Supporting Information:
Mechanisms of Airborne Infection via Evaporating and Sedimenting Droplets Produced by Speaking

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Section A: Diffusive droplet sedimentation without evaporation

The density distribution of droplets that diffuse in a viscous medium (such as air) under the influence of gravitational force is given by the diffusion equation

$$\frac{d}{dt}P(z,t) = D_R \frac{d^2}{dz^2}P(z,t) + V\frac{d}{dz}P(z,t) \tag{A1a}$$

where the stationary velocity is defined as

$$V = \frac{D_R mg}{k_B T} \tag{A1b}$$

and D_R is the droplet diffusion coefficient, m the droplet mass, g the gravitational acceleration, $k_B T$ the thermal energy and $P(z,t)$ is the density of droplets at height z and time t. The fact that droplets do not return to air once they reach the ground at height $z=0$ is accounted for by a vanishing density distribution at the ground, $P(z=0, t) = 0$, which is the absorbing boundary condition. The Laplace-transformed density distribution at time t, given that at time $t = 0$ droplets are placed at height z_0, the so-called Green's function, is given by

$$\tilde{P}(\omega, z|z_0) = \frac{(e^{\gamma_1 z_0} - e^{-\gamma_2 z_0})\ e^{-\gamma_1 z}}{\sqrt{V^2 + 4D_R\omega}} \quad \text{for } z > z_0 \tag{A2a}$$

$$\tilde{P}(\omega, z|z_0) = \frac{e^{-\gamma_2 z_0}(e^{\gamma_2 z} - e^{-\gamma_1 z})}{\sqrt{V^2 + 4D_R\omega}} \quad \text{for } z < z_0 \tag{A2b}$$

where the decay lengths are defined as

$$\gamma_1 = \sqrt{\frac{V^2}{4D_R^2} + \frac{\omega}{D_R}} + \frac{V}{2D_R}$$

$$\gamma_2 = \sqrt{\frac{V^2}{4D_R^2} + \frac{\omega}{D_R}} - \frac{V}{2D_R}$$

The survival fraction of droplets, i.e. the fraction of droplets that have not yet reached the ground, is obtained by the integral over the entire density distribution and given by

$$\tilde{S}(\omega, z_0) = \int_0^\infty dz\, \tilde{P}(\omega, z|z_0) = \left(\frac{4D_R}{V^2}\right)\frac{1 - e^{-\tilde{z}_0[\sqrt{1+\tilde{\omega}}-1]/2}}{\tilde{\omega}} \tag{A3}$$

where rescaled variables $\tilde{z}_0 = \frac{z_0 V}{D_R}$ and $\tilde{\omega} = \frac{4\omega D_R}{V^2}$ are introduced. The inverse Laplace transform reads in closed form

$$S(t, z_0) = \frac{2D_R}{V^2}\left[1 - erf\left(\tilde{t}^{\frac{1}{2}} - \frac{\tilde{z}_0 \tilde{t}^{-\frac{1}{2}}}{4}\right) - e^{\tilde{z}_0} erfc\left(\tilde{t}^{\frac{1}{2}} - \frac{\tilde{z}_0 \tilde{t}^{-\frac{1}{2}}}{4}\right)\right] \quad \text{(A4a)}$$

where $\widetilde{\omega}\tilde{t} = \omega t$, and has for large times the asymptotic decay

$$S(t, z_0) \sim z_0 t^{-3/2} e^{-V^2 t/4D_R} \quad \text{(A4b)}$$

This shows that all higher moments exist. The absorption or sedimentation time distribution is given by

$$K(t, z_0) = -\frac{\partial S(t, z_0)}{\partial t} \quad \text{(A5)}$$

and is normalized. The first moment of the absorption distribution, the mean absorption or sedimentation time, is given by

$$\tau_{sed} = \int_0^\infty dt\, t K(t, z_0) = \int_0^\infty dt\, S(t, z_0) = \tilde{S}(\omega, z_0)|_{\omega \to 0} \quad \text{(A6)}$$

Likewise, the second moment is given by

$$\tau_{sed}^{(2)} = \int_0^\infty dt\, t^2 K(t, z_0) = 2\int_0^\infty dt\, t S(t, z_0) = -2\frac{\partial \tilde{S}(\omega, z_0)}{\partial \omega}|_{\omega \to 0} \quad \text{(A7)}$$

The explicit result for the mean adsorption or sedimentation time therefore reads

$$\tau_{sed} = \frac{z_0}{V} = \frac{k_B T z_0}{D_R m g} \quad \text{(A8)}$$

and is the average time for a droplet falling with constant velocity V to reach the ground.

The thermal equilibrium mean-height of a droplet above the ground (in the absence of absorption) is from the equipartition theorem given by

$$z_{eq} = \frac{k_B T}{mg} \quad \text{(A9)}$$

Using $m = 4\pi R^3 \rho/3$ and numerical constants from Table I, for droplet radii R = 1 nm, 10 nm, 100 nm equilibrium heights of z_{eq} = 100 m, 100 mm, 100 μm are obtained, so it is seen that thermal effects can be safely neglected for all but the smallest droplets. The relative standard deviation of the absorption or sedimentation time follows from eq A7 as

$$\frac{\Delta \tau_{sed}}{\tau_{sed}} = \frac{\sqrt{\tau_{sed}^{(2)} - \tau_{sed}^2}}{\tau_{sed}} = \sqrt{\frac{2z_{eq}}{z_0}} \quad \text{(A10)}$$

Together with the result eq A9, the relative standard deviation is seen to be small for droplets larger than R = 10 nm and for an initial height z_0 in the meter range.

Section B: Stagnant droplet evaporation in the diffusion-limited regime without evaporation cooling effects

In this appendix, convection effects in the air around the droplet due to the finite speed of a falling droplet will be neglected, which increase the speed of evaporation and will be treated in Sections E-H. Also, evaporation cooling effects will be neglected. The water vapor concentration around a spherical droplet at rest is described by the diffusion equation in radial coordinates

$$\frac{d}{dt}c(r,t) = \frac{D_w}{r^2}\frac{d}{dr}r^2\frac{d}{dr}c(r,t) \tag{B1}$$

where D_w denotes the molecular water diffusion constant in air. The stationary density distribution is given by

$$c(r) = c_0\left(1 + \frac{b}{r}\right) \tag{B2}$$

where c_0 is the ambient water vapor concentration. Here the adiabatic approximation is used and the time it takes for the stationary distribution to build up, which can be shown to be small, is neglected. Particle conservation together with the reactive boundary condition at the droplet surface $r = R$ gives for the flux density j

$$j = -D_w\frac{d}{dR}c(R) = k_e c_l - k_c c(R) \tag{B3}$$

where k_e and k_c are the evaporation and condensation reaction rate coefficients, which have units of velocity, and c_l is the liquid water concentration inside the droplet (or, to be more precise, at the droplet surface). Inserting the solution eq B2 into eq B3, the resulting equation can be solved for the coefficient b and the total water evaporation flux, J, is obtained as

$$J = 4\pi R^2 j = 4\pi D_w c_0 b = 4\pi R^2 D_w \frac{k_e c_l - k_c c_0}{D_w + k_c R} \tag{B4}$$

For saturated water vapor with concentration c_g, the evaporation flux must vanish, i.e. $k_e c_l = k_c c_g$, and the evaporation rate coefficient k_e can be eliminated from eq B4 to give

$$J = 4\pi R^2 D_w \frac{k_c c_g (1 - RH)}{D_w + k_c R} \tag{B5}$$

where the relative fractional air humidity is defined as $RH = c_0/c_g$. As expected, the evaporation flux vanishes for $RH = 1$ corresponding to water-saturated air. The condensation reaction rate coefficient k_c is large, since every water molecule that hits the air-water interface basically sticks. From molecular kinetic considerations, it follows that k_c is given by the thermal molecular water velocity

$$k_c = \sqrt{\frac{k_B T}{m_w}} \approx 370 \; m/s \tag{B6}$$

where the water molecule mass m_w from Table I in the main text was used. The diffusion-limited rate scenario is defined by $k_c R > D_w$, which is realized for droplet radii $R > \frac{D_w}{k_c} = 68\ nm$, where the water molecular diffusion constant in air, D_w at 25°C from Table I in the main text, was used. In this limit, one can neglect the term proportional to D_w in the denominator of eq B5 and obtains the classical diffusion-limited result for the evaporation flux, which is linearly proportional to the droplet radius,

$$J = 4\pi R D_w c_g (1 - RH) \tag{B7}$$

Mass conservation of the droplet means that the evaporation flux is balanced by a decreasing radius, which can be written in terms of the droplet volume as

$$\frac{d}{dt}\left(\frac{4\pi}{3} R^3(t)\right) = -v_w J = -4\pi R D_w c_g v_w (1 - RH) \tag{B8}$$

where v_w is the volume of a water molecule in the liquid phase. The differential equation B8 is easily solved with the result

$$R(t) = R_0 \left(1 - t\frac{2 D_w c_g v_w (1-RH)}{R_0^2}\right)^{1/2} = R_0 (1 - \theta\, t(1 - RH)/R_0^2)^{1/2} \tag{B9}$$

where R_0 is the initial droplet radius and the numerical prefactor is given by

$$\theta = 2 D_w c_g v_w = 1.16\ \times 10^{-9} m^2/s \tag{B10}$$

and has units of a diffusion constant. Note that the calculation neglects cooling effects from evaporation, which substantially change the numerical prefactor, as shown in Section C. The water molecular volume in the liquid phase, v_w, and the water concentration of saturated water vapor, c_g, have been taken from Table I in the main text. It is seen that the shrinking of the radius starts slowly and accelerates over time. The evaporation time down to a radius where osmotic effects due to dissolved solutes and the presence of virions inside the droplet balance the evaporation chemical potential, can thus be approximated as the time needed to shrink the droplet radius to zero, given by

$$\tau_{ev} = \frac{R_0^2}{\theta(1-RH)} \tag{B11}$$

Notably, the evaporation time in eq B11 increases quadratically with the initial droplet radius R_0, while the absorption time in eq 1 decreases inversely and quadratically with R_0. Thus, at a relative humidity of $RH = 0.5$, a common value for room air, a droplet with an initial radius of $R_0 = 1\ \mu m$ has, in the absence of evaporation cooling, an evaporation time of $\tau_{ev} = 1.7$ ms, but takes (neglecting shrinkage of the radius) $\tau_{sed} = 5$ h to fall to the ground, so it will dry out and basically stay floating for an even longer time, depending on its final dry radius.

To calculate the critical initial radius below which a droplet completely dries out before falling to the ground, eq 1 is rewritten in terms of the instantaneous, radius-dependent droplet velocity $v(t)$ and combined with eq B9 gives

$$v(t) = \frac{D_R g m(t)}{k_B T} = \frac{R^2(t)}{\varphi} = \frac{R_0^2}{\varphi}(1 - \theta t(1 - RH)R_0^{-2}) = \frac{R_0^2}{\varphi}(1 - t/\tau_{ev}) \quad \text{(B12)}$$

The distance by which the droplet falls during time t follows by integration as

$$\Delta z(t) = \int_0^t dt'\, v(t') = \frac{R_0^2 \tau_{ev}}{2\varphi}(1 - (1 - t/\tau_{ev})^2) \quad \text{(B13)}$$

By setting $\Delta z = z_0$, the sedimentation time is obtained as

$$\tau_{sed}^{RH} = \tau_{ev}\left[1 - \left(1 - \frac{2\varphi z_0}{\tau_{ev}R_0^2}\right)^{1/2}\right] \quad \text{(B14)}$$

For $RH = 1$ no evaporation takes place and $\tau_{sed}^{RH} = \tau_{ev}$ is recovered. The distance by which the droplet falls during its evaporation time τ_{ev} follows as

$$\Delta z = \int_0^{\tau_{ev}} dt\, v(t) = \frac{R_0^2 \tau_{ev}}{2\varphi} = \frac{R_0^4}{2\theta\varphi(1 - RH)} \quad \text{(B15)}$$

Equating the distance Δz with the initial height z_0, the critical droplet radius follows from eq B15 as

$$R_0^{crit} = (2\varphi\theta z_0(1 - RH))^{1/4} \quad \text{(B16)}$$

Droplets with radii smaller than R_0^{crit} will completely dry out before reaching the ground and the sedimentation time in eq B14 diverges. Note that the calculations in this section neglect the finite solute concentration in the initial droplet, which will be considered in Section K and produces a lower limit to the droplet radius that can be obtained by evaporation.

Section C: Stagnant droplet evaporation in the diffusion-limited regime with evaporation cooling effects

There are several effects that temperature has on evaporation kinetics. The temperature diffusion constant is defined as $a = \lambda/(cC_P)$, where λ is the heat conductivity coefficient, c is the number density of the medium and C_P is the molecular heat capacity of the medium at constant pressure. For air one finds a value $a_{air} = 2.1 \times 10^{-5}$ m²/s, which is very similar to the water diffusion constant in air (see Table I in the main text). Thus, temperature gradient effects cannot necessarily be neglected. The fact that evaporation cooling is relevant can be quickly appreciated. The molecular evaporation enthalpy of water at 25°C is $h_{ev} = 7.3 \times 10^{-20}$ J, the molecular heat capacity of liquid water at 20°C is $C_P^l = 1.3 \times 10^{-22}$ J/K, so one evaporating water molecules cools down 22 liquid water molecules from 25°C to 0°C. The molecular melting enthalpy of water is $h_m = 1.0 \times 10^{-20}$ J, so one evaporating water molecules freezes 7 liquid water molecules. Therefore, cooling due to evaporation needs to be accounted for.

The temperature profile around a spherical heat sink is described by the heat diffusion equation in radial coordinates

$$cC_P \frac{d}{dt} T(r,t) = \frac{\lambda_{air}}{r^2} \frac{d}{dr} r^2 \frac{d}{dr} T(r,t) \tag{C1}$$

where λ_{air} denotes the heat conductivity of air. The stationary temperature distribution is given by

$$T(r) = T_0 \left(1 - \frac{b_T}{r}\right) \tag{C2}$$

where T_0 is the ambient temperature, from which the droplet surface temperature follows as

$$T_s = T_0 \left(1 - \frac{b_T}{R}\right) \tag{C3}$$

The adiabatic approximation is used, meaning that the time it takes for the stationary temperature distribution to build up, is neglected; this is justified since the heat capacity of the droplet is small compared to the evaporation enthalpy, as shown above. After a few water molecules have evaporated, the droplet will have a uniform temperature equal to the air close to the surface. The heat flux into the droplet is given by

$$J_h = 4\pi R^2 \lambda_{air} \frac{d}{dR} T(R) = 4\pi \lambda_{air} T_0 b_T \tag{C4}$$

In a stationary state, the thermal heat flux exactly balances the evaporation cooling rate, which is the water evaporation flux J from eq B5 times the evaporation enthalpy. The energy balance equation reads explicitly

$$J_h = h_{ev}J = 4\pi R^2 h_{ev} D_w \frac{k_e c_l - k_c c_0}{D_w + k_c R} = 4\pi R^2 h_{ev} D_w \frac{k_c c_g (1-RH)}{D_w + k_c R} \tag{C5}$$

In the diffusion-limited scenario this leads to

$$J_h = h_{ev}J = 4\pi R h_{ev} D_w c_g (1 - RH) \tag{C6}$$

Combining eqs C3, C4 and C6, one obtains for the temperature depression at the droplet surface

$$\Delta T = T_0 - T_s = \frac{D_w c_g h_{ev}}{\lambda_{air}} (1 - RH) = \varepsilon_T (1 - RH) \tag{C7}$$

where the numerical prefactor for air is given by $\varepsilon_T \equiv \frac{D_w c_g h_{ev}}{\lambda_{air}} = 55\ K$. This is a surprising result, as it would suggest that the evaporation of a droplet leads to droplet freezing at room temperature at all but very high relative humidities.

The estimate in eq C7 neglects that there are counteracting effects that decrease the evaporation rate with decreasing temperature. Inspection of Table I in the main text and noting that the water vapor concentration is linearly proportional to the water vapor pressure, demonstrates that the dominant temperature effect in eq C6 comes from the saturated water vapor concentration c_g, which is related to the liquid water density according to

$$c_g = c_l e^{\mu_{ex}/k_B T} \qquad (C8)$$

The saturated water vapor concentration c_g thus depends on the liquid water concentration c_l (which depends only slightly on temperature) and exponentially on the water excess chemical potential, which at room temperature is given by $\mu_{ex} = -\frac{27kJ}{mol}$ and decreases significantly with decreasing temperature, suggesting that c_g quickly decreases as the temperature decreases. In the temperature range between 0°C and 25°C, the surface water vapor concentration can be linearly fitted according to

$$c_g^{surf} = c_g(1 - \varepsilon_c \Delta T) \qquad (C9)$$

with a numerical prefactor $\varepsilon_C = 0.032\ K^{-1} = (7.69 - 1.62)/(25 \times 7.69)K^{-1}$, which is obtained by linear interpolation of the water vapor densities at 0°C and at 25°C, see Table I in the main text. The water vapor densities are obtained using the ideal gas law from the water vapor pressures, which are also given in Table I. Replacing c_g by c_g^{surf} in eq C6, the final result for the temperature depression at the droplet surface follows from solving the energy balance equation eq C6 in a self-consistent manner, which gives

$$\Delta T = T_0 - T_s = \frac{\varepsilon_T(1-RH)}{1+\varepsilon_T\varepsilon_C} = 19.9\ K\ (1 - RH) \qquad (C10)$$

One sees that the temperature reduction at the droplet surface, including the temperature dependence of the water vapor concentration at the droplet surface, is less pronounced and for RH = 0.5 is about 10 K. Thus, freezing is preempted and the linearization of the water vapor concentration in eq C9 is valid.

In the presence of evaporation-induced droplet cooling, the diffusional water flux is obtained from eq C6 as

$$J = 4\pi R D_w(c_g^{surf} - c_0) = 4\pi R D_w(c_g(1 - \varepsilon_c \Delta T) - c_0) \qquad (C11)$$

which using eq C10 can be rewritten as

$$J = 4\pi R D_w c_g \left(1 - \frac{\varepsilon_C \varepsilon_T}{1+\varepsilon_C \varepsilon_T}\right)(1 - RH) \qquad (C12)$$

Repeating the steps that led to eq B9 in Section B, one obtains the modified evolution equation for the radius

$$R(t) = R_0(1 - \theta\ t(1 - RH)/R_0^2)^{1/2} \qquad (C13)$$

where the modified numerical prefactor is given by

$$\theta = 2 D_w c_g v_w \left(1 - \frac{\varepsilon_C \varepsilon_T}{1+\varepsilon_C \varepsilon_T}\right) = 4.2 \times 10^{-10} m^2/s \qquad (C14)$$

The factor that self-consistently accounts for the evaporation cooling effect is thus given by $\left(1 - \frac{\varepsilon_C \varepsilon_T}{1+\varepsilon_C \varepsilon_T}\right) = 0.36$. Cooling considerably slows down the evaporation process but does not lead to droplet freezing.

Section D: Stagnant droplet evaporation in the reaction-rate-limited regime

To obtain the evaporation flux in the reaction-rate-limited regime, one starts from eq B5 and assumes $k_c R < D_w$, which is realized for droplet radii $R < \frac{D_w}{k_c} = 68\ nm$ (see discussion after eq B6). In this limit, one can neglect the term proportional to $k_c R$ in the denominator of eq B5 and obtain the reaction-rate-limited result for the flux, which is proportional to R^2 and thus to the surface area of the droplet,

$$J = 4\pi R^2 k_c c_g (1 - RH) \tag{D1}$$

Mass conservation of the droplet reads

$$\frac{d}{dt}\left(\frac{4\pi}{3} R^3(t)\right) = -v_w J = -4\pi R^2 k_c c_g v_w (1 - RH) \tag{D2}$$

The differential equation D2 is easily solved with the result

$$R(t) = R_0 \left(1 - t\frac{k_c c_g v_w (1-RH)}{R_0}\right) = R_0(1 - \alpha^{reac} t(1 - RH)/R_0) \tag{D3}$$

where R_0 is the initial droplet radius and the numerical prefactor is given by

$$\alpha^{reac} = k_c c_g v_w = 8.5 \times 10^{-3} m/s \tag{D4}$$

and has units of a velocity. It is seen that shrinking of the radius now is linear in time, evaporation will stop at a radius when osmotic effects due to dissolved solutes and the presence of virions inside the droplet balance the evaporation chemical potential. The time needed to shrink the droplet to the final state can be approximated by the time needed to shrink the radius to zero, which from eq D3 follows as

$$\tau_{ev}^{reac} = \frac{R_0}{\alpha^{reac}(1-RH)} \tag{D5}$$

For an initial radius of R_0 = 68 nm, which is the upper limit where the stagnant reaction-limited evaporation regime is valid, the evaporation time is $\tau_{ev}^{reac} = 8\ \mu s/(1 - RH)$. Thus, in completely dry air with $RH = 0$ the evaporation of a droplet with an initial radius of R_0 = 68 nm takes 8 μs; for relative humidity $RH = 0.5$, the complete evaporation of a droplet with an initial radius of R_0 = 68 nm takes $16\ \mu s$. This time is completely negligible compared to the sedimentation time of a droplet with a radius of 68 nm, which is many days. The final reaction-rate limited evaporation stage can thus be neglected for droplets significantly larger than 68 nm in radius.

In fact, evaporation cooling effects can approximately be neglected in the reaction-rate-limited regime, since the heat transport stays diffusion limited and thus is faster than the reaction-rate-limited diffusive transport of water molecules.

Section E: Convection effects on evaporation for laminar flow

To estimate air convection effects on the evaporation rate at an analytically manageable level, the concept of a concentration boundary layer will be used. In this section laminar flow will be assumed, finite-Reynolds number effects will be considered in subsequent appendices. In order to obtain the concentration boundary layer thickness, it is necessary to calculate the time it takes for a water molecule to diffuse a certain distance away from the droplet surface. The water diffusion velocity is given by

$$u(r) = \frac{dr(t)}{dt} = \frac{j(r)}{c(r)} = \frac{-D_w dc(r)/dr}{c(r)} = \frac{D_w b/r^2}{1+\frac{b}{r}-\frac{b}{R+\Delta}} \qquad (E1)$$

In the last step the presence of a concentration boundary layer at a radius $r = R+\Delta$ was assumed, where the concentration boundary layer width is defined by Δ and the stationary density distribution is given by

$$c(r) = c_0 \left(1 + \frac{b}{r} - \frac{b}{R+\Delta}\right) \qquad (E2)$$

By this construction, at a radius $r = R+\Delta$, the water vapor concentration is equal to the ambient water vapor concentration c_0. Using again the reactive boundary condition at the sphere surface $r = R$, eq B3, the coefficient b is given by

$$b = R^2 k_c \frac{\frac{c_g}{c_0}-1}{D_w+k_c\Delta R/(R+\Delta)} \cong R^2 k_c \frac{\frac{c_g}{c_0}-1}{D_w+k_c\Delta} \qquad (E3)$$

where $k_e c_l = k_c c_g$ was used. One sees that for a concentration boundary layer larger than the droplet radius, i.e. for $\Delta > R$, the effect of Δ disappears. In the last step therefore the opposite limit, $\Delta < R$, was considered. The total water evaporation flux J follows as

$$J = 4\pi R^2 j = 4\pi D_w c_0 b = 4\pi R^2 D_w k_c c_g \frac{1-RH}{D_w+k_c\Delta} \qquad (E4)$$

The differential equation implied by eq E1 is easily solved and gives

$$(r^3(t) - R^3)\left(1 - \frac{b}{R+\Delta}\right)/3 + b(r^2(t) - R^2)/2 = D_w bt \quad (E5)$$

Using $r = R+\Delta$ and $\Delta < R$, this simplifies to

$$D_w b\tau_{diff} \approx \Delta R^2 \left(1 - \frac{b}{R+\Delta}\right) + b\Delta R \approx \Delta R^2 + b\Delta^2 \qquad (E6)$$

which defines the time it takes a water molecule to diffuse through the concentration boundary layer, which is called the diffusion time τ_{diff}. Inserting the diffusion-limited approximation for b from eq E3, valid for $k_c\Delta > D_w$, the diffusion time becomes

$$\tau_{diff} = \frac{\Delta^2}{D_w(1-RH)} \qquad (E7)$$

This diffusion time has to be compared with the convection time scale

$$\tau_{conv} = \frac{\pi r/2}{v_{air}(r)} \approx \frac{\pi R^2}{2V\Delta} \tag{E8}$$

which is the time the flow needs to travel a quarter circle around the droplet at the boundary layer located at a radius of $r = R+\Delta$ from the droplet center. In eq E8 the air flow profile around the droplet for laminar flow conditions has been used, given approximately by $v_{air}(r) \approx V\frac{r-R}{r} \approx V\Delta/R$, where V is the stationary droplet velocity from eq A1b.

By equating the diffusion and convection time scales τ_{diff} and τ_{conv} the concentration boundary layer thickness results as

$$\Delta = \Delta_0(1 - RH)^{1/3} \tag{E9}$$

with

$$\Delta_0 = \left(\frac{9\,\pi\eta D_w}{4\rho g}\right)^{1/3} = 69\,\mu m \tag{E10}$$

Interestingly, the concentration boundary layer thickness does not depend on the speed with which the droplet falls to the ground, i.e., it does not depend on the droplet radius R. This result means that for radii larger than $R = 69$ μm and for dry air, one has $\Delta/R < 1$ and thus convection cannot be neglected and will accelerate evaporation.

The diffusion-limited rate scenario is defined by $k_c\Delta > D_w$, which using eqs B6 and E9, is satisfied for relative humidity $RH < 1-10^{-9}$, which is certainly always true. It transpires that convective evaporation in the laminar flow regime is basically always diffusion limited. Thus, the term proportional to D_w in the denominator of eq E4 can be safely neglected and the diffusion-limited flux is obtained

$$J = 4\pi R^2 D_w c_g(1 - RH)/\Delta \tag{E11}$$

Mass conservation of the droplet can be written in terms of the droplet volume as

$$\frac{d}{dt}\left(\frac{4\pi}{3}R^3(t)\right) = -v_wJ = -4\pi R^2 D_w c_g v_w(1 - RH)/\Delta \tag{E12}$$

The differential equation E12 is easily solved with the result

$$R(t) = R_0\left(1 - t\frac{D_w c_g v_w(1-RH)^{2/3}}{\Delta_0 R_0}\right) = R_0\left(1 - \alpha^{conv}t(1 - RH)^{2/3}/R_0\right) \tag{E13}$$

where R_0 is the initial droplet radius and the numerical factor is given by

$$\alpha^{conv} = D_w c_g v_w/\Delta_0 = 8.4 \times 10^{-6} m/s \tag{E14}$$

and has units of a velocity. It is seen that shrinking of the radius is linear in time. Again, the evaporation time to a radius where osmotic effects due to dissolved solutes and the presence

of virions inside the droplet balance the evaporation chemical potential can thus be approximated by the time needed to shrink the droplet radius down to zero, given by

$$\tau_{ev}^{conv} = \frac{R_0}{\alpha^{conv}(1-RH)^{2/3}} \qquad \text{(E15)}$$

Thus, at a relative humidity of RH = 0.5, a droplet with an initial radius of R_0 = 1 mm has an evaporation time of τ_{ev}^{conv} = 189 s, but takes (neglecting the shrinking of the radius) τ_{sed} = 0.017 s to fall to the ground from a height of 2 meters, so it will not dry out before being sedimenting to the ground. To accurately calculate the critical initial radius below which a droplet completely dries out before falling to the ground, eq 1 is rewritten in terms of the instantaneous droplet velocity and combined with eq E13 to give

$$v(t) = \frac{D_R g m(t)}{k_B T} = \frac{R^2(t)}{\varphi} = \frac{R_0^2}{\varphi}\left(1 - \alpha^{conv} t(1 - RH)^{\frac{2}{3}}/R_0\right)^2 \qquad \text{(E16)}$$

The distance by which the droplet falls during its evaporation time, τ_{ev}^{conv}, follows by integration as

$$\Delta z = \int_0^{\tau_{ev}^{conv}} v(t) = \frac{R_0^3}{3\alpha^{conv}\varphi(1-RH)^{2/3}} \qquad \text{(E17)}$$

Now equating the distance Δz with the height z_0, the critical droplet radius is

$$R^{crit} = \left(3\varphi\alpha^{conv}z_0(1 - RH)^{2/3}\right)^{1/3} \qquad \text{(E18)}$$

which for an initial height of z_0 = 2 m and for RH = 0.5 gives R^{crit} = 65 μm. This is about the same radius where convection effects are relevant from eq E9. Thus, convective evaporation effects do not play a significant role for droplets that are released from a height of z_0 = 2 m. For an initial height of z_0 =2 km it follows from eq E18 that droplets with a radius smaller than 650 μm evaporate before they fall to the ground.

The results derived in this section neglect effects due to non-linear hydrodynamics, finite Reynolds number effects will be discussed in the following appendices. Evaporation cooling effects have not been treated explicitly in this appendix, but can be approximately accounted for multiplying the water vapor density c_g in eq E14 by the correction term $\left(1 - \frac{\varepsilon_C \varepsilon_T}{1+\varepsilon_C \varepsilon_T}\right)$ derived in Section C.

Section F: Non-laminar flow effects and flow boundary layer

Within the laminar flow boundary layer around an object, viscosity is relevant and laminar Stokes flow develops, outside this flow boundary layer potential flow is obtained. The flow boundary layer thickness scales as

$$\delta = \left(\frac{vx}{V}\right)^{1/2} \qquad \text{(F1)}$$

as a function of the distance x that the flow has moved along the object, where V is the velocity of the object and $\nu = \eta/\rho_{air}$ is the kinematic viscosity. The density of air is denoted as ρ_{air} and given in Table I in the main text. The kinematic viscosity has units of a diffusion constant and characterizes the diffusivity of momentum or vorticity, its value in air is $\nu = 1.6 \times 10^{-5}$ m^2/s and thus it is about half the value of the water diffusion constant in air. This shows that momentum diffuses slightly slower than water in air, thus the flow boundary layer δ is expected to be smaller than the concentration boundary layer Δ. As result, flow boundary layer effects are expected to be relevant.

A simple estimate of the importance of momentum diffusion for a droplet moving in air is obtained by asking whether the flow boundary layer thickness δ is smaller than the droplet radius R for a flow that has travelled by a distance that corresponds to a quarter circle $x = \pi R/2$ around the droplet, i.e.

$$\delta = \left(\frac{\nu \pi R/2}{V}\right)^{1/2} < R \qquad\qquad \text{(F2)}$$

which can be rewritten as

$$Re/\pi \equiv \frac{2\,RV}{\pi\nu} = \frac{2\,RV\rho_{air}}{\pi\eta} = \left(\frac{R}{R_*}\right)^3 > 1 \qquad\qquad \text{(F3)}$$

and is equivalent to the condition that the Reynolds number Re is larger than π. The characteristic radius is defined as

$$R_* = \left(\frac{9\pi\eta^2}{4g\rho_{air}\rho}\right)^{1/3} = 59\mu\text{m} \qquad\qquad \text{(F4)}$$

The radius at which the Reynolds number becomes larger than π and the flow around the sphere will not be laminar anymore is therefore given by $R_* = 59\mu$m. The typical momentum boundary layer thickness given in eq F2 can be rewritten in terms of the characteristic radius as

$$\delta = \left(\frac{R_*^3}{R}\right)^{1/2} \qquad\qquad \text{(F5)}$$

and is for a sphere with the characteristic radius given by R_* itself. Since for not too humid air R_* is smaller than the concentration boundary layer width $\Delta_0 = 69\ \mu m$ from eq E10, it follows that convection effects are modified in the presence of a flow boundary layer, which will be treated in Sections G and H.

Section G: Convection effects on evaporation: Double boundary layer scenario in humid air

In this section the double boundary-layer problem will be addressed, where both concentration and fluid flow boundary layers with widths Δ and δ are present. It will be assumed that $\Delta < \delta$, which a posteriori will be shown to correspond to humid air; the opposite case $\Delta > \delta$ for dry air will be treated in Section H.

Due to the presence of the flow boundary layer, the air flow field around the droplet is compressed by a factor R/δ and is approximately given by

$$v_{air}(r) \approx V \frac{R}{\delta}\left(\frac{r-R}{r}\right) \ . \tag{G1}$$

The convection time scale is defined as the time the flow needs to travel a quarter circle around the droplet at the concentration boundary layer located at a radius of $r = R+\Delta$ from the droplet center and is given by

$$\tau_{conv} = \frac{\pi r/2}{v_{air}(r)} \approx \frac{\pi R \delta}{2V\Delta} \tag{G2}$$

which is smaller than the result for laminar flow in eq E8 by a factor of δ/R. By equating the diffusion time scale τ_{diff} in eq E7, which is not modified by flow boundary-layer effects, with the convection time scale τ_{conv} in eq G2, the boundary layer thickness results as

$$\Delta = \Delta_0(1 - RH)^{1/3}\left(\frac{R_*}{R}\right)^{1/2} \tag{G3}$$

where $\Delta_0 = 69 \ \mu m$ is the laminar concentration boundary layer width from eq E10 and $R_* = 59$ μm is the characteristic radius for non-laminar flow effects from eq F4. To check whether the assumption $\Delta < \delta$ used in this Appendix is satisfied, eq G3 is divided by the momentum boundary layer width δ from eq F5 to obtain

$$\frac{\Delta}{\delta} = (1 - RH)^{1/3}\frac{\Delta_0}{R_*} \tag{G4}$$

It transpires that the assumption $\Delta < \delta$ is satisfied for humid air with a relative humidity larger than

$$RH > 1 - \left(\frac{\Delta_0}{R_*}\right)^{-3} = 0.36 \tag{G5}$$

When is the double-boundary-layer evaporation regime entered? This question is equivalent to asking when the boundary layer width Δ as given by eq G3 becomes smaller than the radius R. For an intermediate humidity that coincides with the threshold value eq G5, it is found that the double-boundary-layer evaporation regime is entered for radii $R > R_* = 59$μm, i.e., as soon as boundary flow effects occur. For more humid the threshold radius increases and follows from eq G3.

The diffusion-limited rate scenario is defined by $k_c\Delta > D_w$, which using eqs B6 and G3, is satisfied for radii

$$R < (1 - RH)^{2/3}\left(\frac{\Delta_0^2 R_*}{(D_w/k_c)^2}\right) = (1 - RH)^{2/3} \ 59 \ m \tag{G6}$$

It transpires that double-boundary layer convective evaporation in humid air is always diffusion limited. Thus, the differential equation eq E12 that is valid in the diffusion limit can be used, which in conjunction with eq G3 yields

$$R(t) = \left(R_0^{1/2} - t\frac{D_w c_g v_w (1-RH)^{2/3}}{2\Delta_0 R_*^{1/2}}\right)^2 = \left(R_0^{1/2} - \alpha^{dbl1} t (1-RH)^{2/3}\right)^2 \qquad (G7)$$

where R_0 is the initial droplet radius and the numerical factor is given by

$$\alpha^{dbl1} = \frac{D_w c_g v_w}{2\Delta_0 R_*^{1/2}} = 5.4 \times 10^{-4} m^{1/2}/s \qquad (G8)$$

It is seen that the shrinking of the radius slows down over time. Evaporation cooling effects have not been treated explicitly, but can be approximately accounted for by multiplying the water vapor density by the correction term $\left(1 - \frac{\varepsilon_C \varepsilon_T}{1+\varepsilon_C \varepsilon_T}\right)$ derived in Section C.

Section H: Convection effects on evaporation: Double boundary layer scenario in dry air

In this section, it will be assumed that the concentration boundary layer width is larger than the flow boundary layer width, i.e. $\Delta > \delta$, which a posteriori will be shown to correspond to dry air. The calculation closely follows Section G.

At the concentration boundary layer, since $\Delta > \delta$, the air flow field around the droplet is unperturbed by the presence of the droplet and given by $v_{air}(r) \approx V$. The convection time scale is therefore given by

$$\tau_{conv} = \frac{\pi r/2}{v_{air}(r)} \approx \frac{\pi R}{2V} \qquad (H1)$$

which is smaller than the corresponding result in eq G2. By equating the diffusion time scale τ_{diff} in eq E7, which is not modified by flow boundary-layer effects, with the convection time scale τ_{conv} in eq H1, the boundary layer thickness results as

$$\Delta = (1-RH)^{1/2} \left(\frac{\Delta_0^3}{R}\right)^{1/2} \qquad (H2)$$

where $\Delta_0 = 69\ \mu m$ is the laminar concentration boundary layer width from eq E10. To check when the assumption $\Delta > \delta$ used here is satisfied, eq H2 is divided by the momentum boundary layer width δ from eq F5 to obtain

$$\frac{\Delta}{\delta} = (1-RH)^{1/2} \left(\frac{\Delta_0}{R_*}\right)^{3/2} \qquad (H3)$$

It transpires that the assumption $\Delta > \delta$ is satisfied for dry air with a relative humidity smaller than

$$RH < 1 - \left(\frac{\Delta_0}{R_*}\right)^{-3} = 0.36 \qquad (H4)$$

When is the double-boundary-layer evaporation regime entered? This question is equivalent to asking when the boundary layer width Δ as given by eq H2 becomes smaller than the radius R. For an intermediate humidity that coincides with the threshold value eq H4, it follows that

the double-boundary-layer evaporation regime is entered for radii $R > R_* = 59\mu m$, i.e., as soon as boundary flow effects according to eq F4 occur.

The diffusion-limited rate scenario is defined by $k_c\Delta> D_w$, which using eqs B6 and H2, is satisfied for radii

$$R < (1 - RH)\left(\frac{\Delta_0^3}{(D_w/k_c)^2}\right) = (1 - RH)\ 71\ m \qquad (H5)$$

It transpires that double-boundary layer convective evaporation in dry air is always diffusion limited. Thus, the differential equation E12 can be used, which in conjunction with eq H2 yields

$$R(t) = \left(R_0^{1/2} - t\frac{D_w c_g v_w (1-RH)^{1/2}}{2\Delta_0^{3/2}}\right)^2 = \left(R_0^{1/2} - \alpha^{dbl2}t(1 - RH)^{1/2}\right)^2 \qquad (H6)$$

where R_0 is the initial droplet radius and the numerical factor is given by

$$\alpha^{dbl2} = \frac{D_w c_g v_w}{2\Delta_0^{3/2}} = 5.0 \times 10^{-4} m^{1/2}/s \qquad (H7)$$

Evaporation cooling effects have not been treated explicitly, but can be approximately accounted for by multiplying the water vapor density c_g by the correction term $\left(1 - \frac{\varepsilon_C \varepsilon_T}{1+\varepsilon_C \varepsilon_T}\right)$ derived in Section C.

In summary, for droplet radii larger than about $R_* = 59\ \mu m$, non-linear hydrodynamic effects become important and produce a finite, so-called flow boundary layer, around the falling droplet. Inside the flow boundary layer viscous effects are relevant and laminar flow is obtained, outside the flow boundary layer viscous effects can be neglected and potential flow is realized. At about the same range of radii, the stagnant approximation becomes invalid, because convection speeds up the evaporation process. This effect can be described by a concentration boundary layer. The problem is thus a double-boundary-layer problem and involves a concentration and a flow boundary layer. Whether the concentration boundary layer or the flow boundary layer is smaller and thus more relevant, depends on the relative air humidity. It turns out that the evaporation in the presence of convection is diffusion limited. For humid air with a relative humidity $RH > 0.36$, the concentration boundary layer is evaporation-rate limiting and the time-dependent radius decrease is given by eq G7. For drier air with a relative humidity $RH < 0.36$, the flow boundary layer is evaporation-rate limiting and the time dependent radius decrease is given by eq H6. The results for the humid and dry boundary layer scenarios thus look quite similar, but the physical mechanisms behind the evaporation process are very different. At high Reynolds numbers the friction experienced by a falling droplet is reduced due to a combination of boundary layer effects, boundary-layer separation effects and turbulence effects. In Section I it is shown that the Stokes expression for the friction force acting on a falling spherical droplet is accurate for radii below about 160 μm.

Section I: Falling speed for large Reynold numbers

The Stokes approximation used for calculating the stationary falling speed of droplets in eq A1b is a low-Reynolds number approximation. An empirical formula for the settling velocity of a sphere in air, that is valid over the entire range of Reynold numbers, is

$$V = \sqrt{\frac{8gR\rho}{3\rho_{air}c_D}}$$

(I1)

where the resistance coefficient is given by

$$c_D = \frac{24}{Re} + \frac{4}{Re^{1/2}} + 0.4$$

(I2)

and the Reynolds number Re is defined in eq F3. The result of eq A1b is reproduced by eq I1 when only the first term in eq I2 is used. The accuracy of this low-Reynolds number approximation can be checked by comparing the first and third terms in eq I2, which become equal for a Reynolds number of about Re=60, which corresponds, using again eq F3, to a radius of about $(\frac{60}{\pi})^{1/3}R_* = 157$ µm. This suggests that the falling speed according to eq A1b is quite accurate for radii below 157 µm. For larger radii the falling speed will be reduced and thus the sedimentation time will be increased. Therefore, the sedimentation times presented in this note are lower estimates for radii larger than about 157µm.

Section J: Internal mixing effects

The calculations so far assumed that diffusion inside the droplet is sufficiently rapid, so that the water concentration at the droplet surface does not differ significantly from the mean water concentration in the droplet. It will turn out that this is a limiting factor for the maximal droplet size that can evaporate at the speed predicted here. According to the diffusion law, the time it takes for a water molecule to diffuse over the droplet radius R inside the droplet is

$$\tau_{mix} = \frac{R^2}{2D_w^l}$$

(J1)

where D_w^l is the molecular water self-diffusion constant in liquid water. The mixing time within a droplet of radius $R = 10$ µm is $\tau_{mix} = 25$ ms and inside a droplet of radius $R = 100$ µm it is $\tau_{mix} = 2.5$ s. Equating τ_{mix} with the mean sedimentation time Eq. (1), the mixing time inside the droplet is only shorter than the sedimentation time for radii smaller than about 100 µm. For larger droplets the internal diffusion will slow down evaporation. Convection effects inside the droplet, due to shear coupling to the outside air flow field, will counteract this effect, but are not considered here. Also, the increase of the internal droplet viscosity due to increasing solute concentration and the possible presence of a solid solute phase is not considered and will further slowdown the diffusion inside droplets.

Section K: Solute-induced vapor pressure reduction effects

Any solute present in the aqueous droplet decreases the water vapor pressure. This colligative effect is basically due to the dilution of the liquid water and can be derived in the following fashion:

Water chemical potential in a two-component liquid system:

The entropy of a liquid two-component system consisting of N_w water molecules with molecular volume v_w and N_s solutes molecules with molecular volume v_s is given up to an irrelevant constant by

$$\frac{S}{k_B} = -N_w ln\left(\frac{N_w}{N_w v_w + N_s v_s}\right) - N_s ln\left(\frac{N_s}{N_w v_w + N_s v_s}\right) \qquad (K1)$$

where ideal mixing and ideal volume additivity is assumed. The water chemical potential in the liquid follows as

$$\frac{\mu_w^l}{k_B T} = -\frac{\partial S}{k_B \partial N_w} + \frac{\mu_{ex}}{k_B T} = ln\left(\frac{1-\Phi}{v_w}\right) + \Phi\left(1 - \frac{v_w}{v_s}\right) + \frac{\mu_{ex}}{k_B T} \qquad (K2)$$

where $\Phi = N_s v_s / (N_w v_w + N_s v_s)$ is the solute volume fraction. In the limit $\Phi \to 0$ this can be rewritten as

$$\frac{\mu_w^l}{k_B T} \approx ln\left(\frac{1-\Phi v_w/v_s}{v_w}\right) + \frac{\mu_{ex}}{k_B T} \qquad (K3)$$

When the solute volume fraction is finite and in particular when the water and solute molecular volumes are similar to each other, one can instead rewrite eq K2 as

$$\frac{\mu_w^l}{k_B T} \approx ln\left(\frac{1-\Phi}{v_w}\right) + \frac{\mu_{ex}}{k_B T} \qquad (K4)$$

Water chemical potential in a multi-component liquid system:

The entropy of a liquid many-component system consisting of N_w water molecules with molecular volume v_w and N_i solute molecules of type i with molecular volume v_i each, where $i=1 \dots M$, is given up to an irrelevant constant by

$$\frac{S}{k_B} = -N_w ln\left(\frac{N_w}{N_w v_w + \sum_i v_i N_i}\right) - \sum_i N_i ln\left(\frac{N_i}{N_w v_w + \sum_j v_j N_j}\right) \qquad (K5)$$

where again ideal mixing and ideal volume additivity was assumed. The water chemical potential in the liquid follows as

$$\frac{\mu_w^l}{k_B T} = -\frac{\partial S}{k_B \partial N_w} + \frac{\mu_{ex}}{k_B T} = ln\left(\frac{1-\Phi}{v_w}\right) + \sum_i \Phi_i\left(1 - \frac{v_w}{v_i}\right) + \frac{\mu_{ex}}{k_B T} \qquad (K6)$$

where $\Phi_i = N_i v_i / (N_w v_w + \sum_i v_i N_i)$ is the solute volume fraction of species i and $\sum_i \Phi_i = \Phi$. In the limit $\Phi \to 0$ this can be written as

$$\frac{\mu_w^l}{k_B T} \approx ln\left(\frac{1-\sum_i \Phi_i v_w/v_i}{v_w}\right) + \frac{\mu_{ex}}{k_B T} \qquad (K7)$$

On the other hand, when the solute volume fraction Φ is finite and the sum $\sum_i \Phi_i\left(1 - \frac{v_w}{v_i}\right)$ is small, one can instead write

$$\frac{\mu_w^l}{k_B T} \approx ln\left(\frac{1-\Phi}{v_w}\right) + \frac{\mu_{ex}}{k_B T} \qquad (K8)$$

which is the approximation that will be used in the following.

Water vapor concentration and evaporation rate in presence of solutes:

From the ideal expression for the water vapor chemical potential

$$\frac{\mu_w^g}{k_B T} = ln(c_g) \qquad (K9)$$

and the equality of chemical potentials, $\mu_w^g = \mu_w^l$, the equilibrium vapor concentration in the presence of solutes follows from eq K8 as

$$c_g^{sol} = \left(\frac{1-\Phi}{v_w}\right) e^{\mu_{ex}/k_B T} = (1 - \Phi)c_l e^{\mu_{ex}/k_B T} \qquad (K10)$$

which depends exponentially on the water excess chemical potential, at room temperature given by $\mu_{ex} = -\frac{27kJ}{mol}$, and where the liquid water concentration in the absence of solute is denoted as $c_l = 1/v_w$. Assuming that initially the volume fraction of solutes is Φ_0 and the initial radius is R_0, the water concentration in the liquid droplet with reduced radius R follows as

$$c_l^{sol} = \frac{1}{v_w}\left(1 - \Phi_0 \frac{R_0^3}{R^3}\right) \qquad (K11)$$

Similarly, one obtains for the water vapor concentration in the presence of solutes

$$c_g^{sol} = c_g \left(1 - \Phi_0 \frac{R_0^3}{R^3}\right) \qquad (K12)$$

Here, c_g represents the water vapor concentration in the absence of solutes. Non-ideal effects can be included via the excess chemical potential and would be described by an activity coefficient different from unity, which is not pursued here. Replacing c_g by c_g^{sol} in eq C12, one arrives at

$$J = 4\pi R D_w \left(1 - \frac{\varepsilon_C \varepsilon_T}{1 + \varepsilon_C \varepsilon_T}\right)\left(c_g^{sol} - c_0\right) \qquad (K13)$$

Together with eq K12, one obtains the modified diffusive water flux in the presence of solutes as

$$J = 4\pi R D_w \left(1 - \frac{\varepsilon_C \varepsilon_T}{1 + \varepsilon_C \varepsilon_T}\right)\left(c_g \left(1 - \Phi_0 \frac{R_0^3}{R^3}\right) - c_0\right) \qquad (K14)$$

The mass conservation equation follows as

$$\frac{d}{dt}\left(\frac{4\pi}{3}R^3(t)\right) = -v_w J = -2\pi R(t)\theta\left(1 - \Phi_0\frac{R_0^3}{R(t)^3} - RH\right) \qquad \text{(K15)}$$

where θ is defined in eq C14. Equation K15 gives rise to the differential equation

$$\frac{2RdR}{1-R_{ev}^3/R^3} = -\theta(1 - RH)dt \qquad \text{(K16)}$$

where the equilibrium droplet radius that is obtained in the long-time limit is defined as

$$R_{ev} = R_0\left(\frac{\Phi_0}{1-RH}\right)^{1/3} \qquad \text{(K17)}$$

Here, R_0 is the initial radius and Φ_0 is the initial volume fraction of solutes, including strongly bound hydration water. Only for RH = 0 does a droplet dry out to the minimal possible radius of $R_{ev} = R_0(\Phi_0)^{1/3}$, for finite relative humidity the equilibrium droplet radius is characterized by an equilibrium solute volume fraction of $\Phi_{ev} = 1 - RH$. As an example, for RH = 0.5, the free water and solute (including hydration water) volume fractions in the equilibrium state equal each other. Equation K17 is modified for solutes that perturb the water activity, but for most solutes non-ideal water solution effects can be neglected. To illustrate this: the saturation concentration of NaCl in water is approximately 6 M, which corresponds, for simplicity assuming equal volume of Na$^+$ cations, Cl$^-$ anions and water molecules, roughly to a volume fraction of Φ_∞ =12M/(55+12)M = 0.18, thus suggesting a water vapor pressure corresponding to a humidity of $RH = 1 - \Phi_\infty = 0.82$, which is rather close to the experimental humidity created by a saturated NaCl solution of 0.75. This reflects that the activity coefficient of NaCl is rather close to unity for concentrations close to the solubility limit.

The solution of the differential equation K16 can be written as

$$t(R) = \frac{R_{ev}^2}{\theta(1-RH)}\left[\mathcal{L}\left(\frac{R_0}{R_{ev}}\right) - \mathcal{L}\left(\frac{R}{R_{ev}}\right)\right] \qquad \text{(K18)}$$

where the scaling function is given by

$$\mathcal{L}(x) = x^2 - \frac{2}{\sqrt{3}}\arctan\left(\frac{1+\frac{2}{x}}{\sqrt{3}}\right) - \frac{1}{3}\ln\left(\frac{x^2+x+1}{(x-1)^2}\right) \qquad \text{(K19)}$$

The scaling function exhibits the asymptotic behavior

$$\mathcal{L}(x) \cong x^2 \qquad \text{(K20)}$$

for large arguments and

$$\mathcal{L}(x) \cong +\frac{2}{3}\ln(1 - 1/x) \qquad \text{(K21)}$$

for small arguments $x \to 1$. A quite accurate crossover expression is produced by summing the two limits as

$$\mathcal{L}(x) \cong x^2 + \frac{2}{3} \ln (1 - 1/x) \qquad\qquad (K22)$$

Neglecting the logarithmic term in eq K22 that reflects the kinetic slowing down due to the reduced water vapor pressure, one obtains from eq K18 the limiting result

$$t(R)/\tau_{ev} = 1 - \frac{R^2}{R_0^2} \qquad\qquad (K23)$$

from which an approximate expression for the evaporation time in the presence of solutes follows as

$$\tau_{ev}^{sol} = \tau_{ev} \left(1 - \frac{R_{ev}^2}{R_0^2} \right) \qquad\qquad (K24)$$

The threshold radius below which the presence of solutes becomes important, can be defined by the radius where the function $t(R)$ changes curvature. The second derivative of $t(R)$ in eq 10 is given by

$$\frac{R_0^2 d^2 t(R)/\tau_{ev}}{dR^2} = -2 + \frac{2}{3}((1 - R/R_{ev})^{-2} - (R/R_{ev})^{-2})$$

and vanishes at a radius of $R/R_{ev} = 1.54$. Thus, according to this curvature criterion, droplets enter the solute-dominated evaporation regime for radii smaller than $R = 1.54 \, R_{ev}$, independent of the initial droplet radius R_0.

Sedimentation time in the presence of solutes:

The sedimentation of small enough droplets proceeds in two stages: First, the droplets shrink down to a radius given by eq K17, second, the droplets sediment for an extended time with a fixed radius. The distance by which the droplet falls during its evaporation time τ_{ev}^{sol} follows in analogy to eq B15 as

$$z_{ev} = \int_0^{\tau_{ev}^{sol}} v(t) = \frac{R_0^2 \tau_{ev}}{2\varphi} (1 - (1 - \tau_{ev}^{sol}/\tau_{ev})^2) = \frac{R_0^2 \tau_{ev}}{2\varphi} \left(1 - \left(\frac{R_{ev}}{R_0} \right)^4 \right) \qquad (K25)$$

Thus, the total sedimentation time is given by

$$\tau_{sed}^{sol} = \tau_{ev}^{sol} + \frac{\varphi(z_0 - z_{ev})}{R_{ev}^2} \qquad\qquad (K26)$$

where the first term is the time it takes for the droplets to shrink down to the equilibrium radius R_{ev} and the second term is the time it takes to sediment from the height $z_0 - z_{ev}$ to the ground as given by eq 1 in the main text. Using eq K24 and K25 the final result for the sedimentation time can be written as

$$\tau_{sed}^{sol} = \frac{\varphi z_0}{R_{ev}^2} - \frac{\tau_{ev}}{2} \left(\frac{R_0}{R_{ev}} - \frac{R_{ev}}{R_0} \right)^2 \qquad\qquad (K27)$$

For droplets that are so large that they do not reach the radius R_{ev} before they hit the ground, eq B14 describes the sedimentation time accurately. The crossover between the two sedimentation time regimes occurs when z_{ev} as described by eq K25 equals z_0, the critical droplet radius follows as

$$R_0^{crit} = \left(\frac{2\varphi\theta z_0(1-RH)}{1-(\Phi_0/(1-RH))^{4/3}} \right)^{1/4} \tag{K28}$$

which constitutes a generalization of eq B16 in the presence of solutes. Droplets with radii smaller than R_0^{crit} will reach their equilibrium radius before sedimenting to the ground and the sedimentation time is given by eq K27.

Section L: Surface tension effects

The large surface tension of water increases the vapor pressure produced by droplets. The surface free energy of a droplet is given by

$$F = 4\pi\gamma R^2 \tag{L1}$$

The chemical potential contribution, the so-called Kelvin potential, reads

$$\mu_{Kel} = \frac{dF}{dN} = \frac{dR}{dN}\frac{dF}{dR} = \frac{2\gamma v_w}{R} \tag{L2}$$

where the number of water molecules inside the droplet is taken as $N = 4\pi R^3/(3v_w)$. Inserting numbers, the rescaled Kelvin potential reads

$$\frac{\mu_{Kel}}{k_B T} = \frac{10^{-9}m}{R} \tag{L3}$$

which is significant compared to the water excess chemical potential only for droplet radii smaller than one nanometer.
To avoid confusion: the Laplace pressure

$$P_{Lap} = -\frac{dF}{dV} = -\frac{dR}{dV}\frac{dF}{dR} = \frac{2\gamma}{R} \tag{L4}$$

is significant and reaches 1 bar for a droplet radius of 1 μm, but it is unrelated to the vapor pressure.