General Comments

This paper presents the results of laboratory experiments performed to characterize the hygroscopicity of water-soluble organic carbon isolated from ambient filter samples collected at several U.S. national parks. Overall, the authors show convincingly that the WSOC makes an important contribution to the hygroscopic properties of the aerosols, even if it is a relatively minor contributor to the PM mass. The findings are significant, and certainly relevant for ACP. The paper is concise, well-organized, and the writing is polished. I highly recommend it for publication after the following items are addressed:

Specific Comments

- The biggest weakness (or opportunity for improvement) is that the authors do not extend their analyses to estimate the contribution of WSOC (and inorganic salts) to water uptake as a function of RH for each of the locations/seasons. The major finding in the paper, from the conclusions: "Contrasts between the hygroscopic growth of WSOC and total WSM samples from two of the studies suggest that soluble components in ambient aerosol can interact to enhance water uptake at atmospherically relevant RH." But this is only qualitative, in line with the major discussion surrounding this point in the manuscript. It seems that the authors' current results would enable a more quantitative interpretation of the effects of the WSOC on the hygroscopicity of the ambient particles.

- Two relevant studies are not included, but which should be: The introduction could include a brief discussion of Asa-Awuku et al. (2008), which characterized the CCN activity of WSOC isolated from biomass burning samples. Additionally, Guo et al. (2015) analyzed the contribution of organics to aerosol water in the southeastern U.S. Although their methods were different, the results of Guo et al. (2015) can provide some important context and comparison for the present study, especially the samples from Great Smoky Mountains National Park.

- Should the WSOC concentrations presented throughout the paper have units of μg-C m^{-3}? If not, how have the authors converted from OC to OM?

- I question some of the WSOC concentrations presented in Table 1, especially the samples from GRSM. WSOC contributions of only 4% to the total reconstructed $PM_{2.5}$ mass appear to be unrealistically low.

- I realize that prior papers have presented data from this same study, which the authors cite; however, I think the paper would benefit from added experimental details. Specifically, many of the results compare the pre-hydrated and pre-desiccated scans. Even if this information can be found in another paper, I recommend adding detail to the Methods section to clarify the sequence of the measurements so that the distinction between these measurements is easier to interpret.

- In what ways might the unrecovered WSOC alter the conclusions? The authors have some idea of which types of compounds are likely not recovered – there should be at least a brief discussion of how these compounds could alter the results.

- Finally, in Section 3.6 – what kind of irregular shape do the authors propose? "The hypothesis relies on the desiccated shape produced by drying the atomized aerosol being different than the desiccated shape of the aerosol at low RH in the pre-hydrated measurements." It is not clear how the rate of drying would contribute to this effect? The authors do not believe that they lose WSOC mass in the experimental setup (Section 3.6, line 28). But this at least seems plausible, given recent ambient results (El-Sayed et al., 2016) and laboratory studies (see multiple papers from the De Haan and Turpin groups). Overall, the explanations put forth in this section are shaky, and need further development.

Technical Corrections

none

References

Asa-Awuku, et al.: Investigation of molar volume and surfactant characteristics of water-soluble organic compounds in biomass burning aerosol, *Atmos. Chem. Phys.*, 8, 799-812, 2008.

El-Sayed, et al.: Drying-Induced Evaporation of Secondary Organic Aerosol during Summer, *Environ. Sci. Technol.*, 50, 3626-3633, 2016.

Guo, et al., Fine-particle water and pH in the southeastern United States, *Atmos. Chem. Phys.*, 15, 5211-5228, 2015.