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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Hygroscopic behavior of aerosols generated from solutions of 3-methyl-1,2,3-butanetricarboxylic acid, its sodium salts, and its mixtures with NaCl" *by* Li Wu et al.

Anonymous Referee #2

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In this work, the authors investigated the hygroscopicity of laboratory-generated, micrometer-sized, pure MBTCA and its salts using in-situ Raman microspectrometry (RMS). The authors have clearly demonstrated how interactions between the MBTCA and NaCl could modify the aerosol composition and subsequent hygroscopic behaviors during the hygroscopic measurement. The paper is generally well written. I have minor comments and suggestions for authors' consideration before the paper is accepted for publication.

Comments Line 115, "he particles on the Si wafer were exposed to a hygroscopic measurement cycle, where they experienced dehydration process first (by decreasing

RH from ∼95 to ∼1%), followed by a humidification process (by increasing RH from ∼1 to ∼95%). Can the authors discuss whether an equilibrium state was achieved in their measurements?

Line 139, "Mono-/di-/tri-sodium MBTCA salt solutions were obtained by mixing MBTCA and NaOH (>99.9% purity, Sigma-Aldrich) with molar ratios of MBTCA:NaOH = 1:1, 1:2, and 1:3, respectively." What are the pHs of these aerosols?

Line 251, "The different behavior of the MBTCA particles can be attributed to different nucleation mechanisms, i.e., homogeneous and heterogeneous nucleation, for pure and impure (seed-containing) MBTCA particles, respectively. A similar situation was reported for NH4NO3, NaNO3, and NH4HSO4 particles (Lightstone et al., 2000; Hoffman et al., 2004; Gibson et al., 2006; Kim et al., 2012; Jing et al., 2018; Sun et al., 2018; Wu et al., 2019b). The Si substrates used in this study could also facilitate efflorescence (Eom et al., 2014; Wang et al., 2017)." While the authors have provided possible explanations for the different hygroscopic behaviours of MBTCA particles, it would be important to provide more detailed explanation for each hygroscopic behavior. For example, why did type 1 and type 2 aerosols effloresce at different RH? Any reason why type 3 aerosols were being observed in this work?

Line 284, "NaH2M and Na2HM particles still showed the same shapes and Raman spectra with those at RHs = 3.4% and 2.8%, 285 respectively. These results indicate the non-crystallizable properties and supersaturated amorphous phase state of the particles. The Na3M particles behaved differently as they did not crystallize during the dehydration process. On the other hand, the aerosols exhibited efflorescence at RH = 46.8% during the humidification process (Fig. 3(c)), deliquesced to become a droplet at RH = 53.1%, and grew continuously after that with increasing RH." As mentioned above, can the authors comment:Could the crystallisation of Na3M aerosols attribute to the presence of impurities, the use of Si substrates or other factors? Could the authors rule out these possibilities in their measurements? Can the authors also comment: What is the water uptake (e.g. aerosol water content) of mono-/di-/tri-sodium MBTCA

salt aerosols? Are they different?

Figure 4, unlike the pure MBTCA and mono-/di-/tri-sodium MBTCA salt aerosols, why the dehydration and humidification curves of MBTCA:NaCl = 1:1 and 2:1 do not overlap with each other?

Figure 5, why the dehydration and humidification curves of MBTCA:NaCl = 1:2 and 1:3 do not overlap with each other after deliquescence?

In 3.3.3 Chemical reactivity of aerosols generated from MBTCA−NaCl mixture solutions, the authors have provided a nice discussion of how the chemical composition would evolve due to the chemical reactions between MBTCA and NaCl during the experiments. My question is: do the extent of the reactions depends on the experimental time? Would the duration of the experiments would significantly affect the hygroscopic behaviors observed for different systems in the experiments? Since the chemical compositions evolves over time, can the authors discuss whether an equilibrium state was achieved in their hygroscopic measurements?

Line 472, “Two types of hygroscopic behavior of pure MBTCA particles were observed, corresponding closely to types 1 and 3 aerosol particles in the see-through impactor system.” Can the authors further elaborate why type 1 aerosol is observed in levitated pure MBTCA aerosols?

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