Anonymous Referee #2

We would like to thank the reviewer for the constructive comments. The detailed responses are given as follow:

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1. The authors compared in the text the measured DRH and hygroscopic properties of the organic salts with the previous findings. It may be more straightforward and clearer to include the previous data in the graphs.

Response:
The water cycle of organic salts were measured by Peng and Chan (2001) using an electrodynamic balance (EDB). They used the ratio of the dry solute mass to the solution droplet mass versus relative humidity to present the hygroscopic behavior of organic particles. This is different from the results derived from HTDMA measurements. Therefore, it is not straightforward to put those previous data into the same graphs. But, we compare the DRH and growth factor at 90% in a tabulate form between literature data and our measurements.

Modifications in the MS:
The comparisons in hygroscopic properties of the organic salts are shown in the table 2 in the MS.

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2. (P.7699, section 3.1.1) The authors mentioned that they measured lower DRH than that detected by Peng and Chan (2001) due to different diameters. Although a reference paper is provided, the authors should further explain the issue so that the readers can easily follow.

Response:
One explanation is the Kelvin effect, which may cause two effects influencing the DRH of small particles (Hameri et al., 2000). First, the solubility of a crystal increases as its size decreases, which can be described as a Kelvin effect on a solid particle suspended in the liquid phase. This effect tends to shift the DRH toward lower RH. Second, the solution drop that is formed as deliquescence takes place has an increased water vapor pressure compared with a larger droplet with a similar solute concentration due to Kelvin effect. This tends to shift the DRH toward higher RH. The DRH of may be affected by both of the effects and by water adsorption on dry particles (Hameri et al., 2000). The particles with 20 µm diameter were detected in Peng and Chan's study, while 100 nm particles are measured in our study. Another explanation is the difference in DRH may cause by the measurement uncertainty, which is around +/−1% RH.

Modifications in the MS:
One of reasons may be the difference in particle size between two studies: Particles with 20 µm diameter were detected in Peng and Chan's study, while 100 nm particles are measured in our study. For smaller particles, the Kelvin effect may cause two effects influencing the DRH (Hameri et al., 2000): First, the solubility of a crystal increases as its size decreases, which can be described as a Kelvin effect on a solid particle suspended in the liquid phase. This effect tends to shift the DRH toward lower RH. Second, the solution drop that is formed as deliquescence takes place has an increased water vapor pressure compared with a larger droplet with a similar solute concentration due to Kelvin effect. This tends to shift the DRH toward higher RH. The DRH of may be affected by both of the effects and by water adsorption on dry particles (Hameri et al., 2000). Another reason may be the measurement error with uncertainty of +/−1% RH.

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3. Peng and Chan (2001) reported residual water in sodium acetate and sodium malonate at low RH conditions but this is not observed in the current study. The authors may want to explain the difference between two observations.

Response:
We cannot analyze the residual water of these particles by the HTDMA measurements. The hydration state of the atomizer-generated sub-micrometer particles may be different from that of crystals formed from bulk solutions. For examples, experimental study (Peng and Chan, 2001) shows that Na_2SO_4 particles formed from levitated droplets in an electrodynamic balance (EDB) is anhydrous, but not decahydrate salt, which is the most stable form at 25 ˚C (Cohen et al., 1987). Mensah et al. (2010) also observed that oxalic acid aerosol generated by atomizer show a reduced crystal water content compared to its thermodynamically stable state.

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4. The author reported different deliquescence properties of sodium succinate from those observed by Peng and Chan (2001) and "the reason is unclear" (P7699, section 3.1.4). I would like to see the authors can make some effort on speculating a possible reason, rather than leaving the situation unclear.

Response:
    The solubility of sodium succinicate is strongly temperature dependent (Rozaini and Brimblecombe, 2009). The temperature at which the measurement was performed may cause this discrepancy in DRH determination. In addition, the impurity of solution being measured may be also one possible reason.

Modifications in the MS:
Similar to sodium malonate, no deliquescence point is observed for sodium succinate, as shown in Fig 1. (d). This is different from the observation using an EDB by Peng and Chan (2001), whose result shows the DRH is 63.5-66%. The solubility of sodium succinicate is strongly temperature dependent (Rozaini and Brimblecombe, 2009). The difference in the temperature at which the measurement was performed between two studies may cause this discrepancy in DRH determination. In addition, the impurity of solution being measured may be also one possible reason.

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5. Overall, the authors have made a straightforward report of their measurements and brief comparison between their results and the few literature findings. Given that there are other published works of similar nature and also most of the specific organic salts have been studied previously, they should consider a comprehensive discussion to provide more insight over the existing literature. More specific comments and suggestions are as follows.
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6. Mass transfer effect has been shown to be the limitation of hygroscopic measurements by HTDMA (Chan and Chan 2005 and the references therein). How does the issue affect the current results? The author will need to consider this issue in the comparison of their measurements with the equilibrium measurements using EDB by Peng and Chan (2001) in section 3.1 as well.

Response:
The residence time of particles at nominal RH before entering into the DMA2 is around 2.5 seconds in our system. This residence time may be not significantly long enough for some organic compounds to attain equilibrium state (Chan and Chan, 2005;Peng and Chan, 2001;Duplissy et al., 2008) , for example, Sodium pyruvate which needs longer time to obtain equilibrium (Peng and Chan, 2001). The short residence time may cause underestimation of growth factor for some organic particles, which need longer

time to obtain equilibrium state. We made a comparison between our results and Peng and Chan's, as shown in the table 2. It shows that larger growth factors were observed in Pend and Chan's measurements for sodium acetate, disodium succinate, and sodium pyruvate. The difference in the residence time may partly explain the difference in growth factors between both studies. However, the particle size, detection methods, and measurement uncertainty should also contribute this difference. We will give a discussion in the MS.

Modifications in the MS:
The residence time of particles at nominal RH (90%) before entering into the DMA2 is around 2.5 seconds in our system. This residence time may be not significantly long enough for some organic compounds to attain equilibrium state (Chan and Chan, 2005;Peng and Chan, 2001;Duplissy et al., 2008) , for example, Sodium pyruvate which needs longer time to obtain equilibrium (Peng and Chan, 2001)."

The growth factor of sodium acetate at 90% is 1.79, which is much lower than that (1.91) measured by Peng and Chan (2001). This discrepancy may be caused by the difference in the residence time between two studies as above mentioned. Other factors, such as, the particle size, detection methods, and measurement uncertainty should also contribute this difference.

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8. The statement ": : :lower DRH (73%+/-1) of Mix_mar indicates that ammonium sulfate in the mixture starts to take up some water before reaching its deliquescence point" (P7701, line26) needs to be clarified. It is less convincing without any evidence from suitable characterization for the component phase identity. On the other hand, early water-uptake in mixed particles of ammonium sulfate and organic acids has been characterized in some EDB/Raman studies (e.g. Ling and Chan 2008, Yeung and Chan 2010). The authors may find these papers useful to support the statement.

Response: Yes, we agree!
In Chan et al.'s studies, they investigated partial deliquescence of ammonium sulfate (AS) and dicarboxylic acids mixtures using an electrodynamic balance (EDB) and a Raman spectrometer. In their study, as an example, the lower deliquescence relative humidity (DRH) of AS and malonic acid (MA) mixture was observed compared with pure sulfate. They proposed that MA with lower deliquescence relative humidity (DRH) contributed to the water uptake at low RH, and deliquescence of AS gradually took place starting from 69% RH, then the mixture completely deliquescent at 75%. However, the HTDMA measurement cannot provide the direct information on the phase state of the mixtures of AS and organic salts in our study. The conclusions from Chan et al. will be presented in the manuscript to support our explanations.

Modifications in the MS:
The humidograms of mixtures and pure ammonium sulfate (AS) are shown in Fig. 2. There is a clear shift in DRH to lower RH with increasing organic mass fraction, indicating that these organic salts have significant effects on the deliquescence of AS. With increasing organic mass fraction, a smoothing of the deliquescence behavior is observed. This phenomenon was also observed for mixtures of AS and individual dicarboxylic acids such as citric acid, succinic acid, and malonic acid by previous studies (e.g. Zardini et al., 2008;Hameri et al., 2002). Ling and Chan (2008) and Yeung and Chan (2010) investigated partial deliquescence of AS and dicarboxylic acids mixtures using an EDB and a Raman spectrometer. They observed that malonic acid with lower DRH contributed to the water uptake at low RH, and deliquescence of AS gradually took place starting from 69% RH, then the mixture completely deliquescent at 75%. Some organic acid including glutaric, pinonic, adipic, and phthalic acids with low

water solubility show no influence on the deliquescence behavior of ammonium sulfate (Cruz and Pandis, 2000;Hameri et al., 2002).

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10. One intriguing finding is that the hygroscopic growth of mix_mar and mix_bio particles above 80% RH was found to be close to that of pure ammonium sulfate. The observation implies that the major fraction of organic salt in these particles (ammonium oxalate) should contribute to the water uptake close to that of ammonium sulfate. I would like the authors to elaborate on this observation in the revised paper.

Response:
Thanks for this suggestion. We added this conclusion into the MS.

Modifications in the MS:
The differences in growth factors of Mix_urb with low organic mass fraction and ammonium sulfate are not obvious above their DRHs. For Mix_mar and Mix_bio, growth factors above RH=80% are slightly lower than, but quite close to those of pure ammonium sulfate, as shown in Fig. 2. This observation implies that the major fraction of organic salt in these particles should contribute to the water uptake close to that of ammonium sulfate.

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13. I'm not very sure whether the calculation works in section 3.2.3 is relevant to the rest of the paper. The single hygroscopicity parameter is used to correlate the hygroscopic growth of aerosols with their CCN activity (Petters and Kreidenweis 2007), which is obviously not the objective of this paper. The authors will want to clarify the purpose of the calculation in this section.

Response:
The "κ-Köhler theory" applies over the entire range of relative humidity and solution hygroscopicity (Petters and Kreidenweis, 2007). It can be used to predict particle water content in the sub-saturated regime. The mixing rule for this method is developed based on the Zdanovskii–Stokes–Robinson (ZSR) assumption. To predict the growth factor of mixture using kappa makes no any difference with ZSR method does. In the revised manuscript, we will use the ZSR method instead of kappa mixing rule.

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7. Since the organic salts are formed from the neutralization of their acid forms, one interesting and important implication from this aspect is how the transformations alter the hygroscopic properties of particles. Unfortunately this issue is missing in the paper. The hygroscopic growths of many organic acids have been extensively studied (e.g. Peng et al. 2001, Prenni et al. 2001, Choi and Chan 2002, Braban et al. 2003, Chan and Chan 2003, Parson et al. 2004 and many others). The authors may want to compare the organic acids against their corresponding salts in order to show the significance of organic salt formation.

12. The hygroscopic properties of three mixed particles that simulate the ambient aerosols of different sources (replace carboxylic acids by carboxylic salts) are examined. I would be further interested to know if any differences in term of the hygroscopic properties between the simulate aerosols and the "real ones" (with organic acids). On the other hand, the hygroscopic growth of ambient aerosols from different sources has been extensively studied (e.g. Mcfiggans et al. 2006, Swietlicki et al. 2008). The authors may want to compare their results with the literature findings.

Response:
Both Q7 and Q12 are related to the organic acids. We will answer two questions together.

First, we compared the growth factors at 90% of dicarboxylic acids found in the literature with those of organic salts measured in this study. Afterwards, we predicted the growth factors of mixtures in which organic acids were used instead of their salts by ZSR methods. The detailed answer is given in Q 9-14, see below.

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9. In section 3.2.2, the authors focus on hygroscopic growth of the three mixtures above 80% RH. Why limited to this high RH range? Would data below 80% RH that illustrate the thermodynamics of aerosol drops at high solute concentrations and high ionic strength be more interesting?

11. Further to the point 8, more discussion on the use of a modified Kohler theory are expected (as they mentioned in P.7702, line 15-16). However, I'm not able to find such discussion in section 3.2.3. The information available in that section basically only focuses on the comparison of the hygroscopicity parameters calculated by different methods.

14. Furthermore, I suggest the authors to compare their measurement with the predictions from other well-established thermodynamic model approach (e.g. E-AIM, ADDEM, simple ZSR calculation) over the range of RH. This is a more complete comparison and is also useful to evaluating the model predictions for hygroscopic growth at high solute concentrations.

Response:
Q9, Q11, and Q14 are interrelated. We make the response together. The the Zdanovskii–Stokes–Robinson (ZSR) assumption will be used to predict the hygroscopic growth of mixtures over the whole RH range. The growth factor of mixture of ammonium sulfate and dicarbonxylic acids is calculated using ZSR method based on the literature data. The results are compared with the mixture with organic salts. The comparison shows that if the water soluble organic acids in the atmospheric aerosol are neutralized by ammonium or metal ions, the particle hygroscopicity will decrease. More discussions are given below.

Modification in the MS:
1) **In the section 2, we added on subsection** *2.4 data evaluation* **to briefly describe the ZSR method:**

The hygroscopic growth of mixtures with different components can be predicted using the Zdanovskii–Stokes–Robinson (ZSR) relation for water activities of mixed particles (Stokes and Robinson, 1966). The ZSR method can be presented as:

$$\sum_s \frac{m_s(a_w)}{m_{0,s}(a_w)} = 1 \qquad [6]$$

where $m_s(a_w)$ is the molality of compound s in the mixture, and $m_{0,s}(a_w)$ is the molality of solute s in a solution containing only s at the same water activity as the mixture. Based on the ZSR method, the hygroscopic growth factor of a mixture (GF_{mixed}) can be estimated from the GF_i of the pure components and their respective volume fractions, ε_i (Malm and Kreidenweis, 1997):

$$GF_{mixed} = \left(\sum_i \varepsilon_i GF_i^3 \right)^{1/3} \qquad [7]$$

Here, ε_i are calculated from mass fractions, w_i, of component i in the mixture with the assumption of no change in the volume after compounds are mixed:

$$\varepsilon_i = \frac{w_i / \rho_i}{\sum_k (w_k / \rho_k)} \qquad [8]$$

In this equation, the denominator means the total volume of mixture.

2) **The growth factors of organic acids reported in the literature are listed in the table 2 and compared with their salts.**

Species	GF (90%) in literature	Species	GF (90%) in literature
Ammonium sulfate	1.67-1.72[a]		
Sodium acetate	1.91[b]	acetic acid	NF
Disodium oxalate	1.0[b]	oxalic acid	1.00[d]
Diammonium oxalate	1.0[b]		
Disodium malonate	1.78[b]	malonic acid	1.48[d]
Disodium succinate	1.85[b]	Succinic acid	1.00[d]
Disodium tartrate	NF	tartaric acid	1.39[e]
Diammonium tartrate	NF		
Sodium pyruvate	1.76[b]	Pyruvic acid	NF
Disodium maleate	1.76[b]	Maleic acid	1.51[d]
Humic acid sodium salt	1.18[c]	Humic acid	1.06[c]

a: (Duplissy et al., 2008); b: (Peng and Chan, 2001); c: (Gysel et al., 2004); d: (Wise et al., 2003); e: (Peng et al., 2001)

3) **In section 3.2.2, the comparison between the ZSR prediction and experimental results are discussed.**

3.2.2 Comparison with theoretical predictions

The differences in growth factors of Mix_urb with low organic mass fraction and ammonium sulfate are not obvious above their DRHs. For Mix_mar and Mix_bio, growth factors above RH=80% are slightly lower than, but quite close to those of pure ammonium sulfate, as shown in Fig. 2. This observation implies that the major fraction of organic salt in these particles should contribute to the water uptake close to that of ammonium sulfate.

The *GF*s of mixtures are predicted using the ZSR method with/without taking the solubility of ammonium oxalate into account. The results are shown in Fig. 2. If the contribution to the water uptake by ammonium oxalate is included, the *GF*s predicted by ZSR is closer to those measured by HTDMA than without considering its solubility. Previous study (e.g., Svenningsson et al., 2006) also reported that the ZSR mixing rule can better predict the growth factor of mixture with slightly soluble component when the limited solubility is taken into account.

As shown in Fig. 2, the ZSR predictions agree very well with experimental results above the DRH of ammonium sulfate. However, the ZSR calculations underestimate the water uptake of mixture below the DRH. The similar results were also reported by Zardini A. A. et al. (2008). They showed that the hygroscopicity of mixtures of ammonium sulfate and glutaric acid is well described by the ZSR relationship as long as the two-component particle is completely liquid. In fact, the lower DRH of mixture identified by THDMA measurements indicates that ammonium sulfate starts take up water before full deliquescence. This phenomenon was also experimentally observed using an EDB and a Raman spectrometer (Ling and Chan, 2008;Yeung and Chan, 2010). In the simple mixing rule, it is assumed that ammonium sulfate absorb water until RH reached the DRH. This must underpredict the hygroscopic growth of mixture at lower RH. In previous studies on inorganic and organic mixtures, the ZSR model was found to be useful in predicting the water activity of the mixtures and the growth ratios (e.g., Cruz and Pandis, 2000;Gysel et al., 2004;Svenningsson et al., 2006). However, the detailed deliquescence behaviors of the organic/inorganic mixtures cannot be easily predicted from the hygroscopic properties of the individual components (Choi and Chan, 2002).

In order to compare the effects of organic salts and their acids on the hygroscopic growth on ammonium sulfate, the *GF* of Mix_bio_acid is calculated using the ZSR method and shown in Fig.2. The Mix_bio_acid consists of ammonium sulfate, oxalic acid, malonic acid, succinic acid, maleic acid, and sodium pyruvate (Here, the *GF* of pyruvic acid is not found in the literature). The growth factor of the individual dicarboxylic acid over the whole range of water activity are calculated according to Table 3 and equation (5) in Wise et al. (2003). The volume fraction of individual organic acids is the same with Mix_bio. The *GF* of Mix_bio_acid is lower than that of Mix_bio above the DRH of ammonium sulfate. This implies that if the water soluble organic acids in the atmospheric aerosol are neutralized by ammonium or metal ions, the particle hygroscopicity will decrease.

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Reference:

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Table 2: Polynomial coefficients of the growth curve parametrization as well as *GF*s at RH=90% and Vant't Hoff Factor.

Salts	a_w	a	b	c	GF (90%) This study	GF (90%) In literature	Organic acid	GF (90%) In literature	Van't Hoff Factor
Ammonium sulfate	0.8-0.9	0.23018	0.92875	-0.7282	1.70	1.67-1.72[a]			2.04
Sodium acetate	0.4-0.9	0.51418	0.67867	-0.65864	1.79	1.91[b]	acetic acid	NF	1.80
Sodium oxalate	-	-	-	-	1.00	1.0[b]	oxalic acid	1.00[d]	
Ammonium oxalate	-	-	-	-	1.05	1.0[b]			
Sodium malonate	0.2-0.9	-0.25346	3.20915	-2.5515	1.78	1.78[b]	malonic acid	1.48[d]	2.95
Sodium succinate	0.2-0.9	-0.11095	3.25171	-2.92462	1.69	1.85[b]	Succinic acid	1.00[d]	
Sodium tartrate	0.2-0.9	-0.52175	2.19924	-1.38076	1.54	NF	tartaric acid	1.39[e]	
Ammonium tartrate	0.2-0.9	-0.01105	0.3971	-0.236	1.29	NF			
Sodium pyruvate	0.8-0.9	0.66856	-0.45491	0.28678	1.70	1.76[b]	Pyruvic acid	NF	1.95
Sodium maleate	0.1-0.9	-0.31807	3.41428	-2.69608	1.78	1.76[b]	Maleic acid	1.51[d]	3.30/2.94
Humic acid sodium salt	0.1-0.9	0.10856	0.28154	-0.33565	1.19	1.18[c]	Humic acid	1.06[c]	

a: (Duplissy et al., 2008); b: (Peng and Chan, 2001); c: (Gysel et al., 2004); d: (Wise et al., 2003); e: