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Atmospheric Chemistry and Physics Discussions

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.

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Response to the comments by Benjamin Murray on Atmos. Chem. Phys. Discuss., 11, 4843-4879, 2011

We thank Dr. Murray for taking the time to review our manuscript so carefully. We respond to each of his comments below and have made alterations to the manuscript as indicated.

Scientific Comments: (1) "In the discussion of why it is important to understand the response of aerosol to temperature and RH the role of aerosols in cloud formation is omitted." While our list of the influences of dynamical factors on aerosol properties

was not intended to be exhaustive, the current manuscript could be quite important for aerosol-cloud interactions and this has now been explicitly included, along with the references recommended by the reviewer.

(2) "...can you rule out the presence of one or more crystalline hydrates?" We cannot rule out the presence of crystalline hydrates based on the Raman spectra, but we suggest that the subsequent data and analysis is only consistent with the formation of a glassy state. This has now been noted in the text: "The formation of crystalline hydrates cannot be ruled out at this stage, but the data presented below are consistent with the formation of a glassy state".

(3) "The agreement between model and measurement isn't very good above $\sim$70 % either." We consider that the level of agreement between the model and measurements is unambiguously and systematically worse at the low RH limit than at the high RH limit for the measurements reported in Figure 6. At an RH <10 %, the deviation between measured and modelled growth factors is systematically high by between 0.005 and 0.01 for all 13 measurements, over which range the modelled GF is only $\sim$0.02 above 1. At RHs > 60 %, only 1 point in 6 approaches the level of error of 0.01 in GF; the growth factor is $\sim$0.175 above 1 at this high RH limit. Given the uncertainties in making water activity measurements, we consider that this level of agreement at high RH is satisfactory.

(4) "...the timescale for devitrification is 10-100 s and should be discussed." We agree with the observation made by Murray that the timescale may indeed be important when considering ice nucleation. We have added emphasis to this statement, highlighting the apparent rapidity of the change when the glass transition is crossed from below, but would prefer to not draw too many conclusions from this one set of measurements. We have added the sentence: "This suggests that the ultraslow diffusion of water in the glass may not be the kinetic parameter limiting particle size change once water is able to adsorb to the particle surface and dissolution of the glassy core can proceed towards an equilibrium solution state."

(5) “This discussion is confusing”. As stated in our discussion of the comparison between the experimental measurements and the model, we do conclude that the model is largely successful in reproducing the mass transfer kinetics for the glassy aerosol, reproducing a change in timescale of more than 3 orders of magnitude. However, the three measurements discussed on page 4859-4860 show a size change that is systematically slower than the model, suggesting that the diffusion constant of Zobrist et al. used in the model may still be too large. We have added a statement of further clarification: “This requires further investigation to assess the accuracy of the value of the diffusion constant that has been reported by Zobrist et al. (2011) and used in the simulation of the aerosol optical tweezers measurements.”

(6) “Figure 8. There appears to be a slight discontinuity at $\sim$5000 s,” We do not feel confident in making any assertions from this one experimental measurement, although Dr. Murray’s recognition of the discontinuity may be significant.

(7) In recognition of the referee’s comment, we have added the following statement to the abstract: “When increasing the RH from below to above the glass transition, a particle can return to equilibrium with the gas phase on a timescale of 10’s to 100’s of seconds, once again forming a solution droplet. This is considerably shorter than the timescale for the size change of the particle when glassy and suggests that the dissolution of the glassy core can proceed rapidly at least at room temperature.”

Technical Comments:

(1) We have replaced ‘we’ by ‘Zobrist et al.’

(2) Corrected.

(3) Symbol removed.

(4) There should be no reference to numbered spectra and the text has been corrected to read ‘. . .enhanced Raman scattering in the spectra when the particle is illuminated confirms that the . . .’.

(5) ‘Liquid’ added as suggested.

(6) Figure 4(a) has been broken into 4(a) and 4(b) as suggested. The measured size is unfortunately necessarily sporadic at early times during the rapid change in RH. A comment has been added to the caption to explain this: “The rapidly changing size cannot always be determined from measurements at early time following the sudden decrease in RH due to the rapid translation in WGM wavelengths. This gives rise to the broken time-record of measured size at times of $\sim$10000 s.” Arrows are also included in (c) to indicate which spectrum arises from which image plane.

(7) ‘balance sucrose’ deleted.

(8) The water activity is specified as 0.4 in the first half of the sentence.

(9) We have now stated in the figure caption: “The grey shaded area indicates the RH region which is considered to lie below the glass transition.”

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