

Interactive comment on “Measurements of the timescales for the mass transfer of water in glassy aerosol at low relative humidity and ambient temperature” by H.-J. Tong et al.

Anonymous Referee #2

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This is an excellent experimental study of the response times of micron-sized organic/water particles to changing conditions of relative humidity. The issue is that if the viscosity of such particles is high, as with a glass, then the response times will be long, and the particles will be at disequilibrium with the ambient relative humidity. While the paper is mostly a study of a single component (sucrose) system likely to not be found in the atmosphere, there is direct relevance to the atmospheric community given that such glassy particles may be important for ice nucleation in the atmosphere and potentially for the rates of heterogeneous chemistry as well. The experiments appear to have been done exceedingly carefully by scientists who have a reputation for such work. In particular, optical tweezers are used to constrain the particles for

C797

very long periods of time, allowing for spectroscopic interrogation of particle size and composition. I recommend the paper be published in ACP but I suggest the authors address the following topics in the revised paper.

One suggestion is that a little bit more relevance to the atmosphere and past studies be presented. ACP is an atmospheric journal, but the authors are studying very highly concentrated sucrose particles. Comments of how such particles and those actually present in the atmosphere would be valuable. Do we expect such kinetic limitations at room T in the atmosphere, or only at low T where viscosities may be high? In a related manner, many particles much closer in composition to those in the atmosphere have been studied for their response to changes in RH by tandem DMA, electrodynamic balance and FTIR flow tube techniques (e.g. work of Hameri, Tolbert, Abbatt, Martin, Chan, Kreidenweis, to name just a few groups). These workers have usually observed pretty rapid response of the water content to changing RH, even in the TDMA and FTIR flow tube experiments where the timescales are seconds. It would be valuable to place the present work into the context of the systems for which such slow kinetics are clearly NOT operable. Did such prior studies not observe disequilibrium with the gas phase because they were mostly at room T? Or, is the choice of solutes? Also, what do prior studies that have looked at sulphuric acid at low temperature imply? Is the viscosity of such solutions not so high to lead to these disequilibrium effects?

I would also suggest that the authors give a little bit more thought to surface crystallization and very low mass accommodation coefficients as explanations for the observed behaviour. Personally, I tend to think that the viscosity argument is most valid, but the other two effects have not been ruled out.

For the most part the paper is very well written, however I had trouble following the some of the discussion of the results on pages 4855 and 4856, sometimes not even sure if the correct figure was being referred to. For example, it would help a lot if the figures plotted as a function of time (NOTE: The text uses hours but the figures are in second – please use just one set of units) had also plotted the RH on the upper

C798

x-axis. Could the particle size also be plotted simultaneously? What do spectra 3, 4 and 5 refer to? Can the spectra be labelled? I found the sentences at the top of page 4856 concerning sphericity to be confusing and contradictory, i.e. at one point it is said the particle is spherical but one sentence later on it says that there is a loss of sphericity???

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 4843, 2011.

C799