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Interactive comment on "Low hygroscopicity of organic material in anthropogenic aerosols under pollution episode in China" *by* Juan Hong et al.

Anonymous Referee #2

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The manuscript by Hong et al. presents aerosol hygroscopicity and chemical composition measured near a megacity in south China. Size-resolved hygroscopic growth factors (HGF) and PM1 chemical composition were measured using a Hygroscopic Tandem Mobility Analyzer (HTDMA) and an Aerosol Chemical Speciation Monitor (ACSM), respectively. Based on the closure analysis of the two independent measurements, the authors found that the organic particulate material was primarily hydrophobic during the pollution episode. More specifically, the authors derived a new empirical relation between the HGF for organic material (HGForg) and the O:C elemental ratio. The HGForg values predicted by this new relation appear to be significantly lower than that reported in previous literature. Although I find the new dataset is interesting, the key finding of hydrophobic anthropogenic organic particulate matter seems to poorly supported by the analysis, and can be misleading. One major concern is that the HGForg values derived from the closure analysis are highly uncertain, and the uncertainty is not analyzed in the manuscript. I therefore do not recommend publication in ACP.

Major issues:

1. The closure analysis compares ACSM-derived HGF for PM1 with HTDMA-measured HGF for 100 nm or 150 nm particles. What was the mode diameter (in terms of mass-diameter distribution) of the particle population of PM1? If the mode diameter of PM1 is significantly larger than 150 nm, the estimated HGForg can be significantly underestimated, because the hygroscopicity generally increases with the diameter in the accumulation mode (at least for 50 nm - 250 nm particles as shown in Liu et al. ACP, 2011). Is it possible that the estimated low HGForg is affected by such systematic biases?

2. In line 328-331, the authors mentioned that the measured HGF values for the mixed particles are less sensitive to the relationship of HGForg vs. O:C. I agree with this statement. However, this implies that the derived parameterization of HGForg = 0.3*O:C + 0.87 can be highly uncertain, as it is not well constrained by the measurements. I noticed that including the O:C-dependent HGForg only improves Rˆ2 from 0.5 to 0.51 (Fig. 10 vs. Fig. 9 d). In this case, what is the error associated with the parameterization?

3. Related to the previous comment, the proposed relationship would predict a HGForg value smaller than 1 for O:C < 0.43. This can be misleading. What does a HGForg value smaller than 1 mean? Is it just because of measurement errors?

4. The authors reported a best-fit HGForg value of 1.1. The error bar associated with this value should also be reported. Errors of both HTDMA and ACSM, along with the uncertainties associated bulk vs size-resolved closure analysis should be considered.

5. Even if the HGForg = 1.1 value is accurate, I don't think the authors can assert that "the CCN concentrations would probably be over-predicted as the organic material in

these locations appeared to be close to hydrophobic". Several studies have shown that the CCN activity can be higher than that predicted based on the HGForg at ∼90% RH. For example, alpha-pinene-derived SOA has a kappa value of ∼0.04 at 90% RH, but the CCN-derived kappa value is around 0.1 (see Pajunoja et al., 2015), which is close to the value used in climate models. The low hygroscopicity derived at subsaturation regime does not necessarily indicate a low CCN activity.

Technical issues:

1. The authors should describe how the HTDMA was calibrated and how the data inversion was performed. Did the measurements reproduce literature values for pure compounds (e.g., ammonium sulfate)? Was the RH sensor calibrated? Did the two DMAs have a shift in diameter? Was the DMA transfer function considered in data inversion?

2. The O:C ratio was estimated from the f44 measured by a quadrupole ACSM, presumably less accurate than that measured by the HR-ToF-AMS. What was the uncertainty of the ACSM-derived O:C?

3. Figure 2. Unit of the color bar is missing.

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