Evolution of planar fractures: an experimental and reactive transport modelling study

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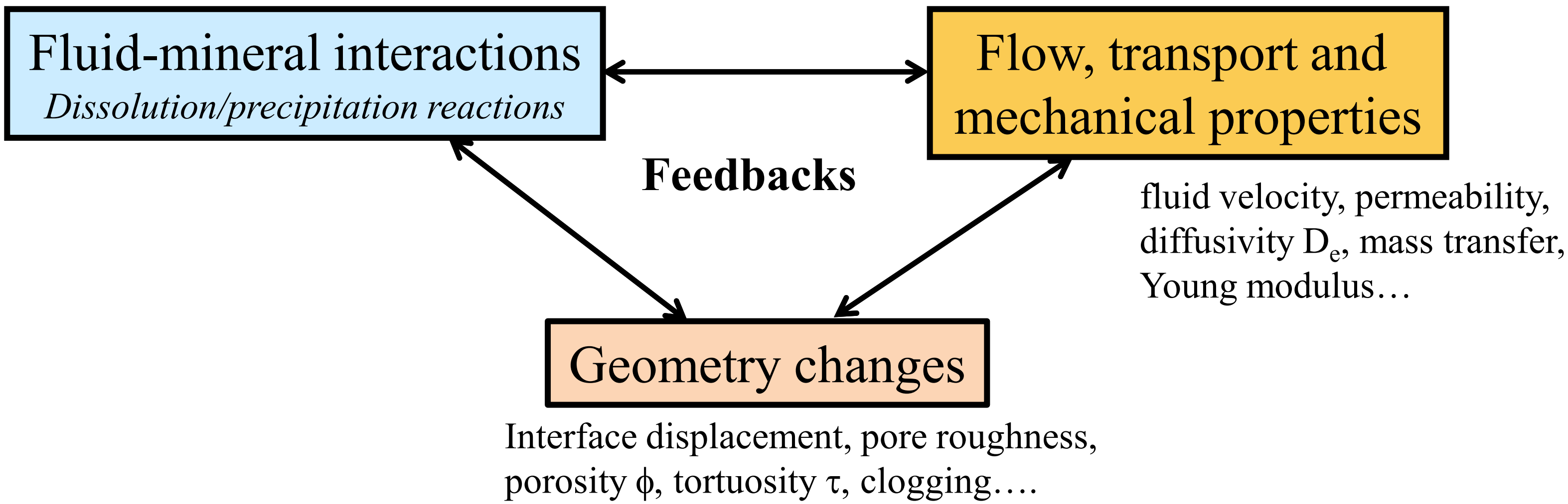
SITRAM workshop, Pau, December 2-3, 2019

Challenging reactive transport modelling

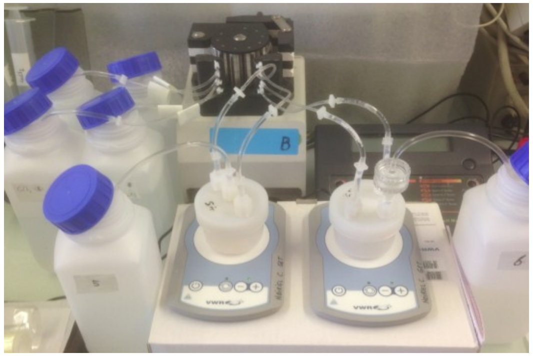
- Dissolution/precipitation reactions → Evolving porous medium / fracture geometry → Changes in flow and transport properties



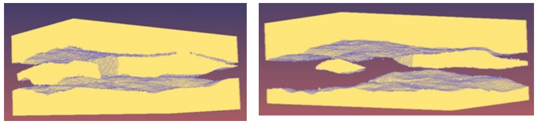
Noiriel C. and Daval D. [2017] Accounts of Chemical Research, vol 50, n°4, p. 759–768

Experimental/numerical approach

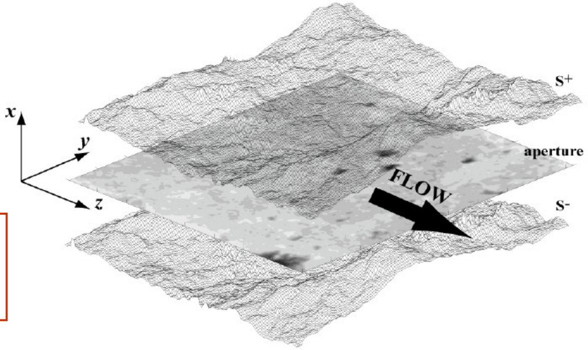
Lab experiments in natural rock / artificial materials (crystals, fractures & porous media)



4D imaging with X-ray micro-tomography
…



Numerical modelling of reactive transport



Observations, characterisation

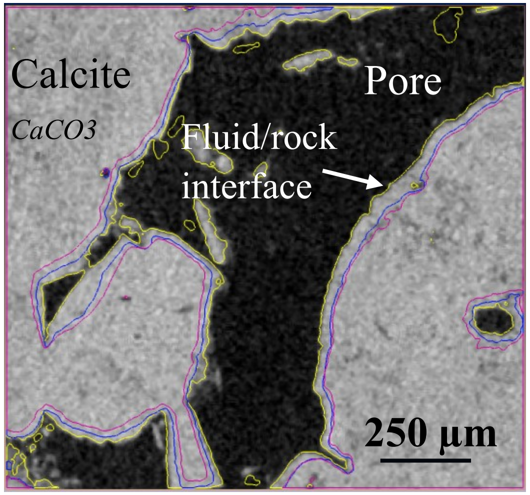
Process quantification

modelling

$$\frac{\partial}{\partial t}(\phi C_i) - \mathcal{L}(\tilde{C}_i) = \mathcal{R}(C_i)$$

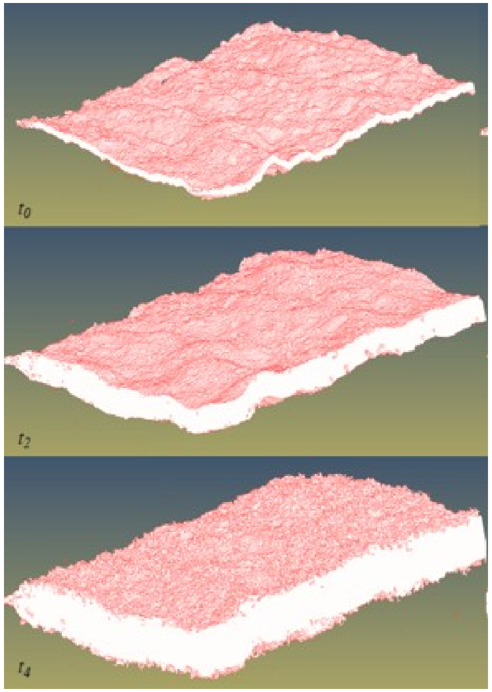
X-ray micro-tomography applications to RT

Dissolution of porous rocks



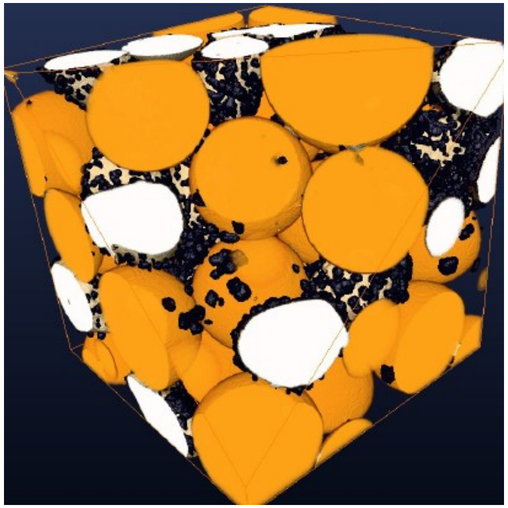
Noiriel et al. [2014] GRL
Noiriel et al. [2015] Oil & Gas Sc. Tech.

Reactive flow into fractures



Noiriel et al. [2013] J. Hydrology

Precipitation in porous media



Noiriel et al. [2012] Chem. Geology
Noiriel et al. [2016] Adv. Water Res.

Crystal reactivity

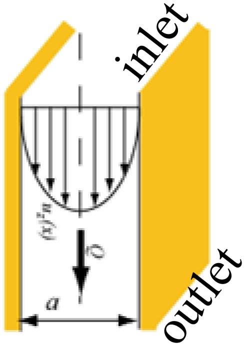
Noiriel et al. [2019] Earth & Space Chemistry

Mineral/structural heterogeneity and instability

- Experiment → injection at pH = 3.8 at different flow rates in a pure limestone

→ ↗ Pe_0

$$Pe = \frac{U\,L^*}{D_m}$$

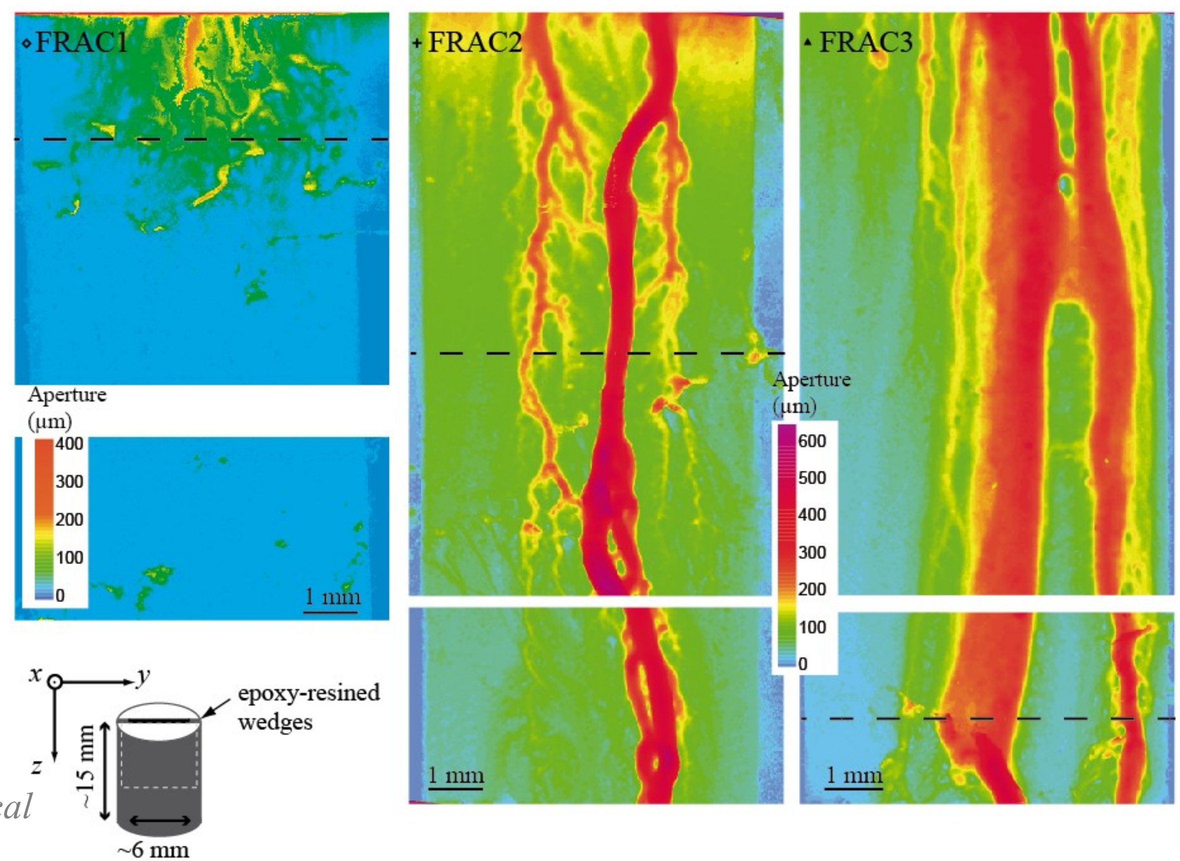


$a_0 = 20\ \mu m$

Q = 1.2 $cm^3.h^{-1}$ Δt = 164.5h

Q = 102 $cm^3.h^{-1}$ Δt = 55h

Q = 300 $cm^3.h^{-1}$ Δt = 26.5h

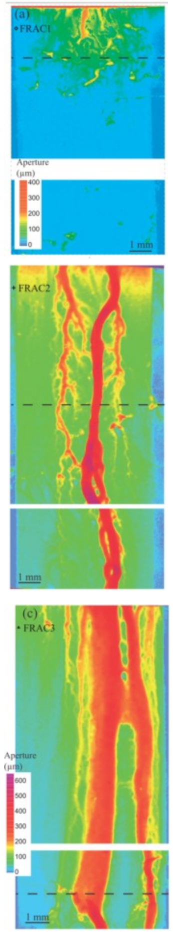


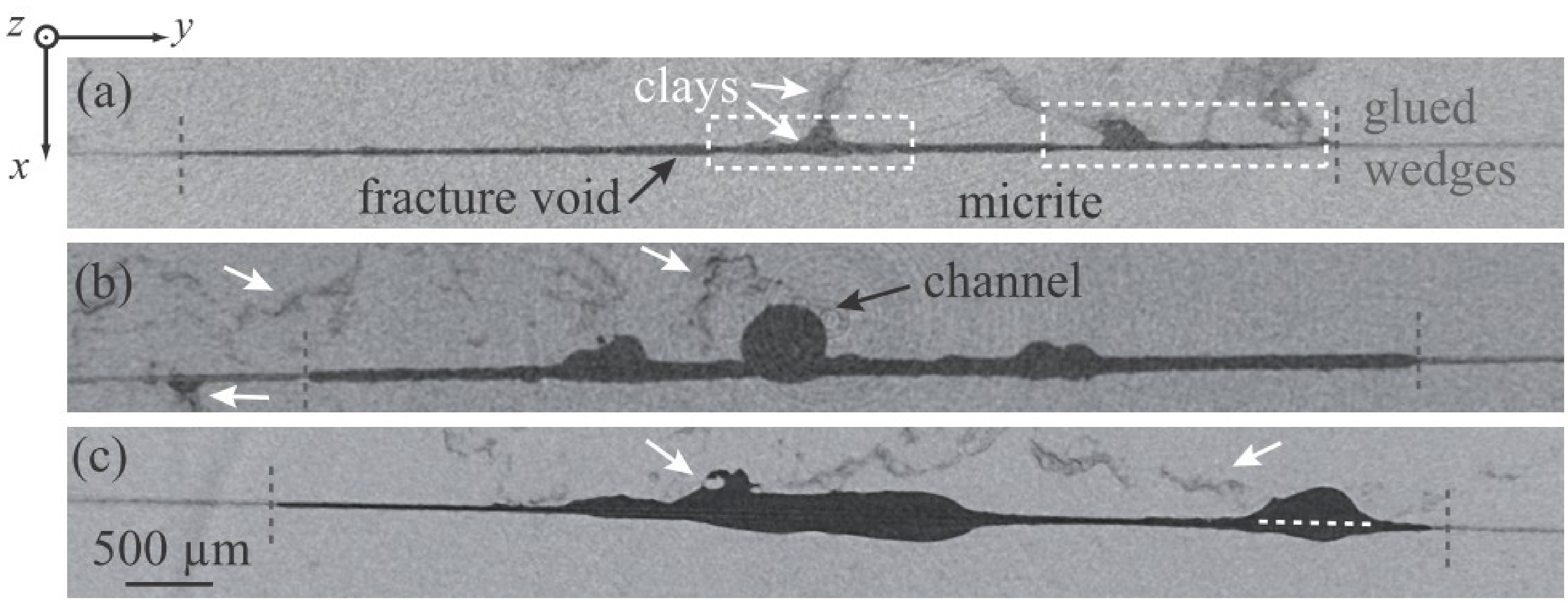
Transition from conical to ramidified to dominant wormhole

Noiriel and Deng [2018] Chemical Geology, vol. 497, 100-114

Evolution of fracture morphology

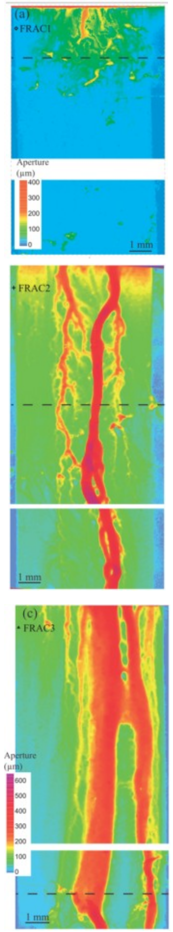
- Heterogeneity developement inherent to the presence of clay spots and clays layers (~1% of the limestone matrix content)

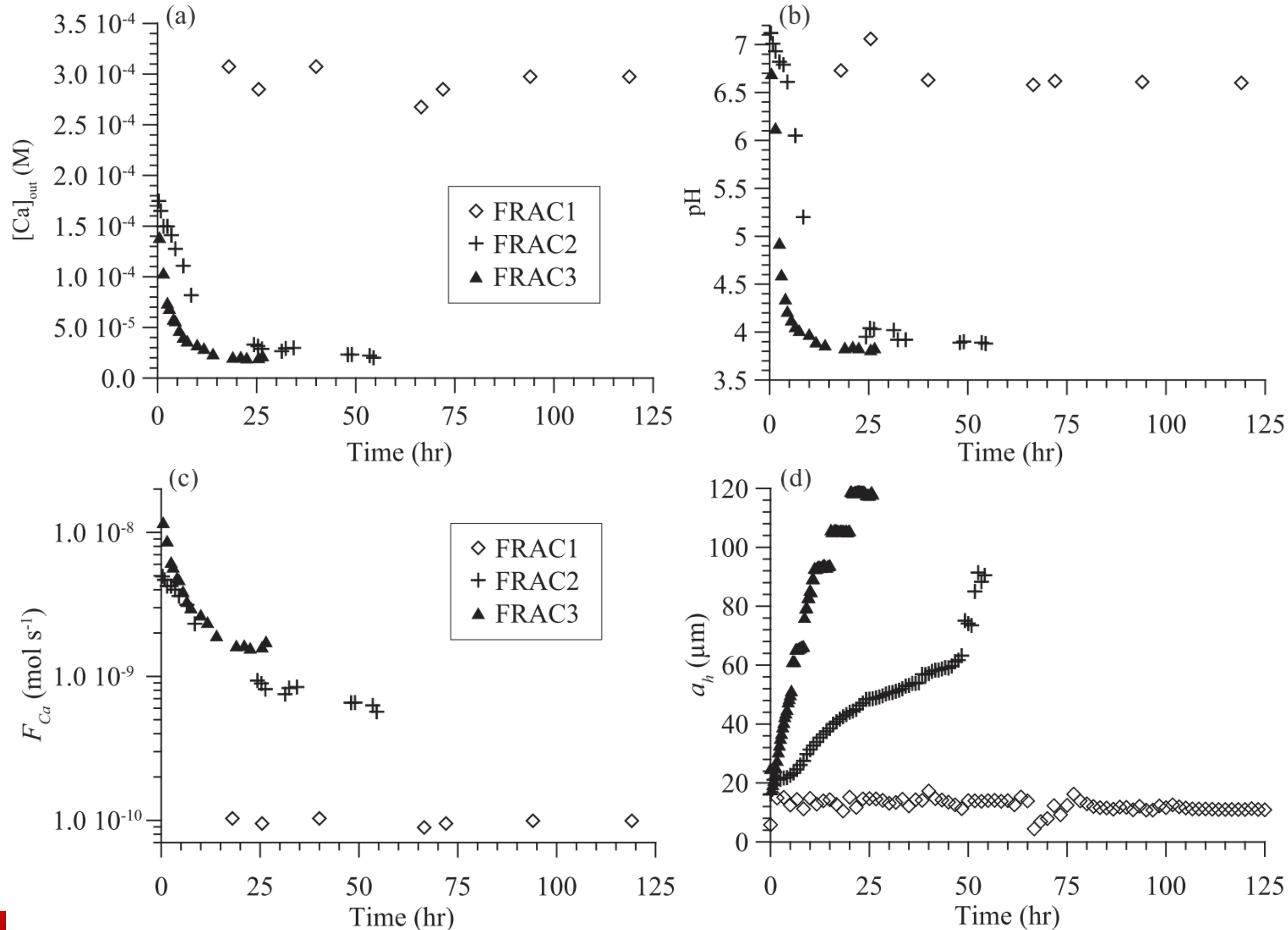




Evolution of fluid chemistry and permeability

- Wormhole breakthrough results in a drop of pH, $[Ca^{2+}]_{out}$, $F_{Ca(out)}$ and increase in permeability





Reactive transport modelling

- 2.5D code derived from Crunchflow and adapted to meshing of the fracture plane
- Grid cell porosity and permeability derived from local aperture

$$\phi_{y,z} = \frac{a_{y,z}}{d_x} \qquad k_{y,z} = \frac{a_{y,z}^3}{12 d_x}$$

- Fluid velocity solved from Darcy's law

$$\phi \, \mathbf{v} = \frac{k \nabla P}{\mu}$$

- ARDE for reactive transport

$$\frac{\partial(\phi \, \Psi_i)}{\partial t} = \nabla \cdot (\phi \, \mathbf{D} \, \nabla \Psi_i - \mathbf{v} \, \Psi_i) - \sum_m \nu_{i,m} R_m$$

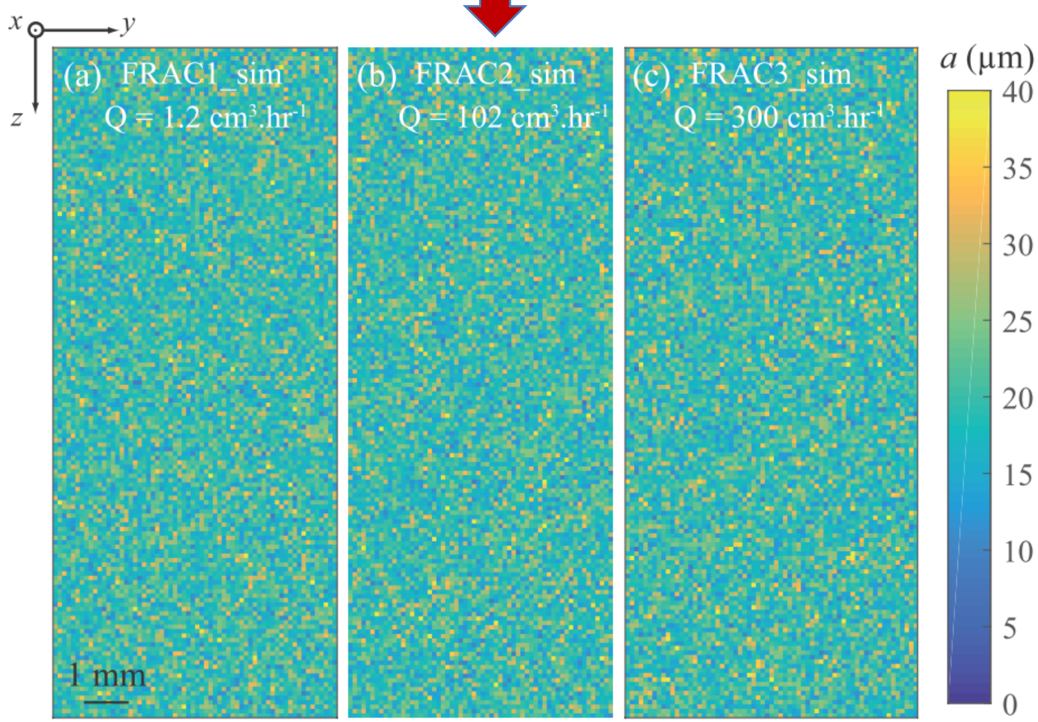
- Thermo-kinetics formulation for calcite dissolution

Initial fracture geometry and boundary conditions

- Random aperture fields (lognormal distribution) $a_0 = 20 \pm 5\mu m$

Qexp

p[HCl]$_0$=3.8



Free flux

Patm

Grid cell 0.1 ×0.1 mm
Δt same as in the experiments

Reactive transport modelling

- Comparison between model and experiments
- Test on 3 parameters:
  - Influence on the reaction rate (kinetic constant k_1 and surface roughness factor SFR)
  - Role of the local heterogeneities (clay spots)
  - Local transport limitations (poor *vs* well-mixed conditions across the fracture walls)

Effect of calcite reactivity

- Kinetic formulation (values of k_1, k_2 and k_3) differs from authors

$$R_m = -\frac{dC_{min}}{dt} = A_m k_{cal} (1 - \Omega)$$

$$k_{cal} = k_1 a_{H+} + \underbrace{k_2 a_{CO2} + k_3}_{\sim 0}$$

  - log k_1 = -0.05 (Chou et al., 1989)
  - log k_1 = -0.3 (Plummer et al., 1978)
  - log k_1 = -1.08 (Deng et al., 2015)

- Surface area is an unknown parameter

$$R_m = -\frac{dC_{min}}{dt} = A_m k_{cal} (1 - \Omega)$$

$$A_m = A_{geo} \cdot SRF = 2 \cdot d_y \cdot d_z \cdot SRF$$

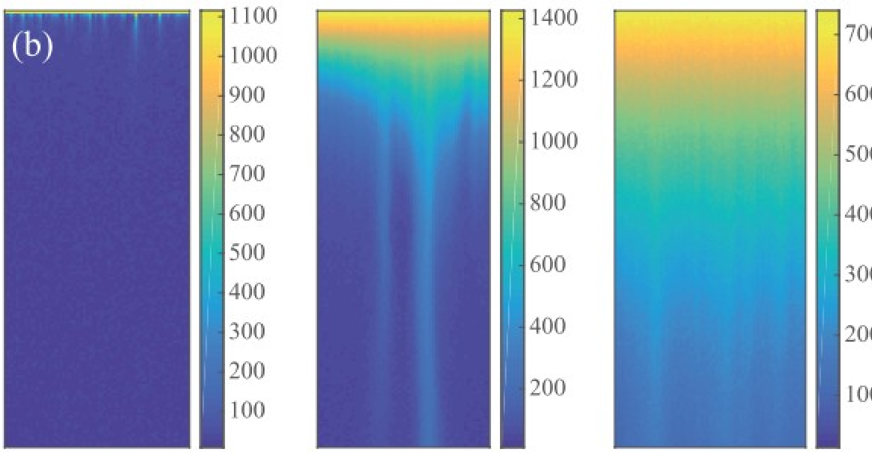
  - Surface roughness factor SFR = 1
  - Surface roughness factor SFR = 4
  - Surface roughness factor SFR = 10

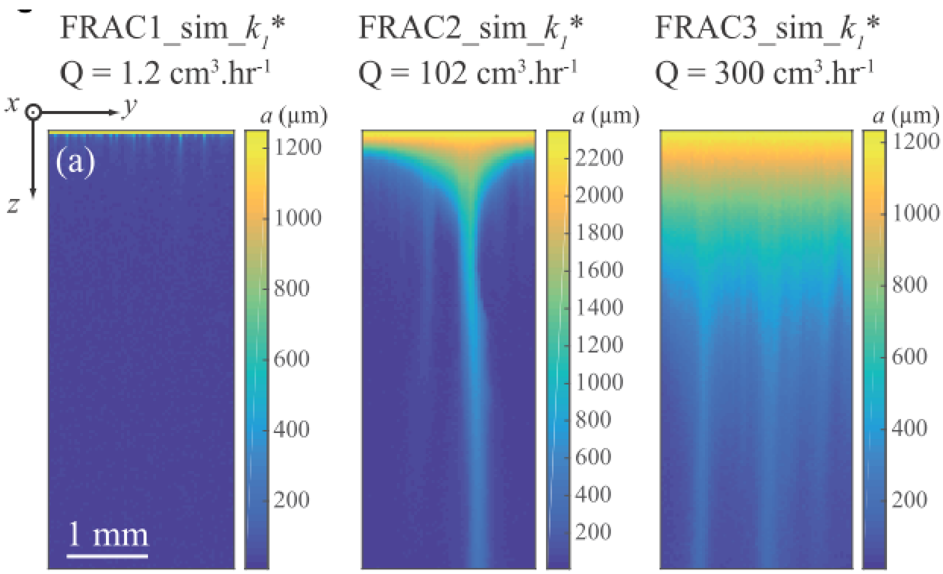
Effect of kinetic constant k_1 (pH-dependence)

- Influence on the spatial dissolution distribution and reactive infiltration instability

SFR = 1 in all simulations

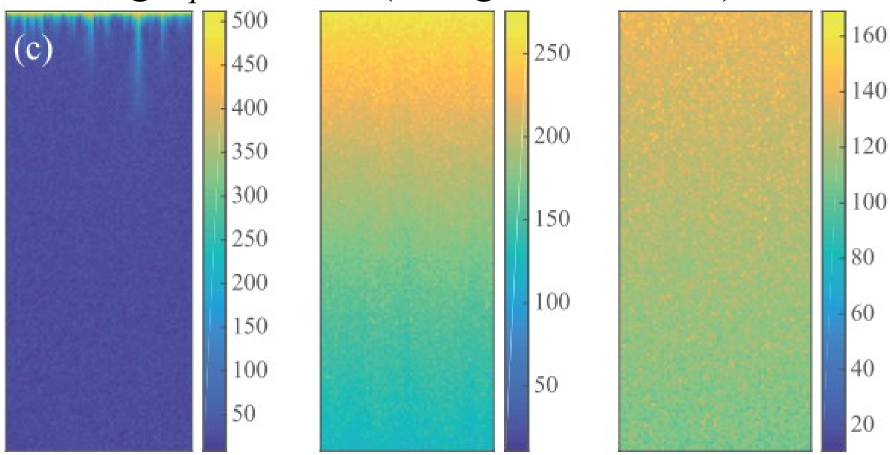
log k_1 = -0.3 (Plummer et al., 1978)





log k_1 = -0.05 (Chou et al., 1989)

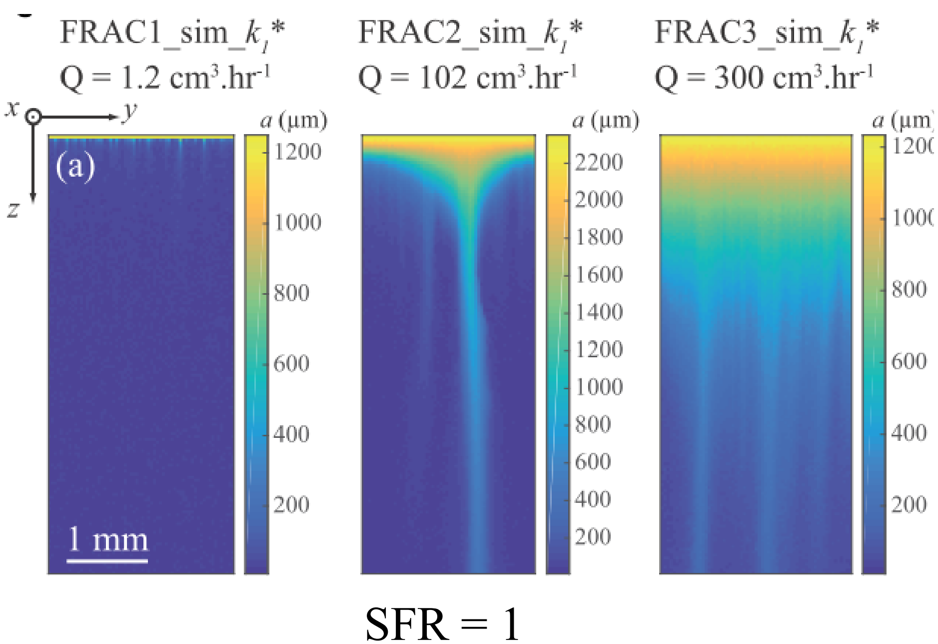
log k_1 = -1.08 (Deng et al., 2015)

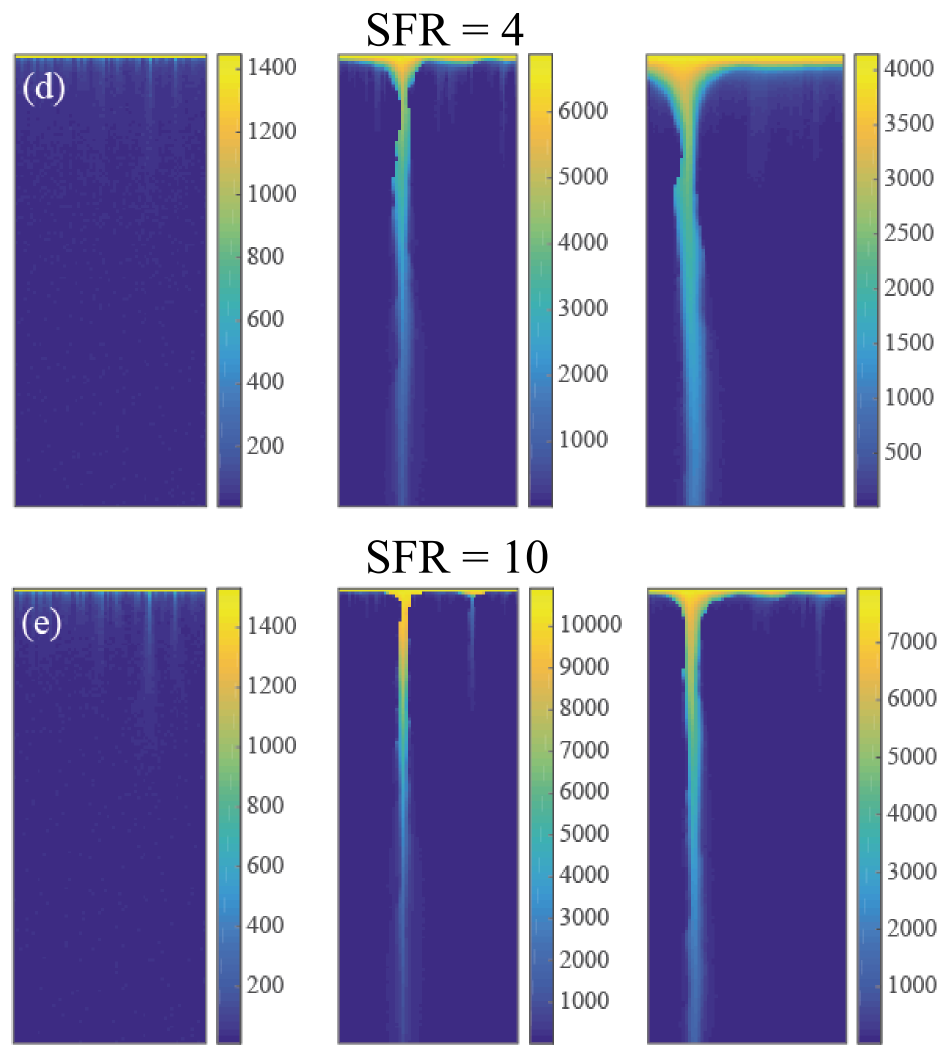


Effect of surface roughness factor

- Influence on the spatial dissolution distribution and reactive infiltration instability

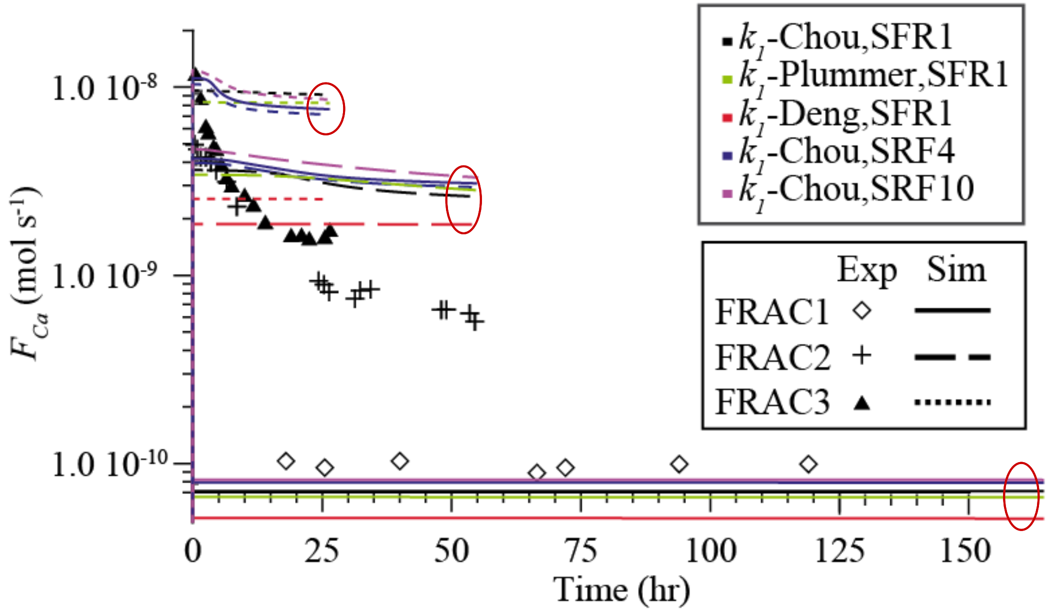
log k_1 = -0.05 (Chou et al., 1989) in all simulations

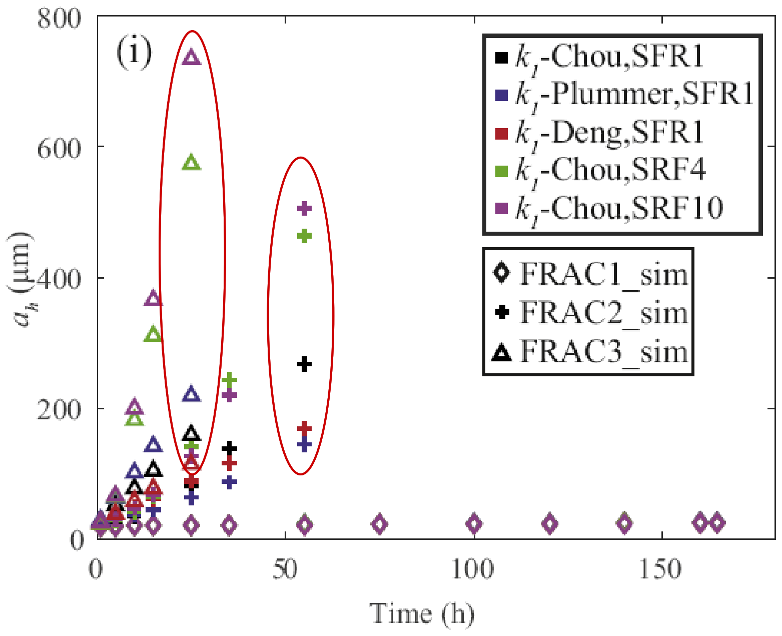




Effect of reaction rate (k_1 and SFR)

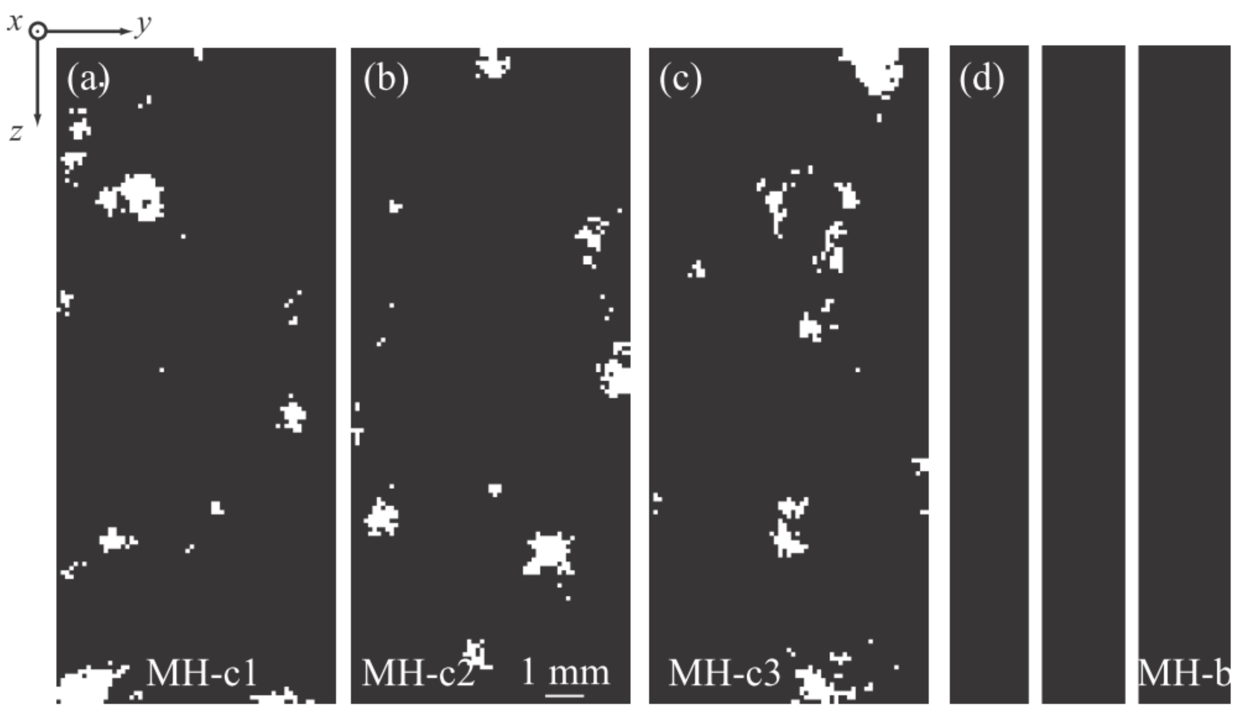
- Small effects on chemical flux but large effect on permeability evolution





Effect of mineral heterogeneity

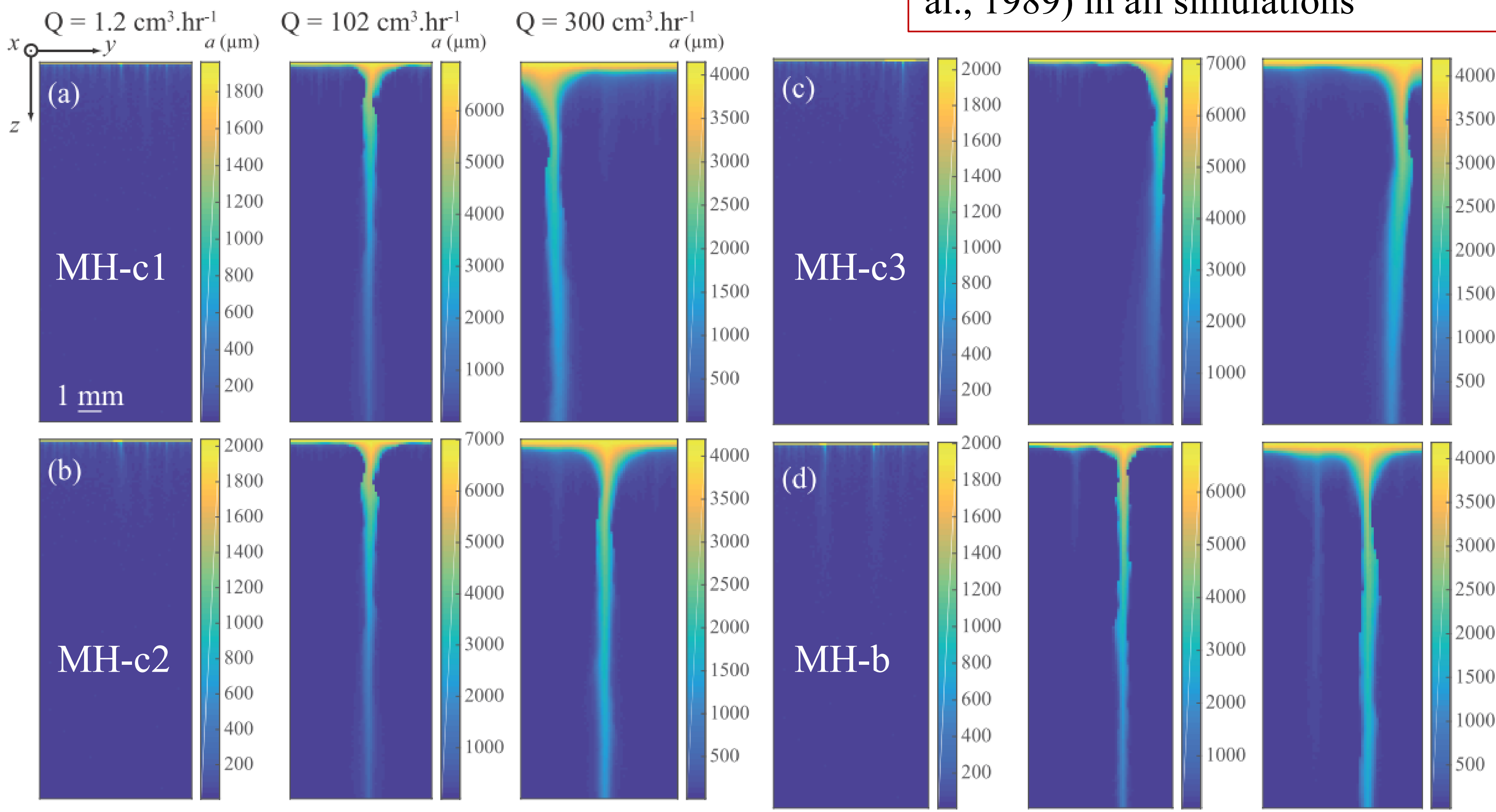
- Mask with different heterogeneity level (from non-reactive isolated clay spots to clay layers) mapped on the aperture field (calcite volume fraction reduced to 60%)



Effect of mineral heterogeneity

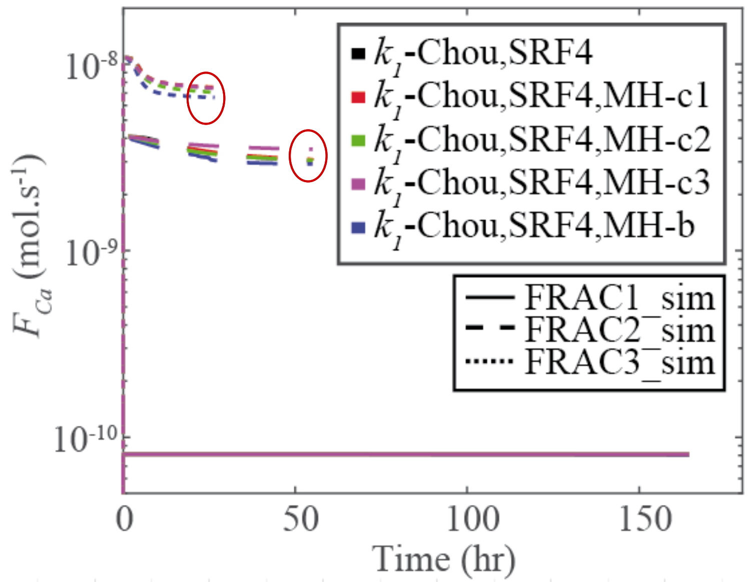
- Influence on the wormhole localisation

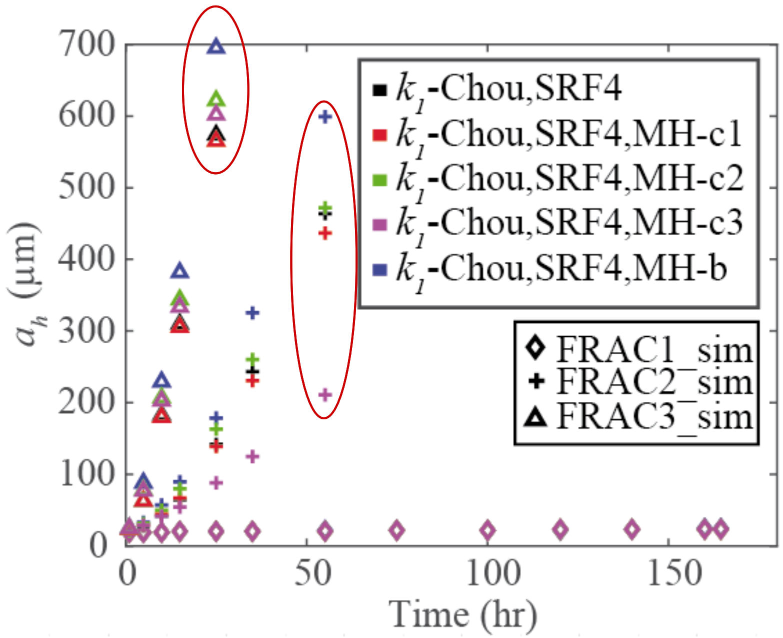
SFR = 4 and log k_1 = -0.05 (Chou et al., 1989) in all simulations



Effect of mineral heterogeneity

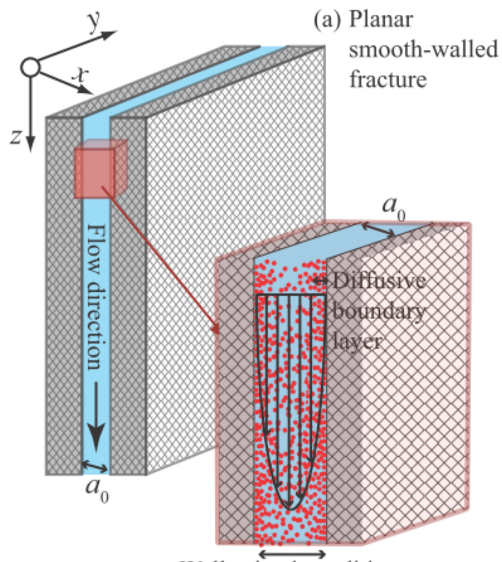
- Very slight effects on chemical flux and moderate to large effects on permeability evolution



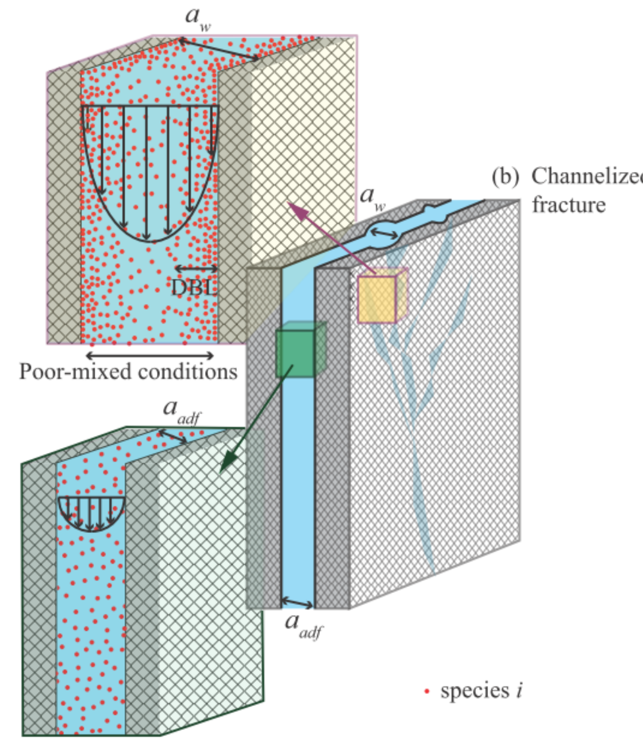


Effect of local transport limitations

- In the initial fractures



- After wormhole formation



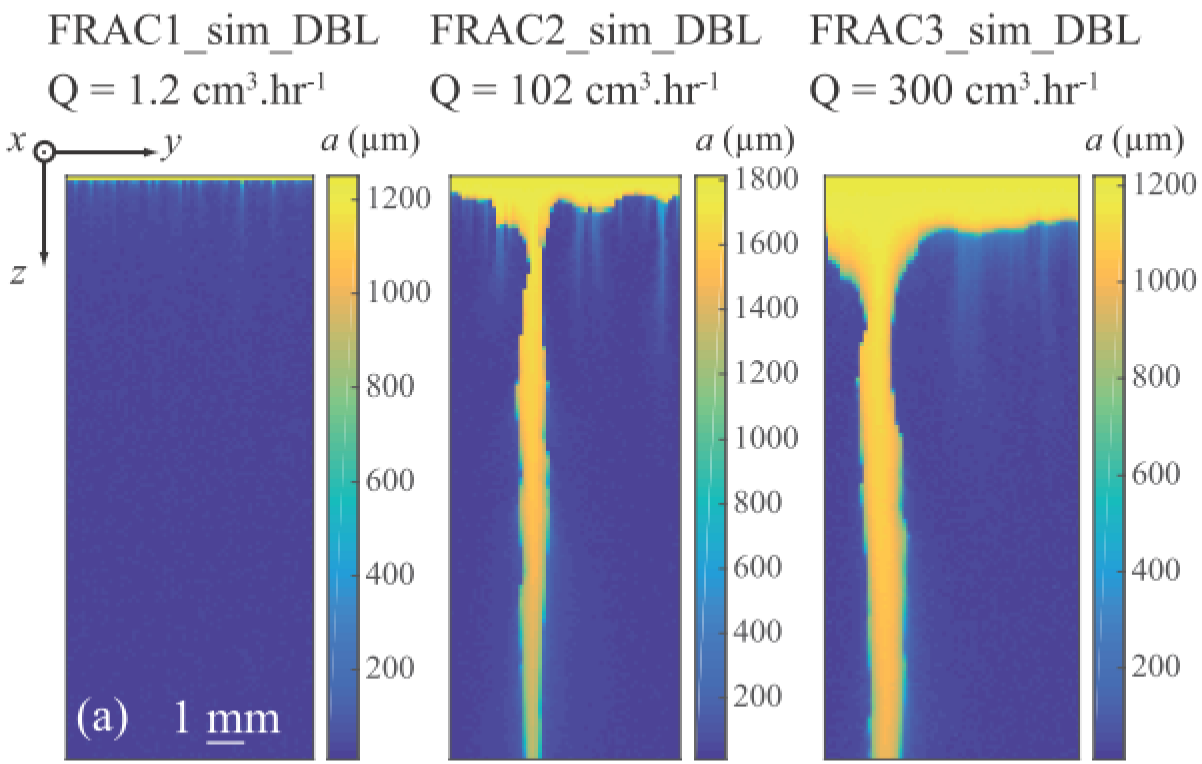
- Diffusion limitation in a boundary layer

$$R_{eff} = \frac{1}{\frac{1}{R_{diff}} + \frac{1}{R_{surf}}}$$

with $R_{surf} = R_m$ and $R_{diff} = \frac{D_0\, Sh}{2a}\left([j] - [j]_s\right)$

Effect of local transport limitations

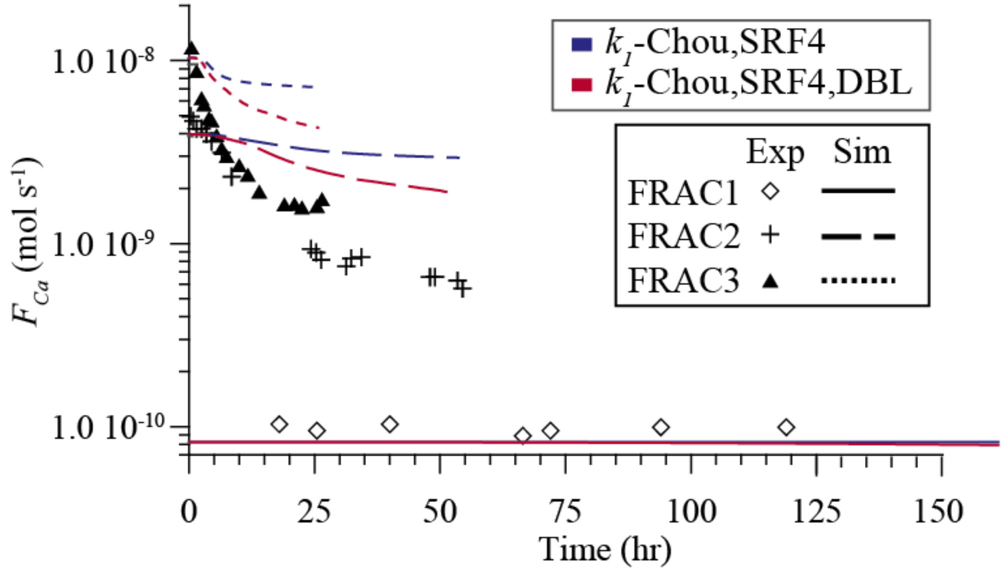
- Favors the fluid localisation and thus the decrease of chemical flux



SFR = 4, log k_1 = -0.05 (Chou et al., 1978) and clay mask Mc-1 in all simulations

Effect of local transport limitations

- Favors the fluid localisation and thus the decrease of chemical flux



Noiriel and Deng [2018] Chemical Geology, vol. 497, 100-114

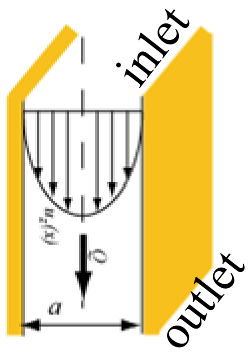
- → Unfortunately fitting with experimental results remains tricky
- → Pore-scale modelling?

Conclusions/perspectives

- Ability of Darcy's scale model to develop reactive flow instabilities
- But wormhole patterns (ramified and dominant) not well reproduced
- Fully coupled model integrating detailed description of flow (Stokes) and reactivity, as well as flow reorganization with moving interface is required. Any idea…?
- Reference: C. Noiriel and H. Deng [2018] Evolution of planar fractures in limestone: the role of flow rate, mineral heterogeneity and local transport processes, Chemical Geology, 497, pp. 100-114, https://doi.org/10.1016/j.chemgeo.2018.08.026

Experimental conditionss

- Injection of HCl into planar fractures at three different flow rates



a_0 = 20 µm

Sample	Experiment duration (hr)	Flow rate Q (cm^3.h^{-1})	Length L (mm)	Width l (mm)	a_0 (µm)	Pe_0	$\bar{v}_0$ (m.s^{-1})
FRAC1	164.5 h	1.2	15.1	6.4	20*	7	2.6 10^{-3}
FRAC2	55 h	102	15.2	6.0	20*	598	2.4 10^{-1}
FRAC3	26.5 h	300	14.9	6.4	20*	1648	6.5 10^{-1}

Flow velocity field

- Calculated from the local cubic law