

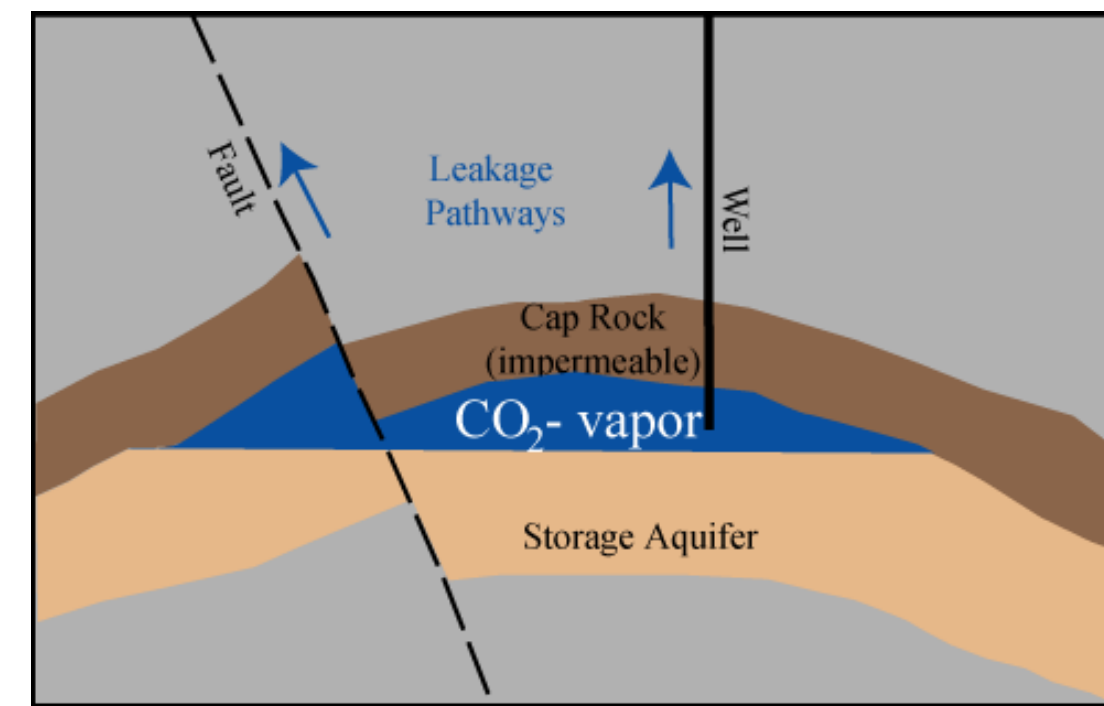
Motivation

Background & Scenario

CO₂ capture and storage is only environmentally sound, if leakage of CO₂ into the atmosphere is limited. Positive buoyancy drives gaseous CO₂ back to the surface along faults or old wells.

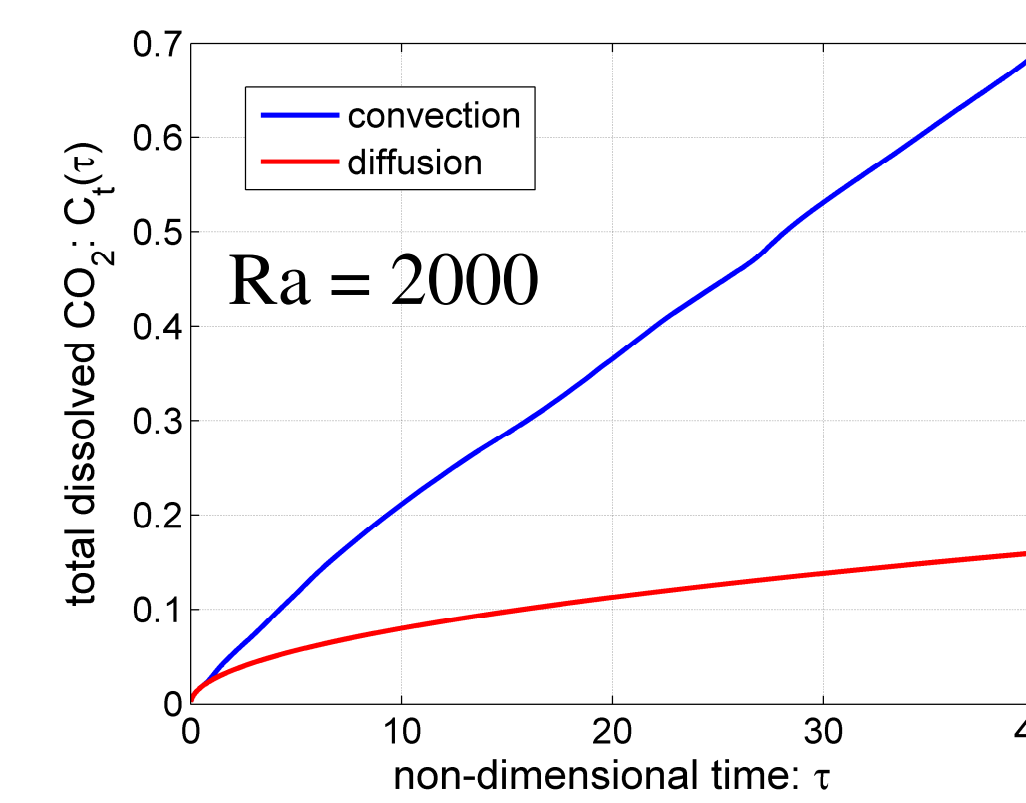
Dissolution of CO₂ into the brine limits the time available for leakage, because dissolved CO₂ has negative buoyancy.

To investigate the time scale for complete dissolution of the injected CO₂, we consider a simplified scenario, of CO₂ in a hydrodynamic trap.



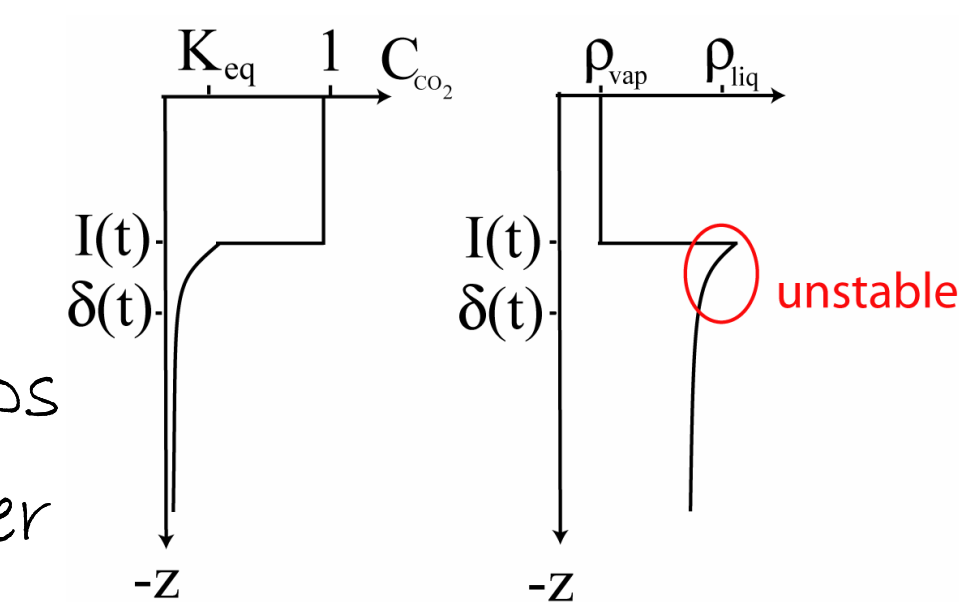
Convection vs. Diffusion

Dissolution is always transport limited, and convective mass transfer significantly increases the amount of dissolved CO₂!



Gravity Driven Convection

Dissolved CO₂ increases the density of the brine. An unstable density gradient develops in the diffusive boundary layer below the CO₂.



Important Questions we are addressing:

- 1) Does convection occur in aquifers?
- 1) How long does it take to get started?
- 2) What is the convective dissolution rate?
- 3) Can we simulate convective transport correctly?

References:

1. Riaz A., Hesse M., Tchelepi H. & Orr Jr. F. (2006): Onset of Convection in a gravitationally unstable, diffusive boundary layer in porous media, *J. Fluid Mech.*, **548**, pp 87-111
2. Hesse M., Riaz A. & Tchelepi H. (2006): Resolving Density Fingering During CO₂-Sequestration: A Challenge for Reservoir Simulation, *Proced. CO2SC Symposium 2006*, 20-22 March 2006, LBNL Berkeley
3. Hesse M., Tchelepi H. & Orr Jr. F. (2006): Natural Convection During Aquifer CO₂ Storage, *Proced. of GHGT 8*, 19-22 June 2006, Trondheim

Onset of Convection

Linear Stability Analysis

At early times the governing equations can be linearized, and the growth rate σ of a disturbance with wavelength λ evolves with time (t) as:

$$\sigma(t; \lambda) = -\frac{1}{t} - \frac{4\pi^2}{\lambda^2 \cdot Ra} + \frac{2\sqrt{\pi}}{\lambda} F(t; \lambda); \quad F(t; \lambda) > 0$$

Initially ($t \rightarrow 0$) the growth rates are negative for all λ and the system is stable. Increasing t and large Ra destabilize the system. At the critical time (t_c) the first λ starts to grow and the system becomes unstable. This first growing λ is the critical wavelength (λ_c) and gives the initial length scale of the instability. The t_c is a lower bound on the onset of convective mass transfer. The analysis gives us t_c and λ_c in terms of average reservoir parameters:

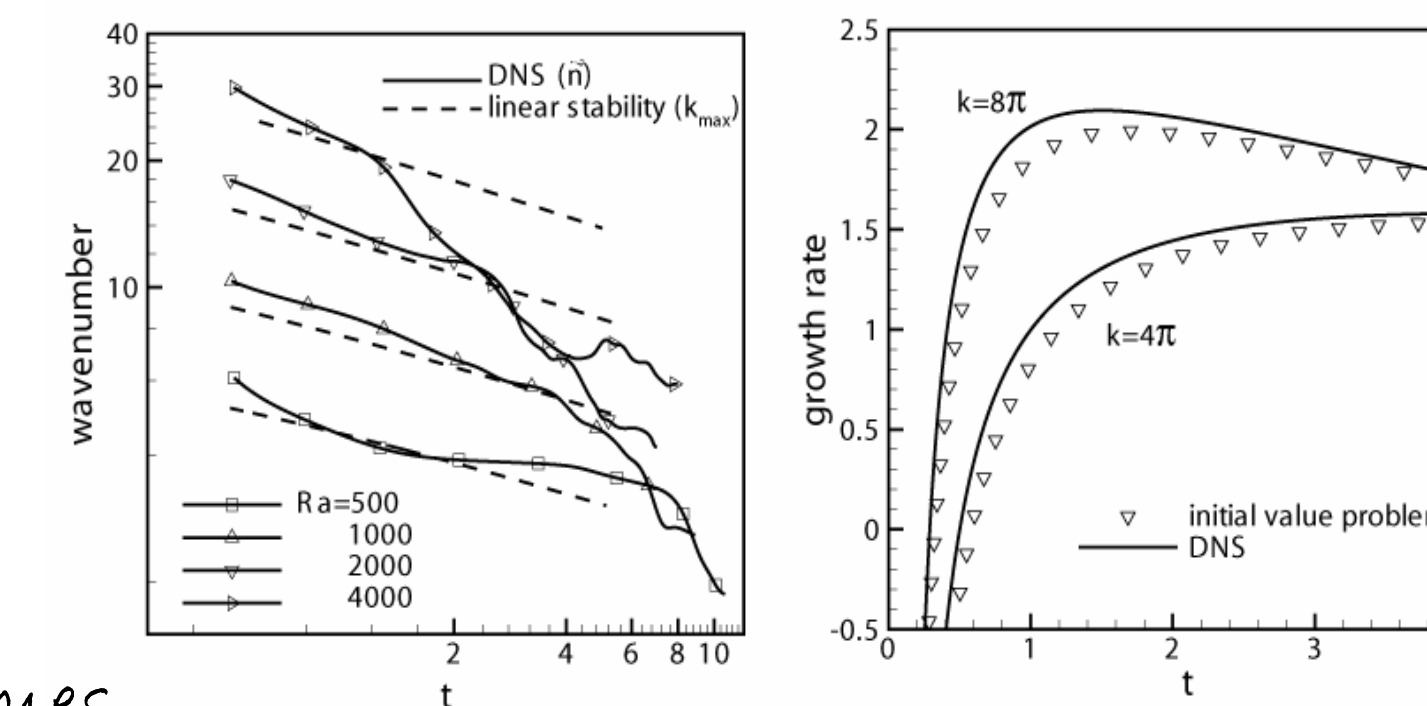
$$t_c = \frac{146\phi\mu^2 D}{(K\Delta\rho g)^2} \quad \lambda_c = \frac{2\pi\mu D}{0.07K\Delta\rho g}$$

K = permeability, ϕ = porosity
 D = dispersion coefficient
 μ = viscosity, $\Delta\rho$ = density diff.

The permeability K has the largest variability so that we can think of t_c and λ_c as functions of K only ($t_c \propto 1/K^2$ and $\lambda_c \propto 1/K$).

Cross-validation of Theory and Numerical Simulation

Comparison of our analytic results with direct numerical simulation (DNS), shows very good agreement at early times where the linearization is valid. This comparison shows



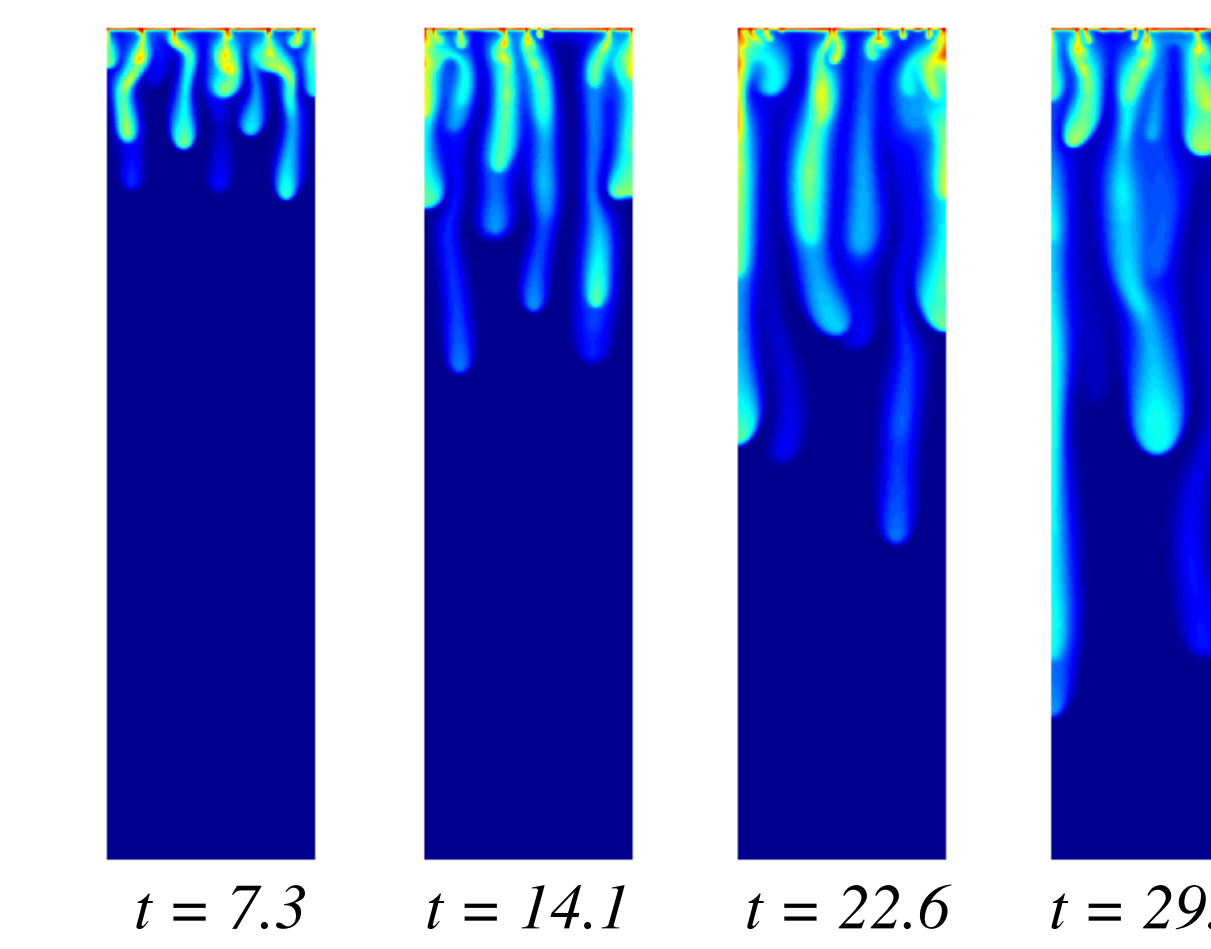
- 1) The linear theory is valid at early times.
- 2) DNS can resolve the very small initial length scales, and is therefore suited to investigate the non-linear evolution.

Implications for CO₂ Storage in Saline Aquifers

- 1) In aquifers with high permeability (Sleipner, ~ 3 Darcy) the critical time is very short $t_c \sim 1$ yr. Convection will increase the dissolution of CO₂ from the beginning, reducing the chance of leakage over long times.
- 2) The initial length scales of convection in high permeability aquifers will be very small $\lambda_c \sim 1$ m. Resolving these small length scales is important to simulate the convective transport, and presents a numerical challenge.
- 3) In aquifers with low permeability (100 mDarcy) have longer onset times $t_c \sim 100$ yrs and larger wavelength $\lambda_c \sim 100$ m. In these settings other processes have to immobilize the CO₂ in the short term (< 100 yrs).

Long Term Evolution

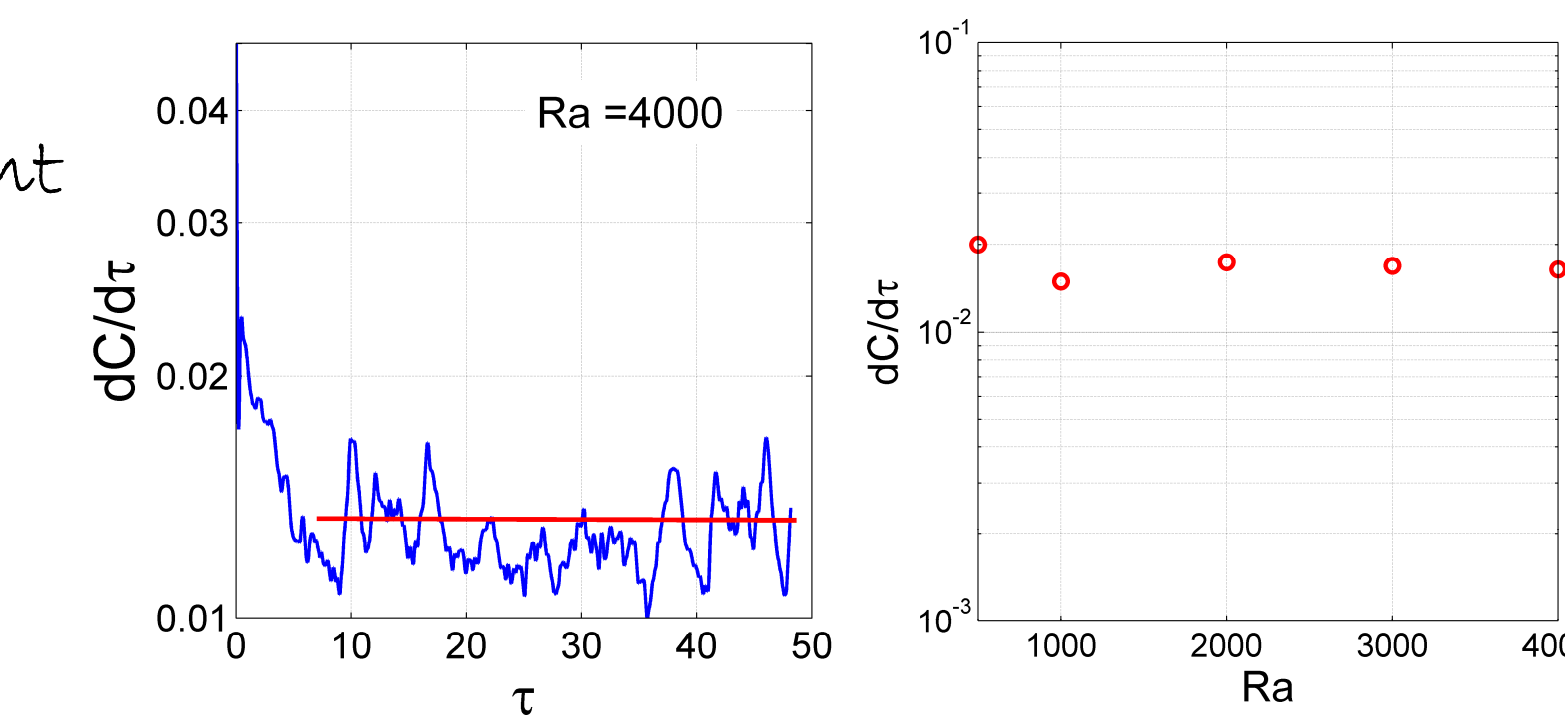
Convection in an Semi-Infinite Domain ($H \ll W \& W=1, Ra = 4000$)



The onset time t_{on} separates the initial diffusive regime from the "infinite acting" convection regime. In the diffusive regime the dissolution rate decays as $t^{-1/2}$, while it is constant during infinite acting convective mass transfer.

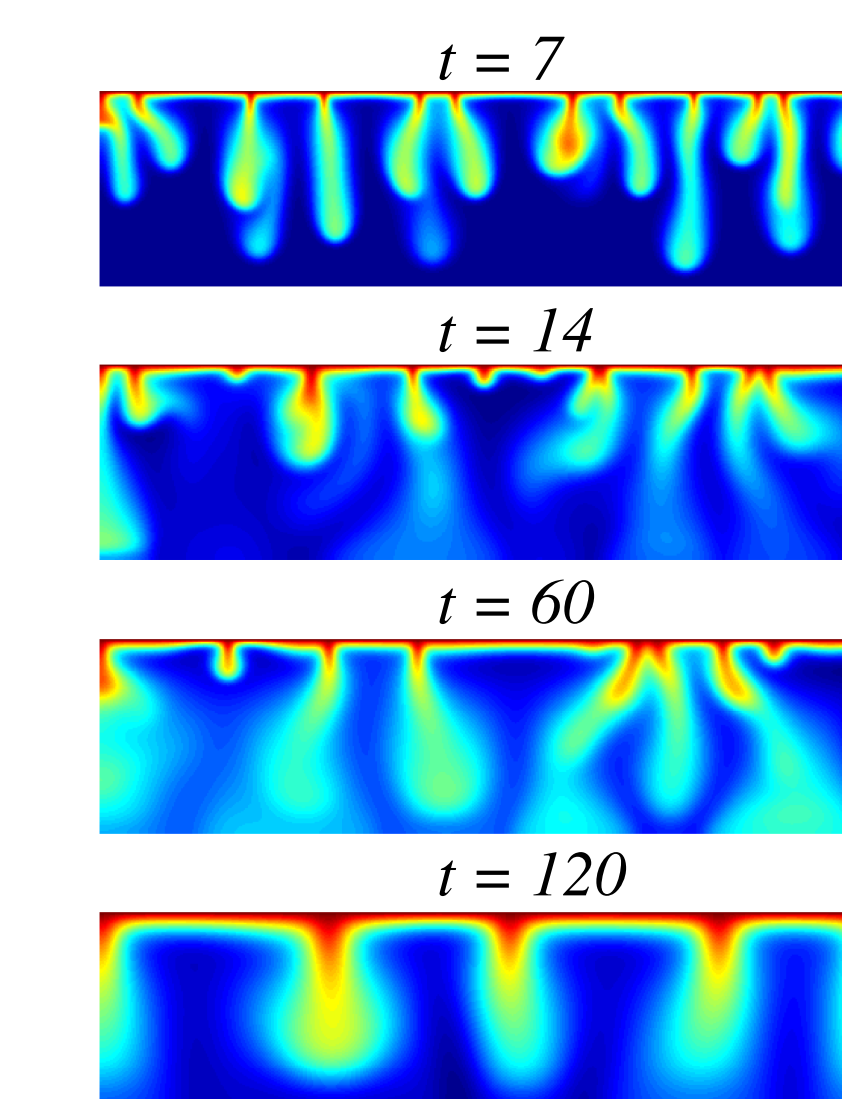
The non-dimensional convective dissolution rate dC/dt is independent of Ra . The dimensional convective dissolution rate is given by:

$$\frac{dC}{dt} \sim 0.017 \frac{K\Delta\rho g C_{CO_2}^{aq}}{\mu\phi}$$



In high permeability aquifers the convective mass transfer will dissolve CO₂ very efficiently, hence reducing the time available for leakage.

Convection in a Closed Domain ($H=1 \& W=4, Ra = 1000$)

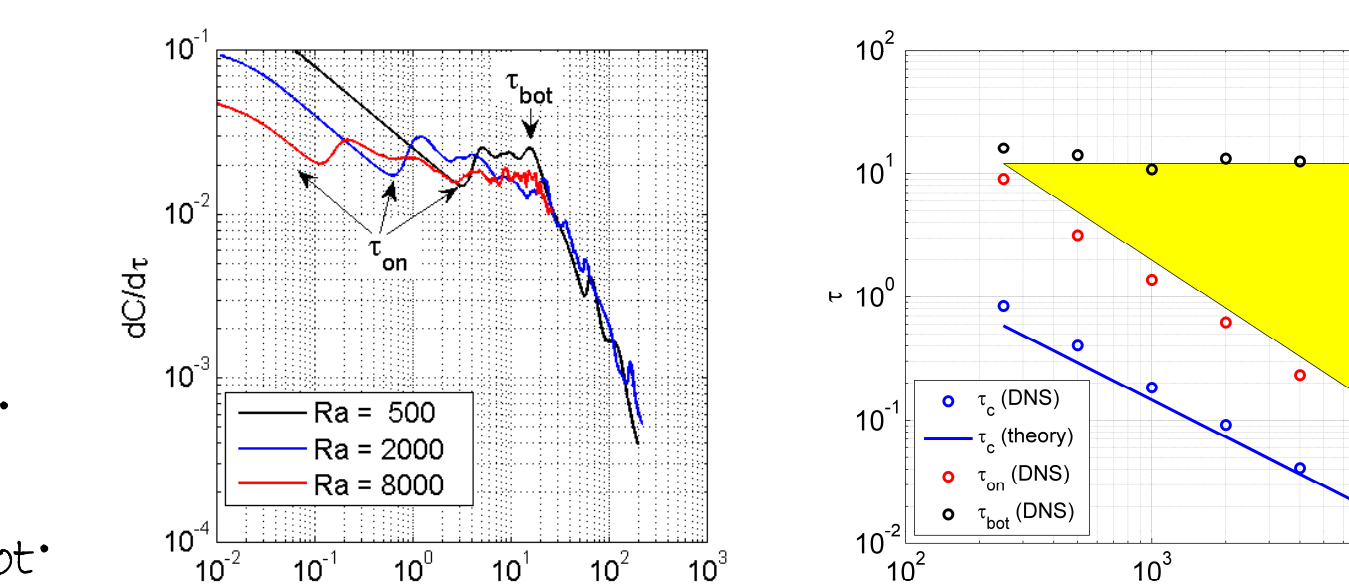


Mass transfer in a closed aquifer can be divided into 3 dynamic regimes.

- 1) initial diffusive regime dissolution rate decays as $t^{-1/2}$
- 2) Infinite acting convection dissolution rate is constant
- 3) Finite acting convection dissolution rate decreases faster than $t^{-1/2}$

The dynamic regimes are separated by 2 time scales:

- Onset time of the convective motion t_{on} .
- Time for the fingers to reach bottom t_{bot} .



The time of effective convective mass transfer is $t_{conv} = t_{bot} - t_{on}$ (shown in yellow).

Convective dissolution is less efficient in finite aquifers, because once the fingers hit the bottom convection slows down and the dissolution rate decreases rapidly.