Competitive Evaporation of Multiple Sessile Droplets

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Droplets are everywhere . . .

. . . but are rarely alone!

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- However, previous experimental and theoretical studies (*e.g.* Lacasta *et al.* 1998, Schäfle *et al.* 1998, Kokalj *et al.* 2010, Sokuler *et al.* 2010, Carrier *et al.* 2016, Castanet *et al.* 2016, Shaikeea *et al.* 2016, Hatte *et al.* 2019, Khilifi *et al.* 2019) and work on closely related problems (*e.g.* Laghezza *et al.* 2016, Michelin *et al.* 2018) have shown the occurrence of **shielding** due to the presence of other droplets.

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- However, there has been relatively little work on the interactions between **multiple** droplets.
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- In this talk we describe two different approaches to this problem, **exact solutions for one and two thin droplets in two dimensions**, and **an asymptotic solution for thin droplets in three dimensions**.

The Diffusion-Limited Model

- The vapour concentration c satisfies $\nabla^2 c = 0$ subject to $c \to c_\infty$ far from the droplet(s),

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- Assuming that the droplet(s) are sufficiently small, they have quasi-static profile

$$\frac{\theta}{2}(R^2 - x^2) \quad \text{with area} \quad \frac{2\theta R^2}{3} \quad \text{in two dimensions, and}$$

$$\frac{\theta}{2}(R^2 - r^2) \quad \text{with volume} \quad \frac{\pi\theta R^3}{4} \quad \text{in three dimensions.}$$

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- The evolution of the droplet(s) satisfies

$$\frac{\mathrm{d}}{\mathrm{d}t}(\text{area or volume of droplet}) = -\frac{1}{\rho}\int_{\text{surface area}} J \,\mathrm{d}S.$$

Part 1:

Exact solutions for one and two thin droplets in two dimensions

Schofield, Wray, Pritchard and Wilson,
"The shielding effect extends the lifetimes of two-dimensional sessile droplets", to appear in *J. Eng. Math.* (2020)

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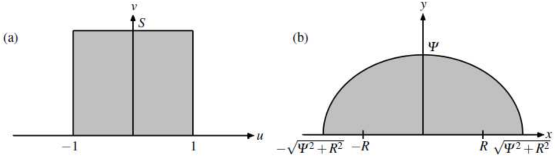


Exact Solution of the One-Droplet Problem

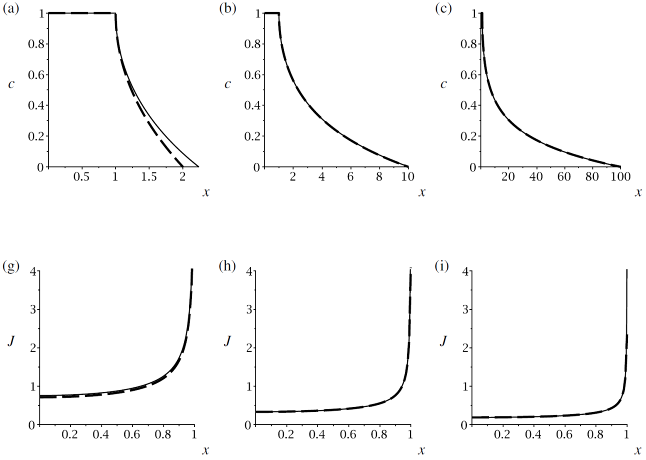
The exact solution in the semi-ellipse is

$$c(x, y) = 1 - \frac{1}{\operatorname{arcsinh}(\Psi/R)} \Im \left[\arccos \left(-\frac{z}{R} \right) \right],$$

and hence

$$J(x) = -\frac{\partial c}{\partial y}(x, 0) = \frac{1}{\operatorname{arcsinh}(\Psi/R)} \frac{1}{\sqrt{R^2 - x^2}} \quad \text{for} \quad |x| < R.$$

Numerical Validation



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- We consider **constant radius (CR)**, **constant angle (CA)**, and **slick-slide (SS)** modes.

Constant Radius (CR) Mode

$$R(t) \equiv 1, \quad \theta(t) = 1 - \frac{3\pi}{2\operatorname{arcsinh}\Psi} t, \quad A(t) = \frac{2}{3}\left[1 - \frac{3\pi}{2\operatorname{arcsinh}\Psi} t\right]$$

Lifetime of droplet

$$t_{\mathrm{CR}} = \frac{2}{3\pi} \operatorname{arcsinh}\Psi$$

Constant Angle (CA) Mode

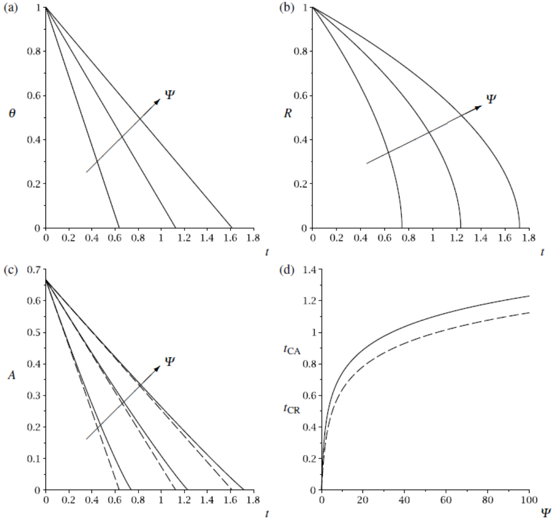
$$t = \frac{2}{3\pi}\left[\text{arcsinh}\Psi - R^2(t)\text{arcsinh}\left(\frac{\Psi}{R(t)}\right) + \Psi\left(\sqrt{\Psi^2+1} - \sqrt{\Psi^2+R^2(t)}\right)\right],$$

$$\theta(t) \equiv 1, \quad A(t) = \frac{2R^2(t)}{3}$$

Lifetime of droplet

$$t_{\text{CA}} = \frac{2}{3\pi}\left[\text{arcsinh}\Psi + \Psi\left(\sqrt{\Psi^2+1} - \Psi\right)\right]$$

CR and CA Modes



Stick-Slide (SS) Mode

In the pinned phase $0 < t < t^*$ droplet is in the CR mode,

$$t^* = \frac{2(1-\theta^*)\text{arcsinh}\Psi}{3\pi},$$

while in the slide phase $t^* < t < t_{SS}$,

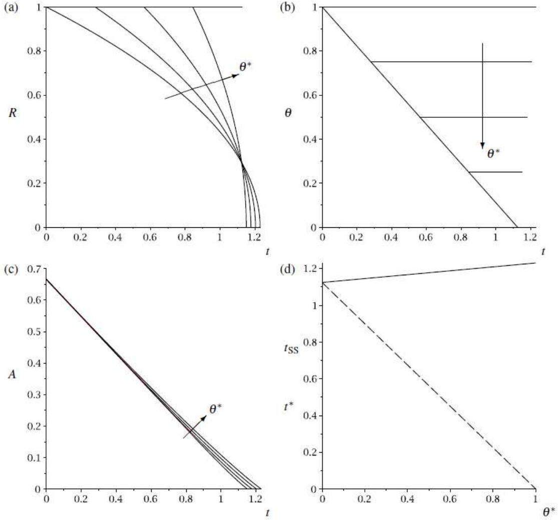
$$t = t^* + \frac{2\theta^*}{3\pi}\left[\text{arcsinh}\Psi - R^2(t)\text{arcsinh}\left(\frac{\Psi}{R(t)}\right)\right.$$

$$\left. + \Psi\left(\sqrt{\Psi^2+1} - \sqrt{\Psi^2 + R^2(t)}\right)\right].$$

Lifetime of droplet

$$t_{SS} = \frac{2}{3\pi}\left[\text{arcsinh}\Psi + \theta^*\Psi\left(\sqrt{\Psi^2+1} - \Psi\right)\right]$$

SS Mode



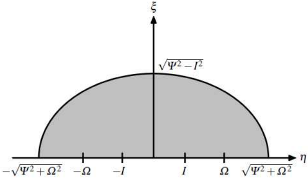
Asymptotic Limit of a Large Domain, $\Psi \gg R$

$$t_{\text{CR}} = \frac{2}{3\pi} \ln(2\Psi) + O\left(\frac{1}{\Psi^2}\right)$$

$$t_{\text{CA}} = \frac{2}{3\pi} \ln(2\Psi) + \frac{1}{3\pi} + O\left(\frac{1}{\Psi^2}\right)$$

$$t_{\text{SS}} = \frac{2}{3\pi} \ln(2\Psi) + \frac{\theta^*}{3\pi} + O\left(\frac{1}{\Psi^2}\right)$$

Exact Solution of the Two-Droplet Problem



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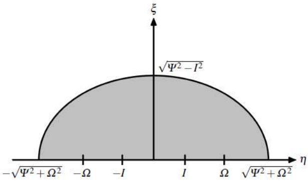


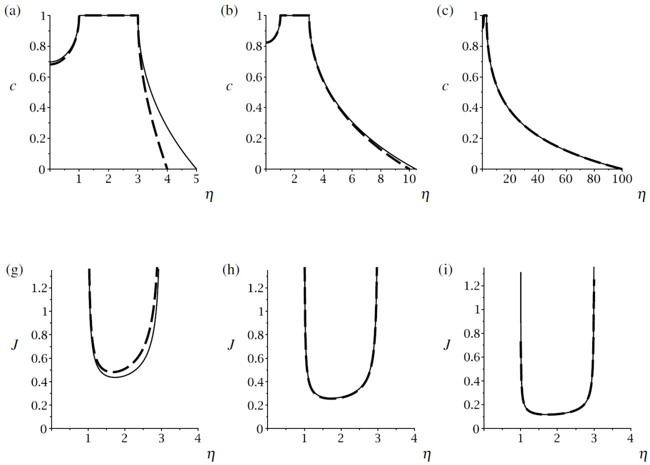
The exact solution is

$$c(\eta,\xi) = 1 - \frac{1}{\operatorname{arcsinh}\left(\Psi/\sqrt{\Omega^2 - I^2}\right)} \Im\left[\arccos\left(-\sqrt{\frac{\zeta^2 - I^2}{\Omega^2 - I^2}}\right)\right],$$

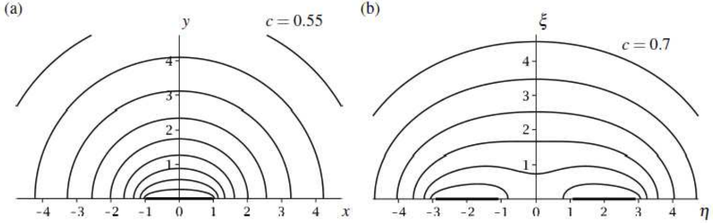
and hence

$$J(\eta) = -\frac{\partial c}{\partial \xi}(\eta, 0) = \frac{1}{\operatorname{arcsinh}\left(\Psi/\sqrt{\Omega^2 - I^2}\right)} \frac{\eta}{\sqrt{\Omega^2 - \eta^2}\sqrt{\eta^2 - I^2}}.$$

Numerical Validation



Exact Solution for Vapour Concentration



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In the two-droplet problem, there is a much richer variety of possible behaviours, including

- constant-inner-and-outer-contact-line (CIO) mode,
- constant-angle-centred (CAC) mode,
- constant-angle and constant-inner-contact-lines (CAI) mode,
- constant-angle and constant-outer-contact-line (CAO) mode.

CIO Mode

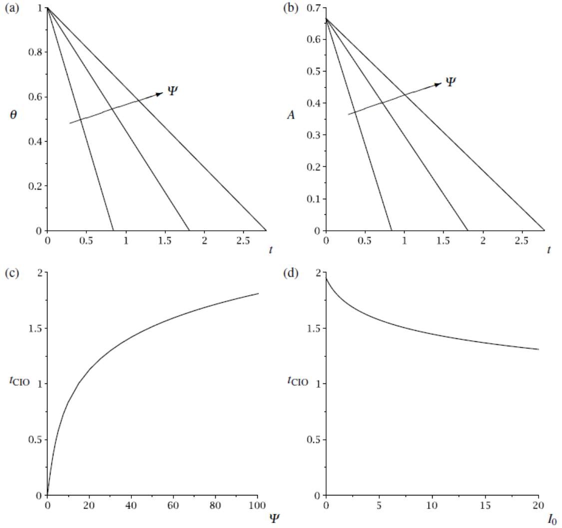
$$\theta(t) = 1 - \frac{3\pi}{4\text{arcsinh}\left(\Psi/\sqrt{\Omega_0^2 - I_0^2}\right)} t,$$

$$A = \frac{(\Omega_0 - I_0)^2}{6}\left[1 - \frac{3\pi}{4\text{arcsinh}\left(\Psi/\sqrt{\Omega_0^2 - I_0^2}\right)} t\right]$$

Lifetime of a pair of droplets

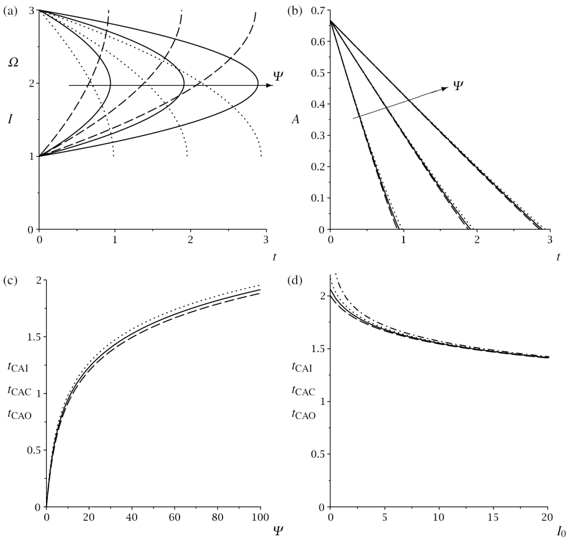
$$t_{\text{CIO}} = \frac{4\text{arcsinh}\left(\Psi/\sqrt{\Omega_0^2 - I_0^2}\right)}{3\pi}$$

CIO Mode



(a)
1
0.8
0.6
0.4
0.2
0
θ
Ψ
0
0.5
1
1.5
2
2.5
t
(b)
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
A
Ψ
0
0.5
1
1.5
2
2.5
t
(c)
2
1.5
1
0.5
0
tCIO
0
20
40
60
80
100
Ψ
(d)
2
1.5
1
0.5
0
tCIO
0
5
10
15
20
I0

CAC, CAI and CAO Modes



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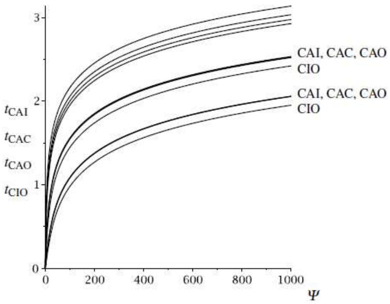

Asymptotic Limit of a Large Domain, $l_0 \ll 1 \ll \Psi$

$$
\begin{aligned}
t_{\mathrm{CIO}} &= \frac{4}{3\pi}\ln(\Psi) - \frac{2l_0}{3\pi} + O\left(l_0{}^2, \frac{1}{\Psi^2}\right) \\
t_{\mathrm{CAC}} &= \frac{4}{3\pi}\ln(\Psi) + \frac{1}{3\pi} - \frac{2l_0}{3\pi} + O\left(l_0{}^2, \frac{1}{\Psi^2}\right) \\
t_{\mathrm{CAI}} &= \frac{4}{3\pi}\ln(\Psi) + \frac{2}{3\pi} - \frac{4l_0}{3\pi} + O\left(l_0{}^2 \log l_0, \frac{1}{\Psi^2}\right) \\
t_{\mathrm{CAO}} &= \frac{4}{3\pi}\ln(\Psi) + \frac{2}{\pi}\left(1 - \frac{4}{3}\ln 2\right) + \frac{4l_0}{3\pi}(1 - 2\ln 2) + O\left(l_0{}^2, \frac{1}{\Psi^2}\right)
\end{aligned}
$$

Asymptotic Limit of a Large Domain, $1 \ll l_0 \ll \Psi$

$$
\begin{aligned}
t_{\mathrm{CIO}} &= \frac{4}{3\pi} \ln\left(\frac{\Psi}{\sqrt{l_0}}\right) - \frac{2}{3\pi l_0} + O\left(\frac{1}{l_0^2}, \frac{l_0}{\Psi^2}\right) \\
t_{\mathrm{CAC}} &= \frac{4}{3\pi} \ln\left(\frac{\Psi}{\sqrt{l_0}}\right) + \frac{1}{3\pi} - \frac{2}{3\pi l_0} + O\left(\frac{1}{l_0^2}, \frac{l_0}{\Psi^2}\right) \\
t_{\mathrm{CAI}} &= \frac{4}{3\pi} \ln\left(\frac{\Psi}{\sqrt{l_0}}\right) + \frac{1}{3\pi} - \frac{4}{9\pi l_0} + O\left(\frac{1}{l_0^2}, \frac{l_0}{\Psi^2}\right) \\
t_{\mathrm{CAO}} &= \frac{4}{3\pi} \ln\left(\frac{\Psi}{\sqrt{l_0}}\right) + \frac{1}{3\pi} - \frac{8}{9\pi l_0} + O\left(\frac{1}{l_0^2}, \frac{l_0}{\Psi^2}\right)
\end{aligned}
$$

Comparison Between Lifetimes

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- Single droplet:

$$\hat{t}_{\mathrm{single}} \sim \frac{2}{3\pi} \ln \left(\frac{2\hat{\Psi}}{\hat{R}_0} \right) \hat{T} \qquad \text{where} \qquad \hat{T} = \frac{\hat{\rho}\hat{\theta}_0 \hat{R}_0^2}{\hat{D}(\hat{c}_{\mathrm{sat}} - \hat{c}_\infty)}$$

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- Pair of droplets with same surface area:

$$\hat{t}_{\text{area}} \sim \frac{1}{3\pi} \ln\left(\frac{2\hat{\Psi}}{\hat{R}_0}\right) \hat{T} \sim \frac{\hat{t}_{\text{single}}}{2}$$

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- Closely spaced pair of droplets with same cross-sectional area:

$$\hat{t}_{\text{close}} \sim \frac{2}{3\pi} \left[\ln\left(\frac{2\hat{\Psi}}{\hat{R}_0}\right) - \frac{1}{2}\ln 2\right] \hat{T}$$

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- Widely separated pair of droplets with same cross-sectional area:

$$\hat{t}_{\mathrm{wide}} \sim \frac{2}{3\pi} \left[\ln\left(\frac{2\hat{\Psi}}{\hat{R}_0}\right) - \frac{1}{2}\ln\left(2^{3/2}\frac{\hat{l}_0}{\hat{R}_0}\right)\right] \hat{T}$$

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- The solutions allow us to quantify the shielding effects the droplets have on each other and, in particular, how they extend the lifetime of the droplets.
- But remember that this is for a slightly artificial two-dimensional problem ... hence motivating Part 2 of this talk on the three-dimensional problem!

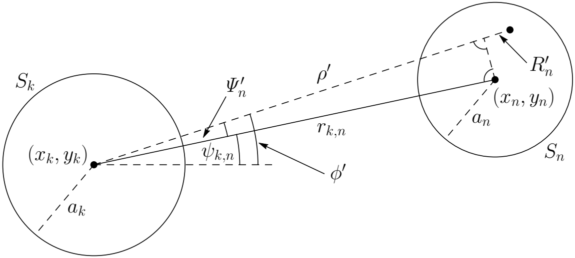
Part 2:

An asymptotic solution for thin droplets in three dimensions

Wray, Duffy and Wilson,
"Competitive evaporation of multiple sessile droplets",
to appear in *J. Fluid Mech.* (2020)

Formulation

Consider N thin droplets on the substrate $z = 0$ whose surfaces S_k have radii a_k and centres (x_k, y_k) for $k = 1, 2, \ldots, N$ with local fluxes J_k and integral fluxes F_k.



Solution

Using a series of integral transformations, Fabrikant (1985) showed that

$$J_k = J_0 \left[1 - \frac{1}{2\pi} \sum_{\substack{n=1,\\ n\neq k}}^{N} \iint_{S_n} \frac{\sqrt{\rho'^2 - a_k^2}\, J_n(\rho', \phi')\, \rho' \mathrm{d}\rho' \, \mathrm{d}\phi'}{\rho^2 + \rho'^2 - 2\rho\rho' \cos(\phi - \phi')} \right]$$

for $k = 1, 2, \ldots, N$, where

$$J_0 = \frac{2}{\pi\sqrt{a_k^2 - \rho^2}}$$

is the flux from the k^{th} droplet in isolation.

The integral flux from the k^{th} droplet (i.e. the integral of J_k over S_k), denoted by

$$F_k = \iint_{S_k} J_k(\rho, \phi)\, \rho\, \mathrm{d}\rho\, \mathrm{d}\phi,$$

is given by

$$F_k = 4a_k - \frac{2}{\pi} \sum_{\substack{n=1,\\ n\neq k}}^{N} \iint_{S_n} J_n\left(\rho', \phi'\right) \arcsin\left(\frac{a_k}{\rho'}\right)\, \rho'\, \mathrm{d}\rho' \mathrm{d}\phi'$$

for $k = 1, 2, \ldots, N$.

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- This is a set of N linear algebraic equations for the F_k which may be solved exactly for any given configuration of droplets.

- Similarly,

$$J_k = J_0 \left[1 - \frac{1}{2\pi} \sum_{\substack{n=1, \\ n \neq k}}^{N} \frac{F_n \sqrt{r_{k,n}^2 - a_k^2}}{\rho^2 + r_{k,n}^2 - 2\rho r_{k,n} \cos(\phi - \psi_{k,n})} \right]$$

for $k = 1, 2, \ldots, N$.

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  for $k = 1, 2, \ldots, N$.
- With the integral fluxes F_k determined, this gives the fluxes J_k explicitly.
- We now analyse one particular configuration, namely a pair of identical droplets.

A Pair of Identical Droplets

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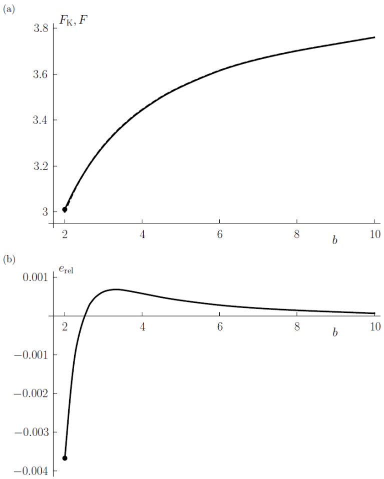
- The flux from the droplet centred at $(-b/2, 0)$ is

$$J_1 = J_0 \left[1 - \frac{F\sqrt{b^2 - a^2}}{2\pi \left(\rho^2 + b^2 - 2\rho b \cos\phi \right)} \right],$$

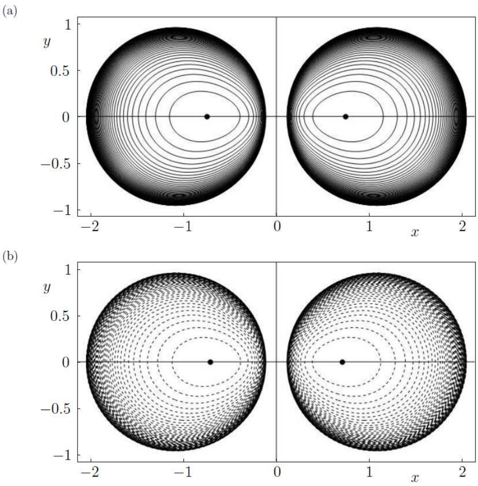
with a corresponding expression for the other droplet.

Numerical Validation



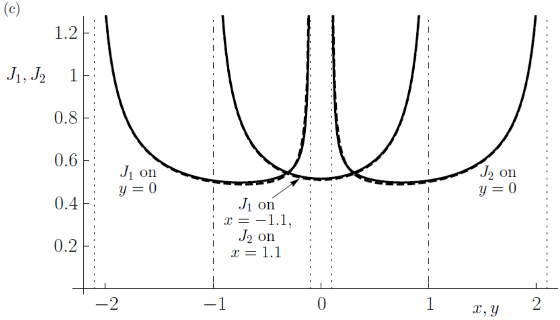
Exact and asymptotic solutions for F.

Numerical Validation



Exact and asymptotic solutions for J_1.

Numerical Validation



Exact and asymptotic solutions for J_1.

Evolutions and Lifetimes

Scale and non-dimensionalise according to

$$\theta_k = \theta_{\text{ref}}\hat{\theta}_k, \quad h_k = a_{\text{ref}}\theta_{\text{ref}}\hat{h}_k, \quad V_k = a_{\text{ref}}^3\theta_{\text{ref}}\hat{V}_k, \quad t = \frac{\rho_{\text{fluid}}\, a_{\text{ref}}^2\theta_{\text{ref}}}{D\,(c_{\text{sat}} - c_\infty)}\hat{t}.$$

For sufficiently small droplets

$$h_k = \frac{a_k\theta_k}{2}\left(1 - \frac{\rho^2}{a_k^2}\right), \quad V_k = 2\pi\int_0^{a_k} h_k\,\rho\,\mathrm{d}\rho = \frac{\pi a_k^3\theta_k}{4}.$$

Evolutions and Lifetimes

Evolution of $a_k = a_k(t)$ and/or $\theta_k = \theta_k(t)$ and $V_k = V_k(t)$ satisfies

$$\frac{\mathrm{d}}{\mathrm{d}t}\left(a_k^3\theta_k\right) = -\frac{4F_k}{\pi}.$$

For a pair of identical droplets with radii $a = a(t)$, contact angle $\theta = \theta(t)$ and volume $V = V(t)$ this gives

$$\frac{\mathrm{d}}{\mathrm{d}t}\left(a^3\theta\right) = -\frac{16a}{\pi\left(1 + \frac{2}{\pi}\arcsin\frac{a}{b}\right)}.$$

Constant Radius (CR) Mode

Setting $a \equiv \bar{a}$ gives

$$\frac{\mathrm{d}\theta}{\mathrm{d}t} = -\frac{16}{\pi \bar{a}^2 \left(1 + \frac{2}{\pi} \arcsin \frac{\bar{a}}{b}\right)},$$

and hence

$$\theta = \bar{\theta} - \frac{16t}{\pi \bar{a}^2 \left(1 + \frac{2}{\pi} \arcsin \frac{\bar{a}}{b}\right)}, \quad V = \frac{\pi \bar{a}^3 \bar{\theta}}{4} - \frac{4\bar{a}t}{1 + \frac{2}{\pi} \arcsin \frac{\bar{a}}{b}}.$$

In particular, the lifetime of the droplets in the CR mode is

$$t_{\mathrm{CR}} = t_{\mathrm{CR}\infty} \left(1 + \frac{2}{\pi} \arcsin \frac{\bar{a}}{b}\right),$$

where $t_{\mathrm{CR}\infty} = \pi \bar{a}^2 \bar{\theta}/16$ is the lifetime of the droplets in isolation.

Constant Angle (CA) Mode

Setting $\theta \equiv \bar{\theta}$ gives

$$\frac{\mathrm{d}a}{\mathrm{d}t} = -\frac{16}{3\pi a\bar{\theta}\left(1 + \frac{2}{\pi}\arcsin\frac{a}{b}\right)},$$

and hence $V = \pi a^3\bar{\theta}/4$ and

$$t = \frac{3\pi\bar{\theta}}{32}\left[\hat{a}^2 + \frac{1}{\pi}\left\{\hat{a}\sqrt{b^2 - \hat{a}^2} - \left(b^2 - 2\hat{a}^2\right)\arcsin\frac{\hat{a}}{b}\right\}\right]_{\hat{a}=a}^{\hat{a}=\bar{a}}.$$

In particular, the lifetime of the droplets in the CA mode is

$$t_{\mathrm{CA}} = t_{\mathrm{CA}\infty}\left[1 + \frac{1}{\pi}\left\{\sqrt{\frac{b^2}{\bar{a}^2} - 1} - \left(\frac{b^2}{\bar{a}^2} - 2\right)\arcsin\frac{\bar{a}}{b}\right\}\right],$$

where $t_{\mathrm{CA}\infty} = 3\pi\bar{a}^2\bar{\theta}/32$ is the lifetime of droplets in isolation.

Stick-Slide (SS) Mode

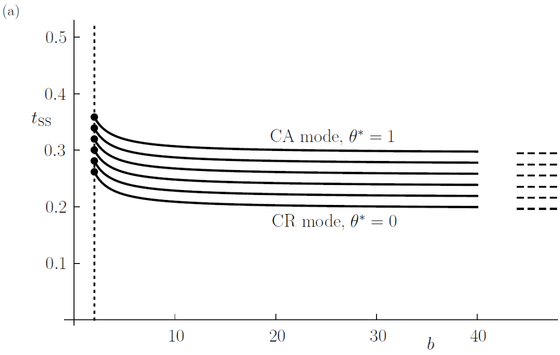
A CR phase with $a \equiv \bar{a}$ is followed by a CA phase with $\theta \equiv \theta^\star$.

In particular, the lifetime of the droplets in the SS mode is

$$t_{\mathrm{SS}} = \left[1 + \frac{\left(2\bar{a}^2\left(2\bar{\theta} + \theta^\star\right) - 3b^2\theta^\star\right)\arcsin(\bar{a}/b) + 3\bar{a}\theta^\star\sqrt{b^2 - \bar{a}^2}}{\pi\bar{a}^2\left(2\bar{\theta} + \theta^\star\right)}\right] t_{\mathrm{SS}\infty},$$

where $t_{\mathrm{SS}\infty} = \pi\bar{a}^2(2\bar{\theta} + \theta^*)/32$ is the lifetime of droplets in isolation.

Droplet Lifetimes



Lifetimes of a pair of droplets evaporating in the CR, CA and SS modes.

Radially Integrated Flux

The radially integrated flux from the k^{th} droplet is given by

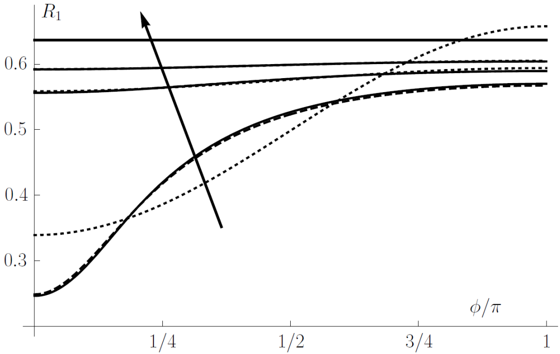
$$R_k = \int_0^{a_k} J_k(\rho, \phi)\, \rho \, \mathrm{d}\rho,$$

and for a pair of identical droplets

$$R_1 = R_0 \left[1 - \frac{F\sqrt{1-k^2}}{2\pi a \sin\phi} \mathcal{I} \left\{ \frac{\log\left[-\left(k\mathrm{e}^{-i\phi} + \sqrt{k^2\mathrm{e}^{-2i\phi} - 1} \right) \right]}{\sqrt{k^2\mathrm{e}^{-2i\phi} - 1}} \right\} \right],$$

where $k = a/b$ and $R_0 = 2a/\pi$ is the radially integrated flux from an isolated droplet.

Radially Integrated Flux



Exact (solid) and approximate (dashed) radially integrated flux.

The Coffee-Ring Effect

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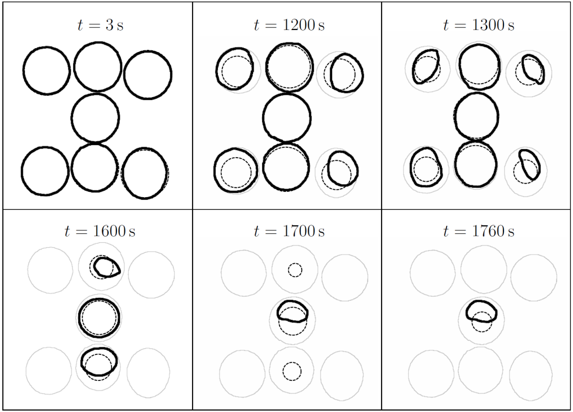
- In particular, the radial directions with the greatest values of R_k have the greatest fluid flux within the droplet, giving the greatest concentration of residue at the contact line.
- Hence, a pair of identical droplets will give rise to **non-homogeneous coffee-rings**, with least residue where the contact lines are closest together and most residue where they are furthest apart.

Comparison with Experiments



Comparison with experimental results of Khilifi *et al.* (2019).

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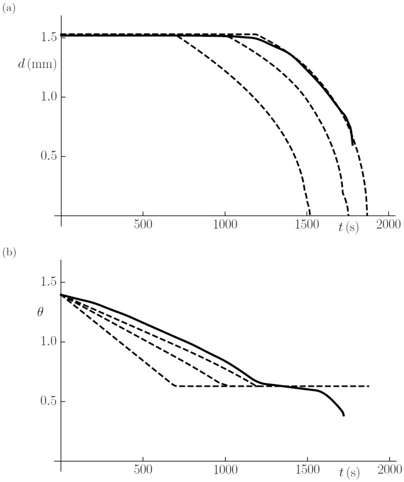

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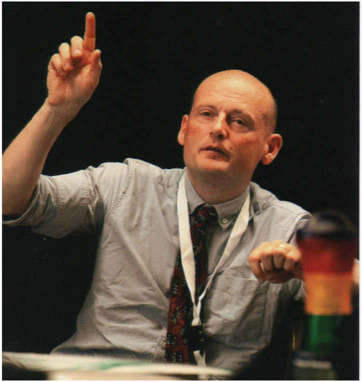
- The predictions of the model were in excellent agreement with the experimental results of Khilifi *et al.* (2019).
- This same method can also be used to solve a wide variety of more complicated configurations and modes of evaporation.



Any questions?

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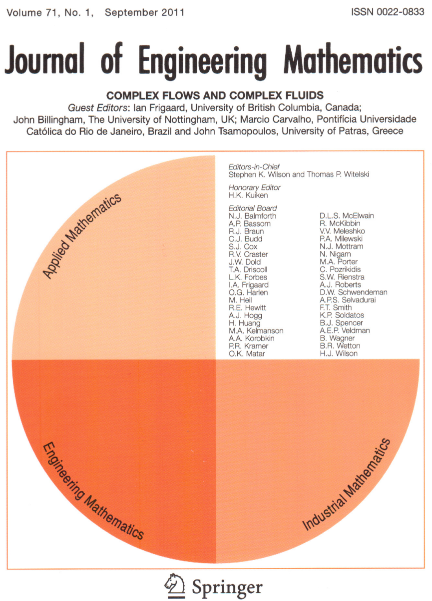

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COMPLEX FLOWS AND COMPLEX FLUIDS

Guest Editors: Ian Frigaard, University of British Columbia, Canada; John Billingham, The University of Nottingham, UK; Marcio Carvalho, Pontifícia Universidade Católica do Rio de Janeiro, Brazil and John Tsamopoulos, University of Patras, Greece

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