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## ***Interactive comment on “Hygroscopic behavior of multicomponent organic aerosols and their internal mixtures with ammonium sulfate” by B. Jing et al.***

**Anonymous Referee #2**

Received and published: 22 September 2015

The authors measure the hygroscopic growth of water soluble organic compounds (single component, multicomponent, and organic/AS) using a hygroscopicity tandem differential mobility analyzers (HTDMA) system. They provide valuable experimental data to evaluate the performance of different models (e.g. UNIFAC, E-AIM and ZSR) on predicting the hygroscopic growth of organic aerosols. Additionally, the attenuated total reflectance Fourier transform infrared (ATR-FTIR) spectroscopy is used to gain information about molecular interactions and phase transitions for some investigated systems. However, I have one major comment. The authors mention that the chemical interactions between the species can affect the hygroscopic growth or phase transition

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of the mixed aerosols throughout the manuscript. For example, in the abstract, “For the internal mixture of oxalic acid with levoglucosan or succinic acid, there is enhanced water uptake at high RH due to positive chemical interactions between solutes.” It is not clear what kinds of chemical interactions are referred to and how these interactions enhance the hygroscopic growth of the mixed aerosols. The authors should explain and address how and what kinds of interactions between the species alter hygroscopicity of the mixed aerosols in details.

#### Minor comments

Page 23362, line 10, “The dry polydisperse aerosol particles were then charged and passed through the first differential mobility analyzer (DMA1), where nearly monodisperse aerosol particles with the desired initial particle diameter of 100nm were selected.” Given that some species are non-crystallizing, how could the author ensure that initial dry aerosols are generated for all investigated systems?

Page 23362, line 15, “The residence time of aerosol particles at a given RH before entering into DMA2 was about 5s in the humidification section.” How could the authors ensure that the single component and multicomponent aerosols reach their equilibrium states within this timescale (5s)?

Page 23368, line 16 “Predictions from UNIFAC model based on MA liquid assumption have a good agreement with measured GF above 50% RH while slightly overestimate hygroscopic growth at lower RH.” Based on MA liquid assumption, what the composition of MA aerosols is predicted by the model? Does the model predict that MA aerosols retain water or have no residual water at low RH?

Page 23370, line 18, “For the 1 : 1 mixed Lev/OA particles (Fig. 3a, b), no deliquescence behavior is observed and the continuous water uptake starting at low 20 RH  $\sim$  10% indicates the initial particles appear to be in a liquid state.” What is the composition of the mixed Lev/OA particles at low RH? Any residual water present in the mixed particles?

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Comment](#)

Page 23372, line 17, “ Both ZSR and ideal solution model could well describe the hygroscopic behavior of this mixture within experiment uncertainty, indicating that no interactions related to hygroscopicity exist and the droplets behave like ideal solution” Please elaborate what is meaning of “there is no interactions related to hygroscopicity exist”.

Page 23373, line 9, “Due to the influence of MA, the OA may also exist in a liquid state at low RH.” What is the composition of mixed particles at low RH? Do the mixed particles are completely dried? Any FTIR data or model results to support the presence of dry particles? If not, how the residual aerosol phase water at low RH affects the Gf calculation and the data analysis?

Page 23373, line 25, “The water uptake by the OA/SA mixture may be contributed to OA. It seems can be explained by the strong interactions among OA, SA and water molecules upon hydration.” Please elaborate what are the strong interactions among OA, SA and water molecules and how the interactions affect the hygroscopic growth.

Page 23374, line 21, “It is clearly that phase behavior of OA is strongly influenced by the coexisting organic species, thus resulting in enhance of interactions between water and organic mixtures containing OA.” Please elaborate how strong interactions between the species enhance the hygroscopic growth.

Page 23376, line 23, “The enhanced hygroscopicity of mixed particles may be attributed to the chemical interactions between levoglucosan and OA.” Please explain why interactions between levoglucosan and OA enhance the hygroscopic growth of mixed particles.

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Interactive comment on Atmos. Chem. Phys. Discuss., 15, 23357, 2015.

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