

Review of “Hygroscopic growth of water soluble organic carbon isolated from atmospheric aerosol collected at U.S. national parks and Storm Peak Laboratory” by Taylor et al. (Manuscript Number: acp-2016-715)

General comments

This paper presents hygroscopic properties of isolated WSOC and total water-soluble matter (WSM) extracted from PM 2.5 filter samples collected at several U.S. sites. The authors measured the hygroscopic parameters of both WSOC isolated and WSM in the laboratory under different RH conditions. They discuss the relative importance of the isolated WSOC and its interaction with inorganic components in terms of particle hygroscopicity. The present work may provide insights into our understanding on aerosol hydration/dehydration process by organics interacting with inorganics. Overall, however, the manuscript lacks quantitative discussion (see comments below), which makes conclusions rather weak throughout the paper. Moreover, most of the discussion are not convincing because supporting data are not shown in most parts of the paper. The discussion too much relies on the data shown in previous works already published, particularly regarding chemical characterization of the WSM. Although the data presented seems to be valuable, there are a number of important issues that need to be worked out before I recommend its publication in ACP.

Specific comments

(1) My major concern is on the exact fraction of WSOC isolated from the WSM compared with “the exact total WSOC” that can be measured without using any XAD columns. In Table 1, although the “WSOC recovery” is shown, it is not clear how the authors calculated/estimated the numbers. It is expected that there might be WSOC components that are not retained either XAD-8 or XAD-4. What is the percentage of these components, and how do these WSOC affect the conclusions? This point should be closely linked to the major conclusion in which the author mentioned the contribution of WSOC to the hygroscopicity of WSM.

(2) How did the authors quantify WSOC? More detailed description on the detection of WSOC is needed including if any syringe filters were used to remove any particulate OC in samples, an instrument used, lower limit of detection, measurement uncertainty, etc. In addition, is the unit of WSOC $\mu\text{g}/\text{m}^3$, or $\mu\text{gC}/\text{m}^3$? If the authors discuss WSOM

in $\mu\text{g}/\text{m}^3$, then how was the mass of WSOC converted to that of WSOM?

(3) Methodology of the hygroscopicity and CCN measurements: The authors should provide more detailed explanations for the settings and calculation with regard to the GF and CCN measurements. Several parameters are not defined in the text, such as water-activity, shape factor, etc. Also, the authors should provide how they derived several important parameters (e.g. κ values), together with some estimation of uncertainty for the GF and CCN measurements. This is important to evaluate the quality of these measurements.

(4) I understand that this study is linked with the previous works in terms of the chemical characterization of aerosols using the same/similar sample sets obtained at the same observational sites. However, almost none of the chemical component in the WSM (besides WSOC) is shown in the manuscript, only referring to the prior papers. The authors should show the amount of each inorganic component (at least SNA) relative to that of WSOC and should add more discussion on that data. Otherwise the discussion is very weak regarding the interaction between WSOC and inorganics, and their contribution to the hygroscopicity.

(5) Table 1: It is not clear how the authors estimated “sulfate cleaning efficiency.”

(6) The authors have used the term “highly soluble WSOC” in several parts of the manuscript (e.g., P.10 L.20, P.11, L.7). What is the definition of this “highly soluble WSOC,” and what is the difference between this term and “isolated WSOC” which the authors measured?

(7) The authors should add more details on the site descriptions: characteristics of each site (rural, mountain, forest, relative influence of anthropogenic vs. biogenic sources, etc.) should be listed maybe in Table 1 or in an additional table. Also why were those sites selected or appropriate to study the hygroscopicity of WSOC? It is really difficult to understand the major difference among the sites for general readers.

(8) In figures 4-6, the authors

Most of readers may not be interested in the sample number, but interested in the exact difference among the samples in terms of their chemical characteristics.

(9) Section 3.6: Discussion on possible effects of aerosol shape is too speculative. Again, the authors only refer to the previous works and do not show any supporting data to convince readers of their hypothesis. For example, if the authors assume a shape factor other than 1, then how can this support the discussion here?

(10) In Table 1, "SO4" should be "SO₄²⁻" or "sulfate"