

Interactive comment on “Hygroscopic growth of water soluble organic carbon isolated from atmospheric aerosol collected at U.S. national parks and Storm Peak Laboratory” by Nathan F. Taylor et al.

Anonymous Referee #3

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The study reports hygroscopic growth factors, kappa and CCN activity of water-soluble aerosol components in the form of re-aerosolized liquid samples from filters originally collected in National Park locations in the US. The methods, in particular the separation of WSOC from WSM, and the separate analysis of WSOC hygroscopicity, are novel and very interesting to the community. The results show that WSOC has large effects on the hygroscopic behaviors of mixed organic/inorganic aerosols. The paper is well-written and the topic is very relevant. The following points should be addressed before publication:

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General comments:

- 1) The complex experimental setup and measurement program deserve a more detailed description, in particular concerning uncertainties and the exact measurement program. What were the ranges of uncertainty and stability of the various RH settings and flow rates? How were the TDMA data inverted and growth factors determined? What were the uncertainties in the determined activation diameters? The section on TDMA and SMPS-CCNC operation would also profit from a better links between the text and Figure 1: What were the setpoints of size and RH at which points in the setup and in time for which measurement series?
- 2) More detail should also be provided on the handling and chemical analysis of the filter samples. How many filters, and which days were combined for each re-aerosolized sample? How might aerosol properties have changed over the course of the days combined into each sample? Also, it appears that some of the filter samples were taken as long as ten years ago - when were they extracted, and when were the laboratory measurements done? Can it be ensured that there are no artefacts from storage due to evaporation or other processes? In the results, fractions of WSOC and OC are reported, as well as WSOC retention percentages. How was this assessed, was there an independent WSOC measurement? How (and when) was OC measured? It would also be much more reader-friendly to present the inorganic composition of the combined samples in this paper, rather than (more or less explicitly) referring to earlier studies (do they feature the same combination of filters to larger samples?).
- 3) The discussion on the enhancement of hygroscopic growth through WSOC could, and should, be more quantitative. “Enhancements” would be more obvious if measured values were compared to reference values - either of measured hygroscopic properties of the inorganic components (not just the total WSM) or of theoretical values/theoretical growth curves of inorganic salts in the Figures. See more specific comments below.
- 4) The discussion of impact of particle shape needs substantiation. It seems a stretch

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to invoke two different mechanisms of particle shape changes as an explanation for both the deliquescence and efflorescence branch of the growth curves. The collapse of particle structures upon hydration is a known effect for aerosol types such as fresh combustion agglomerates, but it is a far less obvious thought for a re-aerosolized WSOC sample. How do the authors know that the atomized aerosol is “irregularly shaped”? A discussion of the growth factor uncertainties deriving from the experimental setup and from GF determination from the DMA2 size distribution should be given. Also, why are the GF<1 not showing in the GRSM GF curve in Figure 2?

Detailed comments:

p.4, Section 1.1: Please give more details on those locations and sampling sites.

p.6, line 20: Why was this exact dry diameter chosen?

p.6, line 24: How often is “periodically”?

p.7, lines 14-16: This should be described earlier, along with more details on the measurement methods and results of the inorganic compounds.

p.7, line 23: Please specify here: which one is the winter study? It is not reader-friendly to have him/her leaf back to the study description, find the abbreviation that refers to the winter study, and then re-locate that abbreviation in Figure 2.

Figure 2: a) dark blue and dark green, as well as light blue and light green are hard to distinguish. How about a different symbol but the same color for WSM and WSOC of the same location? It would also be nice to call “GRSM I”/“GRSM II” “GRSM summer” and “GRSM winter”, to spare the reader repeated leafing back for which one is which. b) This presentation of activation diameters should be commented on in more detail in the text. Alternatively, it could be dropped.

p.8, line 19: what kind of change classifies as “minor”, what as “large”? Please expand (or add a reference) on how a “large” change in kappa indicates a phase change.

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p.8, line 30-31: In Figure 3, GRSM I starts at around 0.3 -I would not call this “near zero”.

p.9, line 8: “complementary enhancement” as compared to what? The inorganic components alone? (Below or above efflorescence?) For this, a reference hygroscopicity value (measured or calculated) of the isolated inorganic components should be given. What is currently shown in Figure 3 is just that total WSM has a higher kappa than the WSOM, which is not surprising. The paper would improve substantially if this analysis could be more quantitative.

p.9, line 6-7: These chemical compositions should be shown in a figure.

p.9, line 20: Where and how is this shown?

p.9, line 26: Where is this argument going?

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