**Atmospheric
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Comment

Interactive comment on "Determination of the evaporation coefficient of D$_2$O" *by* W. S. Drisdell et al.

W. S. Drisdell et al.

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We thank the reviewer for his/her support for publication of this manuscript in ACP and thorough review, including repeating and expanding on our numerical calculations. We also appreciate the reviewer's perspective on the relationship of our work to other recent experiments. In particular, we value the reviewer's statement that our experiments will "lead to a deeper understanding of evaporation in kinetic regimes and effects at interfaces."

The reviewer raised five specific comments and noted a number of technical corrections required. All of these will be addressed in a revised manuscript as described below.

Reviewer Comment 1: Am I correct that the cubical vacuum chamber walls were at

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room temperature?I tried to account for the radiative energy transfer (up to 40 K temperature difference!) and found it negligible, am I correct?

Response: The cubical vacuum chamber walls are indeed at room temperature, but the reviewer is correct in that the radiative heating from the chamber walls is negligible in the experiment.

Reviewer Comment 2: Droplets emerging from VOAG most probably exhibit shape oscillations (also high multipolarity capillary waves) which might lead to a whole family of phenomena significantly influencing evaporation (surface area change, saturation pressure change, cavitation). I checked that these oscillations decay within a few μs which is negligible in the experimental timescale. Similarly I believe the circulation of liquid in the droplet is of no importance. Is fast rotation of the droplets emerging from VOAG possible? In vacuum they would not be damped.

Response: We also believe that shape oscillations decay rapidly and that they are complete prior to the location of our first measurement. Fast rotation of the droplets is unlikely given that the droplets are formed due to coupling with capillary waves; rotation could only result if the capillary waves were asymmetric. Some low-speed rotation is possible but it is unlikely that rotation would be fast enough to affect the measurements (due to Doppler shifts in the Raman scatter, or other such effects).

Reviewer Comment 3: The droplet radius change was accounted for by the authors (page 8572, line6). I understand that it also covers the "moving boundary effect" for internal heat transfer (compare (Fukuta et al., 2007)).

The droplet radius change that we include in our model should indeed account for the "moving boundary" effect discussed by Fukuta and Myers (Fukuta and Myers, 2007). In our model we assume a stationary boundary for the duration of each time step (10^{-10} s) and then re-size the droplet and the concentric shells based on the mass lost due to evaporation. Experimentation with the model showed that the time step used is sufficiently small that slight changes in the step size do not affect the model results. We

thank the reviewer for pointing out our oversight in not discussing the work by Fukuta and Myers in the context of our experiment.

Reviewer Comment 4: A few words about droplet interaction with light field in the laser beam focus might be instructive.

Response: We agree. The laser power is sufficiently low that it does not induce heating or otherwise affect the evaporation.

Reviewer Comment 5: And finally a more general remark. Since this experiment concerned rather vacuum-liquid than gas-liquid interface and additionally an interface off-balance, it would be interesting to address the differences. I find it most promising direction of investigation.

Response: This issue was also raised by reviewer #1. As discussed in more detail in our response to that reviewer (and as we will address in our revision), we do not believe that there is any difference in evaporation kinetics for a vacuum interface or an interface at ambient pressure and RH.

The reviewer also notes a technical correction, stating: "On page 8567, line 14, m is denominated as 'molecular mass'. In view of the formulas used, it might be somewhat misleading. If it is, as I think, the mass of a molecule, then in formulas (6) and (7) $\sqrt{m}$ should be in the numerator. This mistype seems to have propagated from the authors' previous works."

The reviewer is correct in that m is indeed the mass of an evaporating molecule. However, we believe that equations (6) and (7) are correct as written. Equation (6) is derived from equation (3) in the following manner:

Equation (3) is the Hertz-Knudsen equation, expressing the observed evaporation rate as formulated from kinetic gas theory and discussed in the manuscript. It is reproduced here:

$$J_{e.obs} = \gamma_e J_{e.\max} = \frac{\gamma_e p_{sat}}{\sqrt{2\pi m k T}} \tag{1}$$

Here p_{sat} is the saturation vapor pressure, k is the Boltzmann constant, T is the temperature, γ_e is the evaporation coefficient and m is the molecular mass as discussed above. $J_{e,obs}$ has the units of (evaporating molecules * s^{-1} * m^{-2}). When formulating an expression for the change in temperature of the evaporating liquid as a function of time, we write

$$\frac{dT}{dt} = -J_{e,obs}A\frac{\Delta H}{C_pM_d}, \tag{2}$$

where A is the surface area of the evaporating droplet, ΔH is the enthalpy of vaporization, C_p is the specific heat per unit mass, and M_d is the mass of the evaporating liquid. This mass is smaller than the droplet mass since only the surface is being cooled by evaporation; therefore the equation only accounts for the temperature change of the surface of the droplet (i.e. the outermost shell in our evaporation model). In the manuscript, we listed C_p in units of (J * mol^{-1} * K^{-1}) rather than (J * kg^{-1} * K^{-1}), which are the units that should be used in this equation. This will be corrected in the revised manuscript.

Combining Eqn's 1 & 2 above we find:

$$\frac{dT}{dt} = -\gamma_e A\frac{p_{sat}}{\sqrt{2\pi m k T}}\frac{\Delta H_{vap}}{C_pM_d}, \tag{3}$$

which is Equation (6) in the manuscript. The $\sqrt{m}$ term follows directly from substitution of $J_{e,obs}$. We understand that the two mass terms in this equation can easily cause confusion, so we will replace M$_d$ with (density * Volume of outermost shell) in the revised manuscript. This will also make the transition between Equation (6) and Equation (7) in the manuscript easier to follow.

Lastly, the reviewer suggests that a reference to the work by Zientara et al. (Zientara et al., 2008) be added to the manuscript. At the time of submission, this work had not yet been published. Our revised manuscript will include a reference to and discussion of this recent paper.

Fukuta, N., and Myers, M. N.: Simultaneous measurement of condensation and thermal accommodation coefficients for cloud droplet growth in due consideration of a new moving surface-boundary effect, Journal Of The Atmospheric Sciences, 64, 955-968, 2007.

Zientara, M., Jakubczyk, D., Kolwas, K., and Kolwas, M.: Temperature dependence of the evaporation coefficient of water in air and nitrogen under atmospheric pressure: Study in water droplets, J. Phys. Chem. A, 112, 5152-5158, 2008.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 8565, 2008.