Answers to Referee #2

The authors appreciate the critical comments that the reviewer has raised for the revised manuscript. The authors replied all the four major comments and made proper actions to the revised manuscript according to the suggestions by the reviewer.

Major comments:

The main concern in my initial review was that with the approach used by the authors, they only measure hygroscopic growth under subsaturated conditions for all compounds, which crystallize in a binary aqueous mixture. That holds for example for all dicarboxylic acids besides malonic acid. I argued that an organic atmospheric aerosol particle typically contains a multitude, thousands of compounds, which each single compound only being present in a small fraction. In such multicompound mixtures, the organics do not crystallize as have been proven by Marcolli et al. (2004). What is needed to constrain or to compare to thermodynamic models are the data in the liquid, supersaturated (often called sub-cooled) state. If you are interested in a comparison of thermodynamic models in the subsaturate state, bulk experiments yield much more accurate data than HTDMA experiments. The authors reply to these concerns that crystallization does occurs in binary mixtures, which is true, but irrelevant, if you want to constrain or compare to thermodynamic models as written above. The authors further argue that bouncing experiments with SOA particles show that particle are solid or semisolid at intermediate RH. This is correct, but these particles are not crystalline solids, but in an amorphous state. So, you cannot argue that solid means crystalline. An amorphous solid may be slow in taking up water (as your sucrose example) but will not show deliquescence. If you wait long enough, it will reach thermodynamic equilibrium even at low humidity.

Reply: We agree with the reviewer that comparison of current datasets with thermodynamic models might not be appropriate as the data in the liquid state is better to constrain these models. As the major purpose of current study is not to constrain thermodynamic models, we removed the part of comparison of our datasets with different thermodynamic models in our study, but focused more on the comparison of our data with previous data available measured by other studies. Future study measuring the hygroscopicity of compounds in the supersaturated state is seriously considered and carefully planned.

Considered the main concern raised by the reviewer, we made a new section discussing the major limitation of current instrument as well as some other bulk methods in Sect. 4. The aspect that different researchers should pay attention to when using HTDMA data was extended. Future work with combination of different methods was also suggested. The new section is listed below:

4 Instrument limitations

Our laboratory observations show some deviations with previous literature data measured using similar techniques with different residence time or a completely different method. The residence time of humidification inside the instrument were suggested to be one of the reason responsible for this deviation. We noticed that some compounds with a longer time of humidification show higher GF values, for instance, for sucrose at intermediate RHs. This is likely due to the kinetic limitations to water uptake, which is particularly important for viscous aerosols. This discrepancy further indicates the limitation of current method, which extra care should be taken in the future and viscosity information should always be taken into account when dealing with hygroscopicity data. However, for some other compounds, the techniques with longer residence time not always obtained the higher GF values. For instance, using an STXM (Scanning Transmission X-ray Microscopy) with a residence time much longer than our HTDMA, Piens et al. (2016) obtained a lower GF of fructose as compared to ours, which should not be caused by the evaporation losses due to its low volatility. Moreover, for glutamine and serine particles, no deliquescence was observed by Chan et al. (2005) with a residence time of as longer as 40 minutes, while in our study a moderate water uptake for both compounds were observed. These evidences pointed out, for a large suit of different compounds with various complexity, the influence of residence time on the observed water uptake of particles might not be conclusive. We may need to better understand the kinetics of water transport within particles and figure out which factors would introduce variability, for instance, measurement timescale, the choice of compounds and environmental conditions, before we design the measurement targeting the effect of residence time on aerosol hygroscopicity and eventually draw the conclusion.

The different phase transition observed between our study and the ones using the EDB (Chan et al., 2005), for instance, for glutamine and serine, reveals another limitation of current instrument in this study. One is that drying process might not be sufficient. The inadequate drying may not be able to crystallize the generated aerosols if their ERH values are quite low. This will interfere the generation of a correct phase state we are requiring. The other one is that the condition at higher relative humidity, e.g., >90%, is difficult to access. We notice that many organic compounds have their DRH higher than 90%RH and thus measurements at those conditions are quite scarce. Therefore, different means, including both raising the drying capacity and making the elevated RH condition achievable, are all we should consider to break these limitations and improve the system.

These deviations, on the other hand, also reveal that the measurement for size-resolved particles, particularly relevant for atmospheric conditions, is still needed. As we notice, many of these previous literature data were obtained from bulk samples or particles at micron size range. Previous studies pointed out thermodynamic data of bulk samples or particles at micron-size range may not always represent the ones at nanometre range, particularly in the size range of the atmospheric aerosol accumulation mode (e.g., ~ 10^2 nm). For instance, Cheng et al. (2015) suggested that particle size can be an important factor influencing the solid-liquid equilibrium upon phase transition. Pöschl et al. (2015) and Reid et al. (2018) reported that thermodynamic properties of aerosols are strongly related to particle size, for instance, viscosity or diffusion coefficient. This could be another potential reason responsible for the deviation between ours and previous literature data. On the other hand, Rickards et al. (2015) concluded that the timescale of equilibrium for the water transport inside particles is also size-dependent. Larger particles need much more time to reach equilibrium than smaller particles. Then, the aforementioned objective, investigating the effect of residence time on aerosol hygroscopicity, should also take into account of the influence from particle size. Therefore, different approaches with capabilities accessing different particle size ranges, requiring different sample volumes, are still essentially needed.

In the previous section discussing the role of different physico-chemical properties of organics plays in aerosol hygroscopicity, the parameter κ was converted by the measured GF under 90 % RH. For those low or sparingly soluble organic compounds, the GF-derived κ (also known as apparent κ) is RH-dependent (see Fig. S3 as an example) and cannot express their intrinsic κ (expressed by fully dissolved compounds), when compounds are sufficiently soluble in water. As the RH increases, further dissolution of these organic compounds with promoted hygroscopicity is expected. In the real atmosphere, different RH conditions including both sub- and supersaturation can be reached. The measured GF or the apparent κ of ambient aerosols at a certain RH may not be able to reveal their real hygroscopicity under various atmospheric conditions. Further calculations of other variables, for instance the liquid water content (LWC), surface area of wet particles and number concentration of CCN associated with the apparent κ will be significantly biased. If possible, hygroscopicity measurements over large saturation range up to supersaturation, especially with the combination of CCN measurements provide an option to reduce the uncertainties. Therefore, combination of different instruments though with different limitations, should not be discarded but innovated.

Second, the authors fail to compare their data with data available in the literature using different techniques. I tried in my initial review to illustrate this with the data by Salecedo et al. (2006). The authors did a comparison with their data now and provided this figure: However, they do not include it in the revised manuscript. Why? This is exactly what is needed: comparison to previous data (which is available for almost all compounds discussed in the manuscript!) and a critical discussion of such a comparison. In the example for malonic acid shown above, it is not trivial to judge whether the E-AIM fits the complete dataset better of AIOMFAC or UManSysProp. Nor is it clear whether the different data agree within experimental error. Presently, such a discussion is missing in the manuscript. To show another example: In the figure below I plot the data of malic acid of the manuscript together with the data from Peng et al. (2001) and two UManSysProp simulations (I used the density from UManSysProp from Girolami (1994) for the coversion of the Peng et al. data to size growth):

You may argue that all data agree at high humidity (> 85 %), but it is obvious that there is an

increasing deviation between both data sets at lower humidity. (This is definitely larger than any error in the density estimation may cause.) The UManSysProp simulation assuming AIOMFAC activities comes closer to the Peng et al. data while assuming ideality in UManSysProp is closer to the data of the manuscript. It is not clear which data are the correct ones. Are the authors growth factors at low RH limited by kinetic uptake limitations (as in sucrose) also for malic acid? What is however obvious to me is that a comparison of all data in the manuscript with literature data is essential as well as a discussion of discrepancies of these datasets.

Reply: The major suggestion here from the reviewer was to compare our data with data available in literature using different techniques. This was systematically done in the revised manuscript with all the corresponding figures replotted. As the comparison with the thermodynamic models was removed, no judgement about which model fits our results better was given. Detailed discussion for the comparison is listed below for each studied compound. Moreover, we carefully examined their available DRH and ERH values and made thorough discussions on their phase state. A new table (Table 2 in the revised manuscript) was made to summarize all these available information for a better review.

3.1 Hygroscopicity of individual organics

In this section, we summarized the measured hygroscopic properties of the 23 organic species, which are classified into three groups based on their functionality. Particles at the dry size of 200 nm were selected for analysis. Comparison with previous literature data for those compounds were systematically conducted in the following section. The available information with respect to the deliquescence RH (DRH), efflorescence RH (ERH), phase transition, the measured κ at 90% RH for each compound as well as the used instrument in different works are summarized in Table 2.

3.1.1 Carboxylic acids

Carboxylic acids are the most abundant water-soluble components identified in atmospheric aerosols (Chebbi and Carlier, 1996; Mochida et al., 2003; Kundu et al., 2010). Hygroscopic properties of straight-chain dicarboxylic acids have been extensively investigated in previous studies (Chan et al., 2008; Kuwata et al., 2013; Rickards et al., 2013), however, HTDMA data for dicarboxylic acids with additional substitutions and tricarboxylic acids are limited. To achieve a comprehensive overview of the hygroscopicity of carboxylic acids, we measured the water uptake of several common straight-chain dicarboxylic acids in the atmosphere and further extended the hygroscopic measurements for dicarboxylic acids with substitutions and tri-carboxylic acids. Figure 1 & 2 shows the measured humidograms of straight-chain dicarboxylic acids (Fig. 1a-f), dicarboxylic acids with substitutions (Fig. 2a-c) and tricarboxylic acids (Fig. 2d-f), respectively.

Among the studied straight-chain dicarboxylic acids, only **malonic acid** showed continuous hygroscopic growth with increasing RH and the measured GF at 90 % RH was 1.47, shown in Fig. 1a. This agrees quite well with previous studies using other HTDMA systems or other techniques, for instance, EDB, with longer residence time for humidification (Peng et al., 2001; Prenni et al., 2001; Wise et al., 2003; Salecedo et al., 2006; Jing et al., 2016). Previous studies (Swietlicki et al; 2008; Duplissy et al., 2009; Wu et al., 2011; Suda et al., 2013) suggested that the residence time for humidification may also potentially influence the observed water uptake of particles as the measured particles, especially for some organic compounds, may not reach their equilibrium humidified sizes during a quite short time of wetting. However, current results with good consistency with other studies of different residence time suggests that malonic acid may already reach equilibrium of humidification in our system. On the other hand, Braban et al. (2003) reported that the deliquescence RH (DRH) for malonic acid ranges between 69% - 91% RH. However, within this RH range we did not observe any clear evidences for phase transition and our results followed quite well with the ones obtained from the dehydration process by Peng et al. (2001), which further indicated that the measured malonic acid particles in our study existed at liquid state.

The other straight-chain dicarboxylic acids (i.e., **succinic, adipic, suberic and azelaic acids**) did not show any water uptake at RH <= 90 %, as shown in Fig. 1b-e. This non-hygroscopicity was also found for succinic acid in Peng et al. (2001), for adipic acid in Chan et al. (2008) and Dinar et al. (2008) and for azelaic acid in Chan et al. (2008) and Cook et al. (2011), either using a different HTDMA or an EDB. It has to be noted that no previous data was available for suberic acid, suggesting that other measurements using either similar or different methods should be performed for future comparison. Chan et al. (2008) explained that these dicarboxylic acids have quite low-solubility in water and once they crystallized, they would not deliquesce even under high RH conditions (e.g., RH < 90 %). Moreover, we found that the measured GFs of azelaic and suberic acids were less than 1, which could be attributed to the adsorption of a small amount of water at the particle surface, leading to the rearrangements of the microstructure and compaction of the particle (Mikhailov et al., 2004; Mikhailov et al., 2009). It is interesting to note that we observed a slightly weak hygroscopicity for **pimelic acid** (Fig. 1f), while Chan et al. (2008) reported that pimelic acid particles was non-hygroscopic at RH < = 90%, and thus their DRH would be even higher. Based on current comparison with limited data available, we were not be able to confirm which data is more accurate, especially as Chan et al. (2008) measured the particles at micron size range using a different technique with longer residence time. Thus, this deviation indicates that more data for this compound is needed, particularly for dealing with size-dependent hygroscopicity and the influence from residence time of humidification.

The humidograms of the three dicarboxylic acids with substitutions (i.e., double bond or hydroxy group) are illustrated in Fig. 2a-c. The continuous water uptake of **maleic acid** (Fig. 2a) indicates that these particles may be at liquid state under dry conditions, which agrees well with the results of Suda et al. (2013) using another HTDMA at a humidification residence time of 6s. However, the results of Choi et al. (2002), using an EDB with humidification time of 40 minutes, show that maleic acid started to take up water at RH around 75% and this deliquescence process continued until the RH reached to 85%, after which similar growth factor values as ours were observed. Using nonisopiestic method, both Wise et al. (2003) and Marcolli et al. (2004) reported that the DRH of maleic acid would be around 89% RH, being different from Choi et al. (2002) using the EDB as well as the ones by HTDMA. Such a large variation in the phase state as well as phase transition process for this compound provides a note of caution for other researchers when using different data sets. However, the good agreement in GF at 90% RH among different studies suggests that further analysis in Sect. 3.2 using GF at 90% RH should be still reliable.

Continuous water uptake was also found for **malic acid** and **tartaric acid**, similar to those measured by Peng et al. (2001) using an EDB with longer residence time or Cook et al. (2011) using a different HTDMA. However, for both compounds there was a clear and considerable difference in GF at RH lower than 80% between our study and the ones by Peng et al. (2001) during hydration process. This could be due to the short residence time of our HTDMA system, limiting the water uptake of aqueous malic acid. However, previous studies (Apelblat et al., 1995; Marcolli et al.,, 2004; Clegg & Seinfeld, 2006) suggested that both compounds may deliquesce during wetting process, having the DRH potentially ranging between 75% to 82% RH for malic acid and 77% to 78% RH for tartaric acid . Then, their conclusion was not consistent with the results by Peng et al. (2001), where no phase transition was reported. In our study, a small leap of the GFs from 80% to 85% RH was observed for malic acid, implying that these particles were only partially deliquesced and further dissolution occurred at elevated RH. This, on the other hand, partly supported those studies suggesting the deliquescence of malic acid during hydration. Taking both, we are then not sure which factor, either different residence time of humidification or the partial deliquescence, should be considered to be responsible for the difference in GF at intermediate RHs and thus further evidence from other measurements is essentially needed.

Gradual water uptake upon hydration was observed for **citric acid** (Fig. 2d), which is consistent with other studies (Peng et al.,, 2001; Wang et al., 2018). However, our measured GF at any certain RH was higher than the ones by Wang et al. (2018) using another HTDMA, but lower than the ones by Peng et al. (2001) using an EDB. The residence time of wetting inside both instruments were longer than our system but caused opposite effect on the measured GFs, indicating that the influence of residence time on the observed water uptake of particles is not conclusive based on current datasets. Continuous hygroscopic growth over the studied RH range was observed for the other two tricarboxylic acids (e.g., **aconitic acid and tricarboxylic acid**, Fig. 2e-f), indicating no obvious phase change for these particles upon hydration. Based on their GF values at 90% RH, both compounds can be considered as more hygroscopic substances. It has to be noted that it is, to our knowledge, for the first time to report their hygroscopicity within aerosol community. These new datasets, though also need to be further validated with other measurements, might benefit the elaboration of current data pool with extended coverage of atmospherically relevant compounds.

3.1.2 Amino acids

Figure 3 shows the measured humidograms of the 5 amino acids and their individual comparison with previous studies. Chan et al. (2005) reported that the DRH for **alanine and glycine** was 96.9% and 93.2% RH, respectively. Therefore, generally no water uptake at RH <=90% was observed for both compounds in our study as well as theirs. Particularly for glycine (see Fig. 3b), a continuous shrink in wet particle size was observed from 30 % RH to 80 % RH, and above 80 % RH the GFs increased slightly. Previous studies have reported that glycine particles started to absorb water above 60 % RH (Chan et al., 2005; Marsh et al., 2017; Darr et al., 2018) prior to deliquescence due to capillary effect, which could be the potential reason for the shrinkage in particle size of glycine in our study as a result of the microstructural rearrangement of particles upon humidification. The sizing of these structurally-rearranged particles, especially at lower RH range, will be erroneous as the volume change of the particles upon wetting may not only due to the water absorption but also the compaction of the original particles. This phenomena complicates the accurate estimation of the actual water amount absorbed by the particles due to their intrinsic hygroscopicity. In a recent study by Nakao et al. (2014), in order to avoid the influence from particle restructuring upon wetting, they sized wet particles without drying after generation and studied their droplet activation using a wet CCN. This approach they introduced might be an easier attempt, offering an unique solution for current problem from particle restructuring during the hydration processes.

Continuous water uptake was observed for particles of **aspartic acid** (Fig. 3c), indicating that there was no phase transition occurred during the hydration cycle. This pattern was consistent with the ones measured by Hartz et al. (2006) but with slightly lower GF values. As Hartz et al. (2006) used an EDB to measure the hygroscopicity of aspartic acid, the potential reason for the different GF would more likely be the different residence time of humidification between different techniques, though future data should be obtained to further confirm this estimation.

Chan et al. (2005) reported that the DRH values for **glutamine and serine** were 98.8% and 99.1% RH and thus those compounds were considered as non-hygroscopic at RH below 90%. Our observation deviated significantly from theirs, as gradual hygroscopic growth, indicating liquid state, was found for those two compounds. Previous study (Gregson et al., 2020) pointed out that particles without sufficient drying after generation, would not crystallize, for instance, the sample RH after drying was higher than their efflorescence RH (ERH). However, Chan et al. (2005) also reported that the ERH for glutamine and serine were

72.1-74.7 % and 27.6-30.8% RH, respectively, being much higher than the RH of our dried aerosols. Then, this effect with respect to drying process is not the plausible reason for the continuous water uptake of both compounds in our study. Ma et al. (2020) reviewed the ERH values for a series of inorganic and organic compounds and found that particles at micron size range exhibited different ERH as compared to those of submicron particles. On this basis, we speculated that the ERH values for glutamine and serine particles at nanometre range might be different, for instance, lower than the ones reported by Chan et al. (2005) and thus crystallization of these compounds did not occur in our setup. However, the conclusion from Ma et al. (2020), on the other hand, provides a hint that the DRH values for particles at micron size range could also be different than those at nanometre range. Based on this, the DRH of glutamine and serine as well as their phase state might also be size-dependent, potentially explaining the significant difference in the hygroscopic growth between our studies and the ones in Chan et al. (2005).

3.1.3 Sugars and Sugar alcohols

The continuous hygroscopic growth and the measured GF values for particles of **fructose and mannose** (Fig. 4a-b) agree well with the results of Chan et al. (2008) during both hydration and dehydration processes. The only difference is we observed a small leap in GF at RH between 70% and 75% for both compounds. However, considering the measurements uncertainties, this phenomena was not obvious that no further inference can be given. Based on the pattern of their continuous water uptake, we considered these particles exist as liquid state at dry conditions, as no DRH or ERH values were found for those compounds in previous studies. For **sucrose** (Fig. 4c), at intermediate RHs, our measured GF were lower than the ones reported by Estillore et al. (2017) using a different HTDMA with a much longer residence time (40s) as well as the ones by Hodas et al. (2015) based on DASH-SP (Differential Aerosol Sizing and Hygroscopicity Spectrometer Probe) with a residence time of 4s. And the measured GF among different studies show a strong dependence with the residence time of different instruments. As a viscous compound, the transport of water molecules within the particles as well as the particles to reach equilibrium with respect to their surrounding humid air might be quite slow. Thus, the short residence time inside the HTDMA may strongly limit the water uptake for aqueous sucrose. However, our measured GF at high RH conditions, e.g., 90%RH, was quite close to theirs, suggesting equilibrium at this point (i.e., 90%RH) might be already reached or the influence of residence time on the measured hygroscopicity is largely reduced. Therefore, analysis in Sect. 3.2 using GF at 90% RH should still be sound.

For **xylitol and arabitol**, good agreement in the measured GF in general was found between our study and the ones by Bilde et al. (2014), though presented in an AGU abstract, with a small deviation in GF for arabitol at relatively high RH. According to the measured GF, both compounds were considered as more hygroscopic and likely existed as liquid state under dry conditions. From the measured humidogram, **mannitol** with GF less than unity at 90%RH is non-hygroscopic, which also suggested by previous literature (Ohrem et al., 2014; Martău et al., 2020). It has to be noted that no practical data were presented in their work and thus only our measured data was illustrated in Fig. 4f.

Table 2. Summary of the measured properties with respect to their DRH, ERH, phase transition, hygroscopicity parameter (90%RH) obtained in this work and previous literature.

Organic groups	Compounds	DRH (%)	ERH (%)	Phase transition	κ (90% RH)	Instruments	References
Dicarboxylic acids	Malonic acid	n. o.	-	n. o.	0.26	HTDMA	This work
		n. o.	n. o.	n. o.	0.29	EDB	Peng et al., (2001)
		n. o.	<5	n. o.	0.52	HTDMA	Prenni et al., (2001)
		69-91	6	n. o.	-	AFT-FTIR; SMC	Braban et al., (2003)
		71.9	-	n. r.	0.25	Nonisopiestic method	Wise et al., (2003)
		70.9	-	n. r.	0.28	Nonisopiestic method	Salecedo et al., (2006)
		n. o.	-	n. o.	0.39	HTDMA	Jing et al., (2016)
	Succinic acid	n. o.	n. o.	n. o.	0.002	HTDMA	This work
		>90	55.2-59.3	n. o.	0.002	EDB	Peng et al., (2001)
	Adipic acid	n. o.	-	n. o.	-0.003	HTDMA	This work
		>90	>85	n. o.	$2.71 e^{-4}$	EDB	Chan et al., (2008)
		98.5	98.5	n. o.	$4.52 e^{-4}$	HTDMA	Dinar et al., (2008)
	Azelaic acid	n. o.	-	n. o.	-0.007	HTDMA	This work

		>90 81	>85 -	n. o. n. o.	0.003 -	EDB HTDMA	Chan et al., (2008) Cook (2011)
	Suberic acid	n. o. >90	- >85	n. o. n. o.	-0.03 -	HTDMA EDB	This work Chan et al., (2008)
	Pimelic acid	take up water>85 >90	- 51.5-53	n. o. n. o.	0.03 0.092 ± 0.009	HTDMA EDB	This work Chan et al., (2008)
Dicarboxylic acids with substitutions	Maleic acid	n. o. 71-86 88.9 89.1 n. o.	- 48-51 - - -	n. o. Observed n. o. n. r. n. o.	0.30 0.28 0.27 - 0.23	HTDMA EDB Nonisopiestic method Nonisopiestic method HTDMA	This work Choi and Chan (2002) Wise et al., (2003) Marcolli et al., (2004) Suda et al., (2013)
	DL-malic acid	a leap at 80-85 75-82 n. o.	- - n. o. -	Possible partial deliquescence n. r. n. o. n. r.	0.22 - 0.19 -	HTDMA Nonisopiestic method EDB Nonisopiestic	This work Apelblat et al., (1995) Peng et al., (2001) Marcolli et al., (2004)

		80.5	-	n. r.	-	method	Clegg and Seinfeld (2006)
		78.6				Nonisopiestic method	
	Tartaric acid	n. o.	-	n. o.	0.22	HTDMA	This work
		77-78	-	n. r.	-	Nonisopiestic method	Apelblat et al., (1995)
		n. o.	n. o.	n. o.	0.19	EDB	Peng et al., (2001)
		n. o.	-	n. r.	-	HTDMA	Cook (2011)
Tricarboxylic acids	Citric acid	n. o.	-	n. o.	0.18	HTDMA	This work
		n. o.	n. o.	n. o.	0.18	EDB	Peng et al., (2001)
		>90	-	n. o.	0.20	HTDMA	Wang et al., (2018)
	Cis-aconitic acid	n. o.	-	n. o.	0.21	HTDMA	This work
	Butane-1,2,4-tricarboxylic acid	n. o.	-	n. o.	0.14	HTDMA	This work
Amino acids	DL-alanine	n. o.	-	n. o.	-0.009	HTDMA	This work
		96.9	67.3-70.8	n. o.	$6.11 e^{-4}$	EDB	Chan et al., (2005)
	Glycine	n. o.	-	n. o.	0.04	HTDMA	This work

		93.2	53.6-55.2	n. o.	0.04	EDB	Chan et al., (2005)
		60	<35	not clear from data	-	ATR-FTIR	Darr et al., (2018)
	L-Aspartic	n. o.	-	n. o.	0.14	HTDMA	This work
		n. o.	n. o.	n. o.	0.18	EDB	Hartz et al., (2006)
	L-Glutamine	n. o.	-	n. o.	0.16	HTDMA	This work
		98.8	72.1-74.7	n. o.	0.006	EDB	Chan et al., (2005)
	L-Serine	n. o.	-	n. o.	0.19	HTDMA	This work
		99.1	27.6-30.8	n. o.	0.002	EDB	Chan et al., (2005)
Sugar and sugar alcohols	Fructose	n. o.	-	n. o.	0.23	HTDMA	This work
		n. o.	n. o.	n. o.	0.18 ± 0.017	EDB	Chan et al., (2008)
	Sucrose	n. o.	-	n. o.	0.11	HTDMA	This work
		>90	-	n. o.	0.11	DASH-SP	Hodas et al., (2015)
		n. o.	n. o.	n. o.	0.11	HTDMA	Estillore et al., (2017)
		-	-	n. r.	0.10	HTDMA	Dawson et al., (2020)
	Mannose	n. o.	-	n. o.	0.14	HTDMA	This work

		n. o.	n. o.	n. o.	0.183 ± 0.017	EDB	Chan et al., (2008)
	Xylitol	n. o.	-	n. o.	0.21	HTDMA	This work
		n. o.	-	n. o.	0.18	HTDMA	Bilde et al., (2014)
	L-arabitol	n. o.	-	n. o.	0.19	HTDMA	This work
		n. o.	-	n. o	-	HTDMA	Bilde et al., (2014)
	D-mannitol	n. o.	-	n. o.	-0.01	HTDMA	This work

κ values were derived from hydration data at around of 90% RH.

n. o. refers to no crystallization was observed.

n. r. refers to not reported in the work.

- refers to not measured in the work.

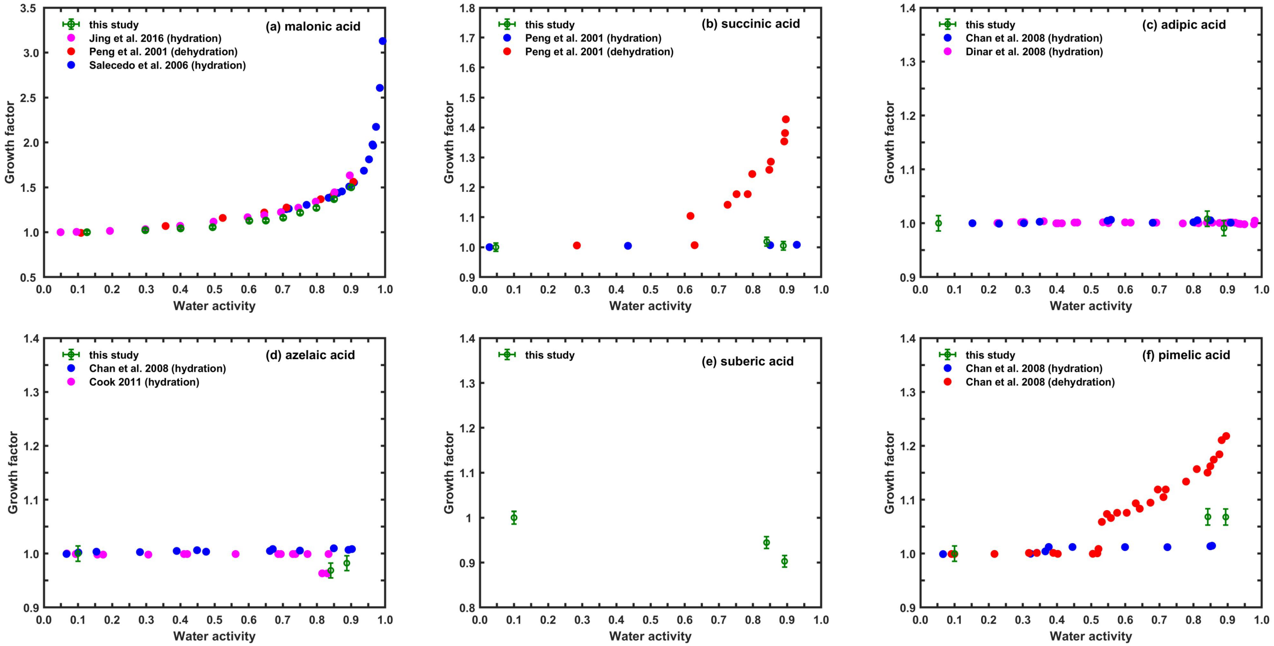

Figure 1. The hygroscopic growth values of straight-chain dicarboxylic acids particles (200 nm) measured in this study and their individual comparison with previous studies.

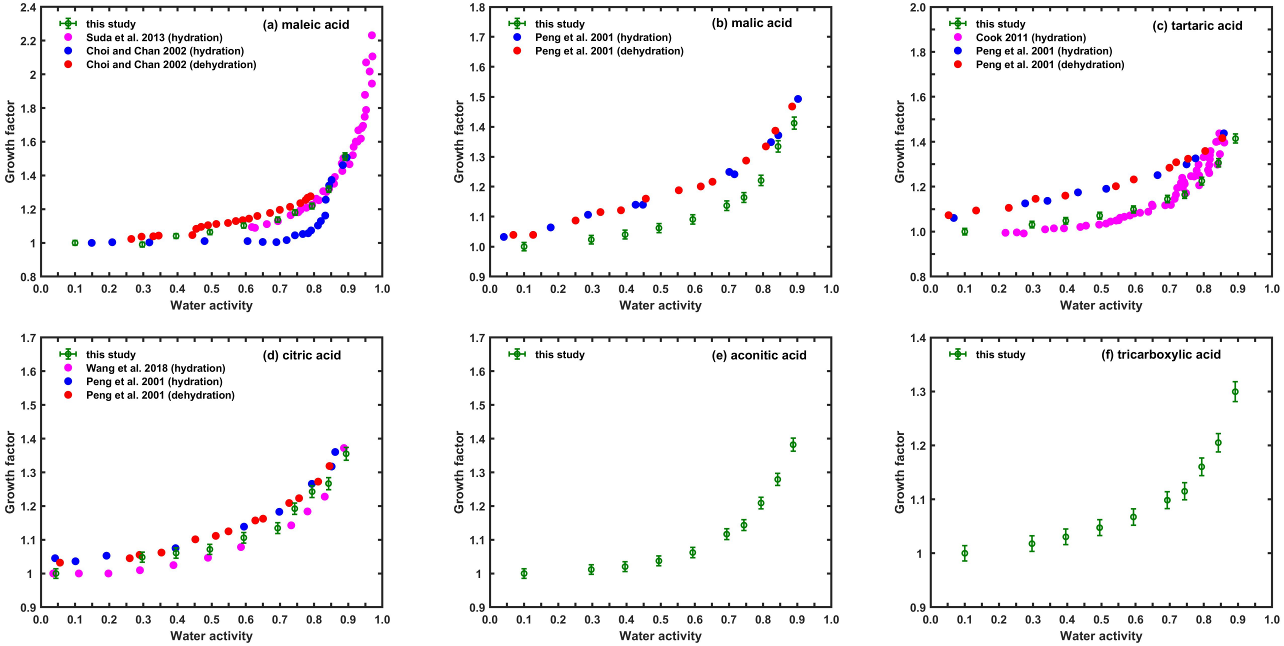

Figure 2. The hygroscopic growth of straight-chain dicarboxylic acids with substitutions (a-c) and tricarboxylic acids (d-f) particles (200 nm) measured in this study and previous studies.

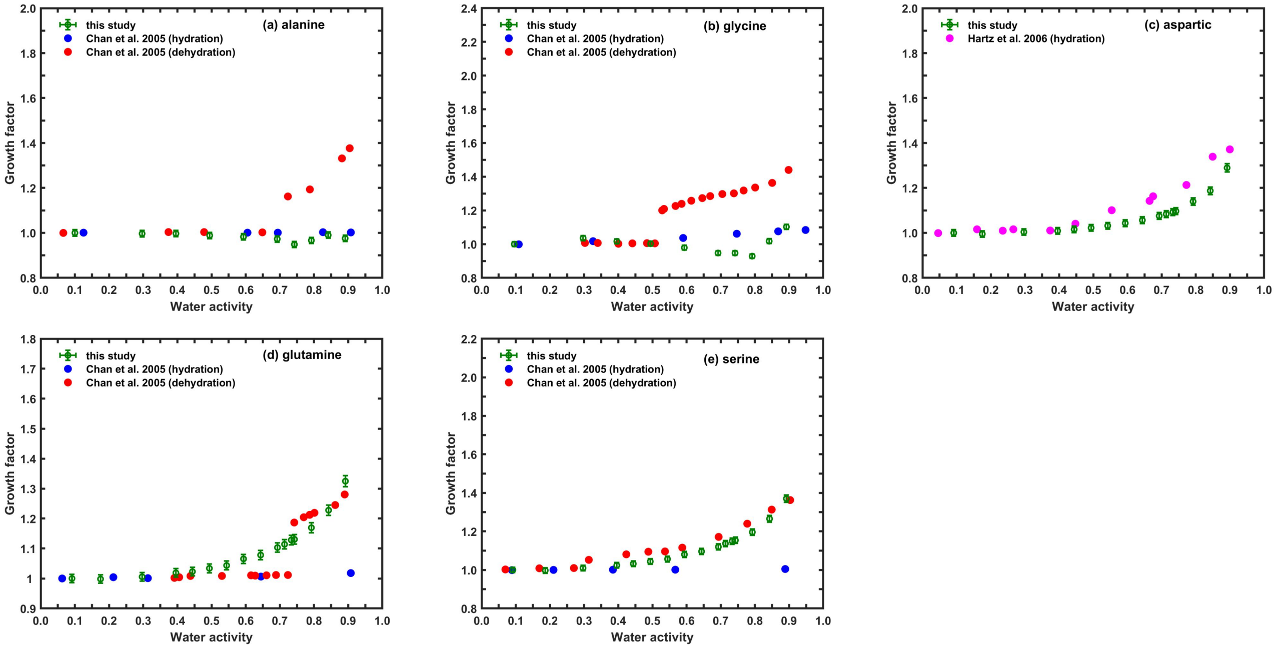

Figure 3. Hygroscopic growth factors of amino acids, and comparison with previous studies.

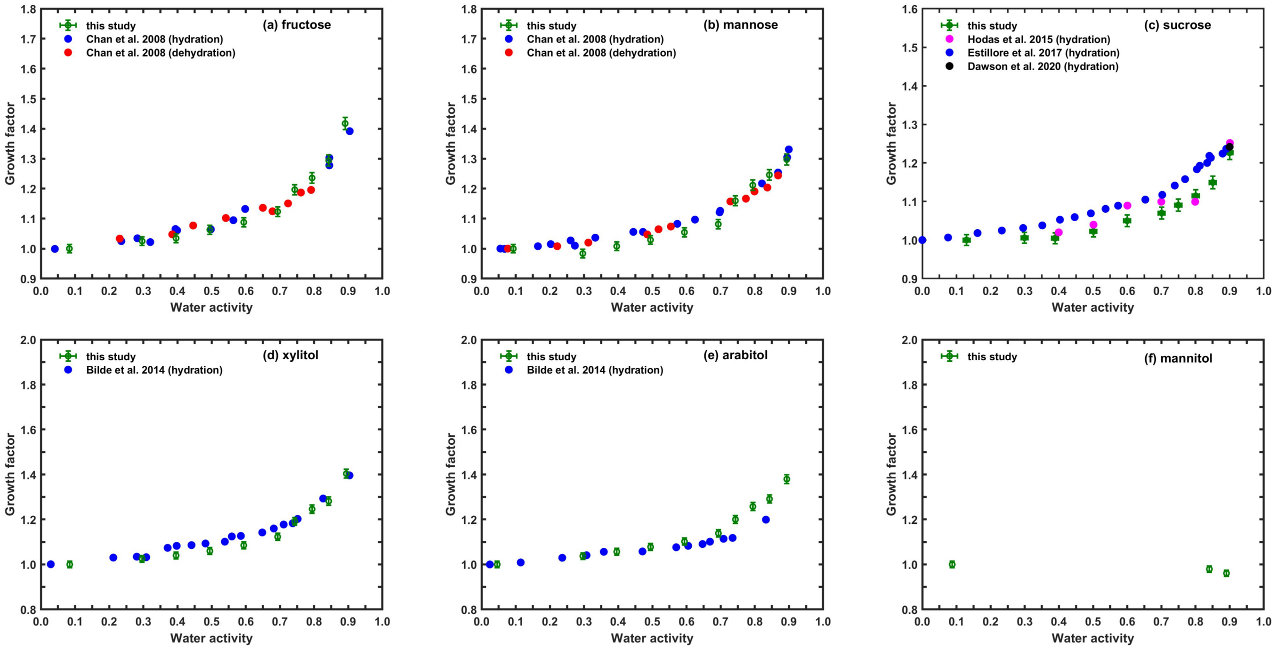

Figure 4. Hygroscopic growth factors of sugars (a-c) and sugar alcohols (d-f), and comparison with previous studies.

Third, I feel the authors did not describe the UManSysProp on-line model well enough. As far as I understand, it is based on AIOMFAC for calculating the water activity (or did the authors assumed it to be ideal?). Hence it is clear that the curves shown in Fig. 1a of the manuscript for the dicarboxylic acid describe the liquid state hygroscopicity and cannot compared to the data if the experiments are done with crystalline particles. Again, a comparison would be possible only if the supersaturated liquid particle were studied experimentally. There are several option for selections with the UManSysProp web-model, for example the method to estimate supersaturated density. The authors do not write which method they used nor why they selected a particular one. I feel, given the considerable discrepancies between the data presented and other datasets, it is not appropriate to judge which thermodynamic model represents the data better as done by the authors for the dicarboxylic acids.

Reply: In the previous version of our manuscript, we used the UManSysProp on-line model assuming non-ideal interactions using AIOMFAC for calculating the water activity. We agree that the results from the UManSysProp describe the liquid state hygroscopicity and cannot be compared to our data, which are for crystalline particles. We are sorry for the limited awareness during the usage of this model and would not give any judgement for their performance in simulating aerosol hygroscopcity until the results of supersaturated liquid particles are obtained. Although we removed the part of model comparison in the revised manuscript, for the reviewer’s interest, we still did a sensitivity analysis of using different supersaturated density for model calculation as suggested by the reviewer. This was particularly done by malonic acid, as we considered this compound exist as liquid state under relative dry condition. The result is listed as below. From the results we notice, there was no obvious difference in the predicted values using different supersaturated density. Moreover, the predicted value did not vary significantly assuming both ideal and non-ideal interactions.

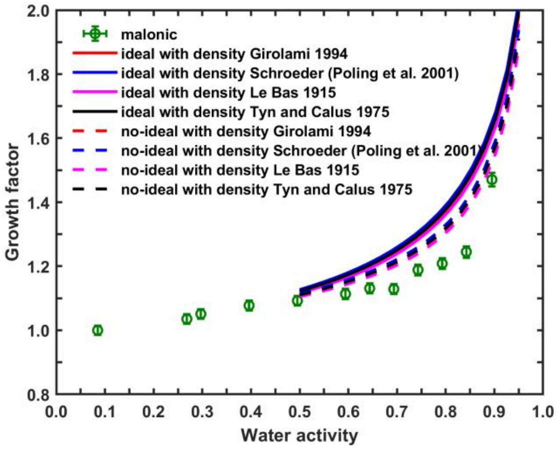

Figure 5: Measured and predicted hygroscopicity of malonic acid. The predicted ones were obtained by the UManSysProp on-line model assuming both ideal and non-ideal interactions using different supersaturated density values.

Fourth, I appreciate the authors address the issue of kinetic limitations to water uptake in their text now, but they do not show how significant the effect could be. In the reply to my question, they provided a nice figure, which I copy here and which in my eyes show the problem unambiguously. It nicely shows that at low RH with high viscosity of the sucrose the authors do not reach water equilibrium while at high RH with water acting as a plasticizer they do. This figure may also explain the differences in the malic acid figure above? However, the author ones more do not include this figure in the revised manuscript. Why? This figure needs to be discussed together with the added text!

Reply: Thanks for the reviewer's suggestion. We included and discussed this figure into the revised manuscript. This was similarly done for malic acid as well as the other compounds.

Overall, I conclude that the manuscript requires at least a major revision with a comparison of all data to the data available in the literature, so that a reader sees the discrepancies and are not left with the impression that there are no other data available and also clearly see the limitations of the technique the authors have been using.

Reply: Thanks for the reviewer's suggestions. We made a thorough comparison with previous data available in the literature, see my response to your second comment. Besides the discussion of the comparison, we also made a summary of the properties with respect to the DRH, ERH, phase transition, hygroscopicity at a certain RH of each compound measured in this work and previous studies. We agree with the reviewer that there remains clearly limitations of current technique, and thus we added a section discussing the limitations of our instrument in Sect. 4, giving the possible atmospheric implications using HTDMA data. This can be seen from the response to your first comment. Overall, we hope the revised manuscript in general could address most of the reviewer's concerns.