Interactive comment on “A study of the phase transition behavior of mixed ammonium sulfate – malonic acid aerosols” by C. F. Braban and J. P. D. Abbatt

Anonymous Referee #2

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General comment: This paper describes an excellent and detailed study on the phase transitions of the ammonium sulfate-malonic acid system using FTIR spectroscopy. While a number of other studies on relevant organic acid/ammonium sulfate particles have been conducted, this work is significant since it is the first work which investigates the phase of each of the individual components as a function of relative humidity (RH). It also brings to light a very interesting point, that considering these complex systems in terms of the deliquescence and efflorescence of the system, as is commonly done, does not present a full description of the aerosol, since at some RHs, solid and solution phase components are simultaneously present in the system.

Specific comments: pg 4 and fig1. You discuss what the spectra looks like at 0.2
fraction MA, but you do not show it. I recommend including it in Figure 1. Also, I disagree with your interpretation of the RH at which transitions are occurring in the case of the NH4+ mode at 1420 cm⁻¹, I clearly see the broadening of the peak, indicative of the salt in solution in Figure 1, C and D, which are 0.49 and 0.75 fraction MA, respectively. However, in 1B, 0.25 fraction MA, the NH4+ appears to still be crystalline. Thus your conclusion, which is that the transition to solution phase is evident at 0.2 fraction MA seems highly unlikely.

pg 9, first sentence. Here you say that there is no hysteresis between the deliquescence and efflorescence (0.25 < fMA< .9). as shown in Figure 3A and 4A. If so, why not show the deliquescence and efflorescence in the same figure, so this would be more obvious? I note that the integrated area of the water band is actually rather different in the deliquescence and efflorescence experiments. For example, for 0.4 fMA at 45

pg 11, last para. Here you mention the longer residence time in the EDB as a possible cause of the differences in efflorescence RH. Did you perform any residence time studies in the AFT in order to confirm that this may present a problem? Unfortunately, if you cannot operation under conditions where you have determined that the residence time is sufficiently long, you are not, in fact, measuring the efflorescence RH and results should not be reported as such.

pg.12, para 3. A rather lengthy description of the work by Brooks et al. is presented, though the Brooks study was conducted using a completely different organic compound. I see no problem with this in general. However, you attempt to use details of your observations to explain their results,... However, this may be due to the partial recrystallization of the ammonium sulfate leaving the maleic acid behind, which would be more consistent with the results we report here.... Given that the two studies are based on different compounds, I recommend removing any reference to MORE or LESS CONSISTENT results.

Overall, this work is a useful and carefully detailed contribution to our understanding of
the phase of atmospheric aerosols and I recommend it for publication.