

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.

H.-J. Tong et al.

j.p.reid@bristol.ac.uk

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Response to the comments by Anonymous Referee 3 on Atmos. Chem. Phys. Discuss., 11, 4843–4879, 2011

We thank the anonymous referee for taking their valuable time to review our manuscript and to make constructive comments. We have addressed the comments in our revised manuscript as specifically outlined below.

1. We have clarified what we mean by environmental conditions in the abstract, specifically stating RH.

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2. We have improved the clarity of the figure captions, but given the number of data sets on each figure, suggest that including legends in the figures would make them too crowded.
3. For the raffinose data set in Figure 1(a), we have increased the line thickness so that it is more apparent.
4. “Figure 2b: In the figure caption it says “and the present data” – does this refer to data obtained in this work – and what is the symbol for these?”. Yes, the estimate of the MFS at the glass transition from this work is shown by the black symbols, with two relationships between the water activity and MFS used to estimate two values. The caption for the figure has been reworded to read: “For the present study, the composition at the glass transition has been estimated from the water activity and either the Norrish water activity/MFS relationship (black star) or the Starzak and Peacock relationship (black triangle).”
5. This text should refer to the volume fraction mixing rule and has been amended accordingly.
6. We have now consistently used seconds throughout the text as the primary unit of time, occasionally also including the time in hours for clarity.
7. We have now stated the glass transition RH and temperature of the measurement at the beginning of Section 4 to improve clarity.
8. As defined in the text, the Zobrist treatment refers to the predicted sizes of the equilibrium state (as for Norrish and ADDEM) and the words ‘predicted at equilibrium’ appear wherever this is used to avoid ambiguity. The deviations from the Zobrist predictions are due to the kinetic limitations imposed on size equilibration by the bulk viscosity increase and the glassy state.
9. “Page 4853: It says that the Norrish and Zobrist models are the most accurate – the Starzak and Pecoock (dashed blue line in figure 2a) seems to be doing as good or better

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that the Zobrist model?” It is true that the Starzak and Peakcock treatment for the water activity/MFS relationship is more accurate than the Zobrist treatment and we have amended the wording of the text to correct this. However, the kinetic model has been benchmarked for the Zobrist thermodynamic parameterisation of water activity. With this in mind, the measurements are compared with the most accurate thermodynamic treatment (Norrish) and the treatment used in the kinetic model. At the end of section 3.1, we have added the clarification: “The Norrish treatment is chosen as it provides the most accurate reflection of the solution thermodynamics. The Zobrist treatment is chosen as the second model for comparison with the experimental data as it provides the underpinning treatment of solution thermodynamics for the kinetic model described below.”

10. We have improved the clarity of Figure 4(a) by separating out into two parts. The experimental data at early time following the sudden decrease in RH is sparse due to the rapidly changing droplet size and translation of the WGMs. We have included a comment to this effect in the caption for Figure 4(a) and do not consider that showing an expanded view of early time will improve the clarity of the figure.

11. We have sought to improve the discussion and clarity of Figure 4 in response to comments made by two of the other referees and detailed in our response to them. We hope that the revised discussion and Figure addresses the referee’s concerns.

12. We have indicated the glass transition on the figure as suggested by the referee.

13. On page 4857, we have removed the ambiguities in the text as suggested by the referee.

14. In response to point (2) made by referee 2, we have expanded our discussion of surface and bulk accommodation at the end of the Conclusions and hope that this has addressed the concerns of the referee regarding the definition of bulk accommodation.

15. In response to point (1) made by referee 2, we have addressed the atmospheric

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implications in greater detail in the Conclusions. We do however feel that it is necessary to avoid drawing too substantive conclusions about real atmospheric aerosol until further measurements have been made.

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

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