

Interactive comment on “A technique for the measurement of organic aerosol hygroscopicity, oxidation level, and volatility distributions” by Kerrigan P. Cain and Spyros N. Pandis

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(1) In this submission, the author combines several established aerosol instruments to measure hygroscopicity, volatility, and oxygen to carbon ratio simultaneously. A new inversion technique to handle the output from the instruments is used. This technique assigns hygroscopicity, and oxygen to carbon ratio to each volatility bin in $\log_{10}(C^)$ space. This requires several assumptions, and no support for the legitimacy of the assumptions are made, however, to do so would be difficult and likely beyond the scope of the submission. There appears to be enough information to reproduce the experiment as well as the inversion routine. The conclusion is original and adds to the discussion*

C1

of oxygen-to-carbon ratio and hygroscopicity as a function of volatility bin. Several studies in this area contradict one another and sometimes theory. The paper's conclusion offers a possibility for previous results to be complimentary, but would require a shift in the proposed theory. The author submits the results of 4 experimental conditions, and the paper uses a single experiment as an example. The results of other experiments appear to generally corroborate the author's conclusions and are located in the supplemental material. All experiments are used to test the noise and limitations of the proposed analysis method. Several comments are listed below. In addition to those comments, a rewrite of the abstract should also be entertained as the first sentences are long and confusing. The main body of the document is clear and mostly precise. No large rewrites are deemed necessary, and I have no major objections.

We appreciate the positive assessment of our work by the referee. We have rewritten the abstract shortening the initial sentences. Detailed responses to the comments can be found below.

(2) Page 1 line 28: Acute not proper for all 3 cites. Miller is a long term study, Dockery is for acute aerosol episodes (not acute mortality), but Brooks seems appropriate.

We have removed the two citations following the suggestion of the referee.

(3) Page 2 line 12: Soften statement. Authors studying cancer causing aerosols may disagree with the “three of the most important properties of organic aerosol.”

We rewrote this sentence to state that these properties are important for the OA lifetime.

(4) Page 2 line 14-15: Hygroscopicity is the measure of a volume of water associated with a unit volume of solute. This is a bit more precise than “ability to absorb water.” Hygroscopicity is not a measure of a particles ability to form cloud droplets.

C2

We have revised this sentence to better define hygroscopicity.

(5) *Page 3 line 7-8: Massoli et al doesn't seem to make those conclusions. The Massoli paper cites Good et al 2010 and Petters et al 2009 which do make the proposed explanations.*

We have added these two citations and removed the Massoli et al. citation to better reflect the proposed explanations.

(6) *Page 3 line 18-19: Insert comma before "and."*

Done.

(7) *Page 3 line 26-29: These two sentences can be combined to reduce repetitive nature.*

We have combined the two sentences.

(8) *Page 5 line 14: Please specify which diffusivity. Mass diffusivity describes the movement of the mass, while thermal diffusivity describes the movement of energy.*

The CCNC exploits water's higher mass diffusivity than heat's thermal diffusivity in air. We have revised the manuscript to make this clear.

(9) *Page 5 line 15: Replace "its" with a proper noun to avoid confusion.*

Done.

(10) *Page 5 line 15: replace "a" with "an"*

Done.

C3

(11) *Page 6 line 10: Confused by the word "adding." Does the author mean that we look up the activation diameter on the log(Sc)-log(Dd) plot? A few more words may be necessary.*

We have revised this sentence describing how we estimated κ from the activation diameters. We plotted the activation diameters at their measured supersaturations on the log(Sc)-log(Dd) plot and then κ 's were estimated by which line of constant κ on this plot fit the activation diameters.

(12) *Page 6 line 14: "higher sensitivity" may not be necessary. If used, should specify that the author desired a higher sensitivity to signal and not mass-to-charge.*

We have removed "higher sensitivity" to avoid confusion.

(13) *Page 8 line 5-7: The thermogram (Figure 2a) displays the evaporation of Ammonium Sulfate below 150 C, but page 7 line 28 says Ammonium Sulfate is involatile below 150 C. In my experience, the thermogram is correct.*

The evaporation of ammonium sulfate particles in a TD expressed as mass fraction remaining depends not only on the TD temperature but also on particle size and residence time of the aerosol in the TD. We have changed the temperature in the manuscript to "above 100° C" to cover all these cases and to be consistent with Figure 2.

(14) *Page 8 line 10-12: Which theory? Kohler theory or previous observations. Appears Ammonium Sulfate is still stable at 100C, but disassociates at higher temperatures. Is the hygroscopicity of disassociated Ammonium Sulfate the same as pure Ammonium Sulfate? What about when including impurities? Are these in the "theory?"*

The activation diameters through the BP and the TD at 50 and 100°C are compared to Köhler theory assuming pure ammonium sulfate aerosol. We have added "Köhler"

C4

before theory to make this clearer. Disassociated ammonium sulfate will have the same hygroscopicity because the ammonium and sulfate ions will interact with water similarly. When aerosols have impurities the overall κ will be higher or lower depending on the κ 's of the impurities due to the mixing rule.

(15) *Page 9 line 23-25: Some may contend that 0.39 is oxidized, especially when compared to alpha-pinene. A comparison may be better.*

We have rephrased this sentence. We now compare the O:C ratio through the BP to the O:C ratio through the TD at 125°C in order to show that the least volatile material is less oxidized than the overall SOA.

(16) *Page 11 line 18-19. In order to use equation 4, you must also assume density is constant between all volatility bins.*

We now state this assumption in the corresponding discussion.

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