Anonymous Referee #1

Shuang Han and co-authors present an experimental study of organic aerosol hygroscopicity for 23 atmospherically-relevant compounds. The hygroscopicity (kappa) is derived from water uptake measurements using an HTDMA. Compounds are grouped as sugars, polyols, di- and tri-carboxylic acids, and amino acids, and are purchased. Two publicly-available chemical equilibrium models are compared with the data. The authors find that, although hygroscopicity varies with organic functional groups, oxidation level (O:C ratio), and molecular weight, these molecular properties are imperfect predictors of water uptake. The models over-predicted hygroscopicity for some, but not all, of the studied compounds. Various possible reasons are discussed. The work is technically sound, and the measurements are neither better nor worse than those of previous studies. This authors have done a good job summarizing previous findings, and they contribute measurements and analysis for new compounds.

This manuscript is a good fit for Atmospheric Chemistry and Physics. The work is laboratory-based but has relevance to modelling and to field studies. The authors have done a good job discussing this relevance.

Although this is an interesting and well-written paper, I cannot recommend publication in its present form. The authors have not adequately described the novelty of the work, and some of their findings are not substantiated by the data. The potential impact is limited because authors have not explicitly identified their contributions. I believe they can nevertheless improve the writing before publication. These and other issues are discussed below. I recommend major revisions.

Major Comments

The authors fall short of establishing/communicating the work’s novelty. Several findings have been explored in past works (functional groups, molar mass, O:C, solubility and deliquescence). The authors have done a good job finding these studies. However, authors should discuss and emphasize their own contributions.

Further comments on the abstract: As stated above, findings summarized here should emphasize the novelty of the current work. The abstract should end with implications.
17- with additional functional groups – addition to what? This is more complicated than just the addition of functional groups. Carbon number matters as well.

18 – It sounds like you mean isomers. This statement is ambiguous.

23 – “moderately”

Uncertainty estimates are needed. As it stands, scatter in the data is used to discuss morphology. Although this is a nice discussion, some error bars and acknowledgement of the limitations of the measurement would lend more credibility to these claims.

Some of the conclusions are not supported by the data. On line 184, the authors discuss the order in which the functional groups contribute to hygroscopicity. Is this statement quantitative? If so, what is the observed partial derivative of kappa with respect to each functional group? The statement seems to have little connection to the data presented in Figure 4.

Restructuring of particles was observed, and this resulted in a negative growth factor. This was shown and discussed in the main text and in Figure S3, which shows severe discontinuities in water uptake for amino acids. Some of the restructuring-sizing error could be avoided by sizing the particles wet, following Nakao et al. (2014). This should be discussed. Nakao et al. (2014), Droplet activation of wet particles: development of the Wet CCN approach, Atmos. Meas. Tech., 7, 2227–2241.

Regarding restructuring, the residence time of the HTDMA is mentioned (2.7 seconds) but the authors do not bring this into the discussion. The authors should mention how this 2.7 s residence time affects particle restructuring, and how this instrument compares to other works.

There is a severe disagreement between the model and the measurements for some compounds, but the authors do not attempt to improve the prediction of water uptake by any calculation or modification to the models.

The authors do not discuss the disagreement between UManSysProp and E-AIM. Why do these models behave differently? There is valuable information here and it should be discussed.

Minor Comments


Implementation of Kohler theory: On line 87, in the equation, \(\frac{(1 - \frac{Ke}{RH})Ke}{RH}\) could be simplified to \(\frac{Ke}{RH-1}\).

Because growth factor as a function of RH is diameter dependent, the results would be more general if the RH is divided by the Kelvin term, here expressed Ke (on line 87). This means that instead of RH you have growth factor as a function of water activity. \(aw = \frac{RH}{Ke}\).

33 – the citation does not match the statement in any way

147,147 – how do you know that the water mass fraction always increased when the growth factor “shrank or grew”? This calculation should be described here or in the
Figures: In general, the thick grey lines behind the data are distracting (all figures)

Figure 4 – Size and style of panel D should be consistent with the rest of the figure

Figure 6 – fonts are difficult to read and should be enlarged. There should be spaces between the words.

The table of contents in the supplement is very confusing. Why are figures each described twice, except for Figure S4? Also: there is no Figure S4. Please clarify this text.

Technical corrections

The references arranged either alphabetically or chronologically (forward or reverse). Do not capitalize the names of organic acids (line 114 and elsewhere).

Caption of Figure 1 – (a) and (b) are listed together but the text differentiates these. Please be more descriptive in the caption as well

Caption of Figure 1 – space between 200 and nm

31 – McFiggans capitalize F

37 – “large” not big

44 – “relies”

47 – “(E-AIM),” add comma, define UManSysProp

55 – “molecular interactions” ... “and water.”

62 – “experimental hygroscopicity data for organics”

66 – “experimentally-determined”

68, 76 – Strike “self-assembled” or replace with, e.g., “home-built”, “custom-made”, “custom engineered”, or “HTMDA built in-house.” “Self-assembled” implies that the instrument assembled itself spontaneously.

74 – “using ultrapure”

Table 1 title: “Substances”

Table 1 header row: “Supplier, purity”

75 – physicochemical –either “physico-chemical” or “physicochemical” – make consistent throughout paper

79 – under dry conditions

80 – “the detailed schematic”

81 – period after (2013)

88 – Mw is erroneously included inside the subscript of sigma

89, 90 – italicize T and R

94 – UManSysProp is defined here – should be defined above, or in both places

97 – please clarify sentence

115 – “dicarboxylic acids”

128 – period after “RH,”

130 – “relatively higher”

131, 132 – “current models have insufficient data” – break sentence into more than one sentence

134 – visible how? Detectable?

136 – “lower RH.”

138, 139 – “Continuous water uptake”

142, 143 – “Previous studies”

144 – remove period after “2018)”

147 – remove comma after “though”

147 – “shrank or grew slightly”

148 – agrees with which previous results? Both of the prior citations? Please elaborate.

148, 149 – “Actually, it cannot be defined” – this sentence is unclear, please rephrase. Remove or replace the word “actually.”
“are generally in better agreement with”
“growth was”
“literature”
please rephrase
“Note that”
“adding an”
209, 210, ... – here and elsewhere, capitalize Hoff
“moderately”
“hygroscopicity”
please clarify these sentences
229-231 – this is a run-on sentence – split the sentence and clarify
“arises”
“good” not “well”
“to our’
“This, on the other hand, indicates”
“whose constitute may be diverged” – this is unclear, rephrase
“groups”
“processes”
“A detailed description of the HTDMA implementation, “