Review on manuscript “Influence of Organic Compound Functionality on Aerosol Hygroscopicity: Dicarboxylic Acids, Alkyl-Substituents, Sugars and Amino Acids” by Marsh et al., submitted to ACP.

The authors reported the hygroscopic data of a series of dicarboxylic acids (DCAs) with subtle molecular structure changes, amino acids, and some sugars and alcohols using the Comparative Kinetics Electrodynamic Balance (CK-EDB). It allows the measurements of a mfs/RH curve within 10 seconds, with the advantage of reducing the loss of volatile organics from the particles. Experimental results have indicated potential improvements for the UNIversal quasichemical Functional group Activity Coefficients (UNIFAC) model and are compared with isotherm models in terms of kappa values. The manuscript was in general well written but some of the wordings are unnecessarily strong that make discussions somewhat confusing or even misleading.

Page 1 line 12: “The dual micro dispenser set up allows for sequential trapping of probe and sample droplets for accurate determination of droplet water activities from 0.45 to > 0.99.” This sentence is not entirely correct. The CK-EDB is based on kinetic measurements and it does have the advantage of fast measurements that reduce evaporation of volatile materials. However, the fast measurements would also potentially lead to non-equilibrium measurements, especially for some organics at low RH. The authors seem to admit a potential shortcoming of the technique on page 11, line 23, “For many compounds, measurements unimpeded by kinetic limitations have not been possible below 80 % RH, and consequently data presented below 80 % do not average to a consistent series of points”. More evidence to demonstrate that the measurements presented in this paper are equilibrium measurements would be needed.

Page 1 line 15: This significance of this sentence is not clear. New data agree better with the UNIFAC predictions than the old data, from which UNIFAC parameters were derived, do. Are the UNIFAC parameters/predictions useful or not?

Page 1 line 22: The authors should discuss the agreement between the measured hygroscopicities and UNIFAC predictions on sugars/alcohols.

Page 8, line 25: "In addition, the short timescale of the measurement ensures that evaporation of the semi-volatile components, such as these dicarboxylic acids, is avoided". Has this been verified or is this merely an assumption? Did they experimentally verify this with some other semi-volatile solutes? Can they really say that evaporation is avoided? In reality, there must be a range of vapor pressure that evaporation is “negligible” but appreciable at larger values.

Page 10, line 1: "In summary, UNIFAC predictions agree well with measurements for simple unbranched dicarboxylic acids with the exception of pimelic acid," Is there any explanation why the UNIFAC cannot predict the hygroscopicity of pimelic acid? Can the authors provide suggestions to make improve the predictions of UNIFAC?

Page 10, line 30: "The model (equation 27 in Dutcher et al. 2013) is fitted to molality experimental data with respect to water activity for finding the parameter value, which results in a significant improvement in the MFS than UNIFAC." Why does the multilayer adsorption isotherm based model from Dutcher et al. (2013) give a better prediction beyond the use of an adjustable parameter? Page 10 line 20. From Figure 8a, it seems L-threonine, rather than L-valine, deviates from the other three compounds most.

Page 11 line 6: Is there any other possible explanation for the discrepancy between measured and literature (Chan et al., 2005) data on those amino acids? Can they rule out the possibility of mass transfer effects or evaporation of solute? They show that their data are consistent with Na et al. (1995), which are compromised by evaporation of solute since they made EDB measurements in vacuum. Furthermore, Chan et al. also made measurements of these amino acids to lower RH to determine the mfs of solid of unity. The assertion that Chan et al. were wrong, which is possible, need to be accompanied by the discussions on how that would affect the mfs of the dried particles. Would the new data provided here yield unreasonable mfs of the dried particles?

Page 11 line 23: It is unclear why kinetic limitations will not affect the hygroscopicity measurements of sugars and alcohols in this study by saying “as established by the RH of the gas phase the droplet is drying in”. Elaborate please.

Page 11 line 26: Would the C4-polyol be described as “long chain”? Would sorbitol be classified as sugar?

Page 12 line 25: The authors discussed on the over-estimation of kappa parameter. However, it seems the UManSysProp model can over-estimate as well as under-estimate the kappa (Figure 12b).

The authors seem to be sending very mixed messages on the reliability of simple parameterized models/equations for predicting hygroscopicity. On one hand, they criticized the limitation of UNIFAC in predicting hygroscopicity of branched acids. On the other hand, they promoted the use of κ values and O:C and N:C ratios based on Figure 11(b) and 12 (a), which do show discrepancies between model predictions and experimental results in κ. When plotted in the form of mfs hygroscopic data, some of these differences are not much smaller than those between the measurements of the branched DCAs and the UNIFAC predictions based on parameterization of simpler acids. Furthermore, the comparison of data and model Kappa parameters are evaluated at 95% but the comparison of UNIFAC related results are in mfs as a function of RH. How would the results look like if Kappa values are evaluated at lower RH? Finally, are these predictive tools considering these very general and smooth relationships really much less computationally expensive than current group contribution methods? κ was calculated by isotherms that require an adjustable parameters. Overall, the comments made by the authors on the use of UNIFAC, Kappa/isotherm models, and more elaborated models such as AIOMFAC and UManSysProp appear not to be unbiased. Elaboration is needed.