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Interactive Comment

Interactive comment on "Determination of the evaporation coefficient of D_2O" *by* W. S. Drisdell et al.

Anonymous Referee #1

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General comments.

Over the years there have been about 50 studies of the process governing water mass accommodation (or evaporation). Still, the value of the mass accommodation (or evaporation) coefficient for gas phase water on liquid water remains unresolved. (The situation is summarized in a 2006 review article Davidovits et al., 2006). To illustrate the main issue: Relatively recent measurements of Li et al. (2001) (Boston College/Aerodyne Group) yield mass accommodation values that are significantly less than unity and display a negative temperature dependence. The experiments of Winkler et al. 2004 (Vienna/Helsinki Group) yield a value of unity independent of temperature. The two groups discuss these results in a joint publication (Davidovits et al. 2004). (The Davidovits et al. (2004, 2006) references are shown at the end of this review. All other references are in the manuscript under review.)

The Boston College/Aerodyne (BC/ARI) group and the Vienna/Helsinki (V/H) group utilized very different methods of measuring mass accommodation. The BC/ARI group measured gas uptake by a train of droplets while the V/H group measured droplet growth from a seed aerosol. Even though it is now evident that the mass accommodation coefficient is sufficiently large not to limit cloud droplet growth, the process is sufficiently important to pursue and clarify it. Certainly new data that could help resolve the outstanding issues would be very useful.

The published work since then has not clarified the picture. Some groups have measured mass accommodation coefficients < 1 (Cappa et al., 2005; Smith et al., 2006; Jakubczyk et al. 2007; Zhang and Leu, 2008, submitted manuscript), while the V/H group re-measured alpha and again found it to be close to 1 (Winkler et al. 2006) and Voigtlander et al., 2007 measured alpha, with average value ˜1 but with error bars extending from 0.3 to 7.

The work described in the current manuscript utilizes the technique that was used by Smith et al. to measure the evaporation coefficient for H2O. That is, the evaporation coefficient is extracted by modeling the cooling rate of droplets in a moving droplet train. The evaporation coefficient for D2O is measured to be 0.57. The method of measurement is novel and interesting. This work could be an important step toward resolving the conflicting results. However, in my opinion, several key issues must be discussed and clarified for the manuscript to be appropriate for publication.

Specific comments.

The precision of the measurements is high but the manuscript does not contain a discussion of accuracy. In my view the greatest uncertainty is introduced by the modeling procedures that yield the evaporation coefficient. While some of the assumptions entailed in the modeling are stated, others are not, and the assumptions are scattered rather than clearly listed. The usefulness of the present manuscript could be raised

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significantly if the authors discussed the effect of the various assumptions that have gone into the formulation of the droplet growth model that connects measurement to the theoretical fitting function. Winkler et al. (2006) do this rather well in section 5 of their publication. The authors might follow their approach. In this connection at least the following should be discussed:

1. What is the effect on the data of the conditions near the droplet exit from the orifice where the vapor pressure is relatively high?

2. What is the effect of the droplet velocity?

3. Is there a vapor sheet that is entrained by the droplet train?

4. The Drisdell et al. experiments are done completely off equilibrium. The surface is likely to be significantly perturbed by the rapid evaporation without the balancing effect of condensation. Under these conditions the water surface is likely to be very different from the surface under near equilibrium conditions found in nature. Could this affect the evaporation coefficient? This is a particularly important point because the near equilibrium case is the one that is relevant for the atmosphere. Therefore, determining if there is a difference is important for climate and meteorological models.

I have in front of me two other articles written by the authors of the current manuscript. They are: Cappa et al., (2005) and Smith et al., (2006). They both measure the evaporation coefficient of H_2O. There are significant discrepancies between the Cappa et al. article on one hand and the Smith et al. (2006) and the current Drisdell et al. (2008) articles on the other hand. The differences between these articles have gone unanswered and this can be very confusing. The present manuscript could serve to clarify or at least point out these differences so that a reader of the articles is not left in a state of puzzlement.

The Conclusion of the Cappa et al (2005) article was that quote: energetic barrier to evaporation exists and consequently, that the evaporation coefficient decreases with

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temperature. Yet in the following Smith et al. (2006) publication and in the Drisdell et al. manuscript under review, the evaporation coefficient is essentially temperature independent. I know that the experiments of Cappa et al. were not identical to those of the two subsequent studies but the modeling procedures were similar. The discrepancy between these two sets of experiments needs to be clarified.

In the Cappa et al. 2005 article it is stated that quote: We find that evaporation coefficient and MA (mass accommodation coefficient) are not equivalent but that evaporation coefficient is proportional (1 minus MA). This is not correct. In the second article and in the current manuscript the relationship is stated correctly i. e. evaporation coefficient = MA. However, the wrong statement in Cappa et al. (2005) is not corrected in either manuscript. To reduce confusion, a simple parenthetical statement would be helpful such as: The relationship between evaporation coefficient and MA given in Cappa et al. (2005) is incorrect. I would be very happy to see this manuscript published if the issues raised were appropriately answered.

References not included in the manuscript: Davidovits, P., D.R. Worsnop, J.T. Jayne, C.E. Kolb, P. Winkler, A. Vrtala, P. E. Wagner, M. Kulmala, K.E.J. Lehtinen, T. Vesala, M. Mozurkewich, Geophys. Res. Lett. 31, 2004, L2211, doi:1029/2004GL020835.

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