S1: Theoretical estimation of mode number in an evaporating sessile droplet

The droplet volume during any stage of CCR mode of evaporation can be estimated as[1]

$$V_{CCR}(t) = \frac{\tau V_0 - 4r_c^3\delta t}{t+\tau} \qquad \text{(S1)}$$

Where $\tau = \frac{16\rho\delta r_c^5}{2DMC_s(1-RH)\left(4\delta r_c^3 + V_0\right)}$ and δ is a fitting parameter which is 1.994 for the present experiments, r_c is the contact radius, D = 2.54 x 10^{-5} m^2/s is the diffusion coefficient of water in air, ρ is the density of water, C_s is the saturated concentration of water vapour at 298 K, RH=0.5 is the relative humidity and M is the molar mass of water. The initial volume V_0 = 5.0 µl is adjusted to account for the delay between the start of oscillation and the deployment of the droplet.

The resonance frequency for a spherical droplet is[2]

$$f_{n,1} = \sqrt{\frac{n(n-1)(n+2)\gamma}{3\pi\rho V}} \qquad \text{(S2)}$$

Where n is the mode number (See Figure 3 of the manuscript), γ is the surface tension of water. Let us denote this as model 1. Taking square on both sides of equation 2 and rearranging the terms we get the cubic equation in n

$$n^3 + n^2 - 2n - k = 0 \qquad \text{(S3)}$$

Where $k = \frac{3\pi f_{n,1}^2\rho V}{\gamma}$ and V=V_{ccr}. Equation S3 is solved using the built-in function “roots” in Matlab. We consider the only real and positive root of the equation, since n is a positive integer by definition. The value of $f_{n,1}$ is chosen as 400 Hz, 700 Hz and 900 Hz.

Sharp et al calculate the resonance frequency of an oscillating sessile droplet as[3] explained in the manuscript

$$f_{n,2} = \sqrt{\frac{2\pi\gamma}{\rho}}\left(\frac{n-{}^{1}/_{2}}{2r\theta_c}\right)^{3/2} \qquad \text{(4)}$$

This is model 2 for the present discussion. The profile length l=$2r\theta_c$ is back calculated from the volume calculate using equation S1. Equation S4 is also a cubic equation in n and is solved in the same way as equation S3. The values of n obtained from both models 1 and 2 are plotted here.

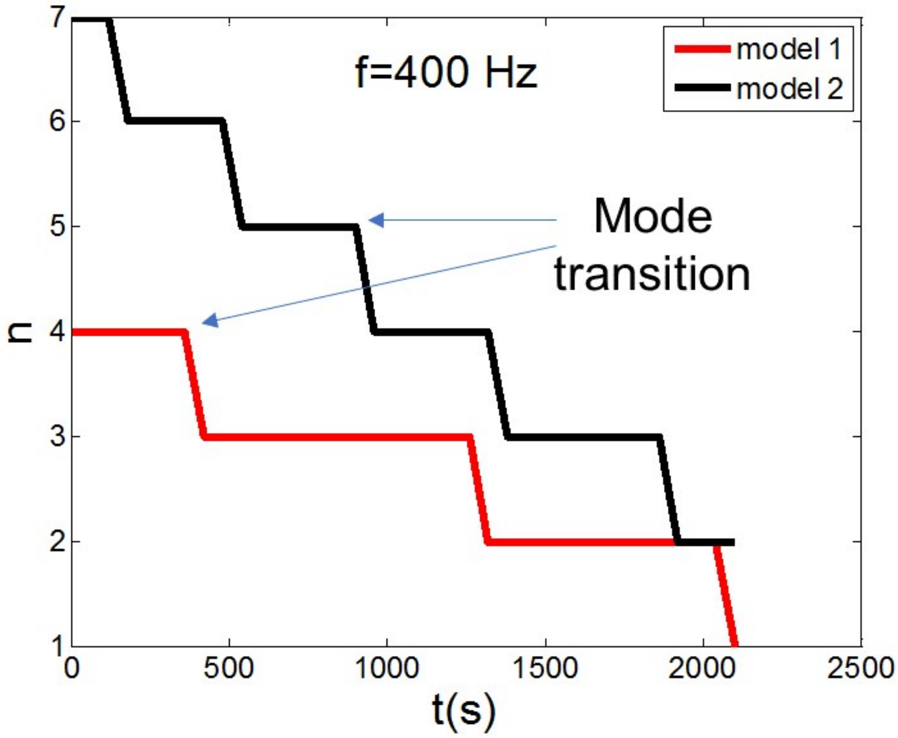

Figure S1: Comparison between models 1 and 2 in estimating the mode number of an oscillating sessile droplet

Model 1 correctly predicts the initial value of the mode number while model 2 overpredicts. Model 1 also correctly predicts the total number of mode transitions during the droplet lifetime. This exercise is repeated for both models at frequencies 700 Hz and 900 Hz. In both cases, the analytical form used in model 1 gives a better result.

S2: Correction factor for PDMS substrate

The method described in S1 is used to predict resonance in droplets on PDMS substrate. Equation 3 of the main manuscript is used to calculate the mass of the droplet undergoing CCA mode of evaporation on PDMS. Using equation S2, the predicted mode number is shown to be overpredicted by one at $f_{n,1}$=400 Hz (Figure S2). However, if equation S2 is modified as

$$f_{n,1} = \alpha\sqrt{\frac{n(n-1)(n+2)\gamma}{3\pi\rho V}} \qquad \text{(S5)}$$

Where α is a correction parameter. Both Sharp et al and Sanyal and Basu report α~0.8. Using this value, the mode number is calculated and shown in Figure S2. We have used this approach in calculating n for the droplet on the PDMS substrate.

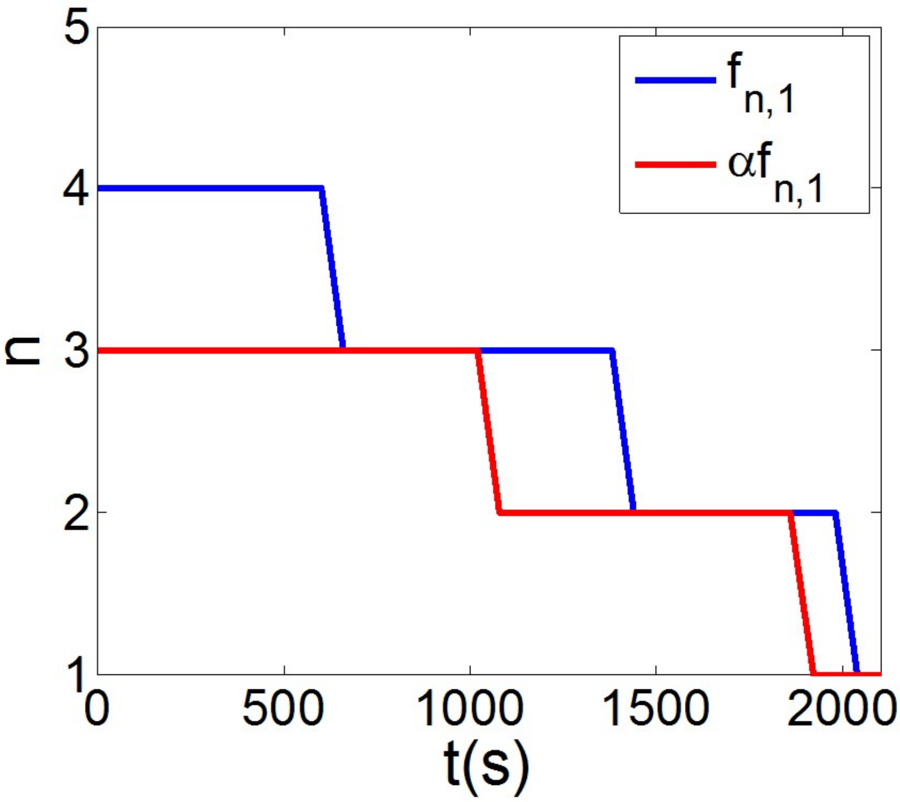

Figure S2: n estimated from $f_{n,1}$ (equation S3) and corrected $\alpha f_{n,1}$ (equation S5) where α=0.8.

References

(1) Schönfeld, F.; Graf, K.-H.; Hardt, S.; Butt, H.-J. Evaporation Dynamics of Sessile Liquid Drops in Still Air with Constant Contact Radius. *International Journal of Heat and Mass Transfer* **2008**, *51* (13–14), 3696–3699. https://doi.org/10.1016/j.ijheatmasstransfer.2007.12.027.

(2) Landau, L. D.; Lifshitz, E. M. *Fluid Mechanics: Landau and Lifshitz: Course of Theoretical Physics*; 1987. https://doi.org/10.1007/b138775.

(3) Sharp, J. S.; Farmer, D. J.; Kelly, J. Contact Angle Dependence of the Resonant Frequency of Sessile Water Droplets. *Langmuir* **2011**. https://doi.org/10.1021/la201984y.