

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 #1

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General Comments:

The study under review investigates the deliquescence and efflorescence properties of internally mixed malonic acid / ammonium sulfate (MA-AS) particles. Several papers on the hygroscopic growth and phase transitions of dicarboxylic acids / ammonium sulfate mixtures have already been published, five treating specifically MA - AS [Choi and Chan, 2002, Brooks et al., 2002, Hameri et al., 2002, Wise et al. 2003, Prenni et al, 2003], one treating ammonium sulfate and malonic acid in a more complex mixture [Marcolli et al., 2004]. Nevertheless, the present study gives new insights into the phase transitions of MA-AS particles: (1) it investigates the whole range of MA-AS compositions; (2) it uses IR spectroscopy to observe the physical state of the compo-

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nents in the mixtures. The presentation of the results is clear. The discussion of the results needs improvement in some instances (see specific comments).

Specific comments:

Introduction, paragraphs 2 & 3 and also later in text: ...binary AS-dicarboxylic acid mixtures at 3 compositions... and ...detailed case studies of a three-component aerosol...: it is not clear to me how the authors use the terms binary and ternary. In one case, they seem to treat water as a component, in the other, they do not. They should make the manuscript more consistent in this respect.

Introduction, paragraph 3: the authors should also mention the works by Wise et al. [2003] and Marcolli et al. [2004].

Section 3.2, paragraph 1: ...the lowest RH...can be calculated and measured...: the authors should specify what kind of calculation they mean.

Section 3.2, paragraph 1: the authors should specify how they predicted EDRH = 73 % for the AS-MA system. They should discuss the accuracy of the prediction. They should also give the measured DRH values for pure AS and MA at 283 K (indicated by the arrows 1 and 2 in Figure 3A).

Section 3.2.2, Figure 3B: Between EDRH and the RH of full deliquescence, crystalline and dissolved AS should coexist in the same particle, giving rise to two ammonium peaks at 1422 cm^{-1} and 1455 cm^{-1} . However, according to Figure 3B, a continuous shift of the ammonium band is observed. Is this a problem of resolution? The authors should comment on this.

Section 4.1: For mixtures of miscible components, deliquescence starts at the EDRH and ends at a higher RH. The results should be discussed in this respect. Specifically, the second sentence of paragraph 1 should be reformulated. Full deliquescence should occur at the EDRH only for the eutonic composition. The authors should try to indicate also the completion of deliquescence as Choi and Chan [2002] have done.

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The sentence In particular it only requires a very low level of AS or MA to shift deliquescence to the EDRH... should be reformulated to: ...to shift the starting point of deliquescence to the EDRH. But this is true for any system.

Section 4.1, paragraph 2, Figure 5: The first hypothesis can not explain the observations: if crystalline AS was present, it should not go into solution below EDRH, which is at 73 % according to the manuscript and between 70.9 and 71.7 % RH according to Brooks et al. [2002]. The Figure caption saying AS observed to go into solution is misleading: In the text it is assumed that AS remains at least partly liquid.

Section 4.2, paragraph 2: The authors should specify the EDRH for the AS-MA system at 303 K and give a reference. It seems unlikely that it is close to the DRH of AS, as stated by the authors.

Section 4.2, paragraph 3: The water uptake starting from 20 % RH described by Brooks et al. [2003] was observed for maleic acid alone. The explanation given by Braban and Abbatt is therefore not valid.

Technical corrections:

Title: The authors should write ...ammonium sulfate - malonic acid aerosol particles, since mixed ammonium sulfate - malonic acid aerosols do not necessarily imply internally mixed particles.

Page 2958, line 14: indicating instead of indication.

Page 2958, line 11: Add particles: ...aerosol particles.

References:

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