**Editor Initial Decision: Publish subject to technical corrections** (01 Mar 2014) by Dr. David Topping
Comments to the Author:

Hi guys.

Thank you for responding to the reviews. Some minor points to quickly discuss with you all.

1) Regarding the response to the use of distinct timescales (referee #1), specifically diffusion half-lives, i think it would act as a great point of reference for the reader to briefly state how you know this is not the most appropriate measure. Given the number of studies on condensed phase diffusion now, we need more anchors for correct terminology. Please feel free to reference your future work in this and some broader recommendations for the community if you feel appropriate. This should also be represented in your summary

We have added a discussion of timescales to section 3.2, which now reads:

“The characteristic half-time for diffusion into a spherical particle of radius $r$ at constant water activity is given by (Seinfeld and Pandis, 2006)

$$\tau_{1/2} = \frac{r^2}{\pi^2 D \cdot \ln(2)} \tag{5}$$

Note that this is the diffusion timescale for water diffusing into a droplet at constant water activity and therefore *D* does not change. It may be of interest to know the timescales over which a droplet would respond to changes in RH under certain atmospheric circumstances. This is not a simple calculation due to the plasticising effect of water in these aqueous solutions – the diffusion coefficient is a function of concentration and would vary radially as a droplet increased or decreased in size. In order to model the response of a droplet to a change in humidity a model of the time response of a droplet undergoing water uptake and loss would be required. This is the subject of ongoing work and beyond the scope of this paper. In this study, we use $\tau_{1/2}$ to give basic information about the rate at which water molecules diffuse within a droplet at a given relative humidity.

Fig. 8(a) shows these timescales for water diffusing into sucrose, levoglucosan and raffinose/M5AS aqueous spherical droplets of radii between 100 nm and 1 mm at 20% RH. For levoglucosan and raffinose/M5AS, water diffusion timescales are faster than 1 s in particles smaller than 250 nm in radius at room temperature. For sucrose these timescales are on the order of minutes to hours. Under these conditions the calorimetry data presented by Zobrist et al. (2008) suggest that aqueous sucrose exists in a glassy state. As Zobrist et al. (2011) point out, even though particles may exist in what is by definition a glassy state, water diffusion is not necessarily arrested. Nevertheless, the kinetic limitation could be important for growth measurements in aerosol instruments which operate on short timescales, such as the HTDMA (Hersey et al., 2013). In addition, diffusion coefficients decrease with temperature and thus under common atmospheric conditions these timescales are expected to be far greater.

Water diffusion coefficients measured in this study have been used to calculate water diffusion timescales as a function of RH in droplet of 100 nm radius, according to Eq. (5); these are plotted as red dots in Fig. 8(b). These timescales have also been calculated using the water diffusion coefficients given in Zobrist et al. (2011) for comparison. To illustrate the magnitude of error that would be encountered when estimating these water diffusion timescales from a viscosity measurement, the Stokes-Einstein relation was used with the viscosity measurements of Power et al.

(2013) to estimate *D* in Eq. (5). The resulting timescales are shown as black crosses. There is a discrepancy of more than 5 orders of magnitude between the two datasets at ~30% RH and ~2 orders at 62% RH. Again, this demonstrates the importance of quantifying diffusion in atmospheric aerosol."

We have also adjusted the second paragraph of the summary as follows:

"Using these measured water diffusion coefficients we predict diffusion timescales for aerosol-sized particles (~100 nm) at room temperature under constant RH conditions, and show that just below the glass transition it takes minutes before equilibrium with water vapour is achieved. This is a vastly shorter time scale than one would expect based on the Stokes-Einstein equation, which has been used to relate diffusion to viscosity in the past. Our results reinforce previous studies showing that the Stokes-Einstein relationship is inappropriate in the regime of the glass transition (Champion et al., 1997; Rampp et al., 2000; Power et al., 2013). Even though the diffusion timescale is much shorter than predicted by the Stokes-Einstein equation, it is still long in comparison to the timescale needed when using thermodenuders and HTDMAs (seconds). Hence, some caution must be applied to the interpretation of results from measurements. In addition, we show for levoglucosan that a high bounce fraction does not necessarily mean long water diffusion timescales."

2) Following the suggestion by referee #1 i would also suggest placing the additional data of Zhu etc on figure 7. Based on your response, it seems that it might be surprising that the data agrees well even though the molecular structures differ. I think this in itself is interesting. A small note based on your response could be added to the paper to this effect.

The diffusion data of Zhu et al. is presented as a function of solute weight percent. As far as we are aware, there is no relationship available in the literature between aqueous maltose water activity and solute weight percent. The only way to compare our diffusion coefficients (measured at known water activities) and those of Zhu et al. in maltose would therefore be to use literature parameterisations to convert from water activity to weight percent, such as those given by Zobrist et al. (2008). We feel that presenting our data in terms of weight percent would make it less easy to relate to the atmosphere. It would also introduce an extra source of error given that it was water activity, rather than weight percent, which we measured. Without the inclusion of an extra figure, the comparison between our data and that of Zhu's is unfortunately not possible.

3) Please do make all fonts larger on all figures.

The fonts on the figures will be of an appropriate size when we upload them for ACP formatting.

Thanks.