A Test-tube Model for Rainfall

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Reporting collaboration with

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(MPI-Dynamics & Self-organisation, Goettingen)

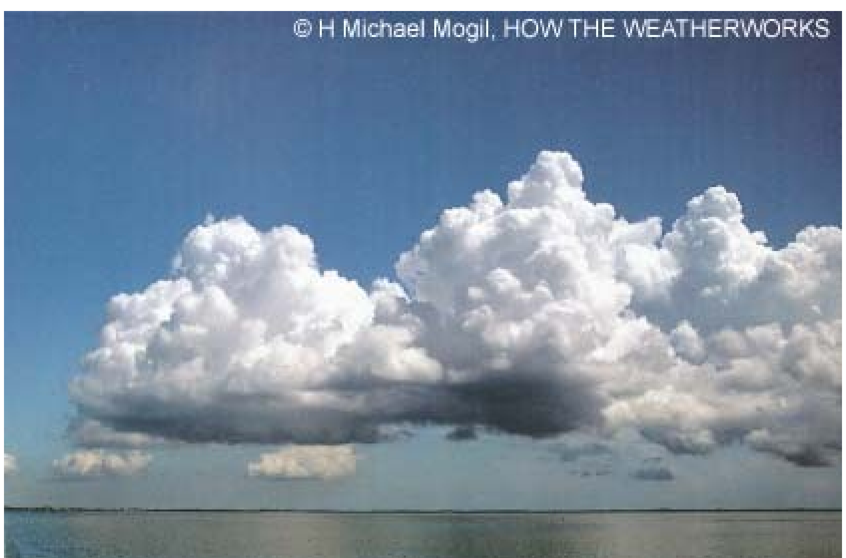
The rain cycle

- Evaporation of water into an air mass.
- Cooling of the air mass causes supersaturation.
- Supersaturation leads to nucleation of water droplets onto dust particles/salt grains, creating clouds.
- The microscopic water droplets in clouds grow by collisions or other processes, until they are heavy enough to fall at a significant rate.
- Falling droplets can sweep up smaller ones and experience a runaway growth. They become rain drops.
- The rain drops sweep the excess moisture our of the air mass.

Types of cloud

There are two major clouds types: *stratus* and *cumulus*:

Stratus clouds: layers in an air mass which is stable against convection. Associated with slow onset of rain from warm fronts. No turbulence.



Cumulus clouds: moisture in clumps of rising air in an atmosphere which is unstable against convection. Associated with showers or thunderstorms from cold fronts. Turbulence.

Kinetics of droplet growth

Droplets may grow by

- Condensation of supersaturated vapour.
- Collisions, leading to droplet coalsecence. The relative velocity may be due to

  a) Brownian diffusion of droplets

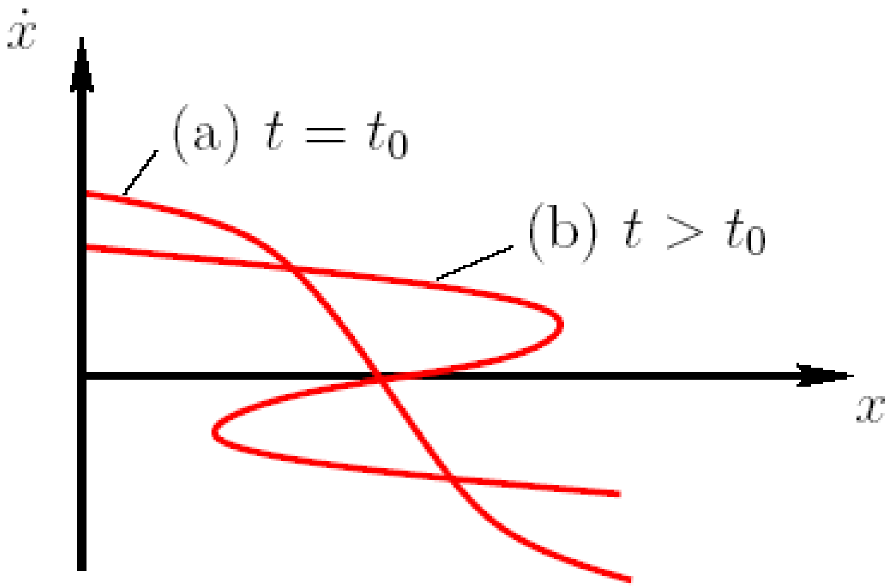
  b) Gravitational settling

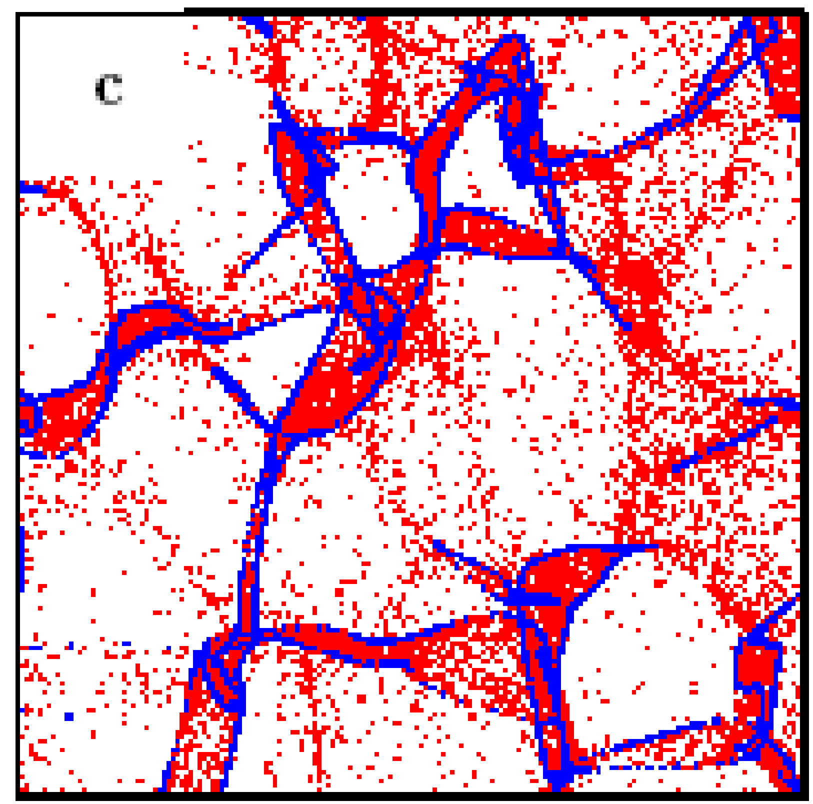
  c) Consequences of turbulence in the cloud.

Because the collision efficiency for small particles is very low, there is a 'condensation-collision bottleneck' which is a barrier to droplet growth in the range $15 - 50\,\mu\mathrm{m}$, and that turbulence is required to bridge the gap.

Caustics

- Velocity field of droplets develops fold caustics as faster drops overtake smaller ones.





Velocity is triple-valued between fold caustics, and relative motion allows collisions.

Caustics in Turbulent Aerosols,
M. Wilkinson and B. Mehlig, *Europhysics Letters*, **71**, 186-92, (2005).

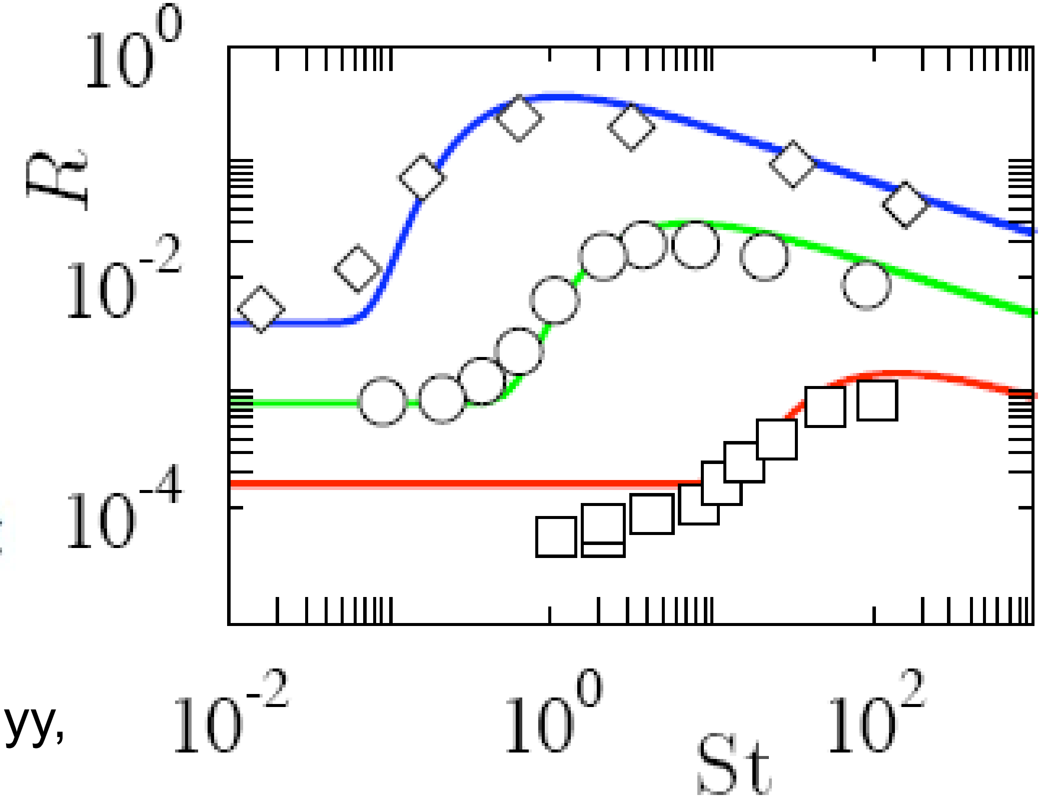
Caustics and collisions

- An interpolation formula gives a very good agreement with numerically computed collision rates: the collision process is 'activated' by the creation of caustics:

$$\ddot{\boldsymbol{r}} = \gamma[\boldsymbol{u}(\boldsymbol{r}(t), t) - \dot{\boldsymbol{r}}]$$

$$\mathrm{St} = \frac{1}{\gamma\tau}$$

$$R = R_\mathrm{a} + \exp(-A/\mathrm{St})R_\mathrm{g}$$



Caustic Activation of Rain Showers,
M. Wilkinson, B. Mehlig and V. Bezuglyy,
Phys. Rev. Lett., **97**, 048501, (2006).

Is turbulence significant?

http://www.usatoday.com/weather/wcumulus.htm

Physical parameters:

R.A.Shaw, *Ann. Rev. Fluid Mech.*, **35**, 183, (2003)

$$a \approx 10\mu\mathrm{m} \qquad n \approx 10^8\mathrm{m}^{-3}$$

$$\nu \approx 10^{-5}\mathrm{m}^2\mathrm{s}^{-1} \quad \mathcal{E} = 0.1\mathrm{m}^3\mathrm{s}^{-2}$$

From these values we find:

$$\eta \approx 3 \times 10^{-4}\mathrm{m} \qquad \tau \approx 10^{-2}\mathrm{s}$$

$$\gamma \approx 500\,\mathrm{s}^{-1}$$

$$\mathrm{St} = \frac{1}{\gamma\tau}$$

$$\ddot{\boldsymbol{r}} = \gamma[\boldsymbol{u}(\boldsymbol{r}(t), t) - \dot{\boldsymbol{r}}]$$

The Stokes numbers may approach unity: here $\mathrm{St} \approx 0.2$

Stokes numbers of order unity imply clustering and other effects, but this only happens for extreme conditions.

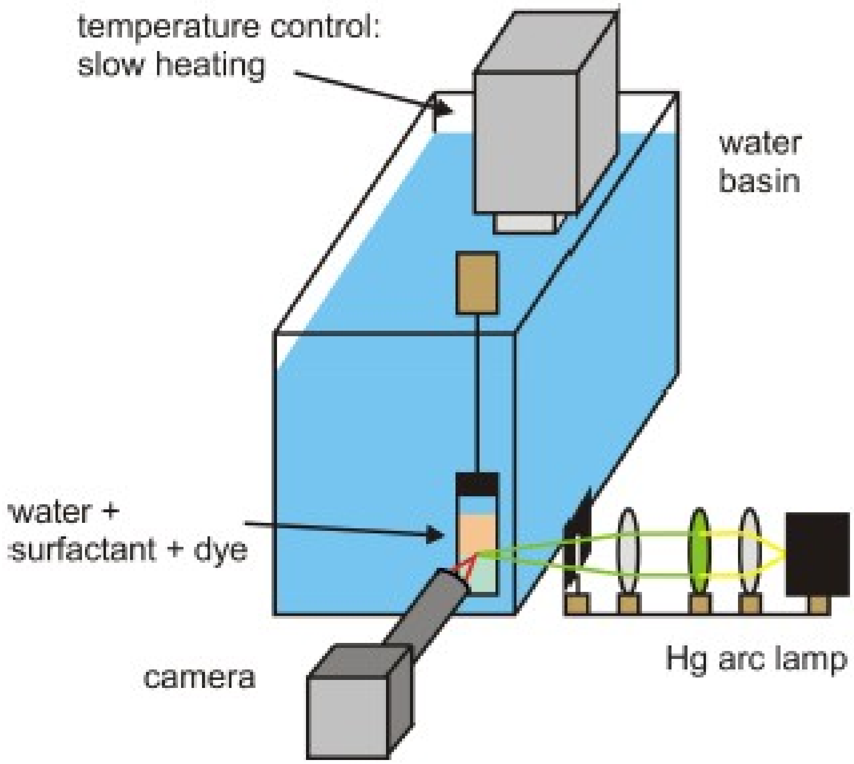
Laboratory models

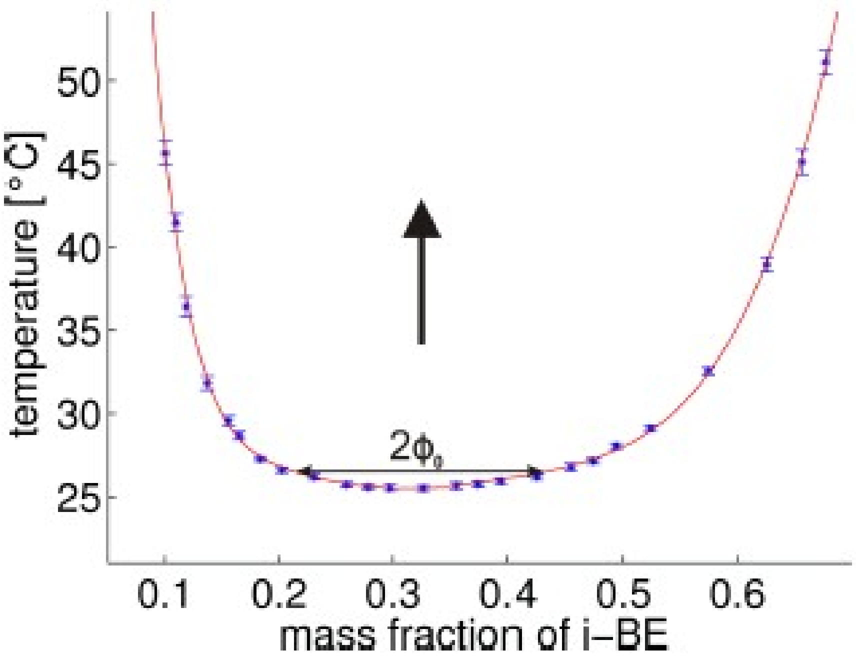
- There are studies of water vapour in large tanks. These are too small to produce rainfall.
- There are reports of rain cycles in liquid sodium reactor cooling circuits.
- An alternative approach, pursued by Juergen Vollmer and co-workers, uses two partly miscible liquids in a test-tube. The temperature is varied smoothly to decrease miscibility. Material comes out of solution in the form of small droplets. These grow and move towards the interface as 'rain'.

  The rain events are seen as in increase in turbidity of the mixture. They are roughly periodic in time, with a period which increases as the rate of cooling decreases.

The Goettingen experiment

Uses water and isobutoxyethanol, which are not fully miscible above $T_c = 25.51°\mathrm{C}$

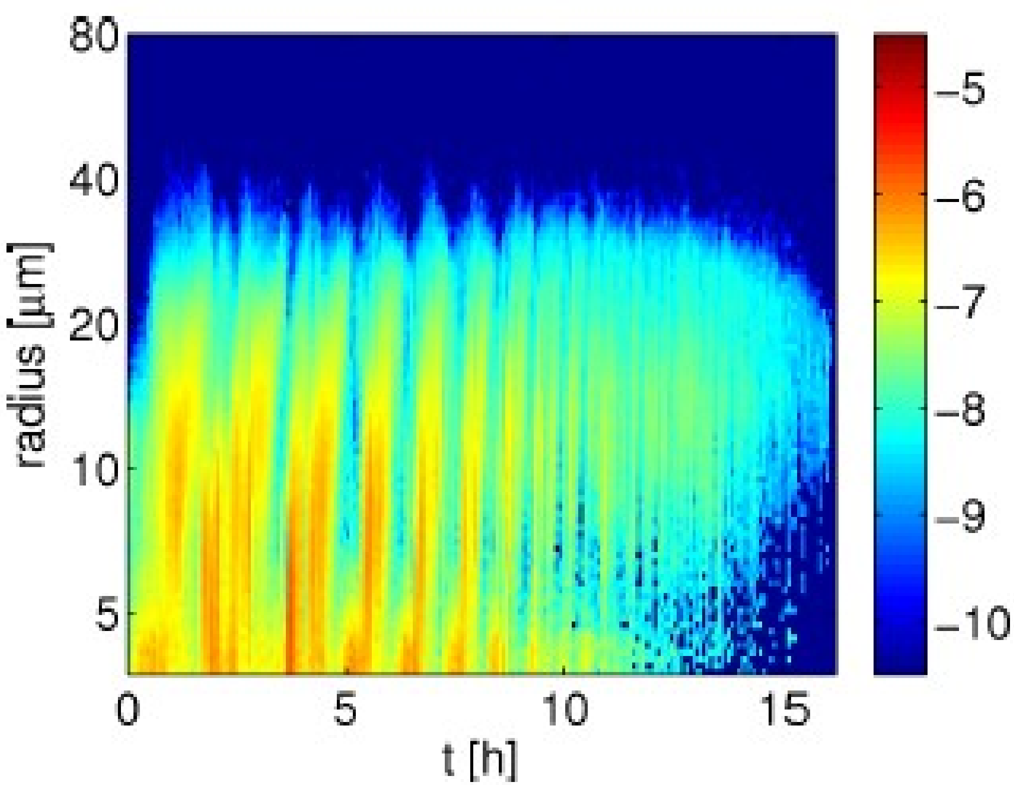




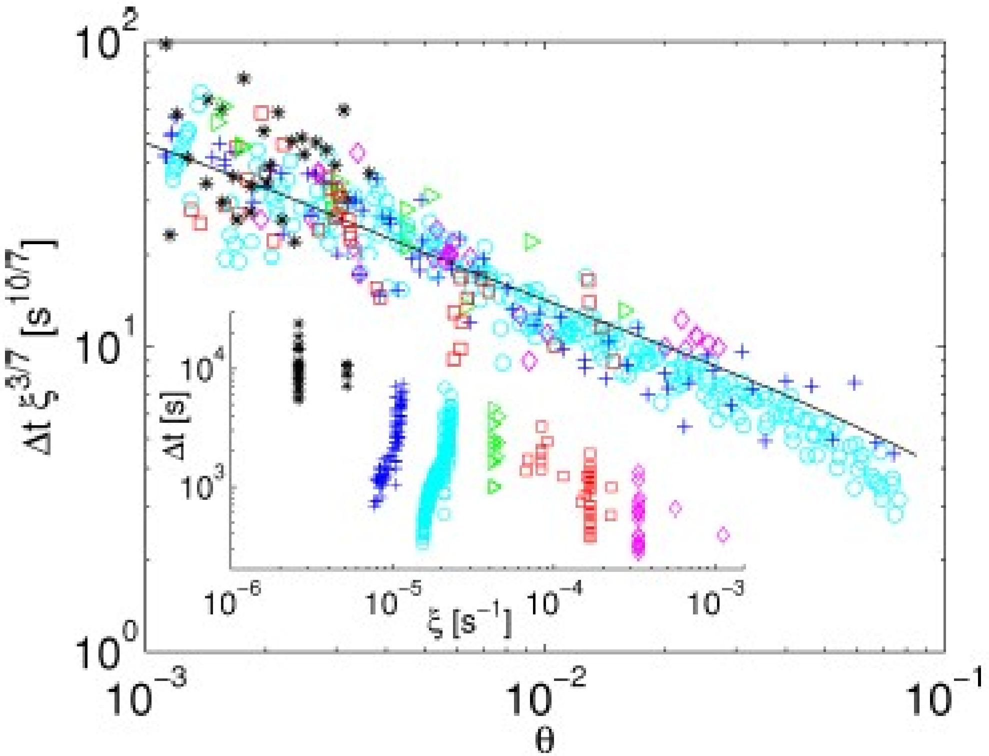
A camera and image-processing software determines the distribution of droplet sizes. Wide range of heating rates and temperatures, allow discriminating test of theories.

Goettingen experiment - results

Time variation of droplet size distribution:



Period as a function of cooling rate (lower layer):



$$\xi_{\mathrm{b}} \equiv \frac{1}{V_{\mathrm{b}}}\frac{\delta V_{\mathrm{d}}}{\delta T}\frac{\mathrm{d}T}{\mathrm{d}t} = \frac{1}{\Phi_{\mathrm{b}} - \Phi_{\mathrm{t}}}\frac{\mathrm{d}\Phi_{\mathrm{b}}}{\mathrm{d}T}\frac{\mathrm{d}T}{\mathrm{d}t} \qquad \theta = \frac{T - T_{\mathrm{c}}}{T_{\mathrm{c}}} \qquad \Delta t \sim \xi^{-3/7}$$

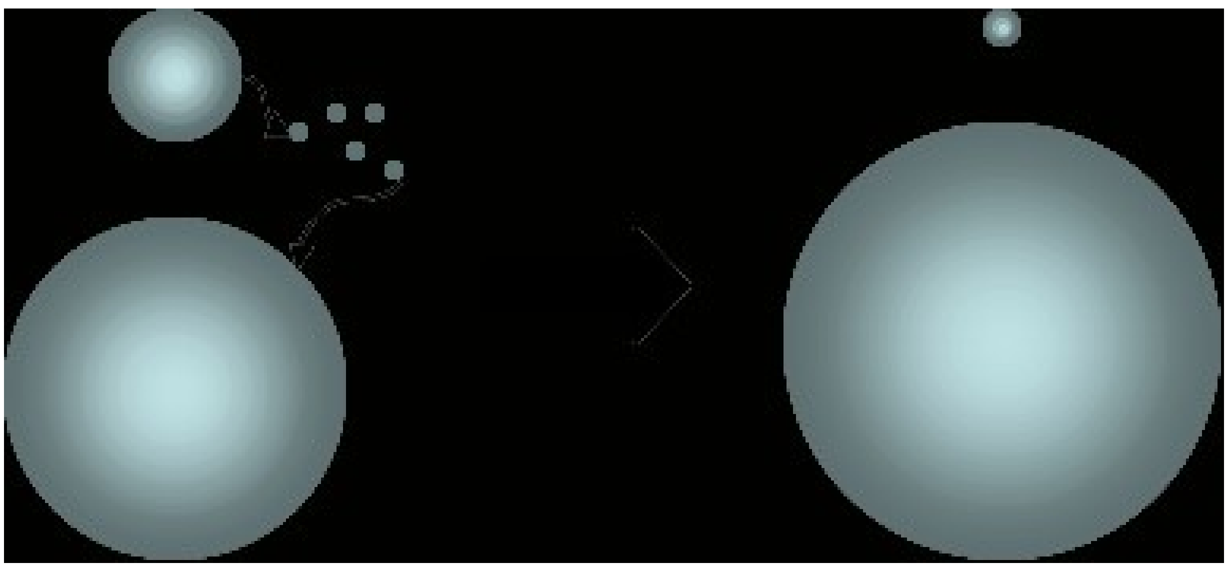
A model for test-tube rain

- After a 'rain' event, negligible supersaturation.
- Heating causes supersaturation, nucleation on dust.
- The microscopic droplets grow by *Ostwald ripening*.
- When droplets are large enough, they drift towards the interface at a significant rate.
- Larger droplets catch up with smaller ones, coalesce, then move faster. There is a finite-time singularity.
- The 'rainfall' sweeps away the excess material, cycle can start again.

  - *All this is similar to 'drizzle' from stratus clouds.*

Ostwald ripening

Consider water droplets in air. The smallest droplets have the highest Laplace pressure. They need a higher level of supersaturation to prevent evaporation. Water vapour from the smallest drops condenses onto the larger ones.



Laplace pressure:

$$\Delta p = \frac{2\gamma}{a}$$

The mechanism is more rapid than collisions due to Brownian motion of droplets, because molecules diffuse faster than droplets.

Lifshitz-Slezov theory

The surface of a droplet is in contact with vapour at a higher concentration:

$$\Phi(a) = \Phi_\mathrm{e} + \frac{\sigma}{a} \qquad \sigma = \frac{2\gamma v \Phi_\mathrm{e}}{kT} = \frac{2\gamma V_\mathrm{m}}{RT}\Phi_\mathrm{e}$$

The concentration obeys Laplace's equation. The concentration gradient at the surface leads to diffusive deposition, which increases the radius:

$$\Phi(r) = \Phi_\mathrm{e} + s + \frac{\sigma - sa}{r}$$

$$\frac{\mathrm{d}a}{\mathrm{d}t} = -D\frac{\partial \Phi}{\partial r}\bigg|_{r=a} = \frac{D\sigma}{a}\left(\frac{1}{a_0} - \frac{1}{a}\right)$$

Droplets smaller than $a_0 = \sigma / s$ shrink and evaporate.

Simplified model for droplet growth

The largest droplets are the important actors. Neglect the evaporation term, add a term for growth by sweeping up smaller particles. Vertical velocity due to buoyancy is:

$$u = \frac{2}{9}\frac{\Delta\rho g}{\rho\nu}a^2 \equiv \kappa a^2$$

Sweeping up material leads to additional term in equation for growth of radius:

$$\frac{\mathrm{d}a}{\mathrm{d}t} = \frac{D\sigma}{a^2} + \frac{\epsilon}{4}\kappa a^2 \xi t$$

ϵ =collision efficiency=unity

Initial growth is by Ostwald ripening, leading to

$$\langle a(t)\rangle = \left(\frac{4}{9}D\sigma t\right)^{1/3}$$

Prediction of period

Growth by Ostwald ripening, collisional term is comparable, at time t_1 .

$$a^3 \sim D\sigma t$$

Then a runaway growth with finite-time singularity, time t_2 .

$$\Delta t = t_1 + t_2$$

Growth equation:

$$\frac{\mathrm{d}a}{\mathrm{d}t} = \frac{D\sigma}{a^2} + \frac{\epsilon}{4}\kappa a^2 \xi t$$

Terms are comparable when:

$$\frac{D\sigma}{\kappa\xi} = a_1^4 t_1 \qquad a_1^3 \sim D\sigma t_1$$

$$t_1 = \left(\frac{1}{D\sigma(\kappa\xi)^3}\right)^{1/7} \qquad t_1 \sim t_2$$

Comparison with experiment

Data on water/i-BE system is available.

$$\sigma = \frac{2\gamma v \Phi_e}{kT} = \frac{2\gamma V_m}{RT}\Phi_e$$

$$\Delta t \approx \alpha \left(\frac{1}{D\sigma\kappa^3}\right)^{1/7} \xi^{-3/7}$$

$$u = \frac{2}{9}\frac{\Delta\rho g}{\rho\nu}a^2 \equiv \kappa a^2$$

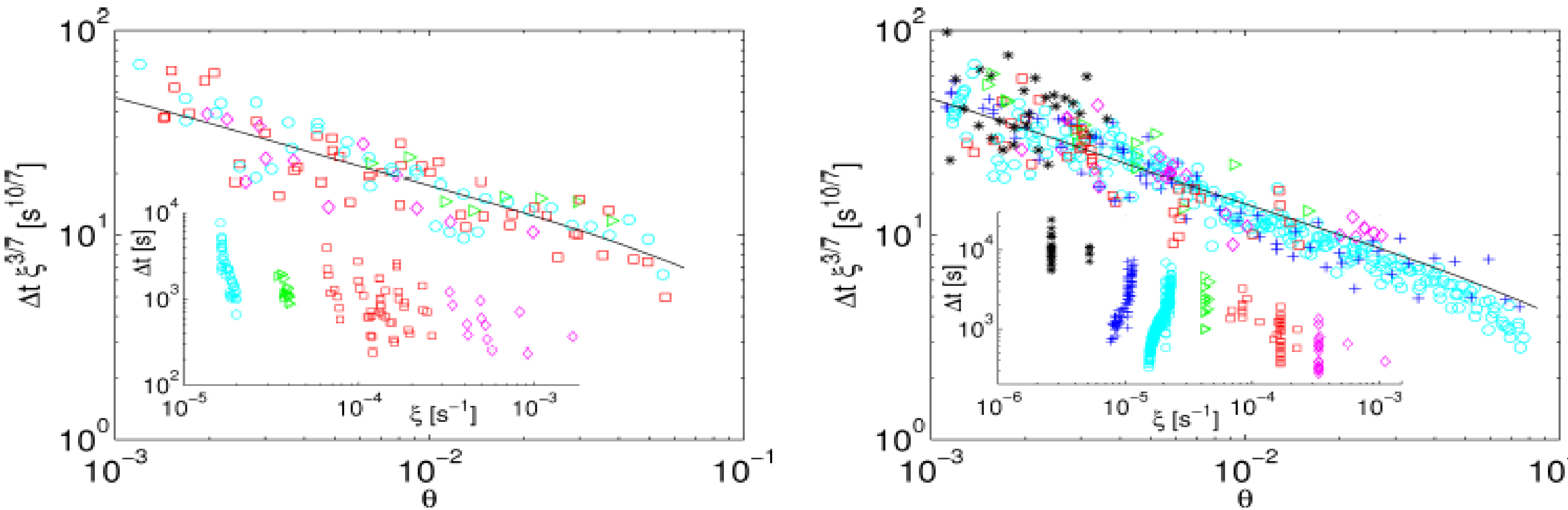


FIG. 3: Plot of temperature dependence of $\Delta t\, \xi^{3/7}$, for the upper (a) and the lower (b) layer. The solid line is the theoretical prediction, eq. (8), with $\alpha = 0.71$. When plotting Δt against ξ (insets) the scatter identifies the considerable dependence of the period on the material parameters. The colors and symbols encode different heating rates ξ: black stars, $\xi < 6 \times 10^{-6}\,\mathrm{s}^{-1}$; blue crosses, $\xi < 1.3 \times 10^{-5}\,\mathrm{s}^{-1}$; cobalt circles, $\xi < 3 \times 10^{-5}\,\mathrm{s}^{-1}$; green triangles, $\xi < 6 \times 10^{-5}\,\mathrm{s}^{-1}$; red squares, $\xi < 3 \times 10^{-4}\,\mathrm{s}^{-1}$; and magenta diamonds, $\xi > 3 \times 10^{-4}\,\mathrm{s}^{-1}$.

Is the theory universally valid?

Agreement is equally good for methanol and hexane, with the same power-law dependence on temperature (a consequence of universality of critical exponents).

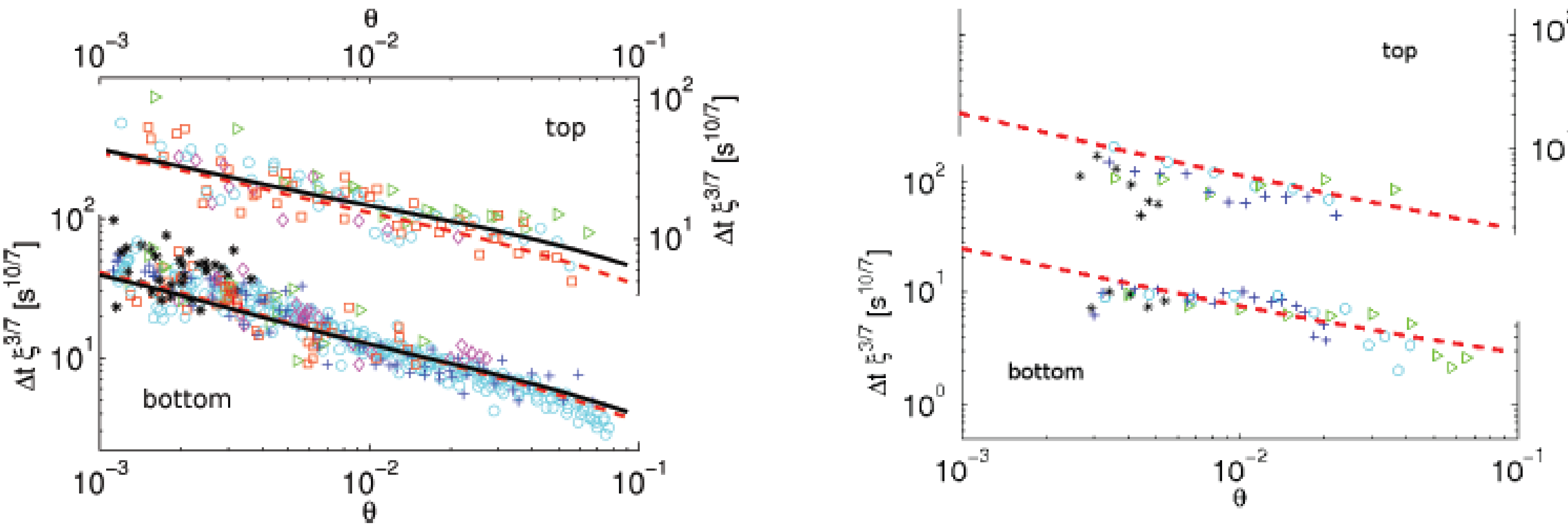


FIG. 2. Plot of temperature dependence of $\Delta t\, \xi^{3/7}$, for the upper and the lower layer of a mixtures of (a) IBE and water and (b) methanol and hexane, respectively. The respective solid lines are the theoretical prediction, eq. (2), with (a) $\alpha = 0.71$ and (b) $\alpha = 1$. The colors and symbols encode different heating rates ξ: black stars, $\xi < 6 \times 10^{-6}\,\mathrm{s}^{-1}$; blue crosses, $\xi < 1.3 \times 10^{-5}\,\mathrm{s}^{-1}$; cobalt circles, $\xi < 3 \times 10^{-5}\,\mathrm{s}^{-1}$; green triangles, $\xi < 6 \times 10^{-5}\,\mathrm{s}^{-1}$; red squares, $\xi < 3 \times 10^{-4}\,\mathrm{s}^{-1}$; and magenta diamonds, $\xi > 3 \times 10^{-4}\,\mathrm{s}^{-1}$. In response to the increase of θ the lag time typically drops by an order of magnitude, ranging between a few minutes for the largest values of ξ till several hours for small values of ξ and θ. (Raw data are shown in the supplementary material.)

Critical exponents

The interfacial tension, density contrast and mutual diffusion coefficient all vanish at the critical point, with universal exponents in the three-dimensional Ising universality class:

$$\Delta\rho \sim \theta^{\beta} \qquad \beta \approx 0.327$$

$$\gamma \sim \theta^{2\nu} \qquad \nu \approx 0.630$$

$$D \sim \theta^{\gamma} \qquad \gamma \approx 1.237$$

This explains the observed power-law dependence of the temperature-dependence of the coefficient:

$$\xi^{3/7}\Delta t \sim \theta^{-(3\beta+2\nu+\gamma)/7} \approx \theta^{-0.498}$$

Any lessons for real clouds?

Ostwald ripening operates in real clouds. It gives a robust timescale, depending only upon temperature.

$$\langle a(t) \rangle = \left(\frac{4}{9} D\sigma t \right)^{1/3}$$

What does this imply for real clouds (at $10^\circ\mathrm{C}$)?

$$D = 2.4 \times 10^{-5}\,\mathrm{m^2 s^{-1}} \qquad \gamma = 7.4 \times 10^{-2}\,\mathrm{Nm^{-1}}$$

$$V_\mathrm{m} = 1.8 \times 10^{-5}\,\mathrm{m}^3 \qquad \Phi_\mathrm{e} = 1.2 \times 10^{-2}$$

$$D_\mathrm{eff} = \frac{D}{1+\Theta} \qquad \Theta = \frac{D_\mathrm{v}}{D_\mathrm{th}} \frac{\bar{m}_\mathrm{a}}{\bar{m}_\mathrm{w}} \frac{L}{RT} \frac{L}{C_p T} \frac{\rho_\mathrm{eq}}{\rho_\mathrm{a}} \qquad \Theta \approx 1.8$$

Growth to $50\,\mu\mathrm{m}$ takes days by Ostwald ripening. This is too slow to be significant. Another mechanism is required.

Aerosols in Rayleigh-Benard cells

Dispersion of droplet sizes may occur due to fluctuations of supersaturation. There is a vast, overparametrised literature on 'entrainment'.

However, the essential features are contained in analysis of an aerosol in a Rayleigh-Benard cell. Rate of increase of variance of droplet radius is related to heat flux:

$$\frac{\mathrm{d}\langle \Delta a^2 \rangle}{\mathrm{d}t} \sim \frac{\Delta T^2 Q}{Ch} \Lambda^2 \qquad \Lambda = \frac{1}{4\pi n_0 a_0^2} \frac{\partial \phi_{\mathrm{eq}}}{\partial T}$$

Summary

We have the first quantitative description of a precipitation cycle. There is a surprising scaling with the cooling rate:

$$\Delta t \approx \alpha \left(\frac{1}{D\sigma\kappa^3} \right)^{1/7} \xi^{-3/7}$$

The theory involves Ostwald ripening, which allows competitive growth of droplets without collisions.

In atmospheric clouds, collisions are not sufficiently frequent and Ostwald ripening is too slow.

An alternative mechanism for convective ripening appears very promising.