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Coupled formulations and coupling algorithms for reactive transport in porous media

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INRIA Rocquencourt

Laboratoire de Mathématiques Appliquées de Compiègne

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SHPCO2

GdRMoMaS

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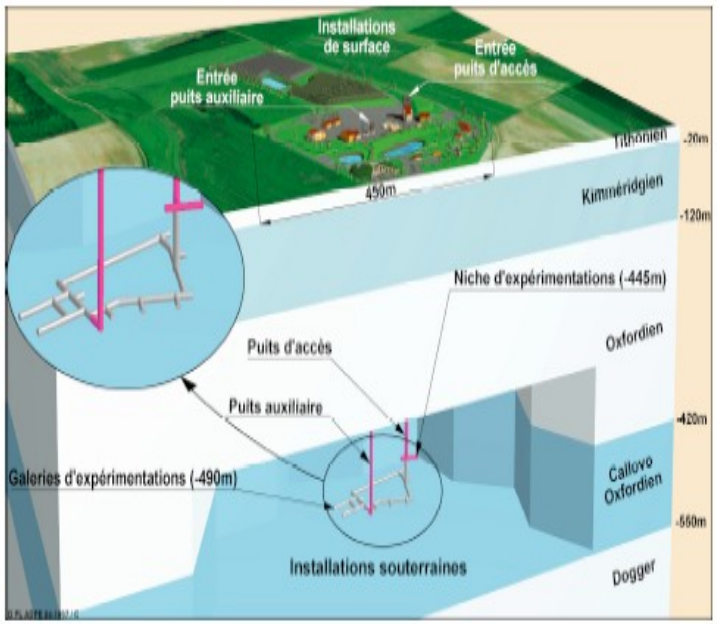
Outline

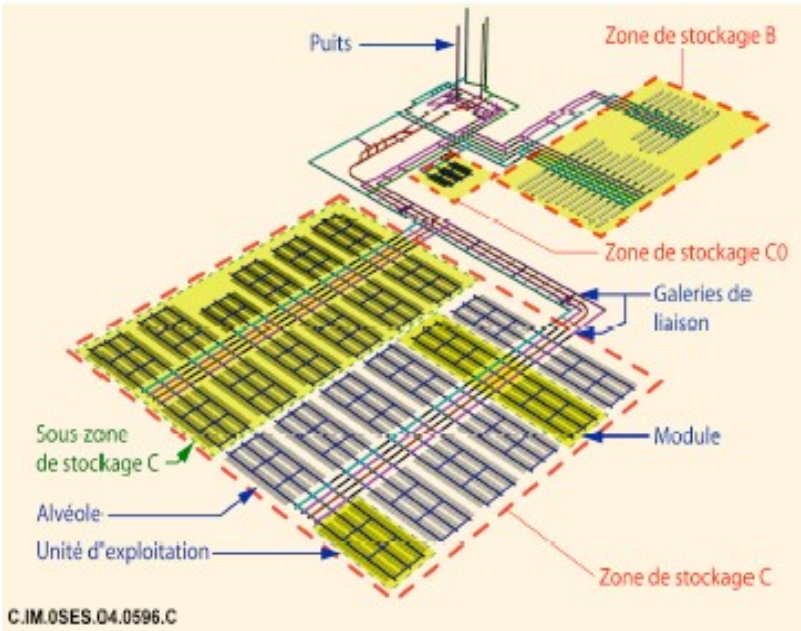
Nuclear waste storage (1)

- Assess safety of deep geological nuclear waste storage (clay layer)
- Long term simulation of radionuclide transport (one million years)
- Wide variation of scales : from package (meter) to regional (kilometers)
- Geochemistry : large number of species
- Strong government regulation
- Main actors : ANDRA, cea, EDF
- Research in mathematical and numerical modeling is conducted in the CNRS MOMAS group (Director A. Ern).

Nuclear waste storage (2)

Present choice in France : a sedimentary geological formation (Bures, in the Meuse region)





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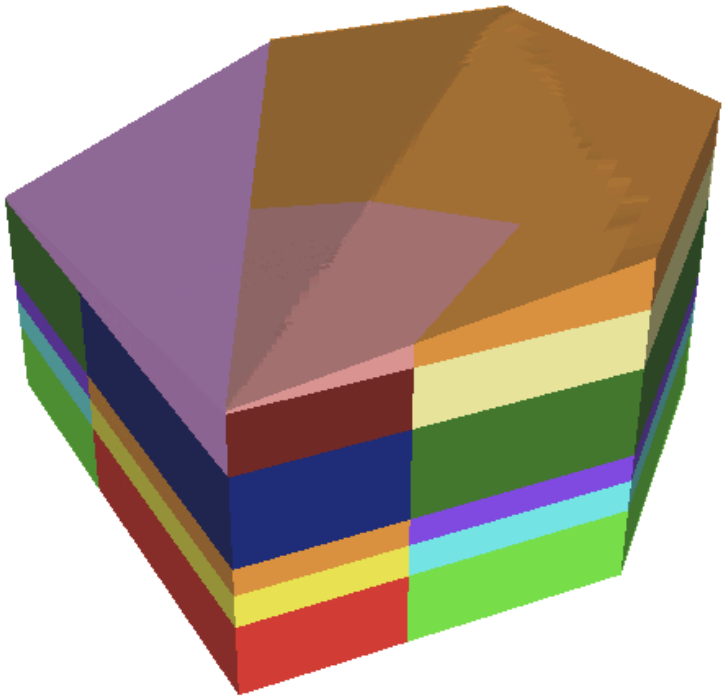
A 3D far field model (V. Martin)



Blown-up 30 times vertically

Difficulties

- Distorted geometry (horizontal $\approx$ 40 km, vertical 700 m)
- Strong heterogenities (permeability varies by 8 orders of magnitude)
- General hexahedral mesh
- Simulation over 500 000 years

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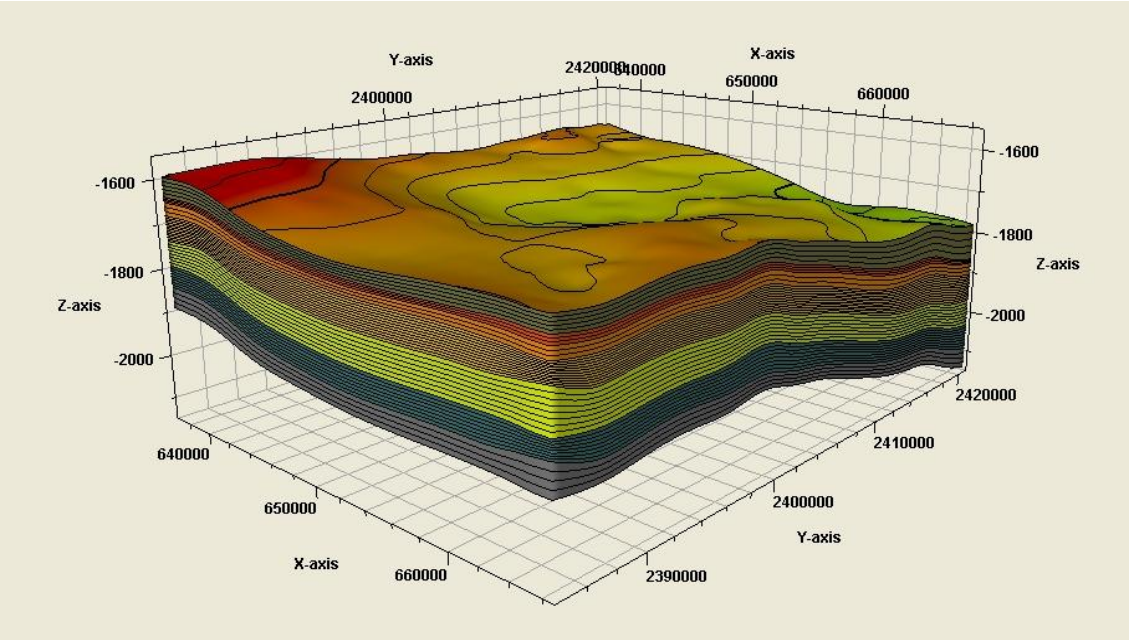
CO_2 sequestration



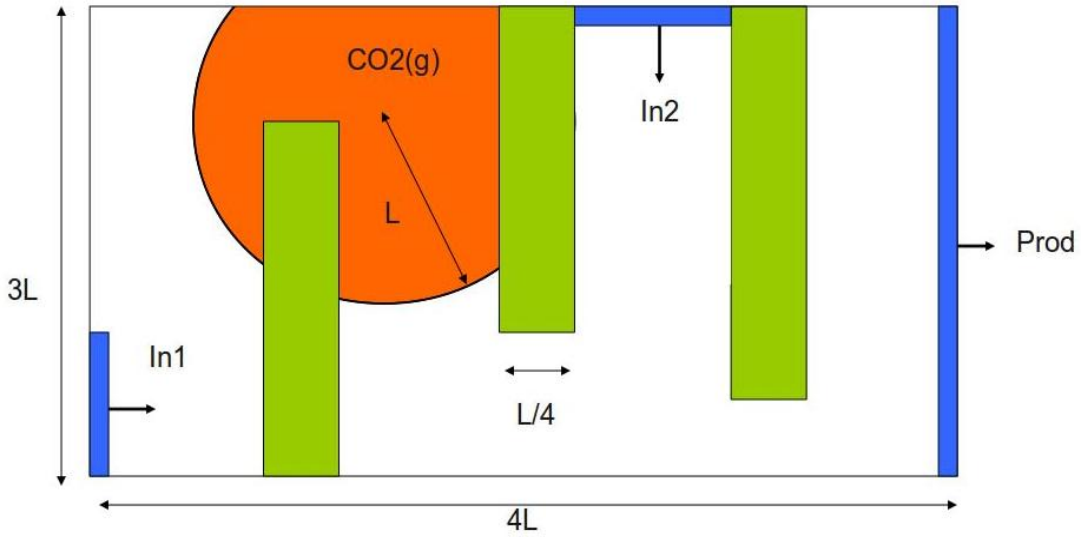
Geological model, BRGM (21 million grid points)

- Long term capture of CO_2 in saline aquifer
- Simulation to understand CO_2 migration through salt
- Coupling of liquid and gas phase, reactive transport

ANR SHPCO2 project High Performance Simulation of CO_2 sequestration

CO_2 sequestration : a synthetic model

Minimal chemical system that still "looks" realistic for CO_2 storage



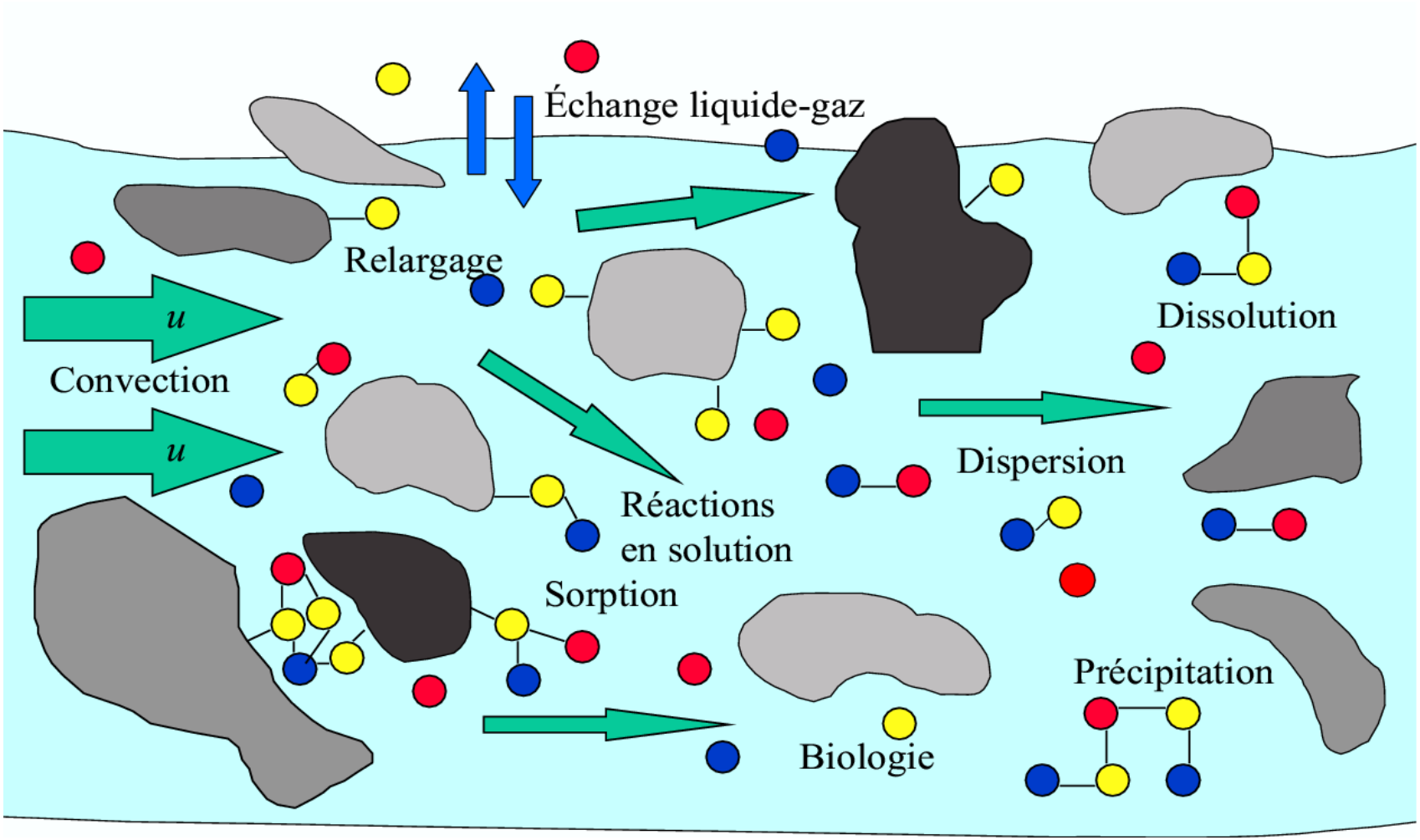
Dissolution of CO_2 in water, dissolution of calcite. Gas assumed immobile (capillary trapping), decouples flow from reactive transport.

Chemical system

- $H_2O \rightleftharpoons H^+ + OH^-$ water dissociation
- $CO_{2\,(g)} \rightleftharpoons CO_{2\,(aq)}$ gas dissolution
- $H_2O + CO_{2\,(aq)} \rightleftharpoons HCO_3^- + H^+$ dissociation of aqueous CO_2
- $CaCO_3 + H^+ \rightleftharpoons Ca_2^+ + HCO_3^-$ Dissolution of calcite

Outline

Transport and chemical phenomena



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Flow model

Flow equations

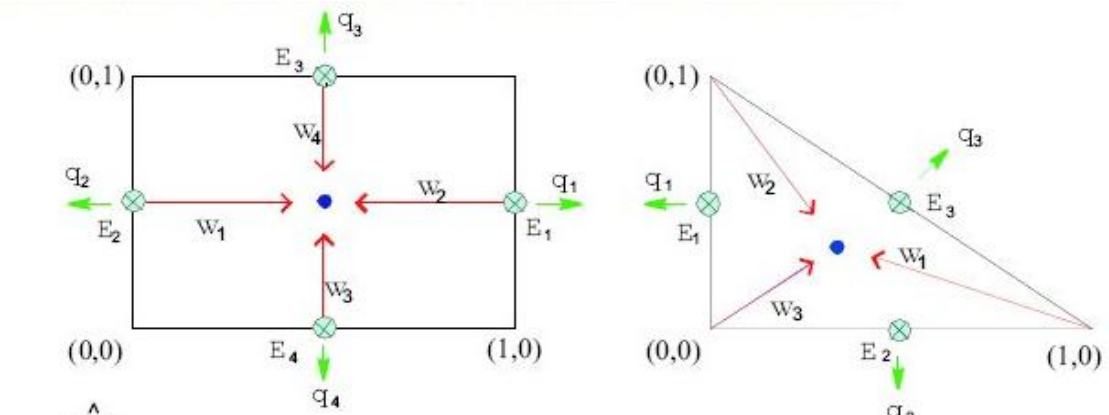
$q = -K\nabla h$ Darcy's law h piezometric head

$\nabla \cdot q = 0$ incompressibility q Darcy velocity

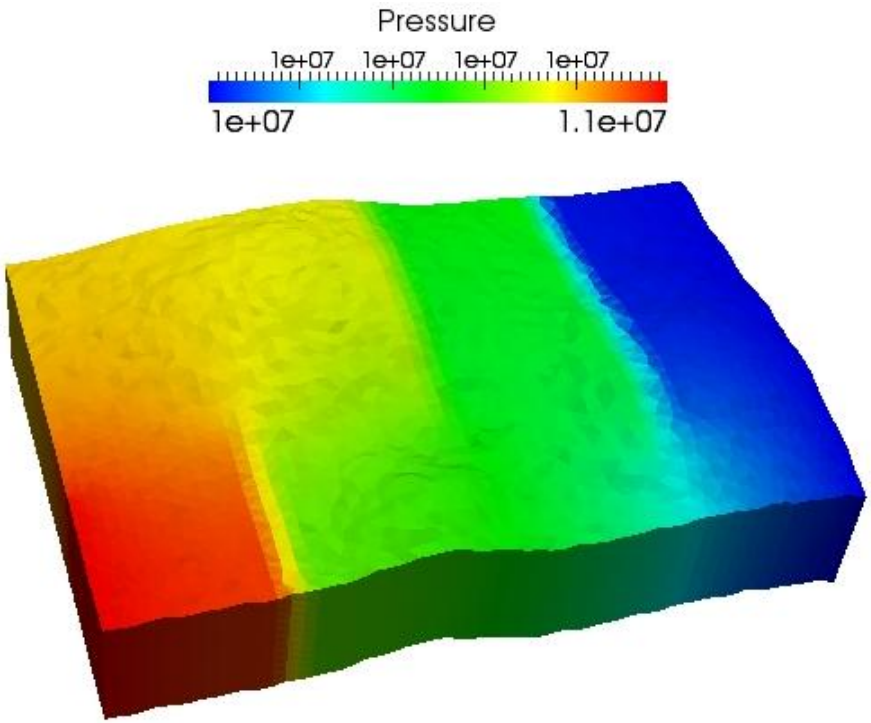
K permeability tensor (heterogeneous, anisotropic)

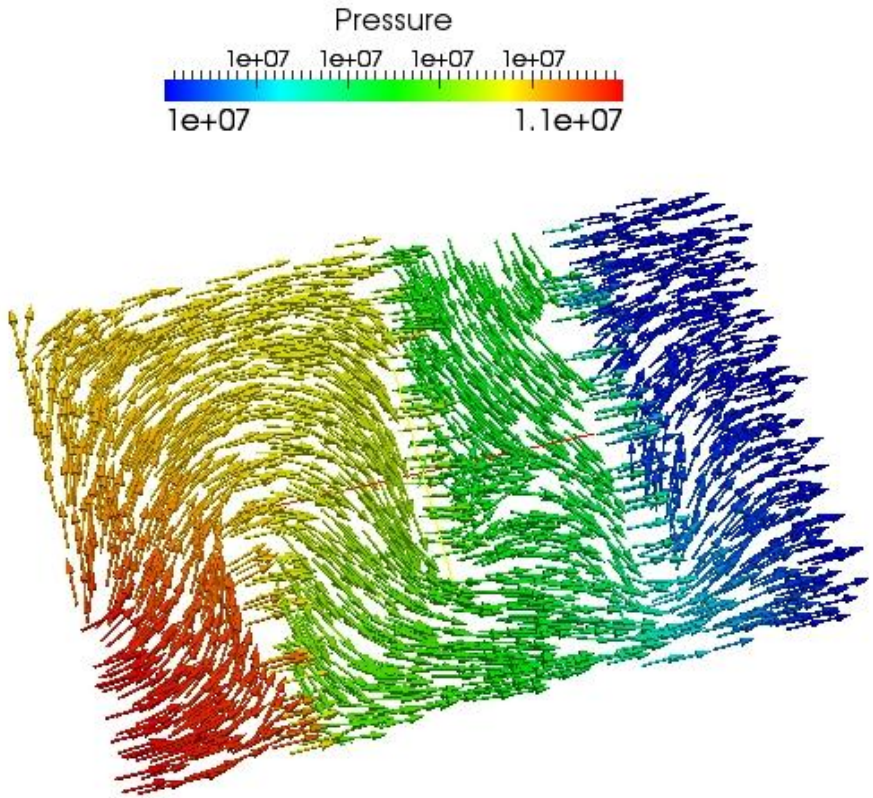
Mixed finite elements

- Approximate both head and velocity
- Continuous flux across element faces
- Locally mass conservative
- Allows full diffusion tensor



Pressure and velocity for CO2 example (A. Fumagalli, M. Franco)





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Transport model

Convection–diffusion equation

$$\phi \frac{\partial c}{\partial t} - \underbrace{\operatorname{div}(\mathbf{D}\operatorname{grad} c)}_{\text{dispersion}} + \underbrace{\operatorname{div}(\mathbf{q}c)}_{\text{advection}} + \phi\lambda c = f$$

- c : concentration $[\mathrm{mol/l}]$
- ϕ : porosity [–]
- λ radioactive decay $[\mathrm{s}^{-1}]$
- $\mathbf{q}$ Darcy velocity $[\mathrm{m/s}]$

Dispersion tensor

$$\mathbf{D} = d_e \mathbf{I} + |\mathbf{q}|[\alpha_l \mathbf{E}(\mathbf{q}) + \alpha_t (I - \mathbf{E}(\mathbf{q}))], \quad E_{ij}(\mathbf{q}) = \frac{q_i q_j}{|\mathbf{q}|}$$

α_l, α_t dispersivity coeff. $[\mathrm{m}]$, d_e molecular diffusion $[\mathrm{m/s}^2]$

Notation $\operatorname{div}(\mathbf{q}c - \mathbf{D}\operatorname{grad} c) + \phi\lambda c \underset{\text{def}}{=} Lc$

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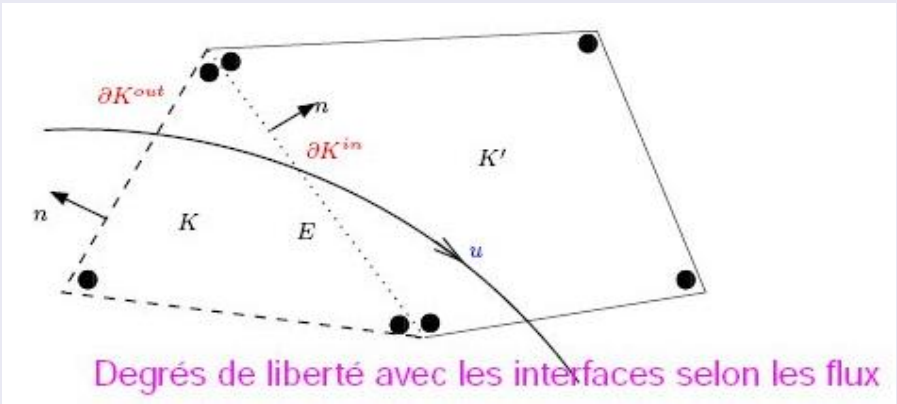
Solution by operator splitting

Advection step

Explicit, finite volumes / discontinuous Galerkine

- Locally mass conservative
- Allows unstructured mesh
- CFL condition : use sub–time–steps



Dispersion step

Like flow equation (time dependent) : mixed finite elements (implicit)

See Ackerer et al., Putti et al., Arbogast et al., ...

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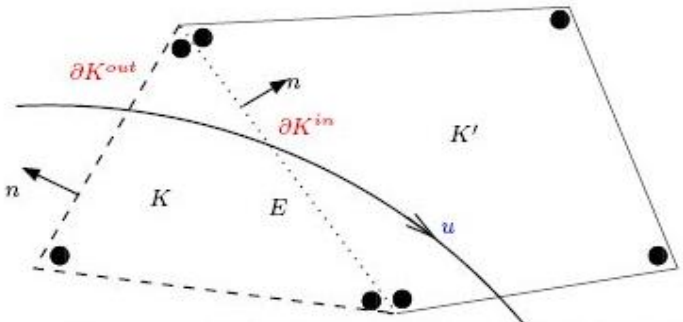

Degrés de liberté avec les interfaces selon les flux

Dispersion step

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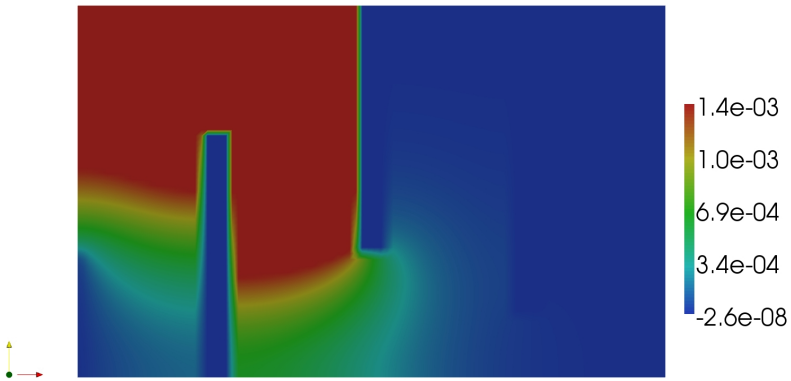
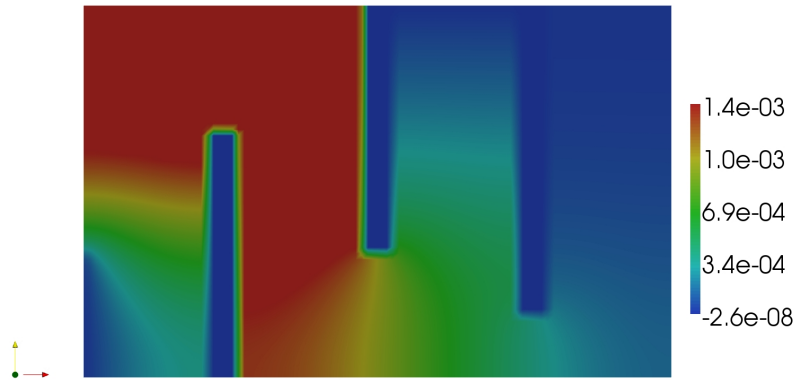
See Ackerer et al., Putti et al., Arbogast et al., ...

Condense transport solver, one time step

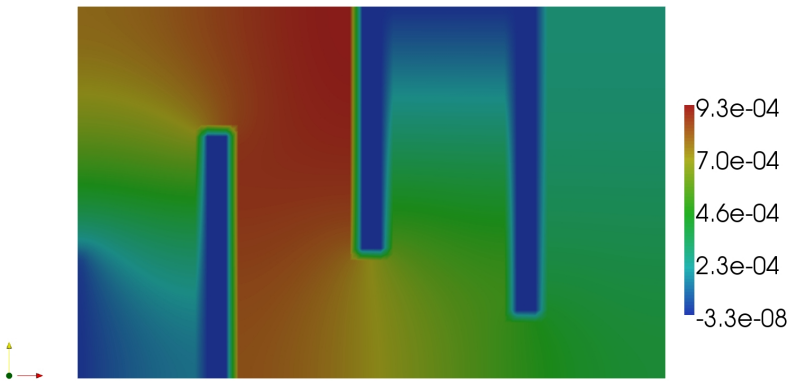
$$C^{n+1} = \Psi_T(f^n, C^n)$$

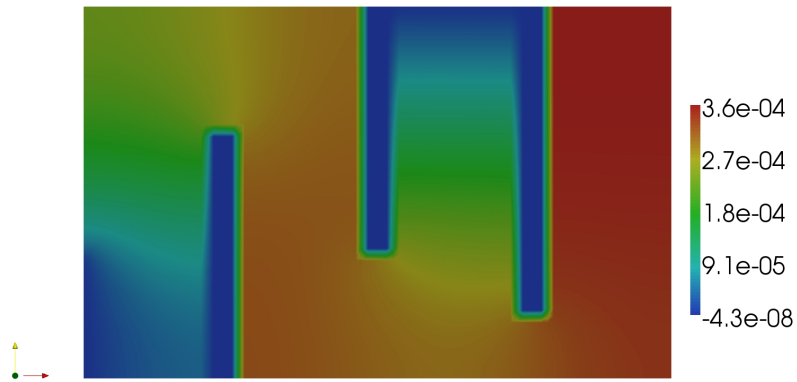
Transport for synthetic CO_2 example





Left $T = 1$ day, right $T = 6$ days





Left $T = 12$ day, right $T = 37$ days

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Classification of chemical reactions

According to nature of reaction

Homogeneous In the same phase (aqueous, gaseous, ...)
Examples : Acid base, oxydo–reduction

Heterogeneous Involve different phases
Examples : Sorption, mineral precipitation / dissolution, gas dissolution, ...

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Fast reactions Reversible, modeled using equilibrium

Depends on relative speed of reactions and transport.

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This talk : only equilibrium reactions

Sorption processes

Definition

Sorption is the accumulation of a fluid on a solid at the fluid–solid interface.

Main mechanism for exchanges between dissolved species and solid surfaces.

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Can be modeled as mass action law

Modeling general equilibrium models

General chemical reactions : N_s species, N_r reactions

$$\sum_{j=1}^{N_s} \nu_{ij} Y_j \leftrightarrows 0, \quad i = 1, \ldots, N_r$$

ν_{ij} stoichiometric coefficients. Matrix equation $\nu Y = 0$

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Partition $\nu = \begin{pmatrix} G & N \end{pmatrix}$, $B \in \mathbf{R}^{N_r \times N_r}$ invertible, $N \in \mathbf{R}^{N_c \times N_r}$. Let $H = -G^{-1}N$

General solution of $\nu Y = 0$: $Y = \begin{pmatrix} x \\ c \end{pmatrix}$, $x = Hc$. $c \in \mathbf{R}^{N_c}$, $x \in \mathbf{R}^{Nr}$.

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Morel formalism :

$(c,\ \bar{c})$ primary species	$(x,\ \bar{x})$ secondary species
(c, x) mobile species	$(\bar{c},\ \bar{x})$ fixed species

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Chemical equilibrium (aqueous and sorption reactions)

Chemical reactions, mass action laws

$$x_i \leftrightarrows \sum_{j=1}^{N_c} S_{ij} c_j, \qquad i = 1, \ldots, N_x,$$

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System of non-linear equations

Mass action law

$$\log x = S \log c + \log K,$$
$$\log \bar{x} = A \log c + B \log \bar{c} + \log \bar{K}.$$

Mass conservation

$$c + S^T x + A^T \bar{x} = T, \qquad T \text{ known from transport}$$
$$\bar{c} + B^T \bar{x} = W, \qquad W \text{ imposed}$$

Mineral reactions

Dissolution of solid, precipitation of aqueous species. Reactions with threshold : which species appear unknown a priori.

Solubility product $\Pi = \log K_p + S_p \log c$

$$\begin{cases} p = 0 & \text{if } \Pi < 0 \\ \Pi = 0 & \text{otherwise} \end{cases}$$

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- Standard procedure : combinatorial search
- Reformulate as complementarity problem
- Interior point algorithm (Saaf et al. ('96), MK (05))
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Remark

Same treatment for gas, with Henry's law

Numerical solution of nonlinear problem

Take concentration logarithms as main unknowns

Use globalized Newton's method (line search, trust region).

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Role of chemical model

Given totals T (and W, known), split into

$$\text{Mobile} \quad C = c + S^T x,$$

$$\text{Fixed} \quad F = A^T \bar{x} + S_p^T p.$$

total concentrations

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total concentrations

Result of chemical problem

$$F = \Psi_C(T)$$

Outline

Reactive transport model

Balance equations

$$\begin{aligned} \phi \partial_t c + L c &= S_{mm}^T R_e^a + S_{mf}^T R_e^h \\ \phi \partial_t \bar{c} &= S_{ff}^T R_e^h \end{aligned}$$

R_e equilibrium rates unknown,

Reactive transport model

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R_e equilibrium rates unknown,

Elimination of equilibrium rates

$$\phi \frac{\partial T^{ic}}{\partial t} + L(C^{ic}) = 0, \quad ic = 1, \ldots, N_c$$

$$T_{ix}^{ic} = C_{ix}^{ic} + F_{ix}^{ic} \qquad ic = 1, .., N_c \text{ and } ix = 1, .., N_x$$

$$F_{ix} = \Psi_C(T_{ix}) \qquad ix = 1, \ldots, N_x.$$

Solution strategies

Fixed point (aka OS) Yeh–Tripathi, Carrayrou et al., Carrera et al.

- + easy to program, code reuse
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DAE formulation Erhel and de Dieuleveult

- + use quality DAE software, accurate
- − expensive

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Elimination technique Knabner et al.

- + Efficient, accurate,
- − difficult to code

A global method from the fixed–point formulation (1)

Discrete non-linear system

$$\begin{cases} C^{n+1} = \Psi_T\left(\phi\dfrac{F^n - F^{n+1}}{\Delta t}, C^n\right) \\ F^{n+1} = \Psi_C(C^{n+1} + F^{n+1}) \end{cases}$$

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uncoupled

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Can be solved by block Gauss Seidel or by Newton's method

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uncoupled

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Residual computation

1. Apply Ψ_T : solve transport for each species,
2. Apply Ψ_C : solve chemistry for each grid cell.

A global method from the fixed–point formulation (2)

- $+$ Non-intrusive approach
- $+$ Precipitation can be included
- $-$ One chemical equilibrium solve for each function evaluation

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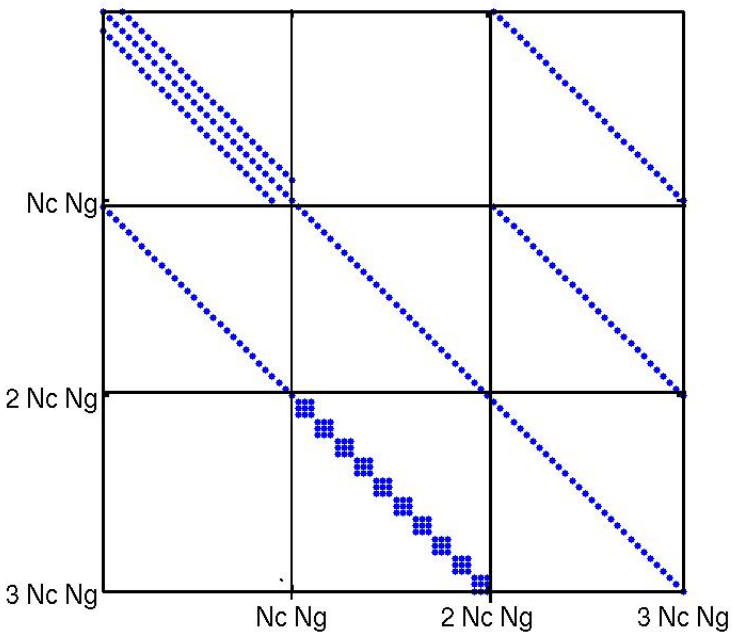


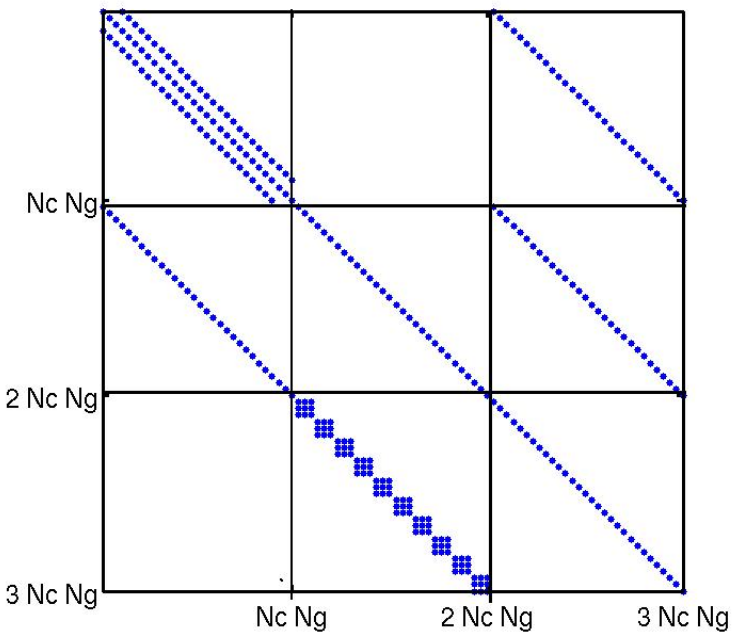
Jacobian structure

A global method from the fixed–point formulation (2)

- + Non-intrusive approach
- + Precipitation can be included
- − One chemical equilibrium solve for each function evaluation

Solution by Newton–Krylov method

- Solve the linear system by an iterative method (GMRES)
- Requires only jacobian matrix by vector products, Jacobian not stored
- Keep transport and chemistry as black–boxes (up to Jacobian computation)



Jacobian structure

Newton–Krylov method

Used for CFD, shallow water, radiative transfer(Keyes, Knoll, JCP 04), and for reactive transport (Hammond, Valocchi, Lichtner, Adv. Wat. Res. 05)

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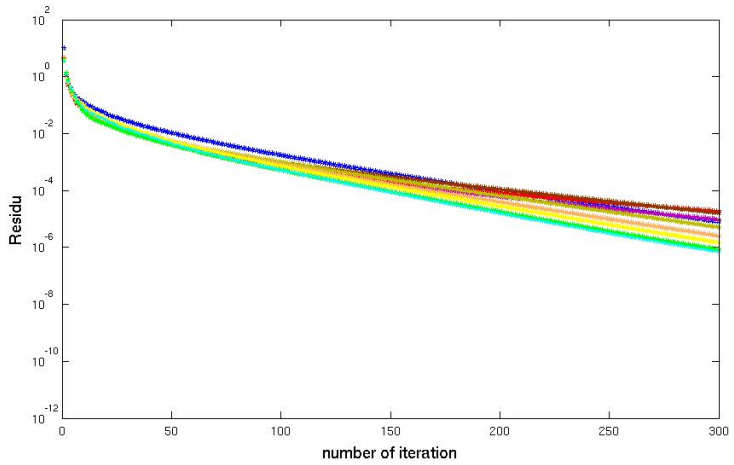
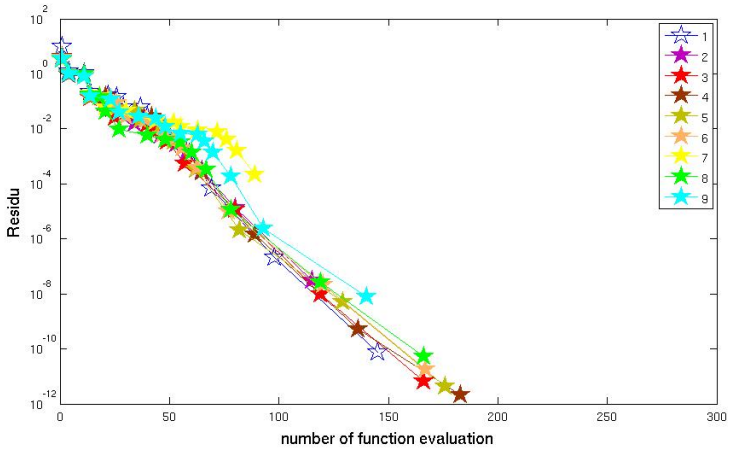
Inexact Newton

- Approximation of the Newton's direction $\|f'(x_k)d + f(x_k)\| \leq \eta \|f(x_k)\|$
- Choice of the forcing term η ?
  - Keep quadratic convergence (locally)
  - Avoid oversolving the linear system
- $\eta = \gamma \|f(x_k)\|^2 / \|f(x_{k-1})\|^2$ (Kelley, Eisenstat and Walker)

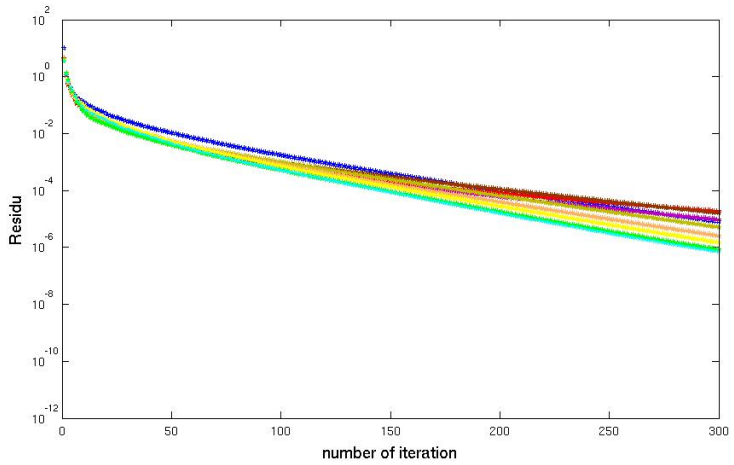
L. Amir's thesis, Amir, MK (Comp. Geosci. 09)

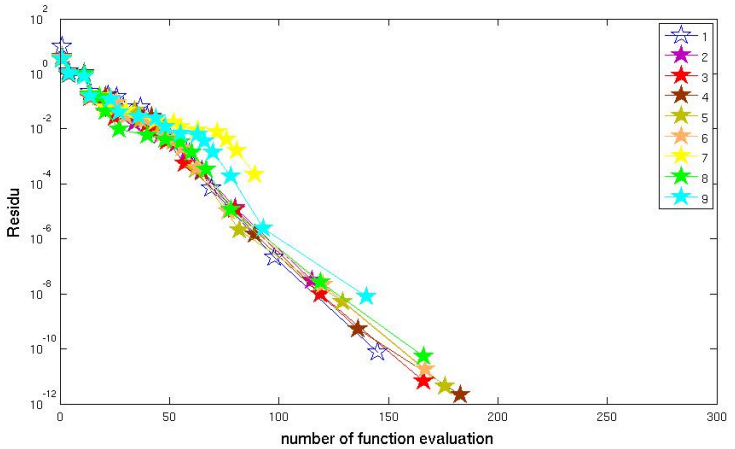
Newton vs fixed-point performance

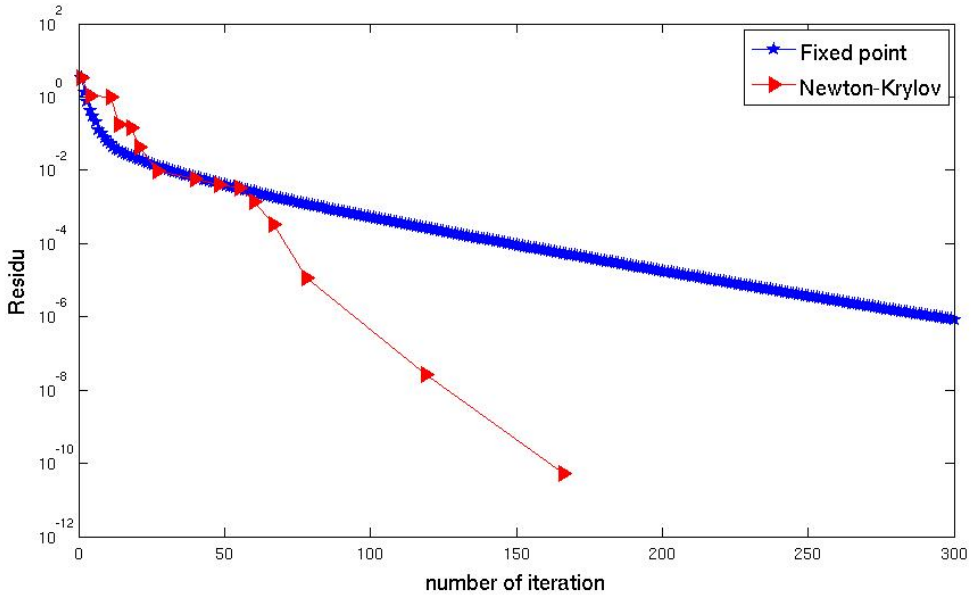




Newton vs fixed-point performance



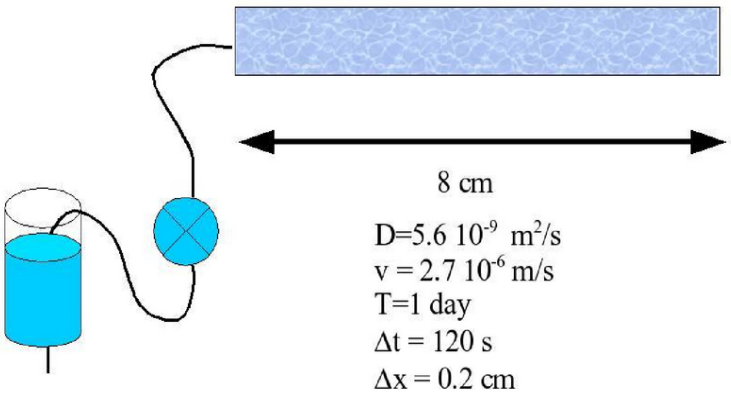




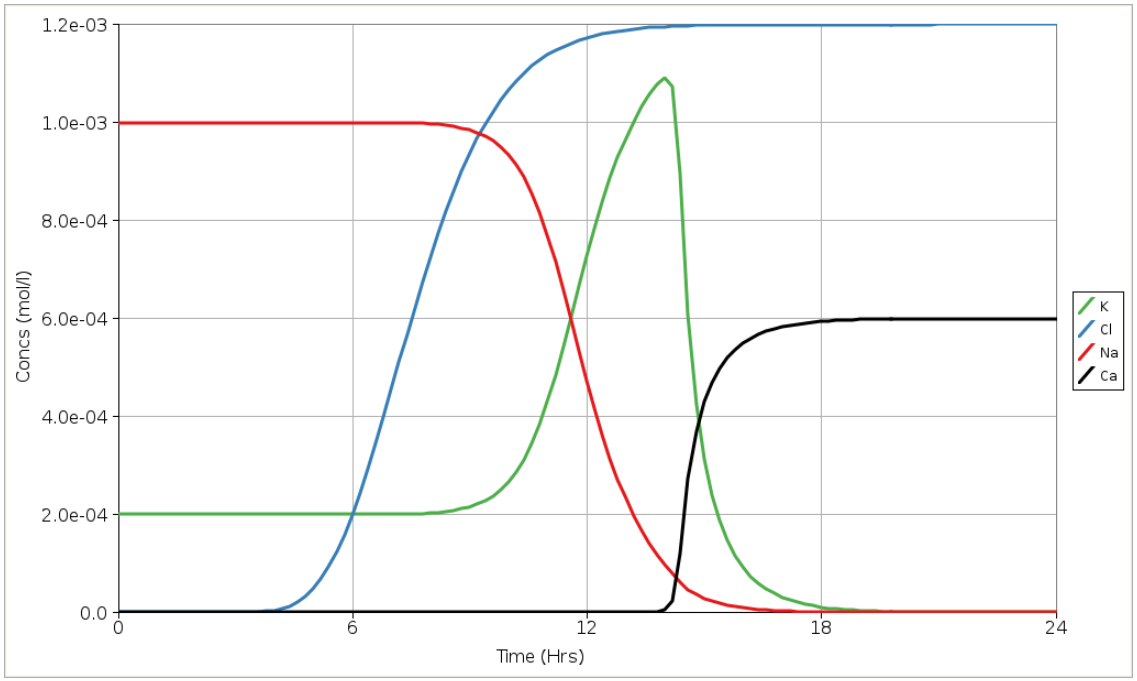
Outline

Example : ion exchange

Column experiment (Phreeqc ex. 11)

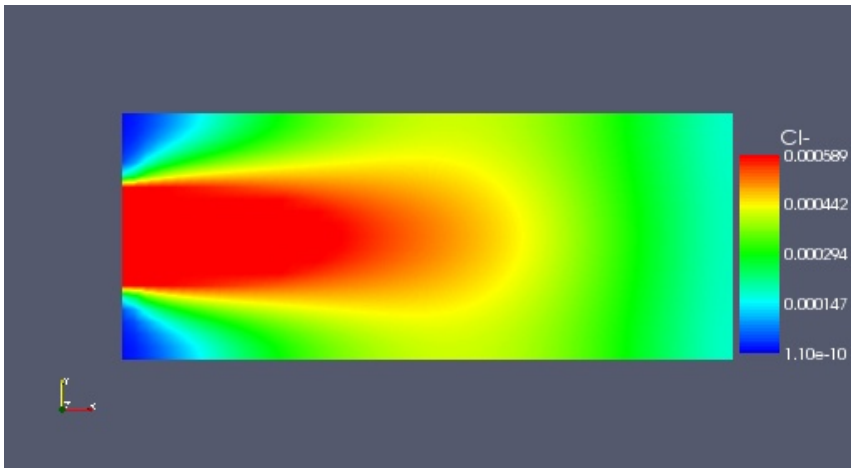


Column contains a solution with $1\,\mathrm{mmol}\,Na$, $0.2\,\mathrm{mmol}\,K$ and $1.2\,\mathrm{mmol}\,NO_3$. Inject solution with $1.2\,\mathrm{mmol}\,CaCl_2$, $CEC = 1.1\,10^{-3}$.

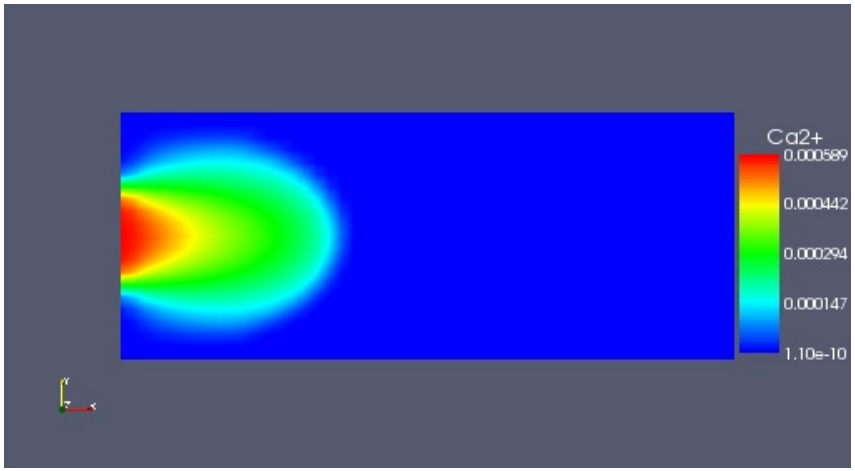


Ion exchange example (ctd)

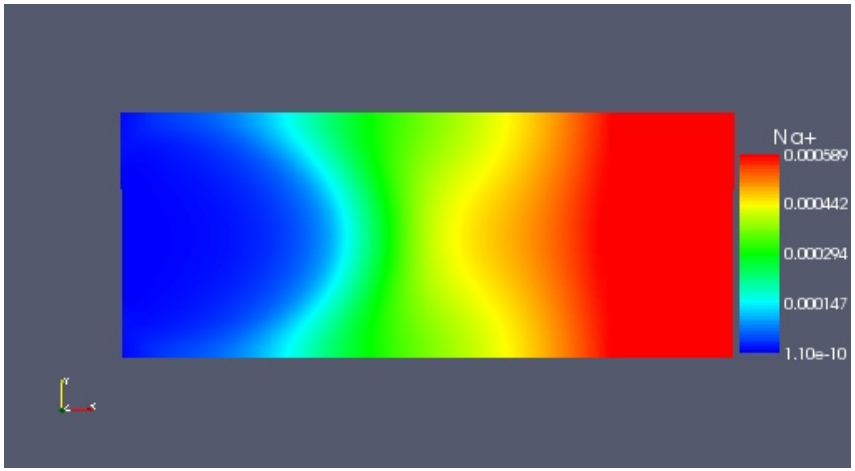
Snapshots at $t = 35$



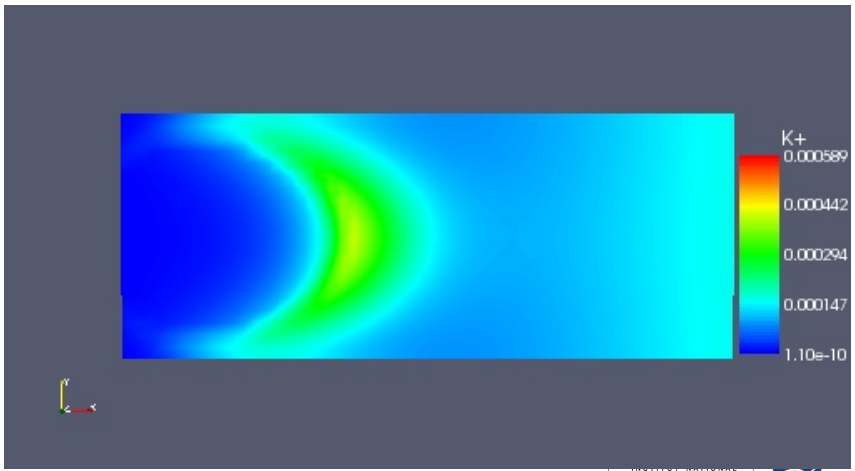
Cl



Ca



Na



K

Application to CO2 model

Simulation code LifeV (EPFL, MOX, INRIA), nonlinear solver Kinsol (LLNL).

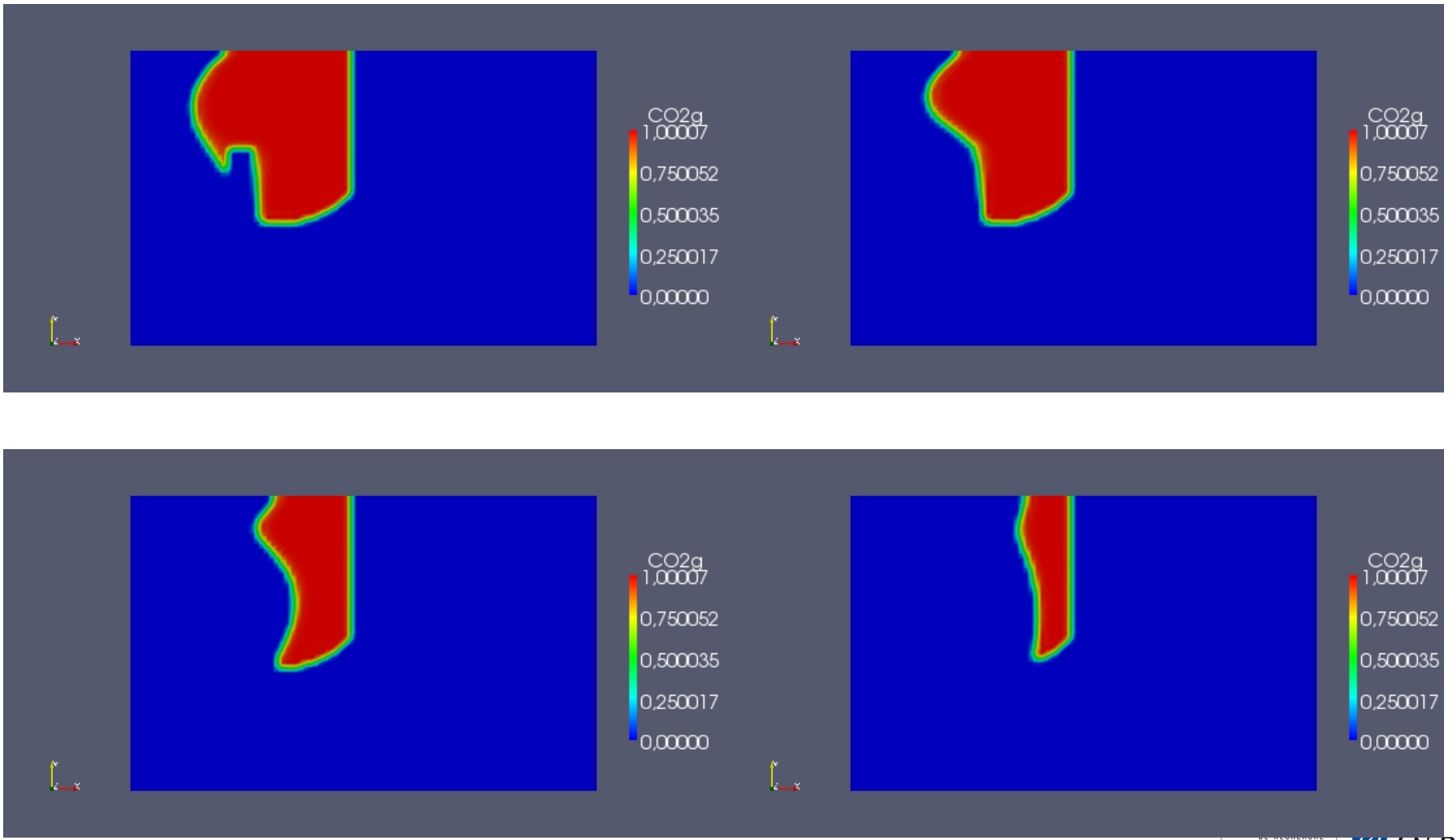
Coarse grid

- 5700 grid points, $\Delta x = 50\mathrm{m}$, $\Delta t = 100$ years
- Simulate 10 000 years
- CPU time 2h10 min

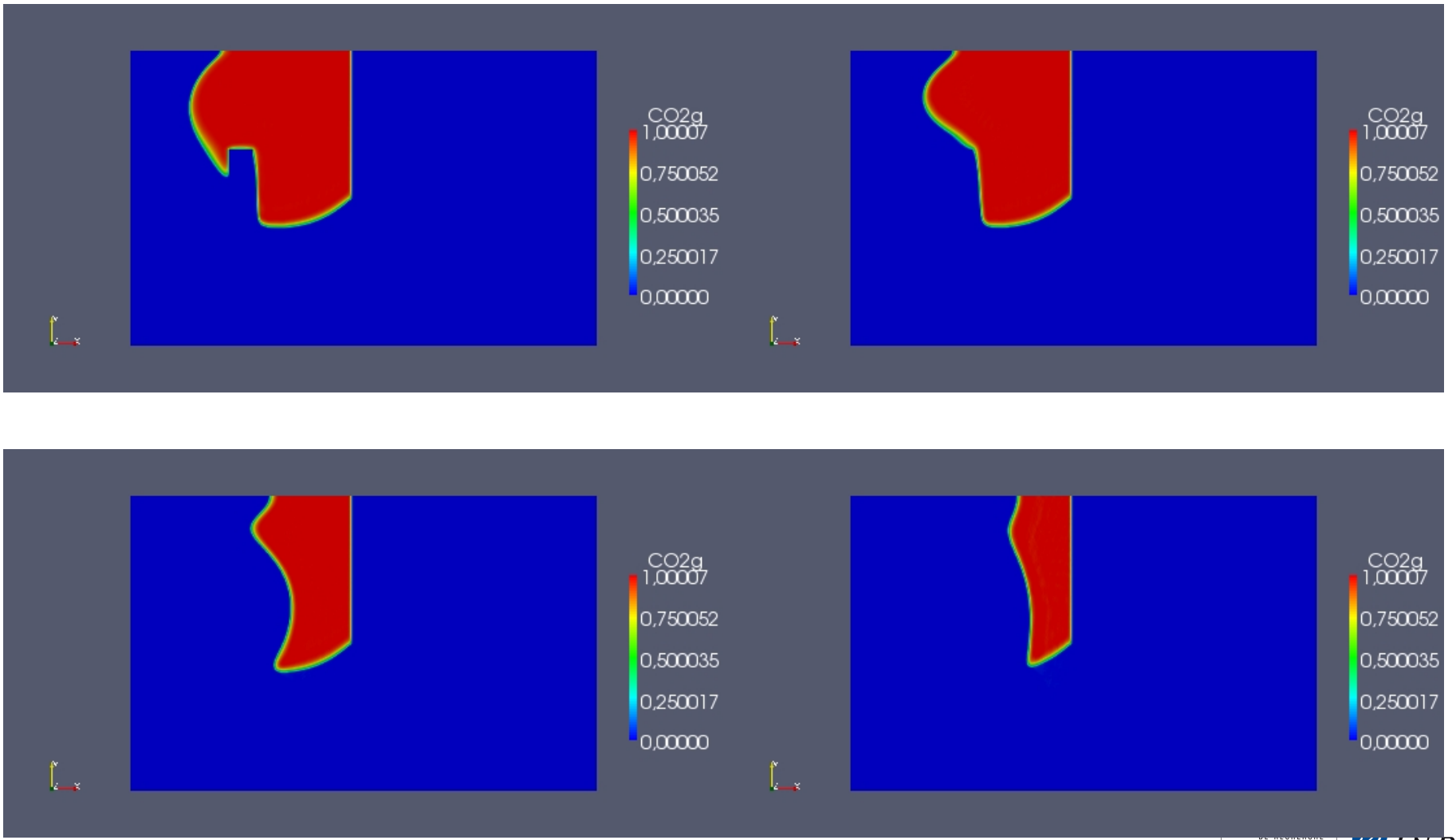
Fine grid

- 142 500 grid points, $\Delta x = 10\mathrm{m}$, $\Delta t = 100$ years
- Simulate 1 600 years
- CPU time 2.5 days

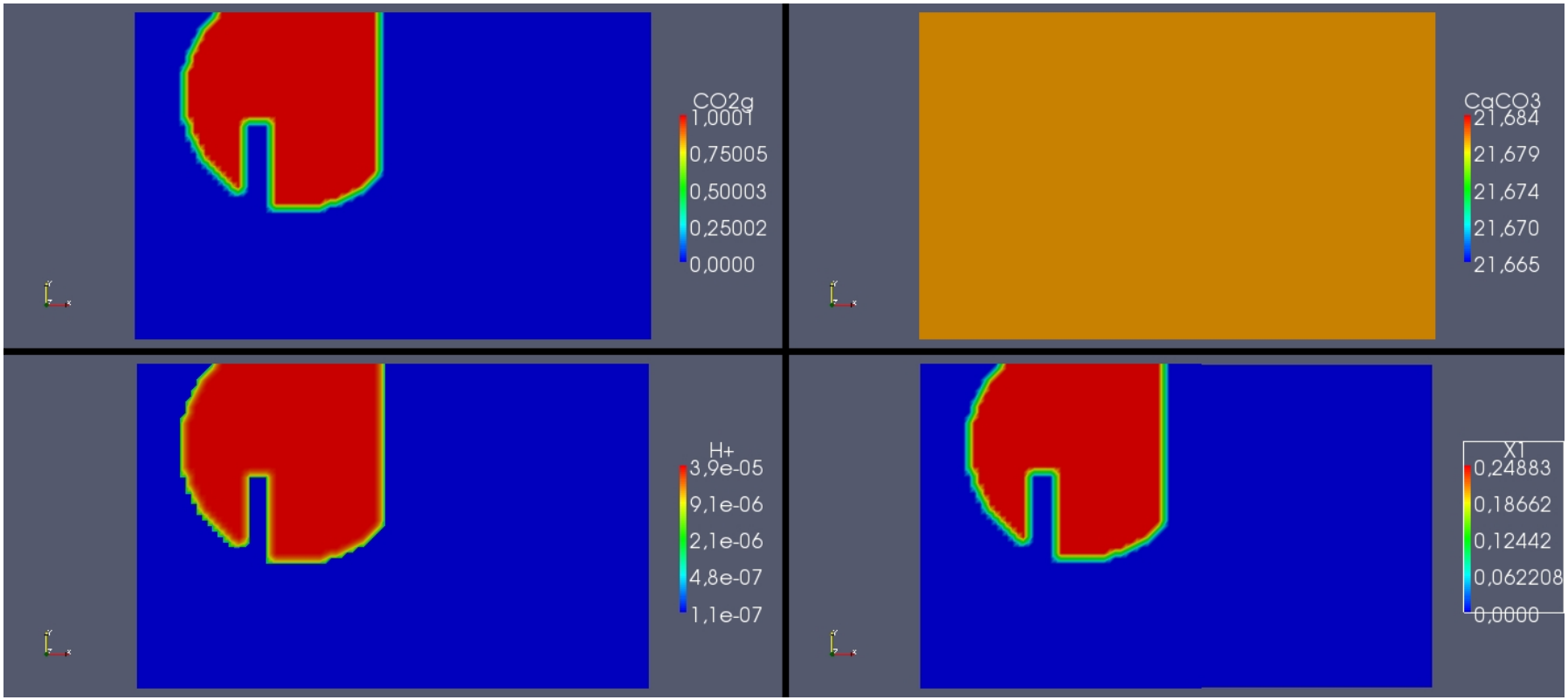
Evolution of CO2 concentration on coarse grid



Evolution of CO2 concentration on fine grid

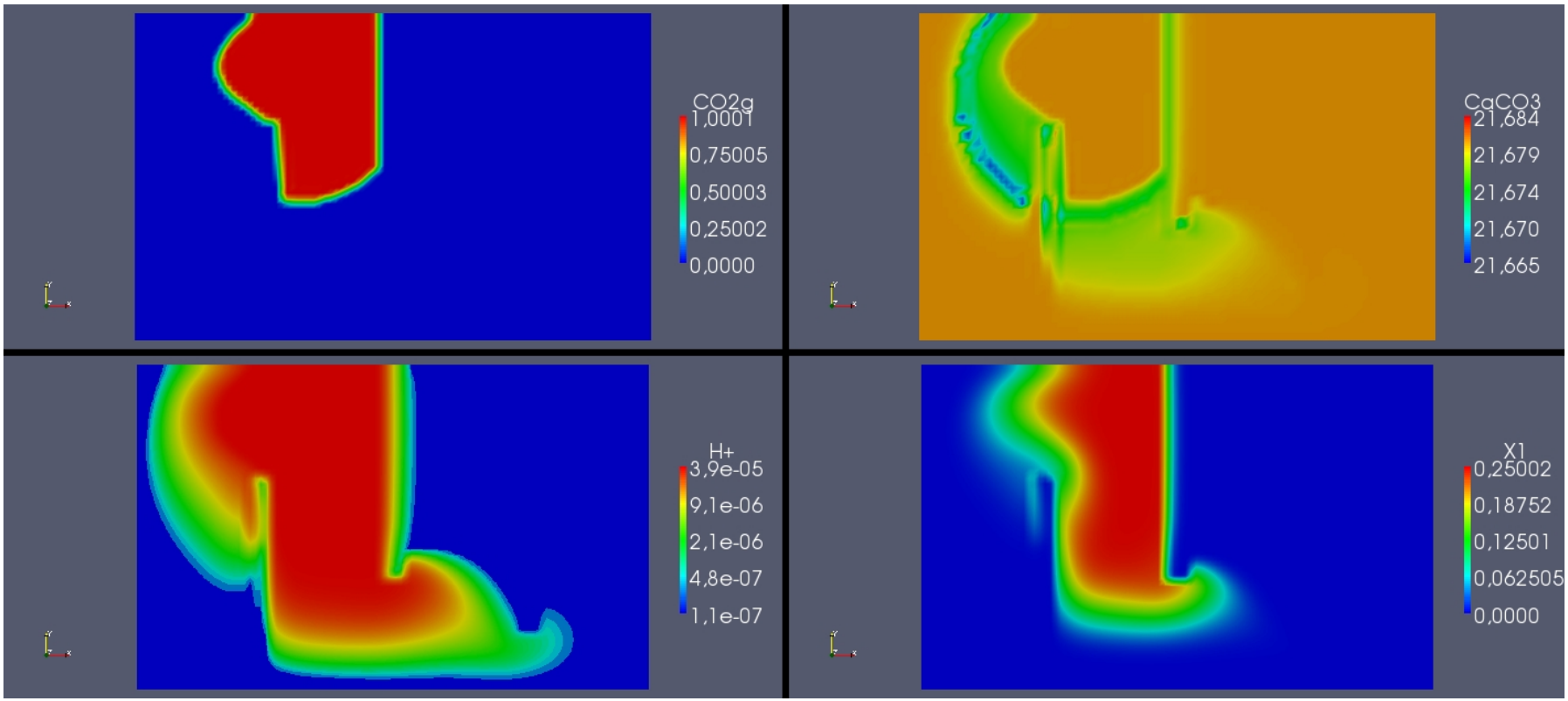


Evolution of concentrations



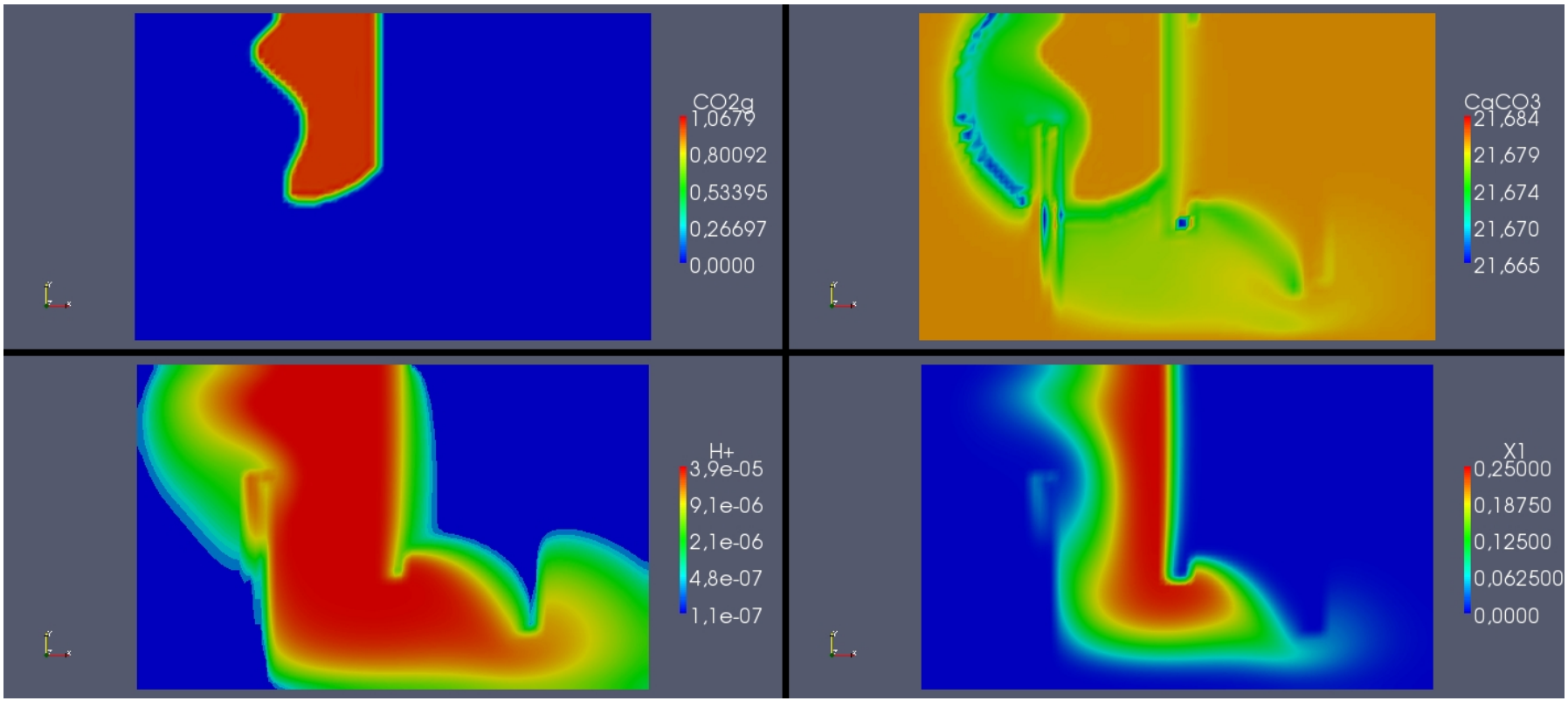
$t = 0$

Evolution of concentrations



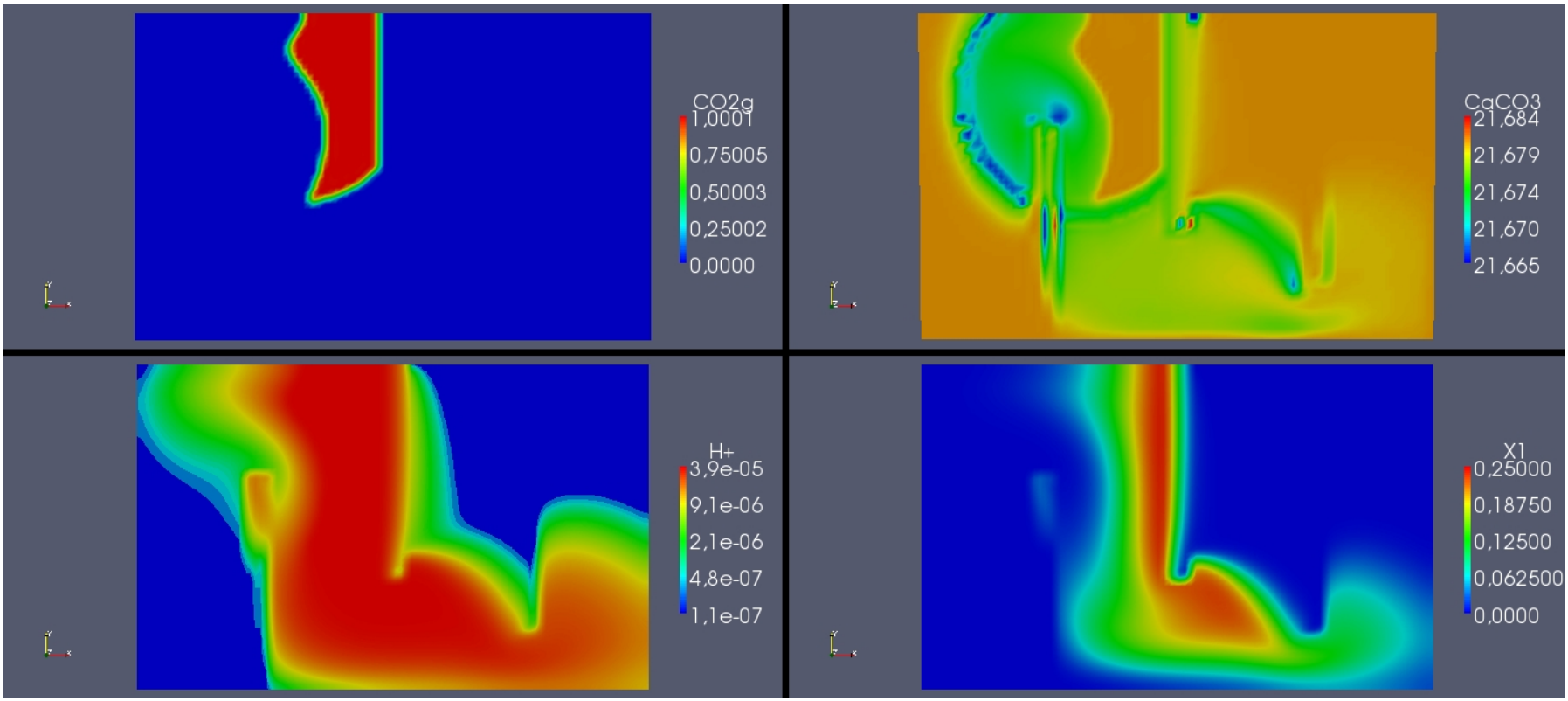
$t = 400$ years

Evolution of concentrations



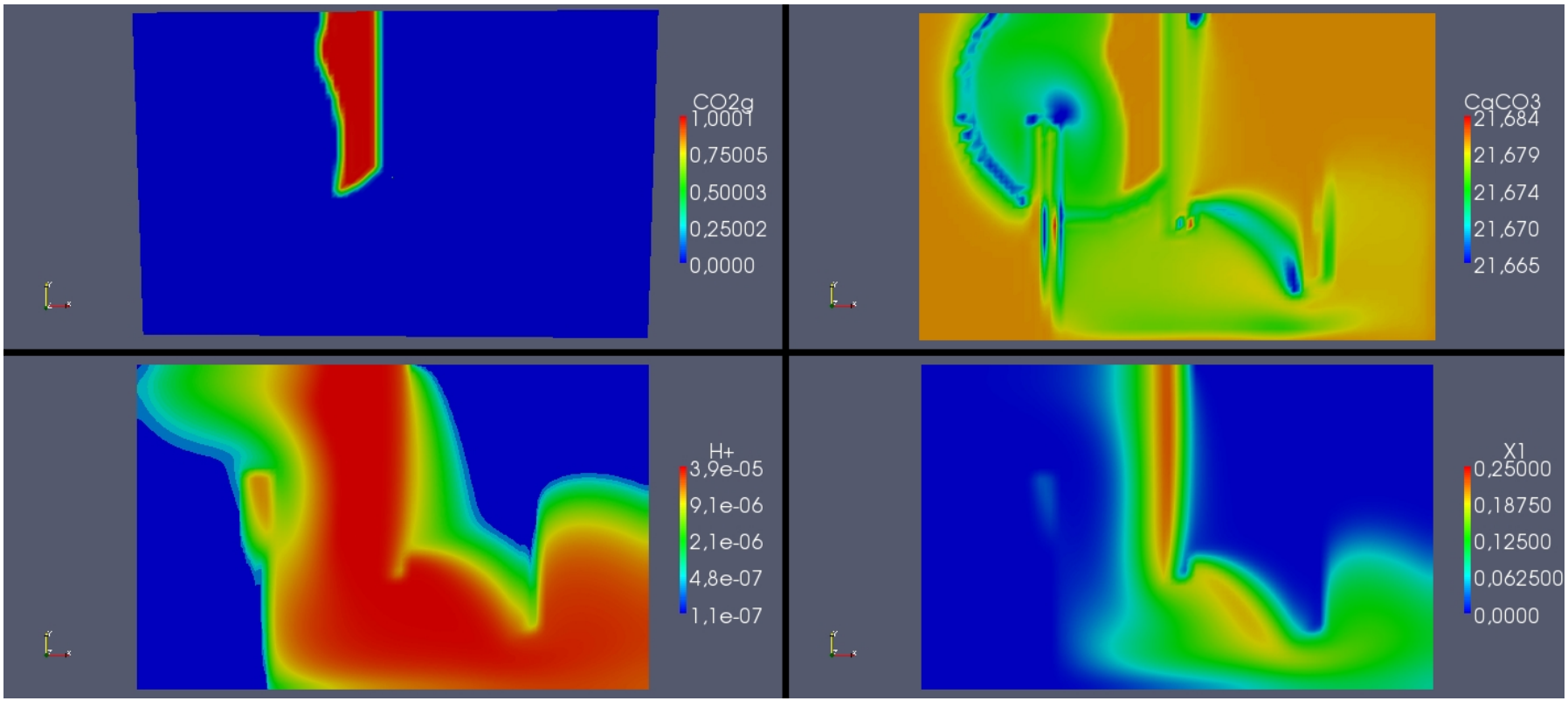
$t = 800$ years

Evolution of concentrations



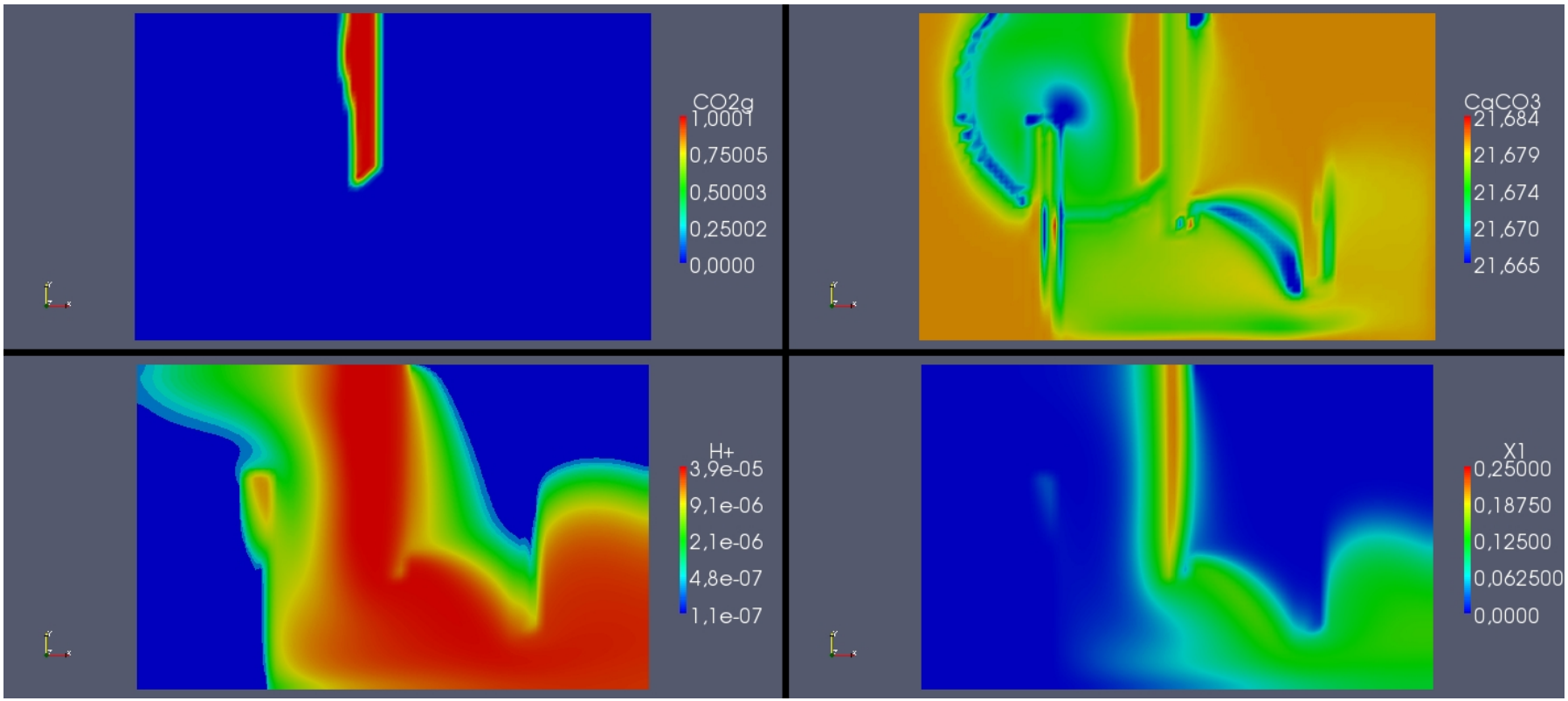
$t = 1200$ years

Evolution of concentrations



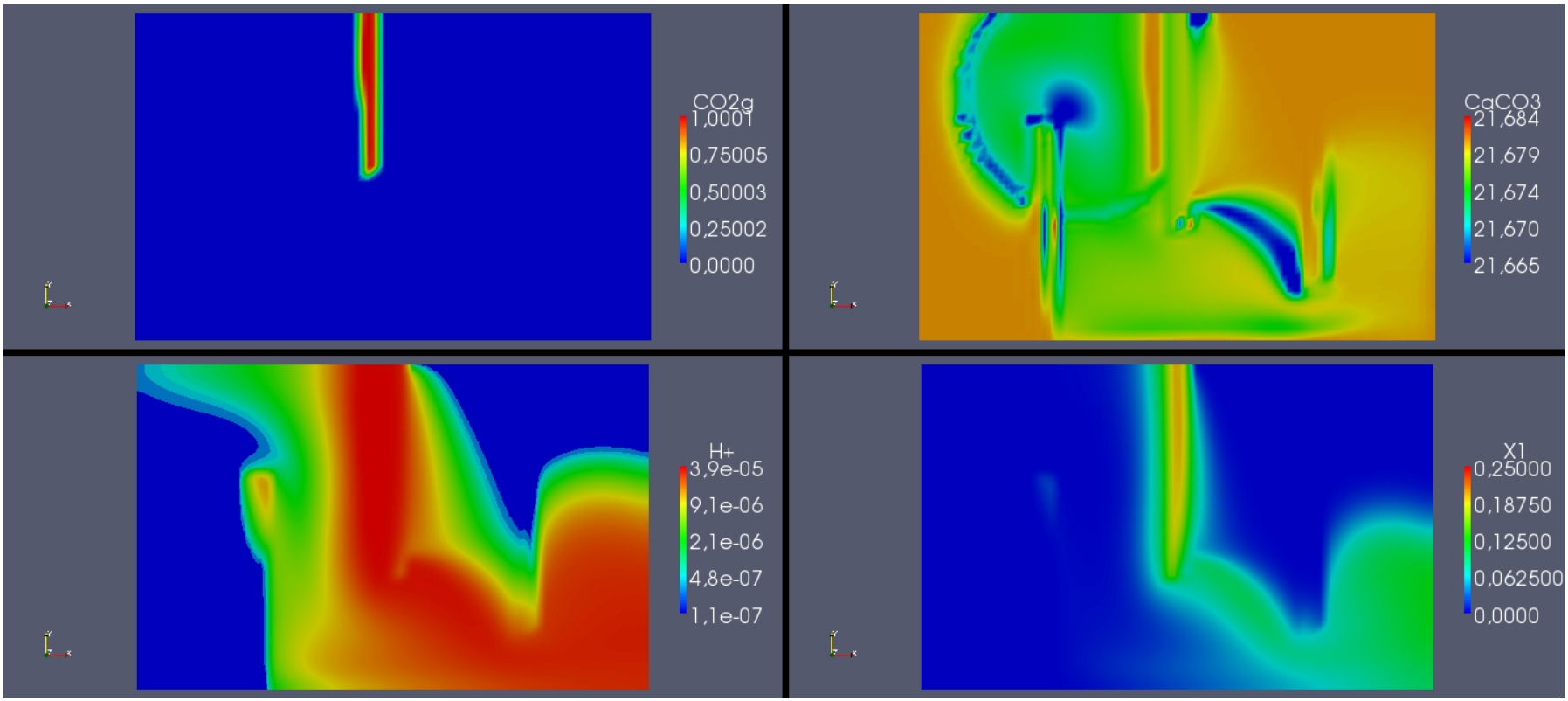
$t = 1600$ years

Evolution of concentrations



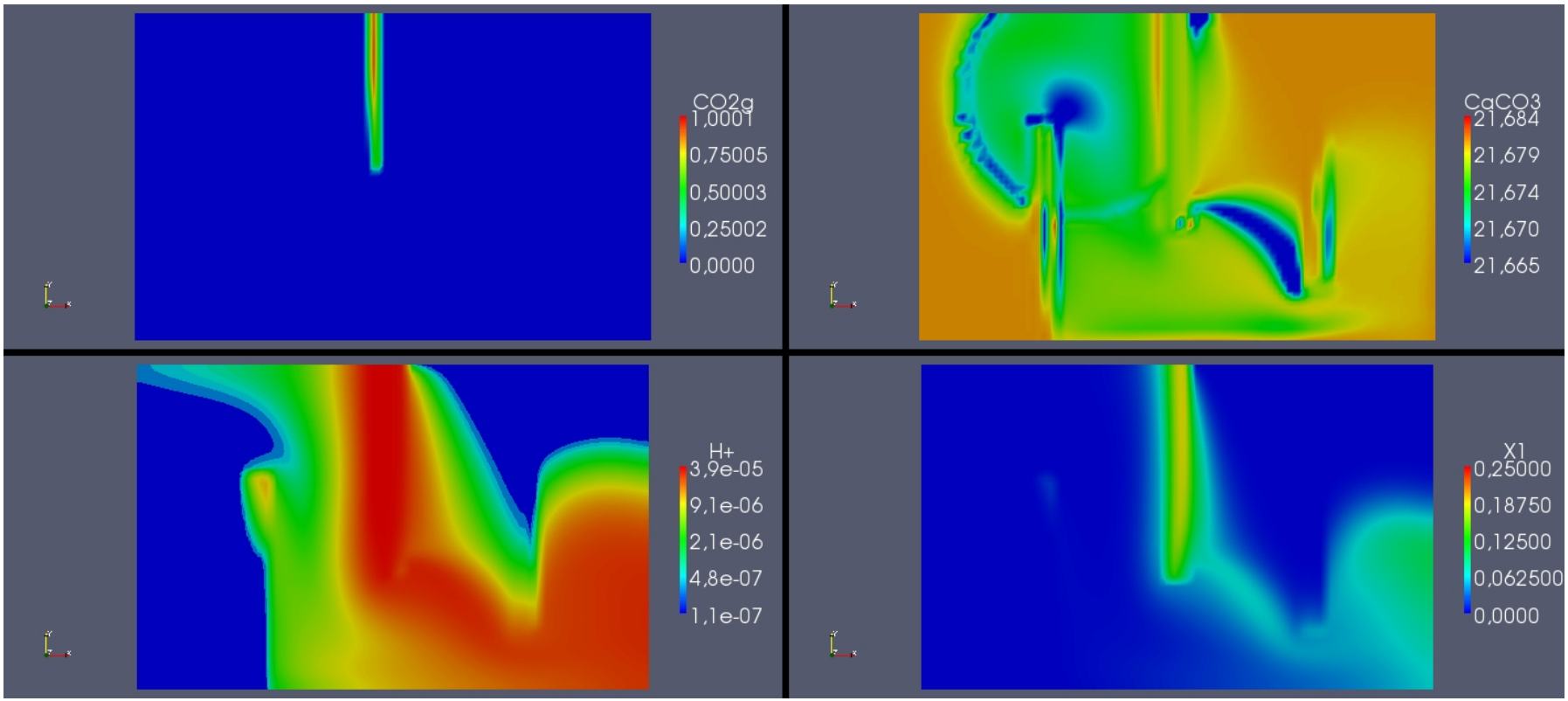
$t = 2000$ years

Evolution of concentrations



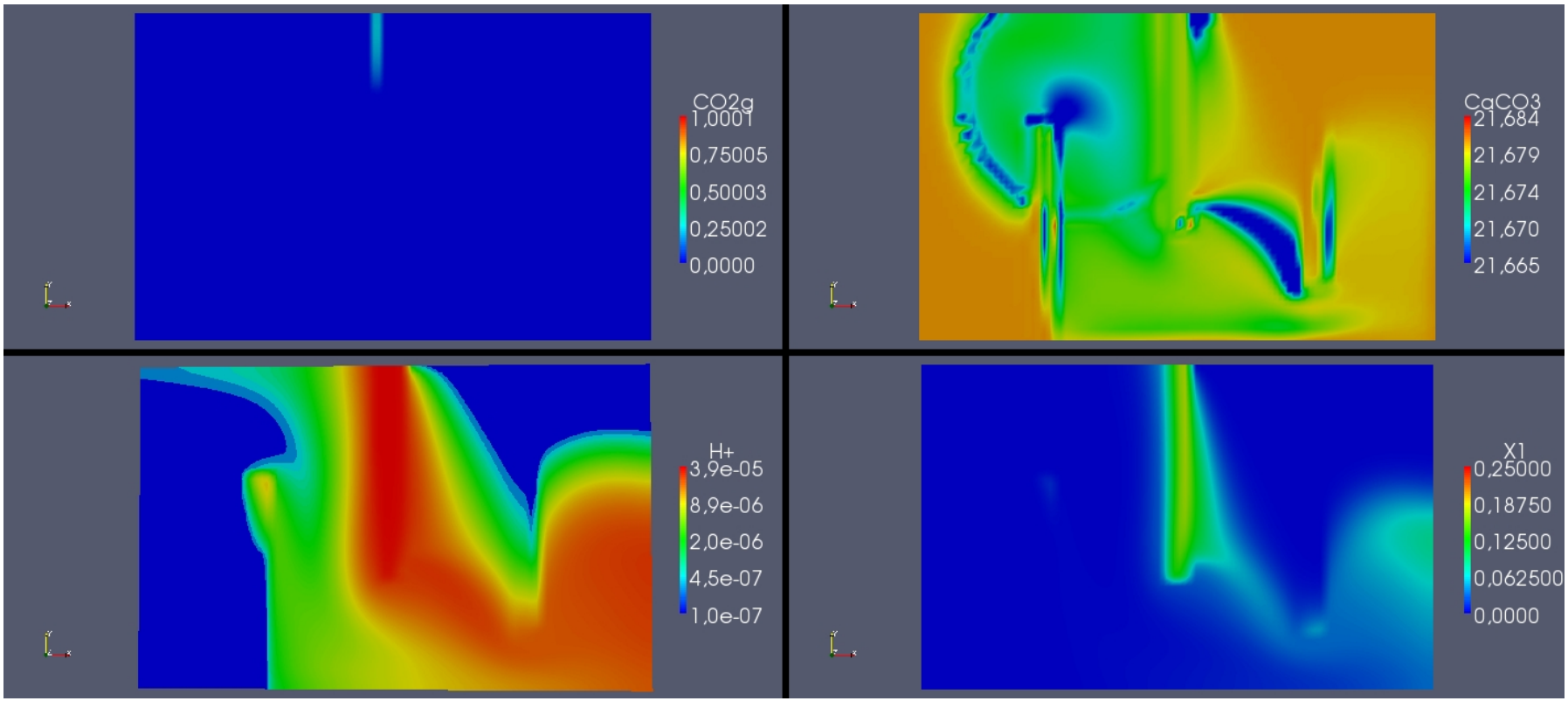
$t = 2400$ years

Evolution of concentrations



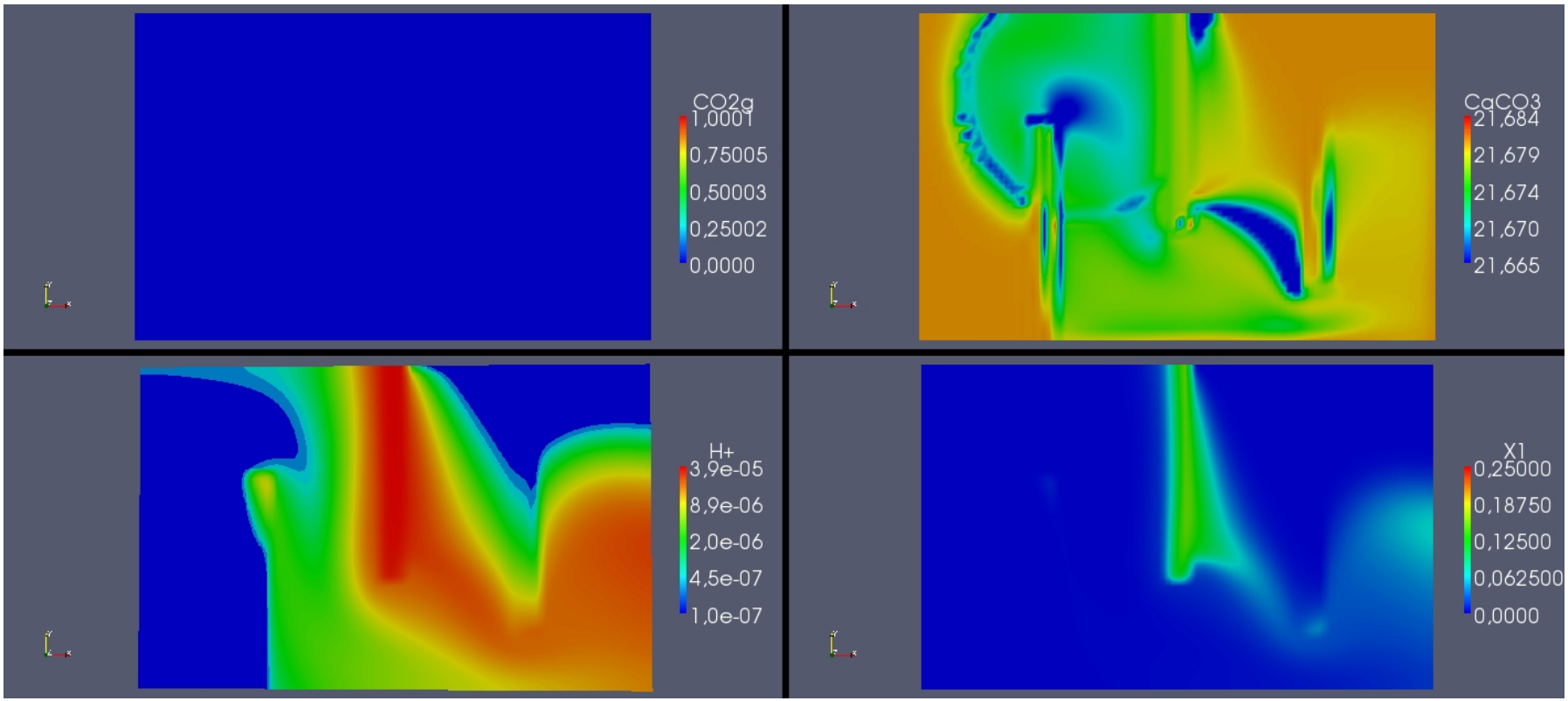
$t = 2800$ years

Evolution of concentrations



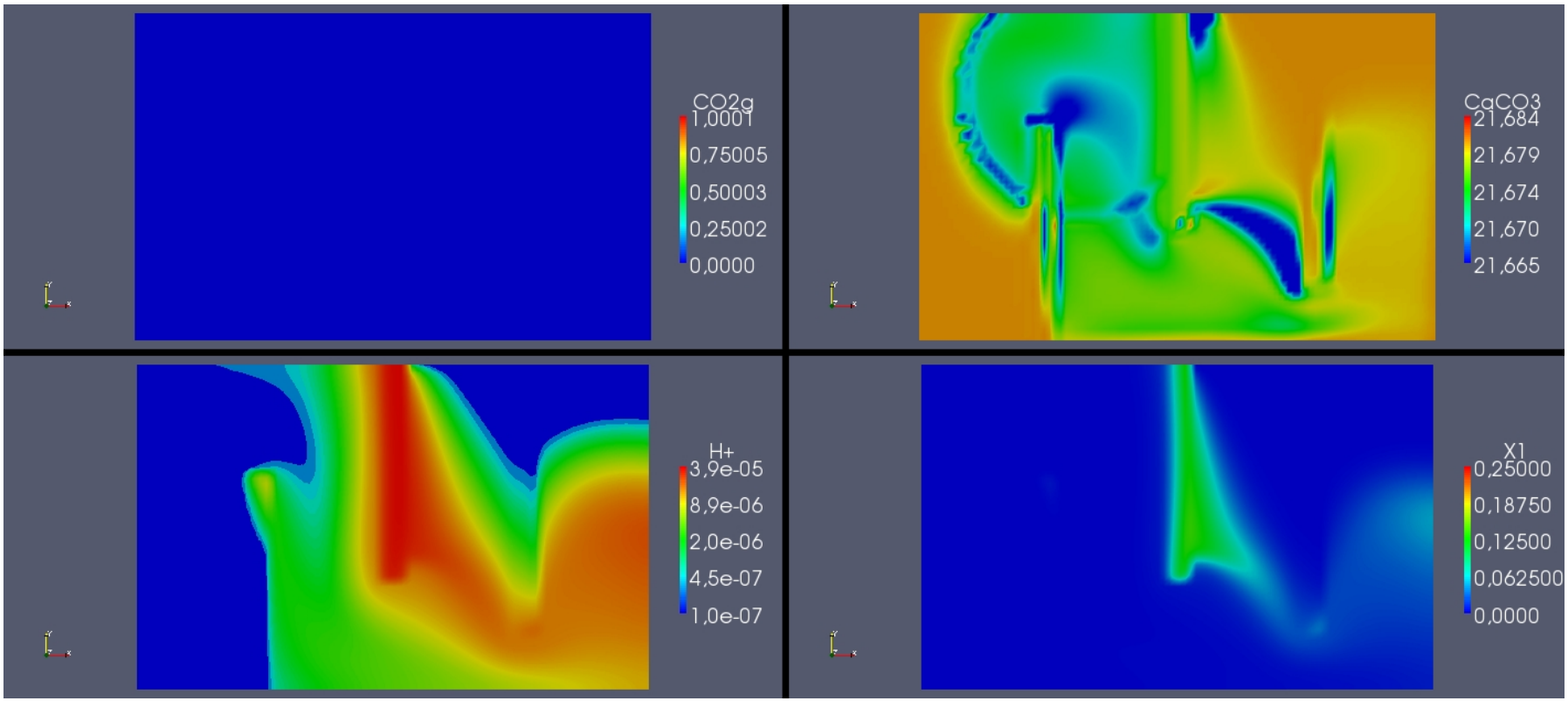
$t = 3200$ years

Evolution of concentrations



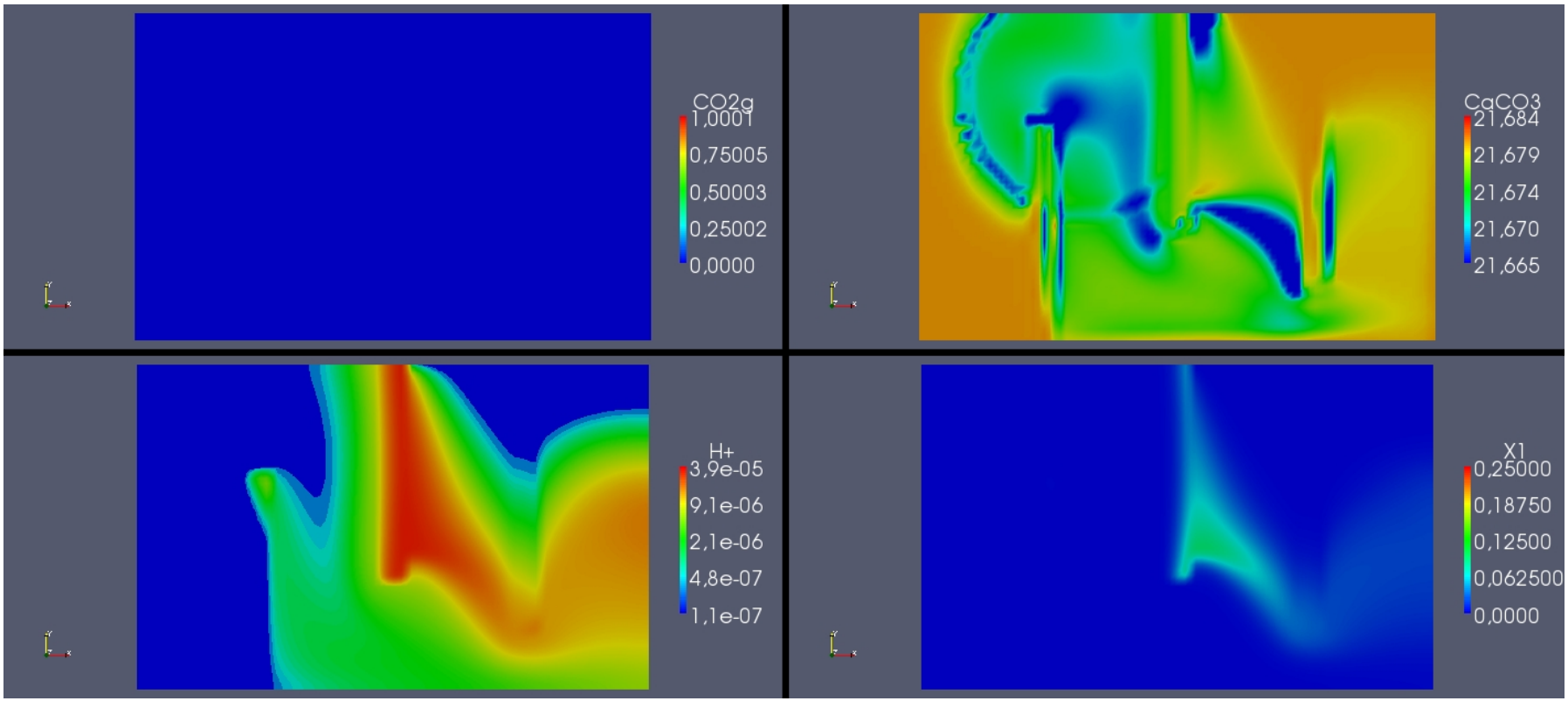
$t = 3600$ years

Evolution of concentrations



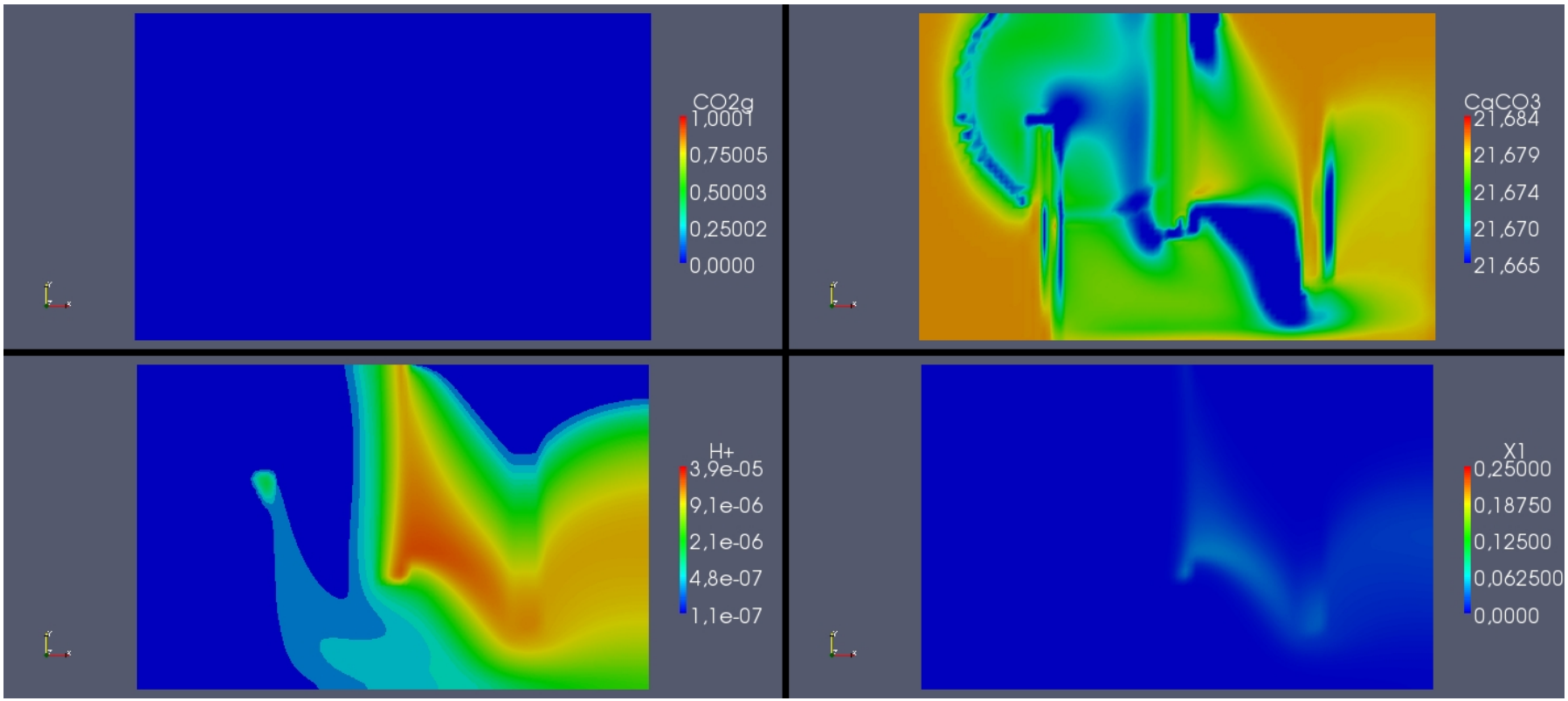
$t = 4000$ years

Evolution of concentrations



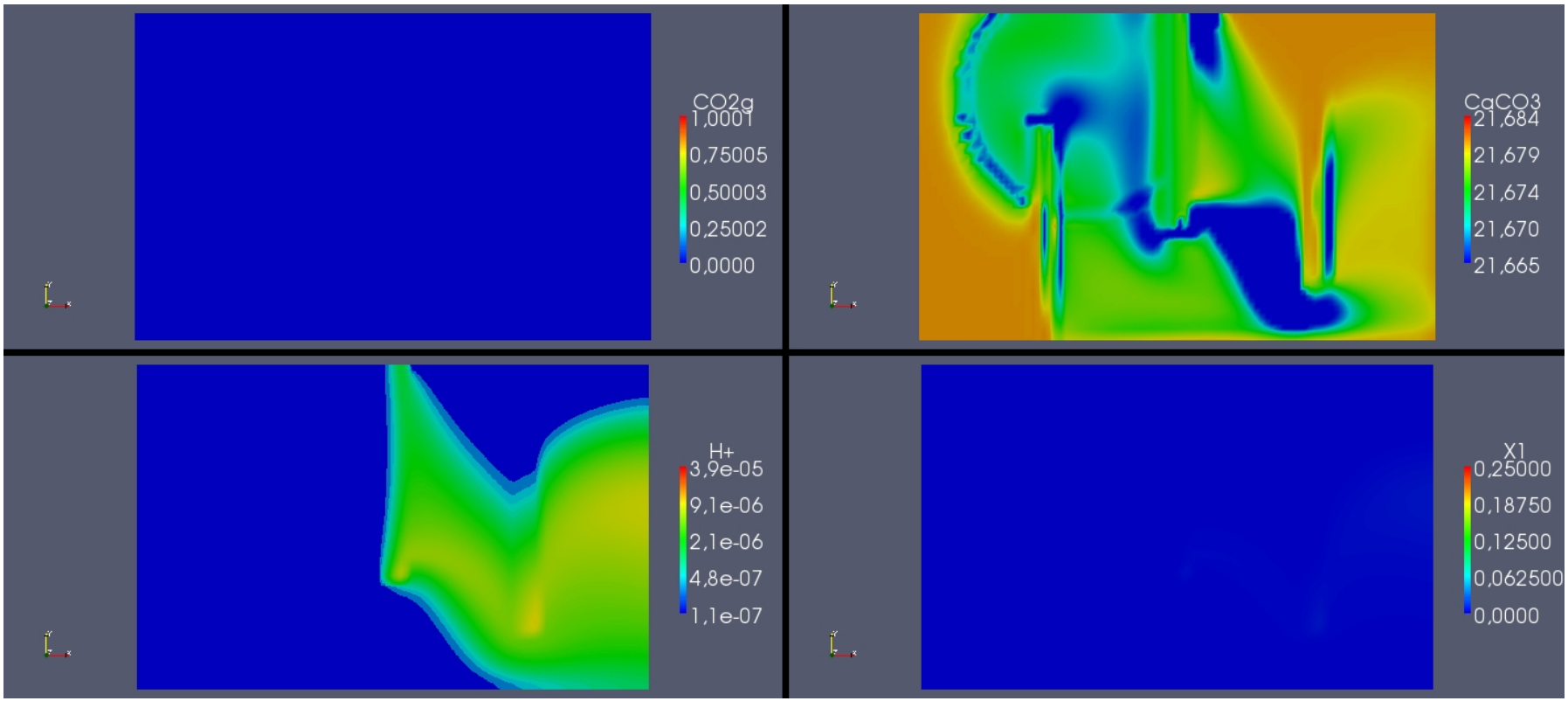
$t = 5000$ years

Evolution of concentrations



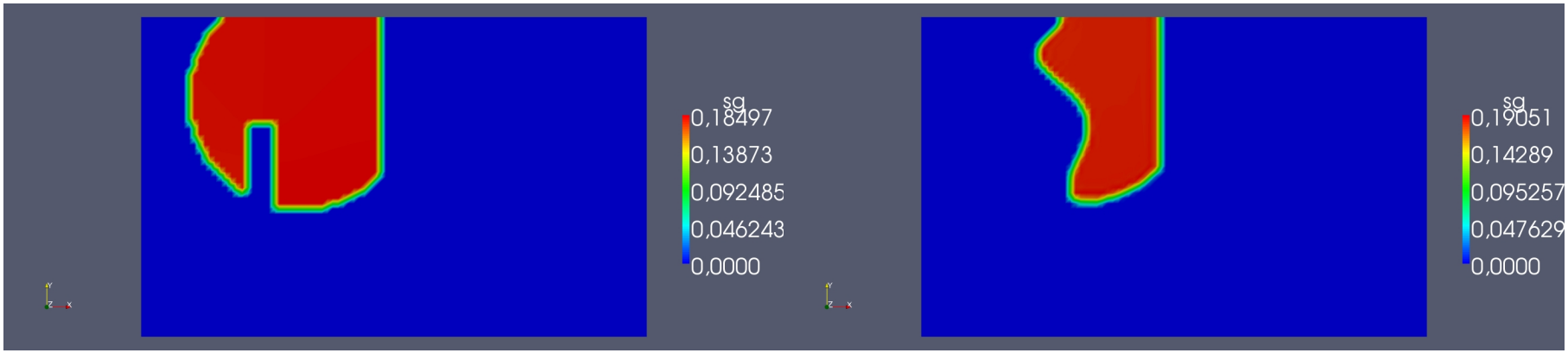
$t = 7000$ years

Evolution of concentrations

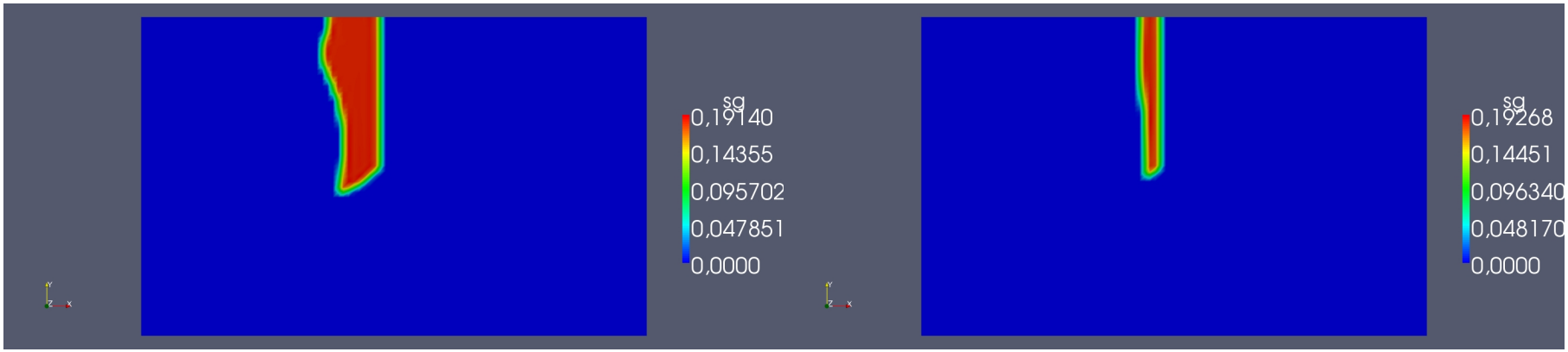


$t = 10000$ years

Evolution of gas saturation



$t = 0$ $t = 800$ years



$t = 1600$ $t = 2400$ years

Outline

Conclusions – perspectives

- Robust methods for flow and transport
- Newton–Krylov as a framework for code coupling
- Extension of chemical solver to handle minerals and gas
- Preconditioner for simplified system, mesh independent convergence

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- Robust methods for flow and transport
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- Implement analytical Jacobian – vector product
- Implement kinetic reactions
- Parallel computing (w. MOX, Milano)
- Extension to multiphase (compositional) flow