

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

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Received and published: 12 July 2004

Specific comments: pg 4 and fig 1. 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 NH_4^+ 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 NH_4^+ 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.

The changes do occur at $f_{MA} > 0.25$, though water uptake at all relative humidities is observed at lower f_{MA} 's. So the point is well taken and the text will be changed.

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Because the ammonium modes remain unchanged up to this MA fraction, it is not necessary to include the additional spectrum at 0.2 fraction MA.

pg 9, first sentence. Here you say that there is no hysteresis between the deliquescence and efflorescence ($0.25 < f_{MA} < .9$). 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 f_{MA} at 45

For clarity the efflorescence and deliquescence experiments were shown on separate figures. The integrated areas are different for the experiments presented in Figures 2 and 3 because they were normalized at different relative humidities so that comparison between the different compositions in that particular mode of experiment could be facilitated.

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.

In a flowing mode, the residence times can be varied only over relatively small timescales, in the order of tens of seconds, in a flow tube of this type. However, we do feel that our measurements of the "efflorescence" RHs are as valid as any others in the literature, given that all experimental techniques have a finite observation time. (What residence time is "sufficiently" long?) Assuming homogeneous nucleation, the measured efflorescence point is determined by both the observation time and the volume of the particles being observed. In flow tube experiments of this type, the observation time is relatively short compared to atmospheric residence times. However, the particle

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volumes are many orders of magnitude smaller than those used in other experiments used to observe efflorescence, such as those in electrodynamic balance and optical microscopy of mounted-particle experiments.

We also note that in the past, a flow tube of this type was operated in static mode. In particular, ammonium sulfate particles at RHs just a few percent higher than the ERH were observed to not effloresce over timescales of up to 30 minutes, after the flow was abruptly shut off and the remaining particles then observed. (Cziczo et al., 1997) This is a validation, to some degree, of using flow tubes to measure ERHs, at least for the ammonium sulfate system.

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.

The sentence: "however..." will be removed, and less emphasis given to the comparison between the two studies will be given in the final manuscript.

References

Cziczo, D.J., J.B. Nowak, J.H. Hu, and J.P.D. Abbatt, *J. Geophys. Res.*, 102, 18843-18850, 1997.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 2949, 2004.

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