding (i.e. more stagnant aerosol and perhaps more aged)?

Line 337-338: The wording here in this sentence and the general paragraph appear to be too strong in my view since the authors did not unambiguously prove that photo-Fenton reactions are even occurring. Aren't these just speculations? I suggest to use less strong language and to differentiate better between proved findings and speculations.

Line 329-359: have the authors considered all other factors that could affect the diurnal

behavior of oxalic acid particles, such as meteorological factors or different emissions types during the day. Can the authors comment on how the profiles of other species look in Figure 7 if they were plotted such as sulfate, EC, nitrate? Is rush hour traffic influential at all in any of the discrepancies between the peak of ozone and the other parameters currently shown in Figure 7? While these other factors may not be important, it is still important to mention that various other potential factors were considered.

Section 3.3: the discussion and analysis surrounding the acid-catalyzed hypothesis is too thin in my view. Were such relationships not observed in the summer, and if not, then why? Could another reasonable explanation be that that precursors of oxalic acid and the organosulfate species are co-emitted? To strengthen this conclusion, the discussion and analysis needs to be more convincing with also more discussion of other relevant words using field data to point to this mechanism of acid-catalyzed formation of SOA.

Line 107-110: This line needs a revision because it is not entirely fair. It should be noted somewhere around this section that quite a bit of work has been done with fast time resolution on aircraft to address the issue of meteorological uncertainty and temporal resolution limitations. These various studies have discussed the formation pathways leading to oxalic acid with detailed in-cloud and out-of-cloud measurements:

Wonaschuetz, A., et al. (2012). Aerosol and gas re-distribution by shallow cumulus clouds: an investigation using airborne measurements, J. Geophys. Res., 117, D17202, doi:10.1029/2012JD018089.

Sorooshian, A., et al. (2006). Oxalic acid in clear and cloudy atmospheres: Analysis of data from International Consortium for Atmospheric Research on Transport and Transformation 2004, J. Geophys. Res. 111, D23S45, doi:10.1029/2005JD006880.

Sorooshian, A., et al. (2007). Particulate organic acids and overall water-soluble aerosol composition measurements from the 2006 Gulf of Mexico Atmospheric Composition and Climate Study (GoMACCS), J. Geophys. Res., 112, D13201,

doi:10.1029/2007JD008537.

Figures: Font size needs to increase in many of the figures for labels.

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