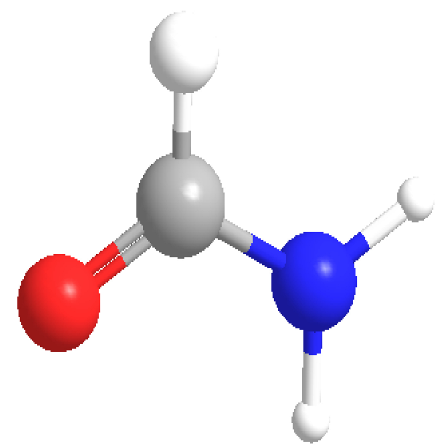
Thermophoresis and the ‘Origin-of-Life



116th General Assembly o

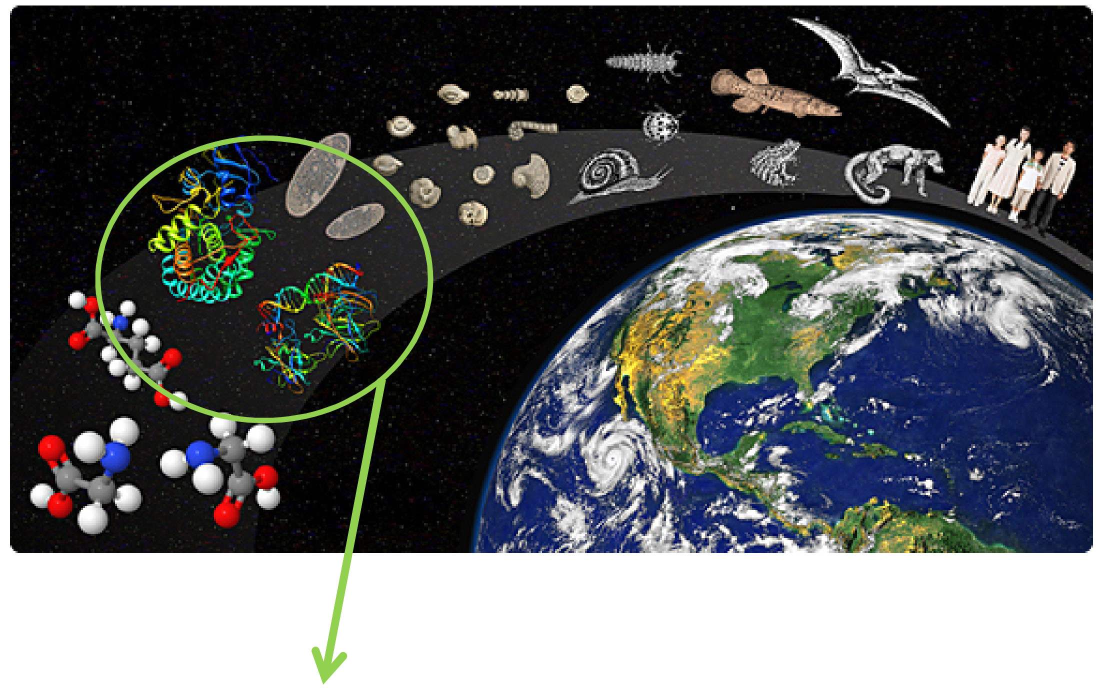
German Bunsen Society

26.05.2017 | Doreen Niether, D. Afanasenkau, J.K.G. Dhont and S. Wiegand

FORSCHUNGSZENTRUM JÜLICH

Looking for the Origin of Life

- Biological evolution: development of complex life forms from simple organisms
- Chemical evolution: formation of organic molecules and proto-biomolecules
- Current research: transition from chemical to biological evolution



Chemical mechanisms
(How can biotic/prebiotic molecules form?)

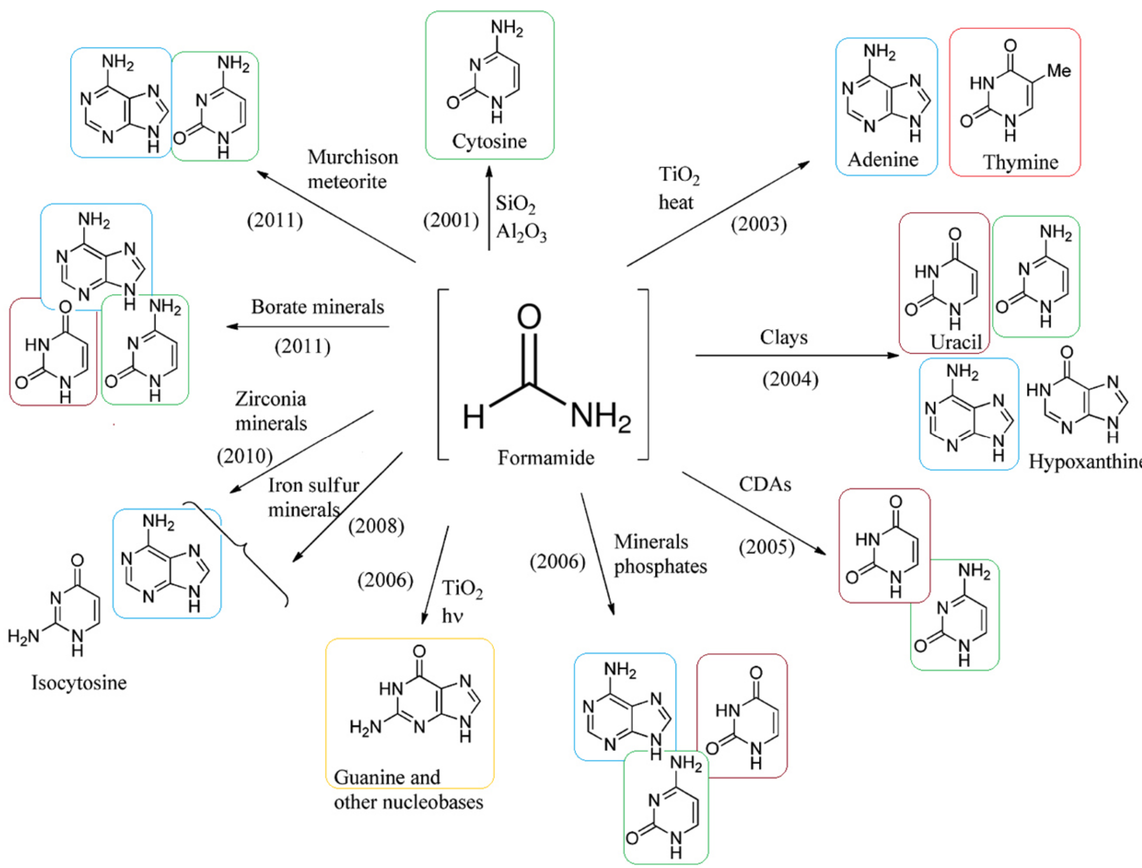
Physical mechanisms
(What were possible reaction conditions?)

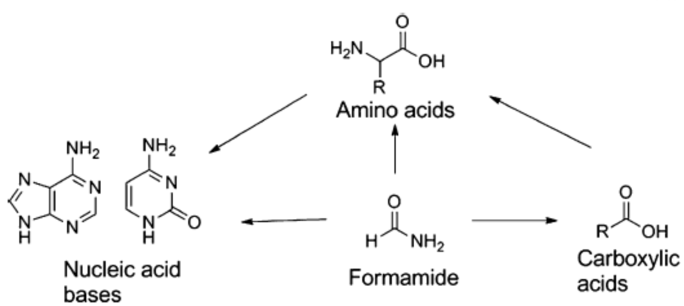
Mitglied der Helmholtz-Gemeinschaft

Formamide - a precurser of life

Starting material for abiotic synthesis of

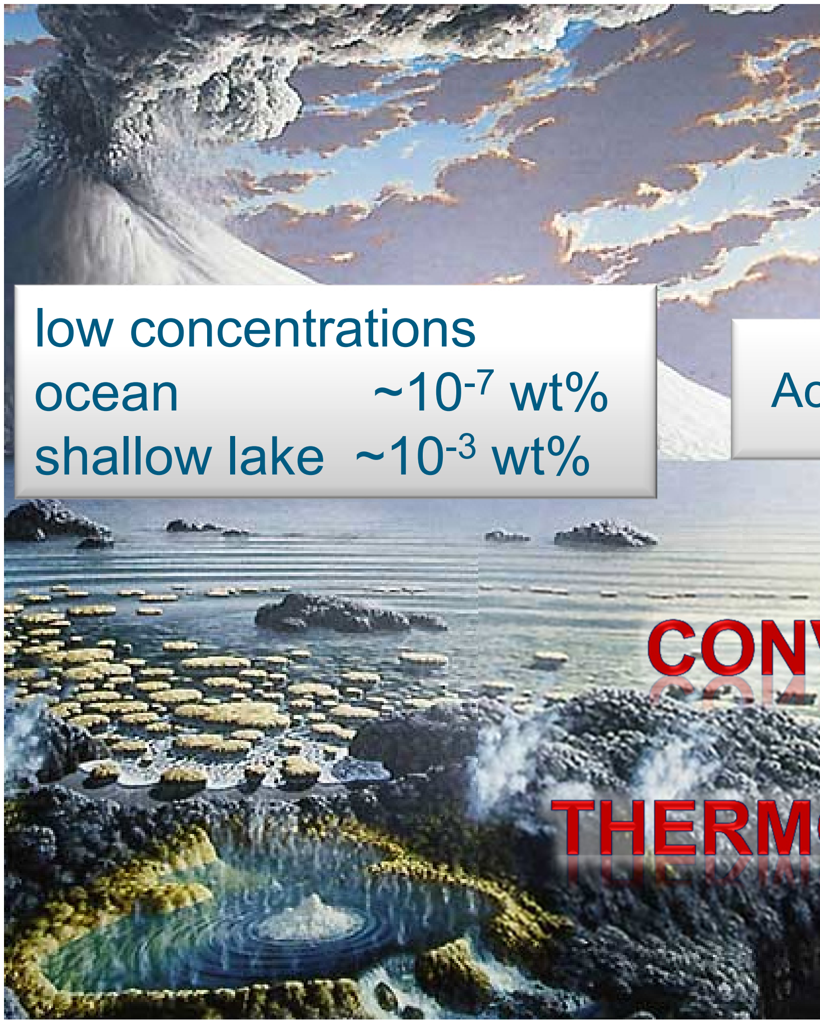
- Prebiotic nucleobases
- Carboxylic acids
- Amino acids

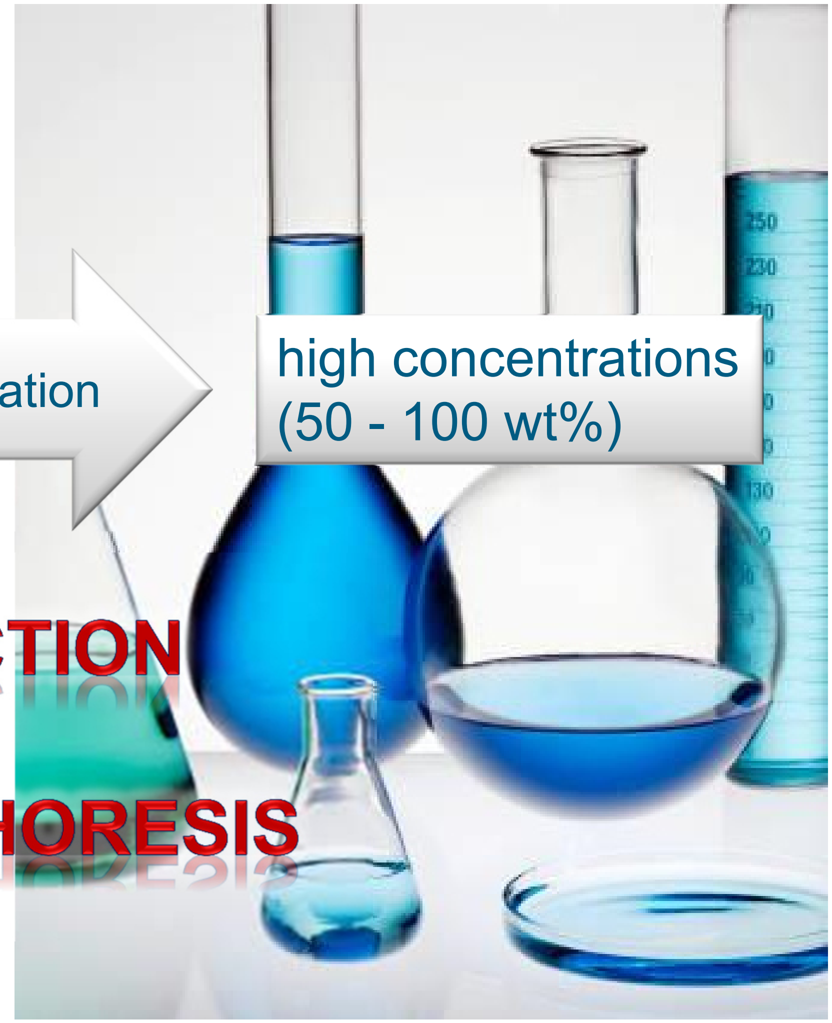




R. Saladino et al./ Physics of Life Reviews 9 (2012) 84-104

The concentration problem





low concentrations
ocean ~10^{-7} wt%
shallow lake ~10^{-3} wt%

Accumulation

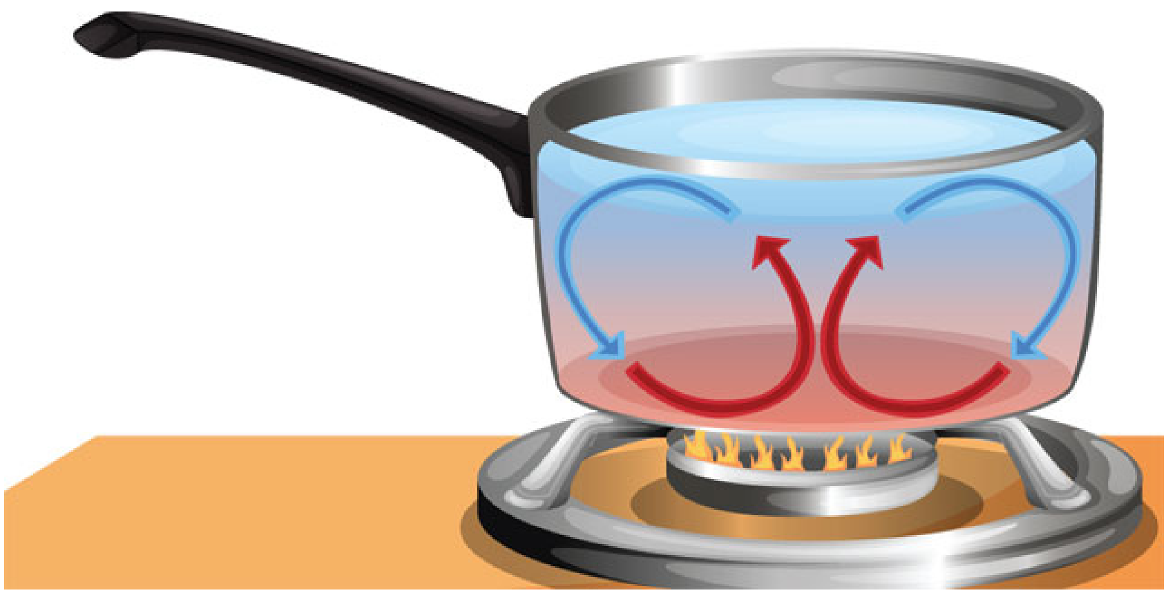
high concentrations
(50 - 100 wt%)

CONVECTION
+
THERMOPHORESIS

Convection

Transport mechanism in gases or liquids

Dependent on density, viscosity, heat capacity, thermal conductivity



temperature difference

different densities

cold (denser) fluid moves down, hot fluid moves up

circular flow

Thermodiffusion

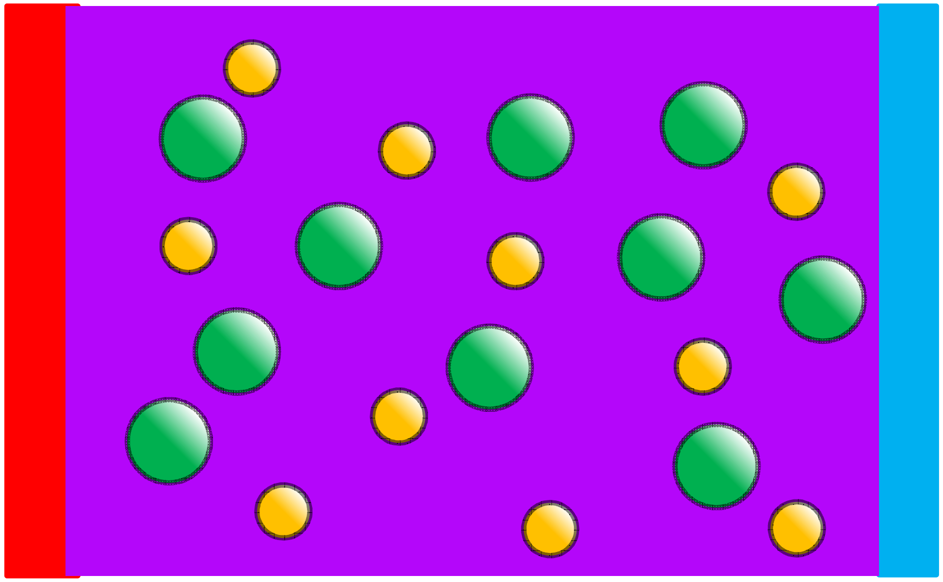
Movement of particles driven by temperature gradient

$$\vec{j} = -D\vec{\nabla}\omega - \omega(1-\omega)D_T\vec{\nabla}T$$

Steady state $\vec{j}=0$

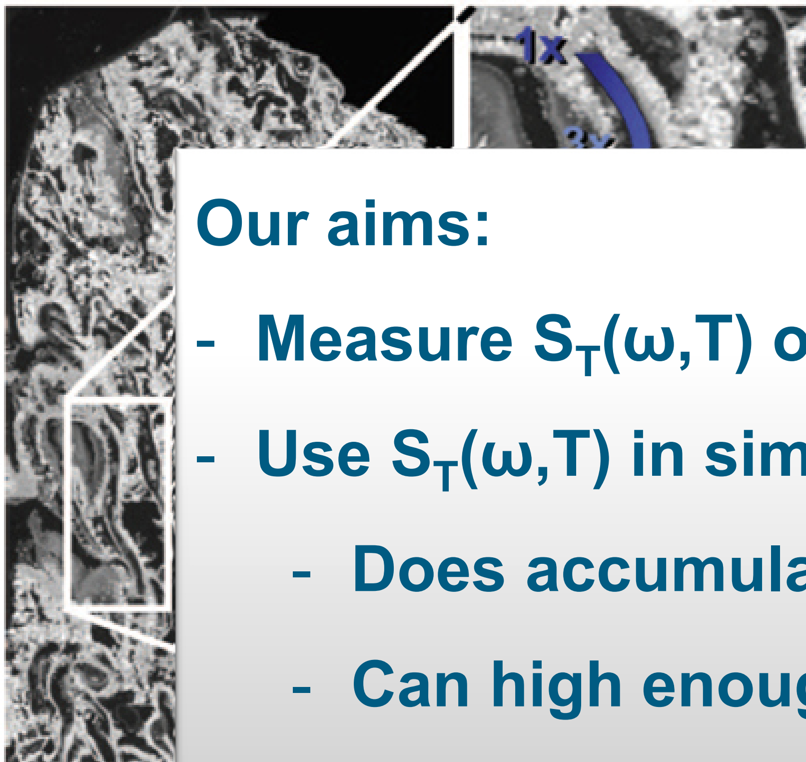
$$\frac{D_T}{D} = -\frac{\vec{\nabla}\omega}{\omega(1-\omega)\vec{\nabla}T}$$

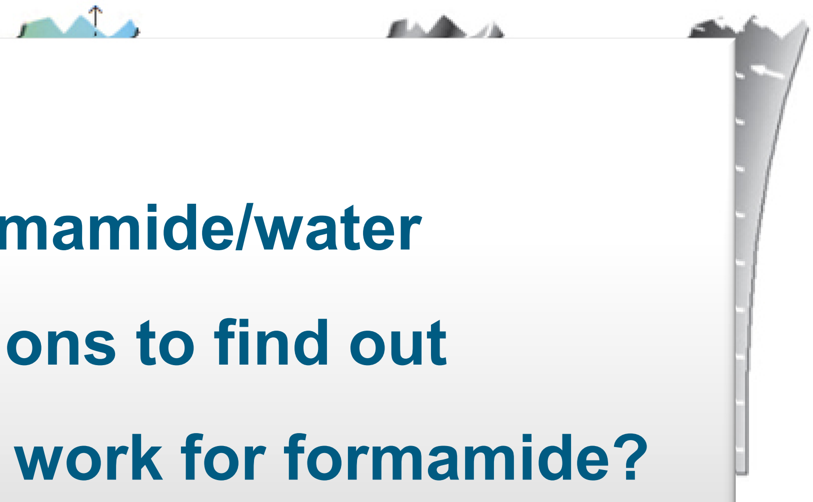
$$S_T = \frac{D_T}{D} \propto \frac{\Delta\omega}{\Delta T}$$



D - diffusion coefficient,
ω - concentration,
D_T - thermodiffusion coefficient,
$\vec{j}$ - flux,
T – temperature
S_T –Soret coefficient

Thermophoretic Accumulation





Our aims:

- Measure $S_T(\omega,T)$ of formamide/water
- Use $S_T(\omega,T)$ in simulations to find out
    - Does accumulation work for formamide?
    - Can high enough concentrations be reached?

- Setting: porous minerals near hydrothermal vents
- Nucleotides and RNA accumulate in pores
- same principle as thermogravitational column, but open (influx of matter)

Baaske et al., PNAS vol. 104, no. 22, 9349 (2007)

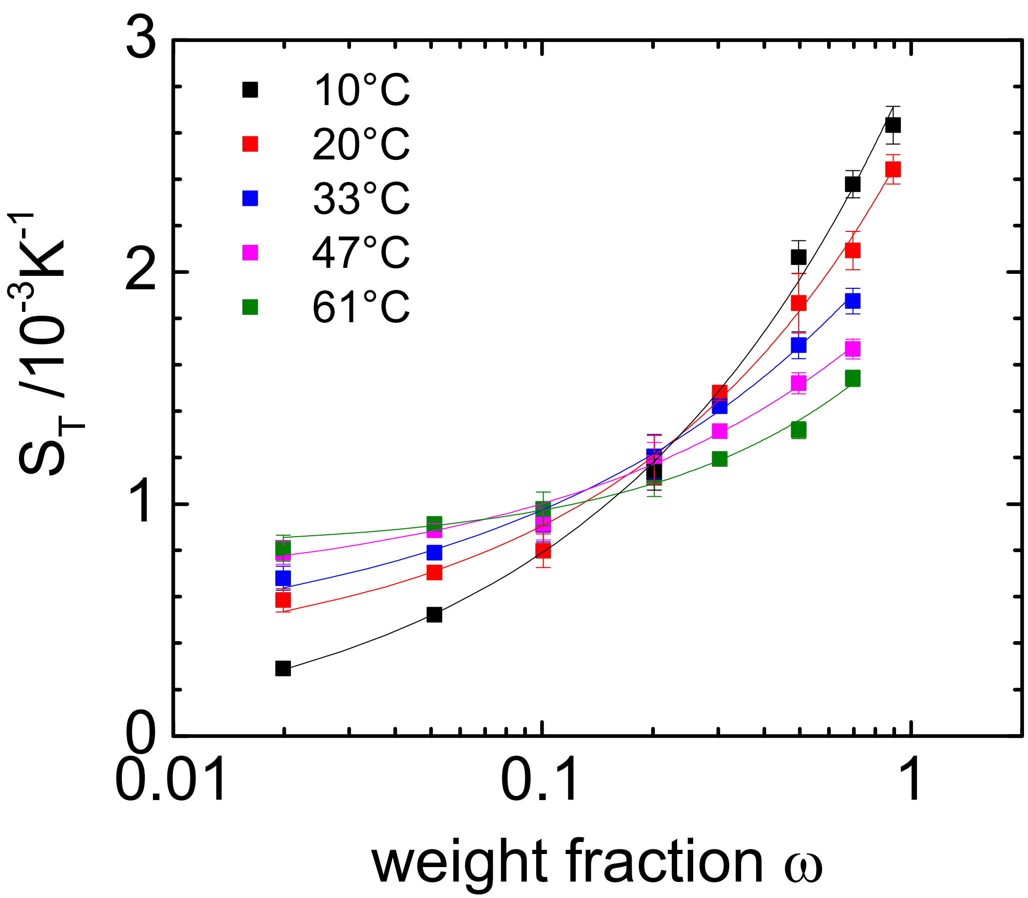
Thermodiffusion of Formamide in Water

Experimental results

- Fit with Hill equation:

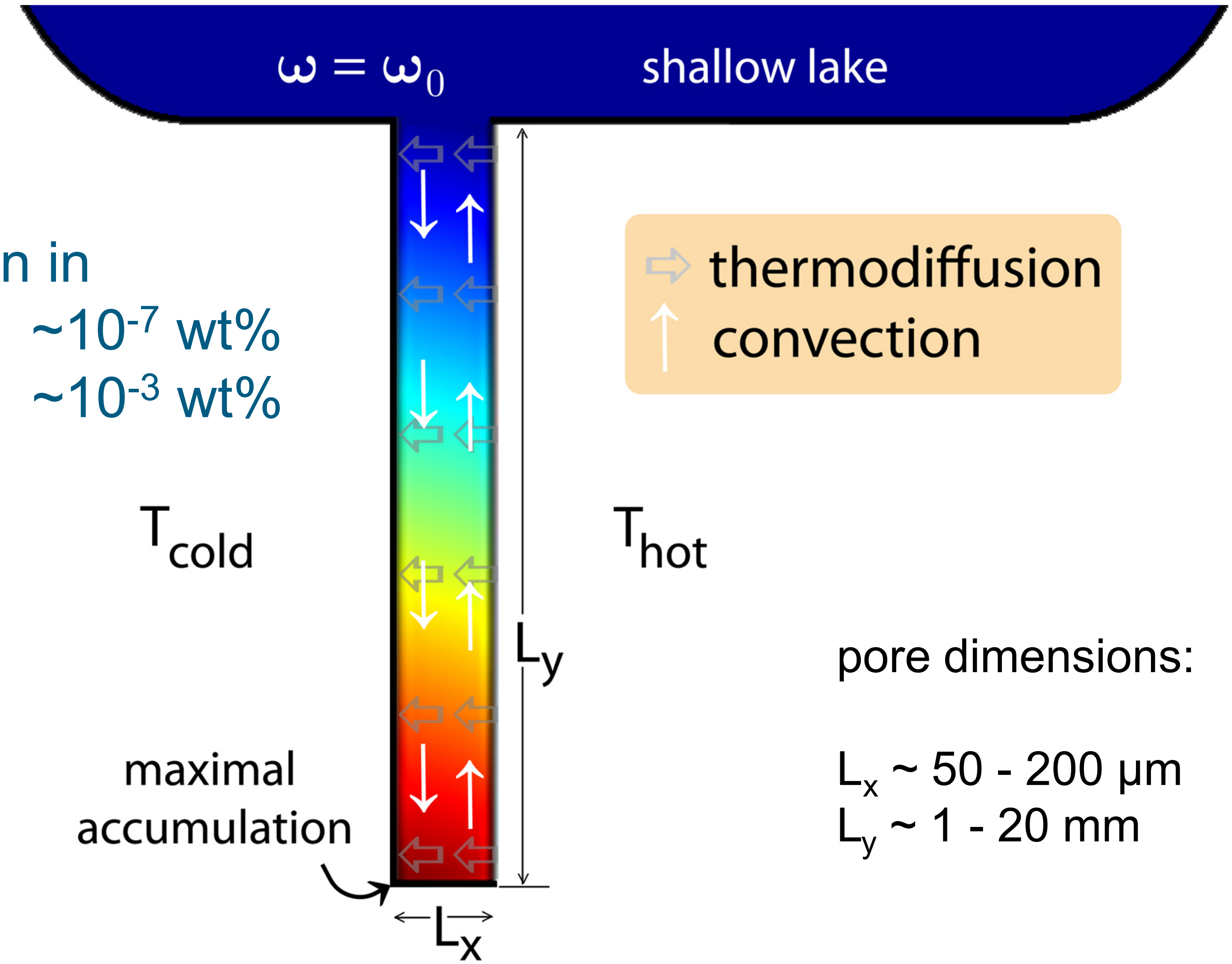
$$S_T(\omega) = \frac{\omega^a}{K + \omega^a} + S_T^0$$

- Approaches constant value for small concentrations
- becomes more thermophobic with increasing concentration



Simulation with COMSOL

concentration in
ocean ~10^{-7} wt%
shallow lake ~10^{-3} wt%



pore dimensions:

L_x ~ 50 - 200 µm
L_y ~ 1 - 20 mm

[D. Niether, D. Afanasenkau, J.K.G. Dhont, SW, P*NAS* **113** (2016) 4272]

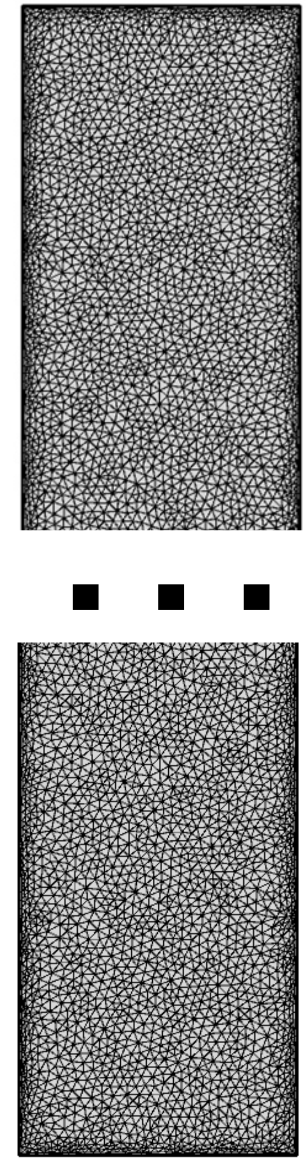
Simulation with COMSOL

temperature and concentration dependant parameters of formamide/water:

- Soret coefficient
- density
- viscosity
- heat capacity
- thermal conductivity

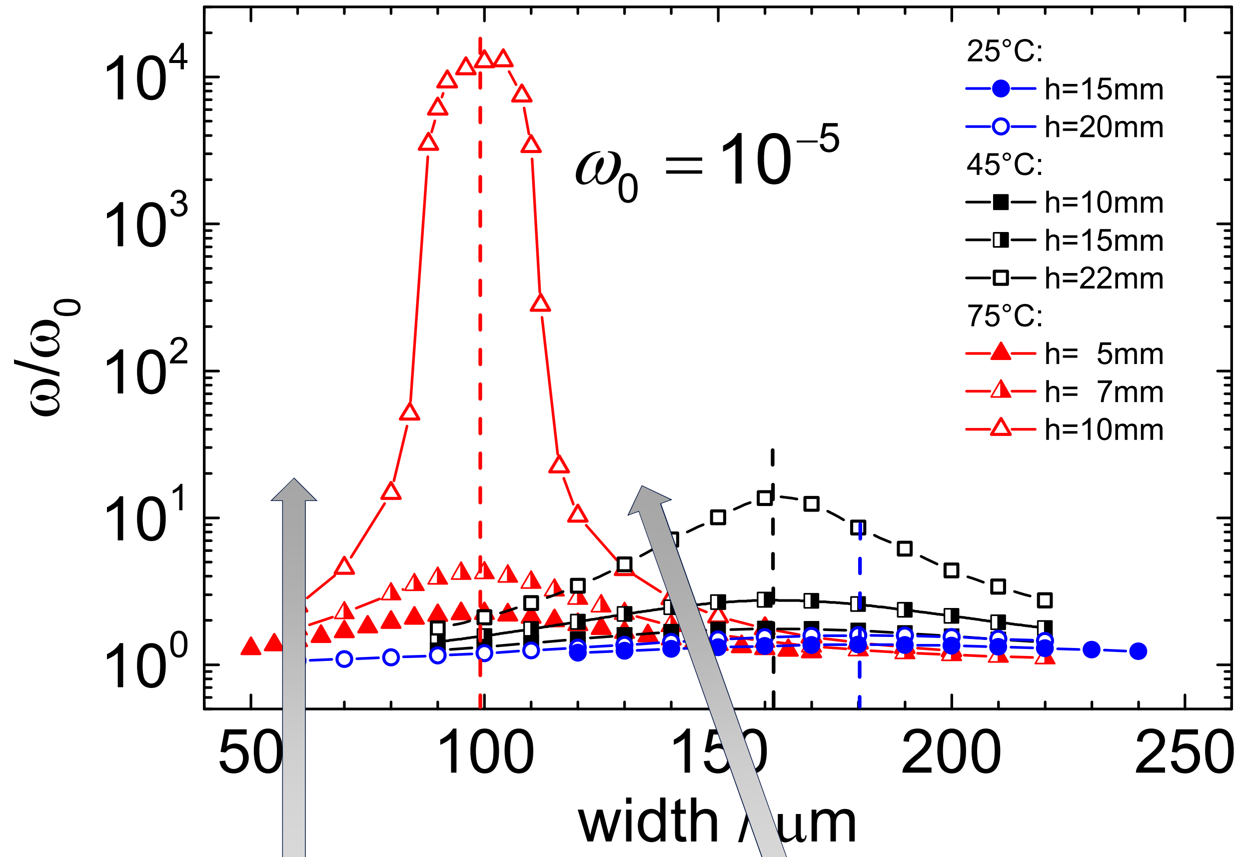
Finite elements simulation:

- Navier-Stokes equations
- Heat transfer equations
- Thermal diffusion/convection



[D. Niether, D. Afanasenkau, J.K.G. Dhont, SW, P*NAS* **113** (2016) 4272]

Geometry and temperature influence

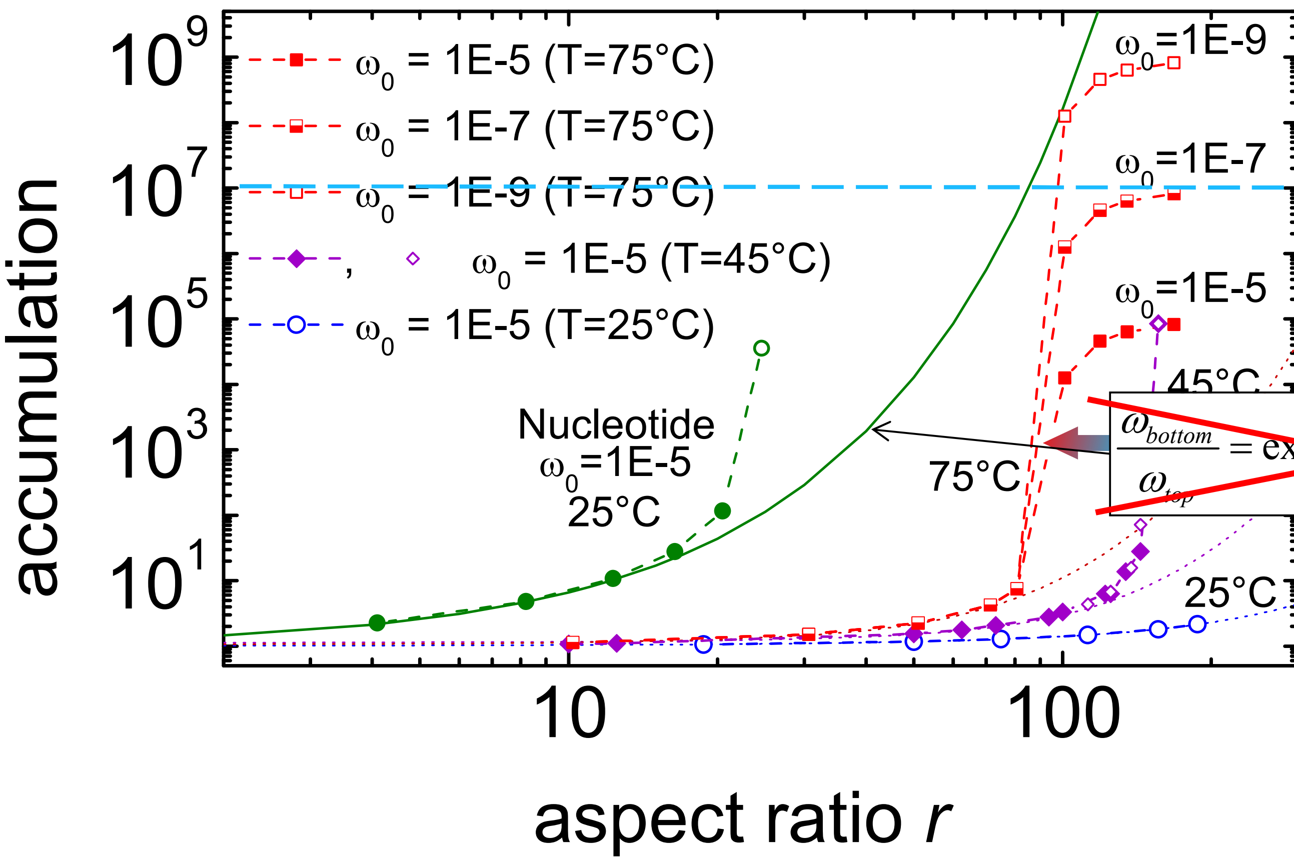


reduced convection

(only effective for larger w_0)

convection too fast

Accumulation of Formamide



$$\frac{\omega_{bottom}}{\omega_{top}} = \exp(0.42 \cdot S_T \cdot \Delta T \cdot r)$$

$$\frac{\omega_{bottom}}{\omega_{top}} \cdot \omega_0 \leq 1$$

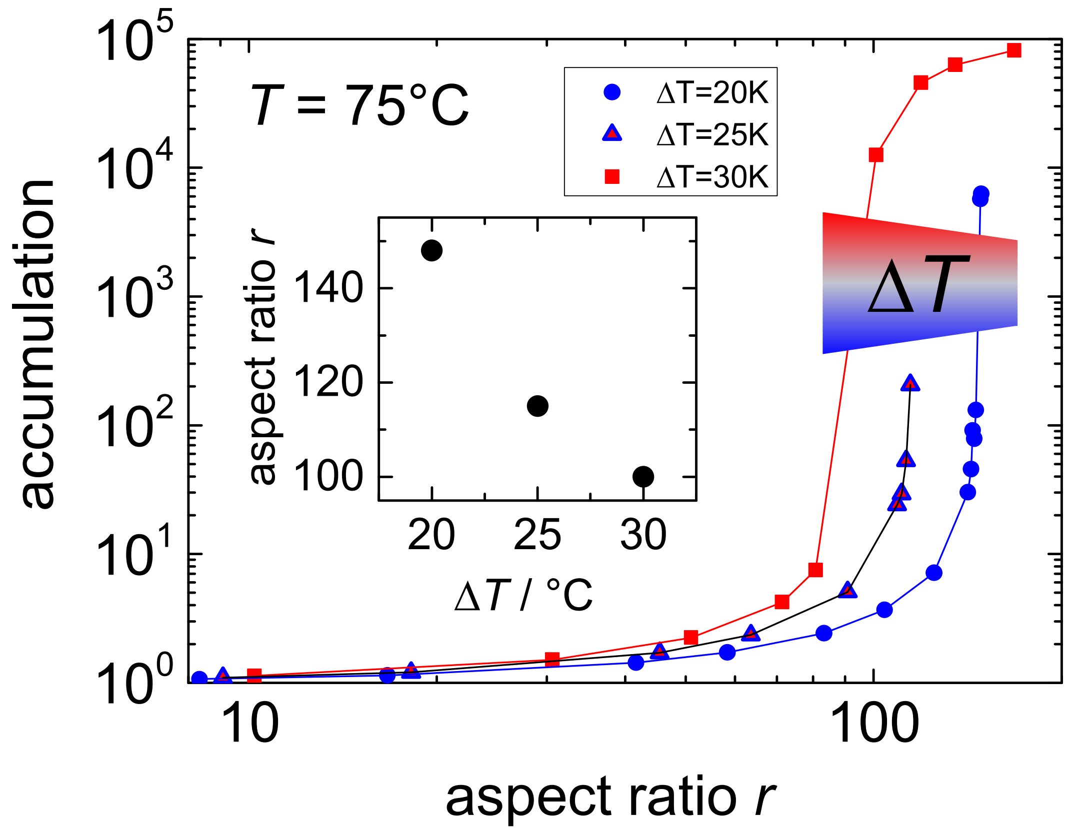
$$r = \frac{height}{width}$$

pore needs a certain height for effective accumulation

- depends on Soret coefficient and ambient temperature

Accumulation of Formamide

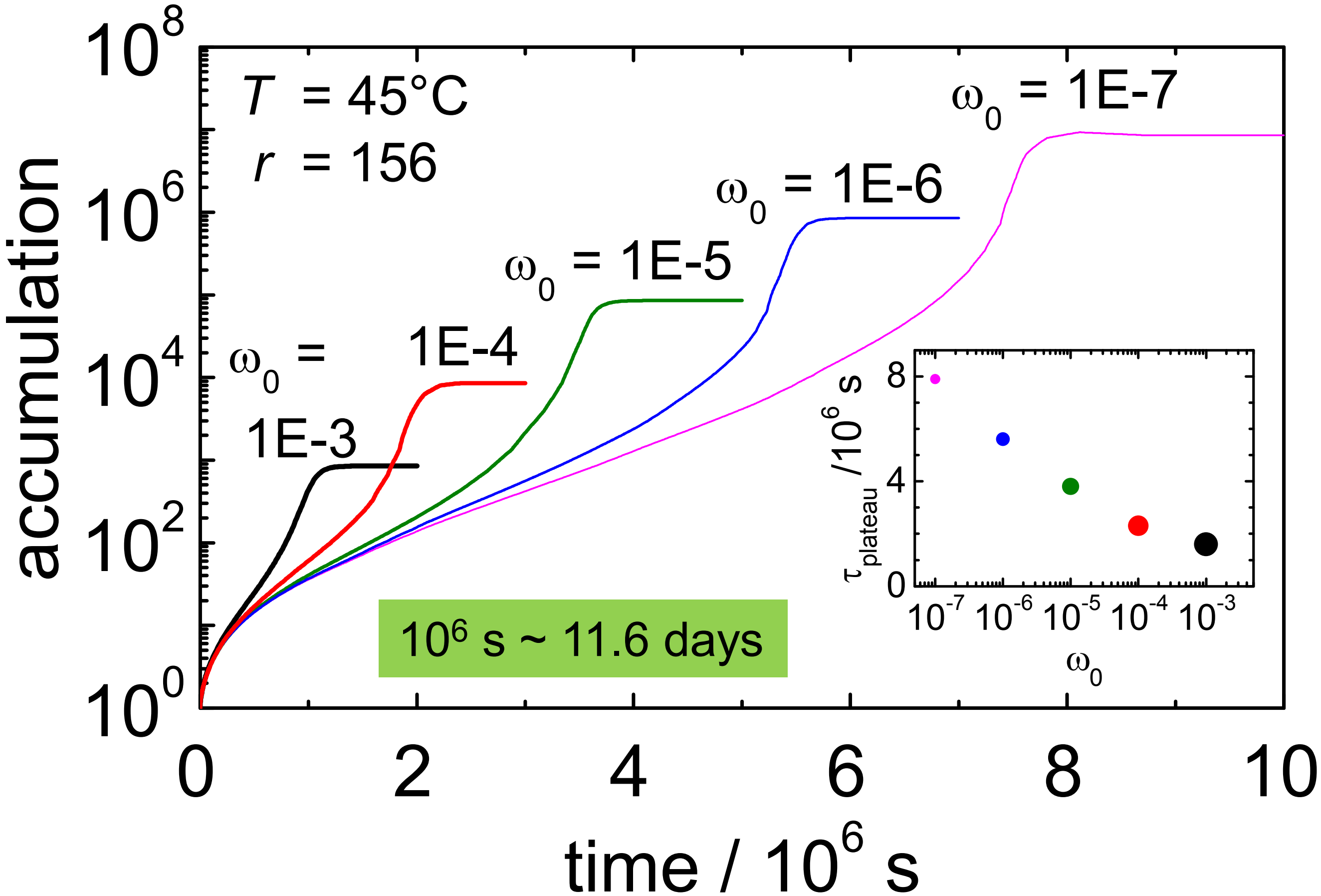
Dependence on the temperature difference



larger temperature difference → accumulation at smaller heights

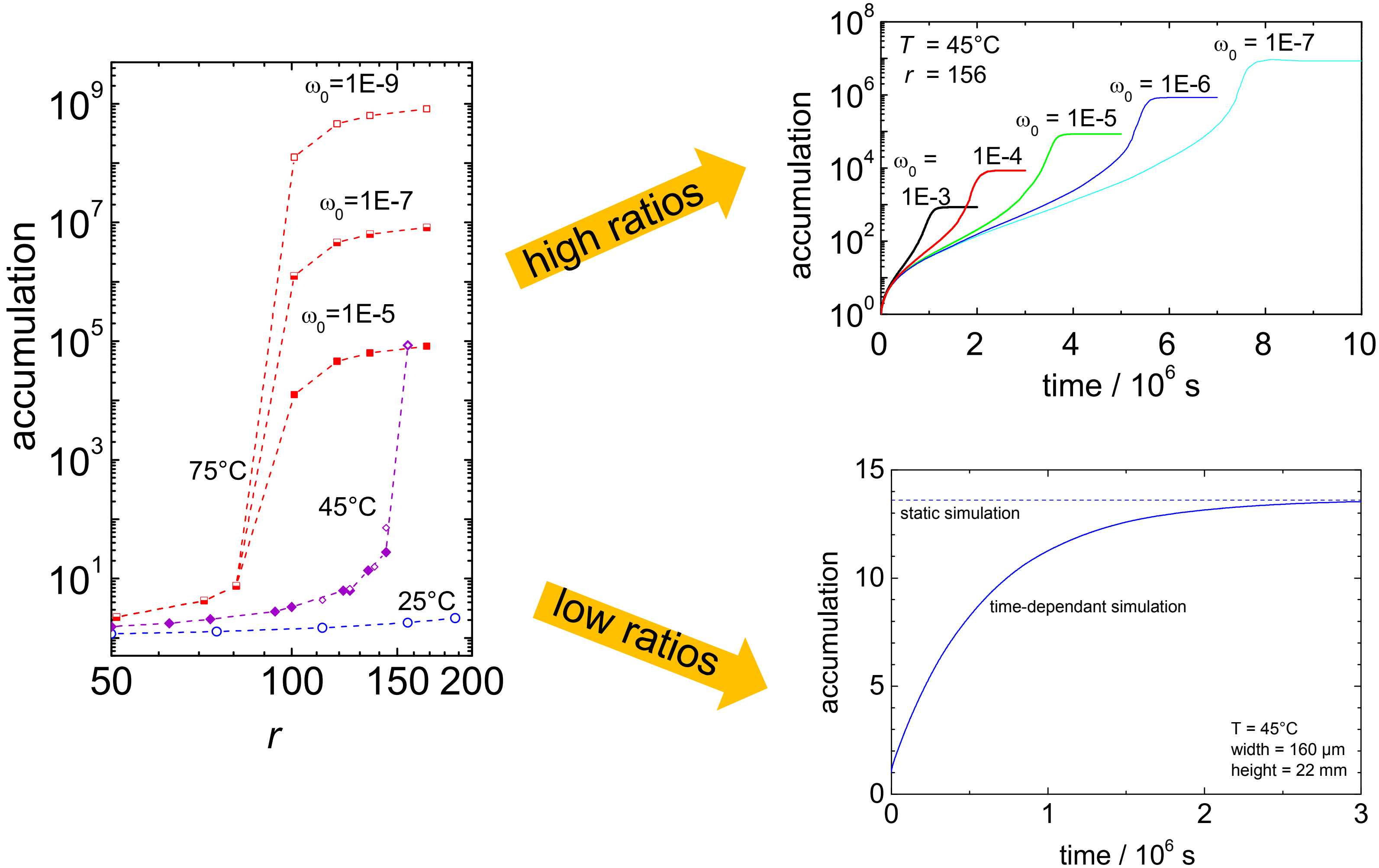
Accumulation of Formamide

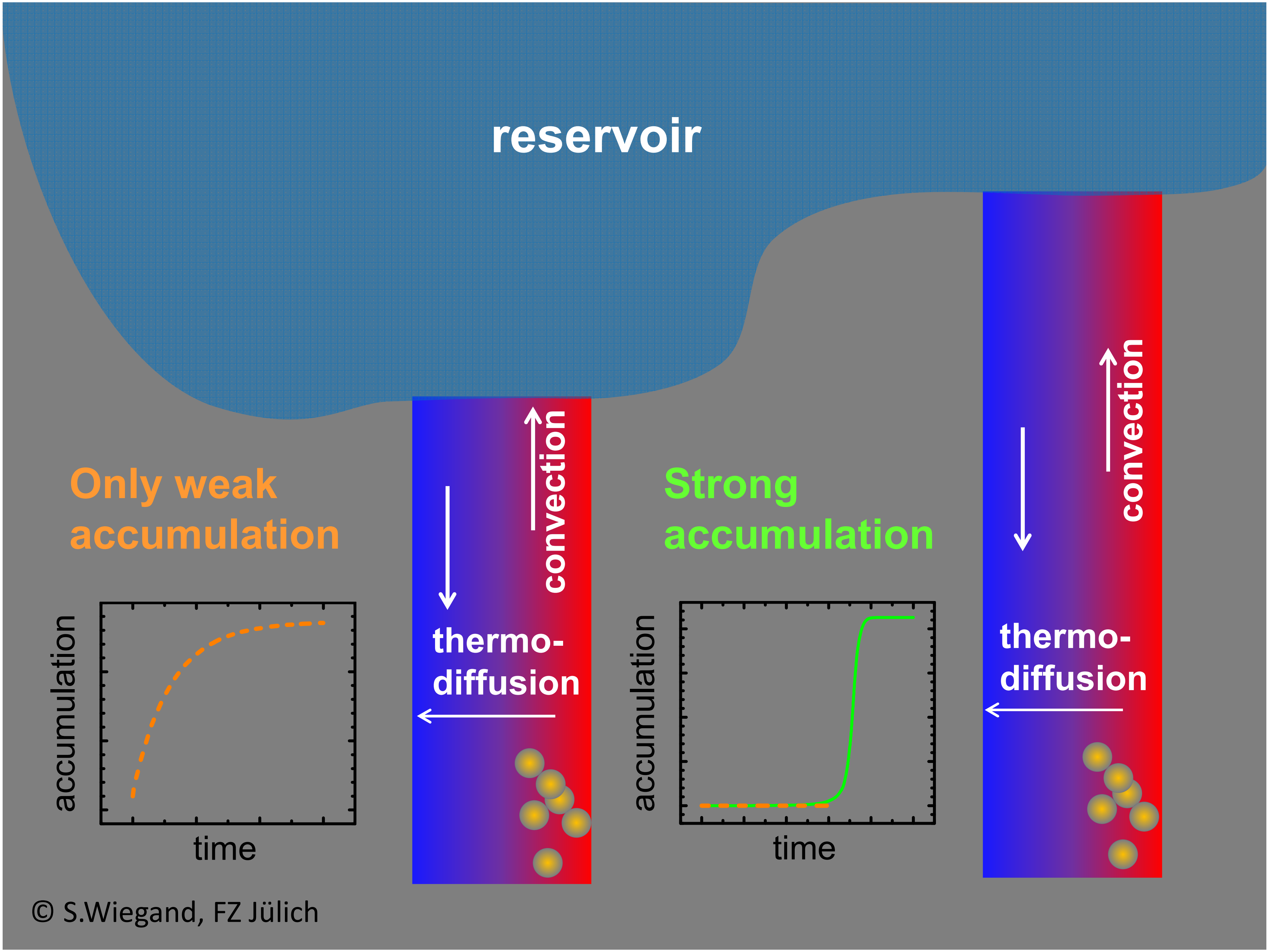
Dependence on the starting concentration



concentration rise to ~85%, independent of starting concentration

Accumulation of Formamide
Dependence on pore height




reservoir
convection
thermo-
diffusion
Only weak
accumulation
accumulation
time
convection
thermo-
diffusion
Strong
accumulation
accumulation
time
© S.Wiegand, FZ Jülich

Can we understand the steep increase?
simple heuristic approach

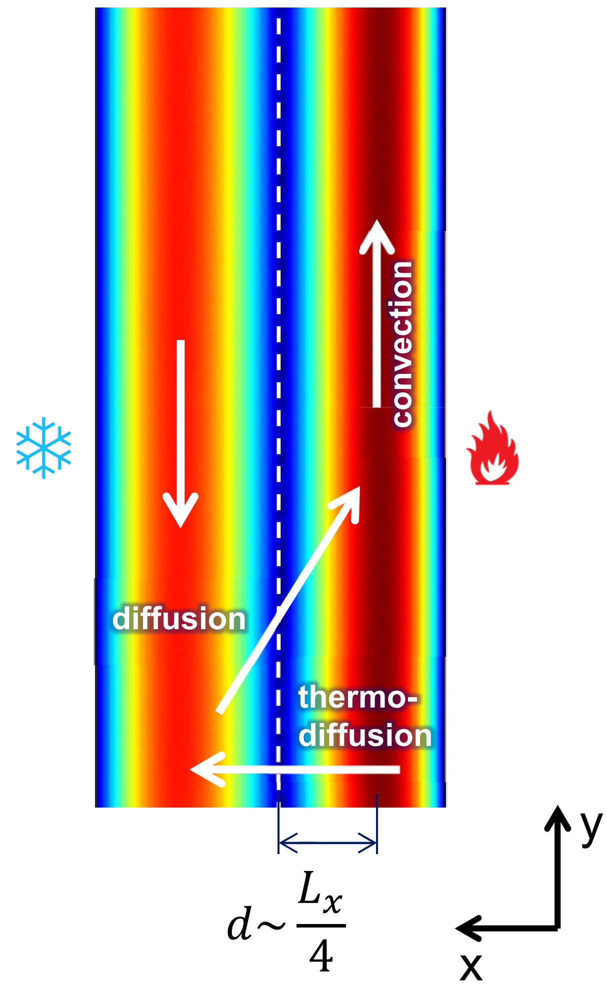
$$v_y = v_{convection} + D\frac{\Delta_y w}{L_y}$$

$$v_x = -D\frac{\Delta_x w}{L_x} + w(1-w)D_T\frac{\Delta T}{L_x}$$

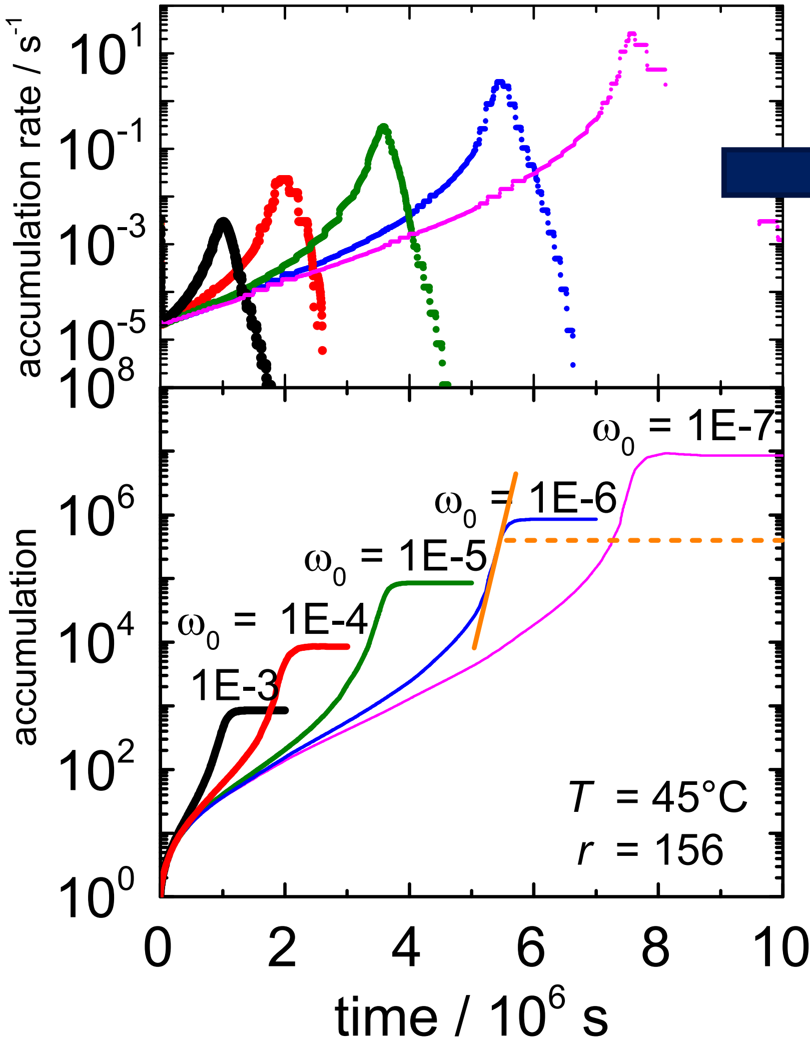
time to reach the top of the column > time to cross the middle of the column

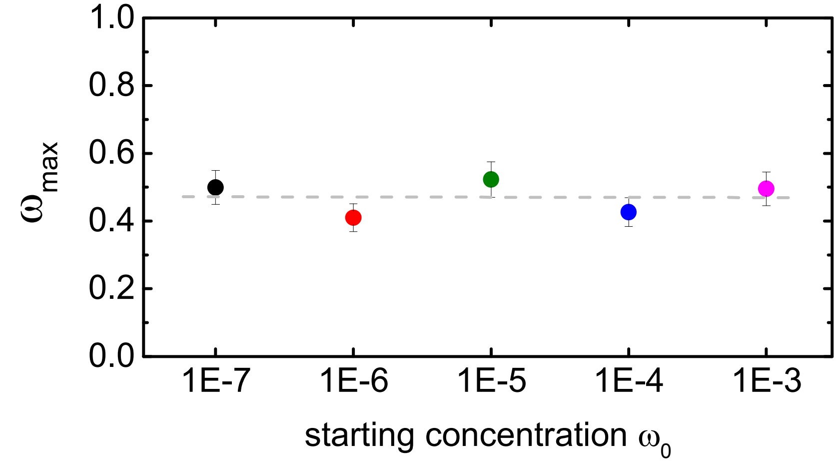
$$\frac{L_y}{v_y} > \frac{L_x}{4v_x} \Rightarrow \frac{L_y}{L_x} > \frac{v_y}{4v_x}$$

$$(r^*)^2 = \left(\frac{L_y}{L_x}\right)^2 > \frac{(L_y \cdot v_{convection} + D \cdot \Delta_y w)}{4 \cdot (-D\Delta_x w + w(1-w)D_T\Delta T)}$$



Simple heuristic approach





r^* has a minimum at ω = 0.5

$$\left(r^*\right)^2 \propto \frac{1}{\omega(1-\omega)D_T\Delta T}$$

Conclusion

- Experimental determination of S_T and D_T
- Simulation of accumulation in pores with regard to the temperature and concentration dependence of substance parameters
  - accumulation possible at different temperatures, temperature differences and starting concentrations
  - very effective at large pore ratios (up to 85 wt%)
  - time varies for different starting parameters (~45-90d)
- Simulations for ideal system (stable conditions, pure formamide/water mixture) – accumulation in real system probably less effective

Publications:

- “Accumulation of formamide in hydrothermal pores to form prebiotic nucleobases”, D. Niether, D. Afanasenkau, J. K. G. Dhont, S. Wiegand, *Proc. Natl. Acad. Sci. USA* 113(16), 4272 - 4277 (2016)
- “Heuristic Approach to Understanding the Accumulation Process in Hydrothermal Pores”, D.Niether, S.Wiegand, *Entropy* 19, 33 (2017)

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Thank you for your attention!