Ten years of reactive transport modeling and simulation

Jocelyne Erhel, Michel Kern

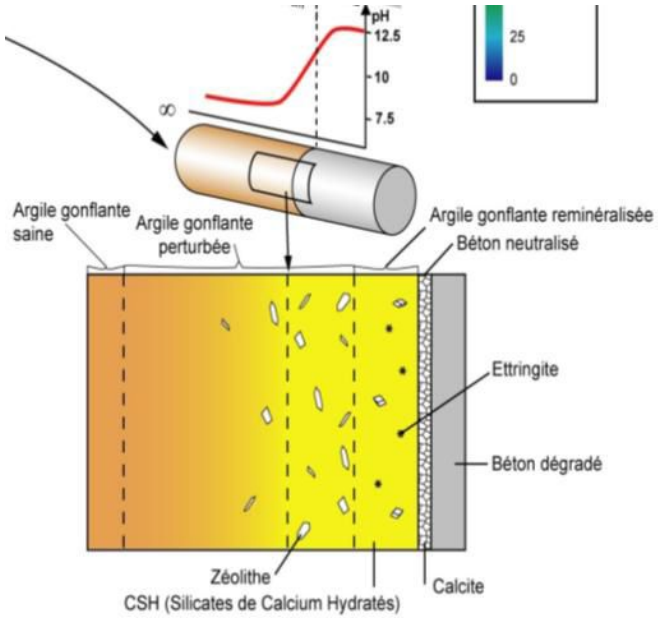
INRIA

Journées Scientifiques du GNR MoMaS
CIRM, Marseille
November 2–4, 2011

Motivations

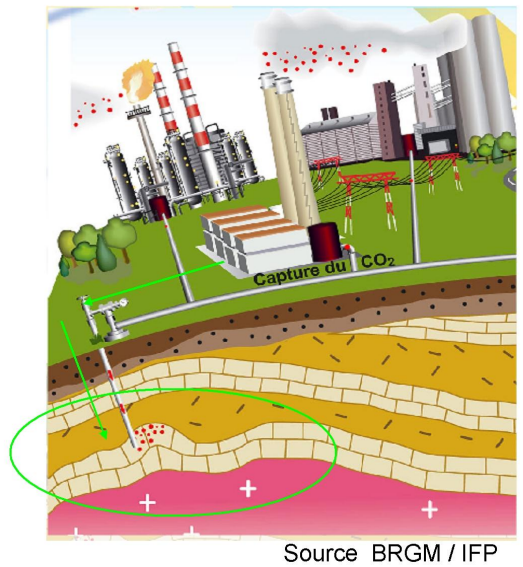
Importance of chemical phenomena in various applications

- Alteration chimique des composants du stockage
- Séquestration du CO2



G. Pépin (Andra)

Carbon Capture and Storage



Capture → Transport → Stockage

Aquifères Salins
Gisements Déplétés
Veines de Charbon

Source BRGM / IFP

A. Michel (IFP)

Plan

Outline

Transport with chemical reactions

Mobile species

$$\omega \partial_t c_i + \underbrace{\nabla \cdot (\mathbf{u} c_i - \mathbf{D} \nabla c_i)}_{L(c)} = \sum_{j=1}^{N_R} \nu_{ij} R_j(c_1, \ldots, c_{N_S}), \quad i = 1, \ldots, N_S$$

- c_i : concentration of ith species $[\mathrm{mol/l}]$
- $\mathbf{D}$ Dispersion – diffusion tensor $[\mathrm{m}^2/\mathrm{s}]$
- R_j reaction term for jth reaction
- ω : porosity [–]
- $\mathbf{u}$ Darcy velocity $[\mathrm{m/s}]$
- ν_{ij} stoichiometric coeff.

Transport with chemical reactions

Mobile species

$$\omega \partial_t c_i + \underbrace{\nabla \cdot (\mathbf{u} c_i - \mathbf{D} \nabla c_i)}_{L(c)} = \sum_{j=1}^{N_R} \nu_{ij} R_j(c_1, \ldots, c_{N_S}), \quad i = 1, \ldots, N_S$$

- c_i : concentration of ith species $[\mathrm{mol/l}]$
- $\mathbf{D}$ Dispersion – diffusion tensor $[\mathrm{m}^2/\mathrm{s}]$
- R_j reaction term for jth reaction
- ω : porosity [–]
- $\mathbf{u}$ Darcy velocity $[\mathrm{m/s}]$
- ν_{ij} stoichiometric coeff.

Condense transport solver, one time step

$$c(t + \Delta t) = \Psi_T(R, c(t))$$

Mass balance for immobile species

$$\rho_s \partial_t \bar{c}_i = \sum_{j=1}^{N_R} \nu_{ij} R_j(c_1, \ldots, c_{N_S}, \bar{c}_1, \ldots, \bar{c}_{\bar{N}_S}), \quad i = 1, \ldots, \bar{N}_S$$

Modeling general equilibrium models

$$\sum_{j=1}^{N_s+\bar{N}_S} \nu_{ij} Y_j \leftrightarrows 0, \quad i = 1, \ldots, N_r \qquad N_s + \bar{N}_S \text{ species }, N_r \text{ reactions.}$$

Mass action law $\nu \log \begin{pmatrix} c \\ \bar{c} \end{pmatrix} + \log K = 0$

Mass conservation $\nu^T \begin{pmatrix} c \\ \bar{c} \end{pmatrix} = \begin{pmatrix} T \\ W \end{pmatrix}$

System of non-linear equations

T known from transport, W imposed

Modeling general equilibrium models

$$\sum_{j=1}^{N_s+\bar{N}_S} \nu_{ij} Y_j \leftrightarrows 0, \quad i = 1, \ldots, N_r \qquad N_s + \bar{N}_S \text{ species }, N_r \text{ reactions.}$$

Mass action law $\nu \log \begin{pmatrix} c \\ \bar{c} \end{pmatrix} + \log K = 0$

Mass conservation $\nu^T \begin{pmatrix} c \\ \bar{c} \end{pmatrix} = \begin{pmatrix} T \\ W \end{pmatrix}$

System of non-linear equations
T known from transport, W imposed

Mineral reactions

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

Solubility product $\Pi = \log K_p + S_p \log c$, $\begin{cases} p = 0 & \text{if } \Pi < 0 \\ \Pi = 0 & \text{otherwise} \end{cases}$

Numerical solution of nonlinear problem

Take concentration logarithms as main unknowns

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

Numerical solution of nonlinear problem

Take concentration logarithms as main unknowns

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

Methods for minerals

- Standard procedure : combinatorial search
- Reformulate as complementarity problem
- Interior point algorithm (Saaf et al. ('96), MK (05))
- Also semi–smooth Newton (Kräutle)

Numerical solution of nonlinear problem

Take concentration logarithms as main unknowns

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

Methods for minerals

- Standard procedure : combinatorial search
- Reformulate as complementarity problem
- Interior point algorithm (Saaf et al. ('96), MK (05))
- Also semi–smooth Newton (Kräutle)

Role of chemical model

Given totals T (and W, known), split into mobile (C) and immobile (F) total concentrations

Numerical solution of nonlinear problem

Take concentration logarithms as main unknowns

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

Methods for minerals

- Standard procedure : combinatorial search
- Reformulate as complementarity problem
- Interior point algorithm (Saaf et al. ('96), MK (05))
- Also semi–smooth Newton (Kräutle)

Role of chemical model

Given totals T (and W, known), split into mobile (C) and immobile (F) total concentrations

Chemistry solver

$$H\begin{pmatrix}\log c\\ \log \bar{c}\end{pmatrix} = \begin{pmatrix}T\\ W\end{pmatrix}$$

$$F = \Psi_C(T, W)$$

Outline

Formulations and solution strategies

Eliminate unkown equilibrium reaction rates by introducing mobile and immobile totals.

$$\phi \partial_t C + \partial_t F + LC = 0 \text{ with } F = \Psi_c(C + F)$$

Formulations and solution strategies

Eliminate unkown equilibrium reaction rates by introducing mobile and immobile totals.

$$\phi \partial_t C + \partial_t F + LC = 0 \text{ with } F = \Psi_c(C + F)$$

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

- $+$ easy to program, code reuse
- $-$ not robust, small time steps

Formulations and solution strategies

Eliminate unkown equilibrium reaction rates by introducing mobile and immobile totals.

$$\phi \partial_t C + \partial_t F + LC = 0 \text{ with } F = \Psi_c(C + F)$$

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

- $+$ easy to program, code reuse
- $-$ not robust, small time steps

Direct subsitution Lichtner et al., Saaltink et al.

- $+$ accurate, robust,
- $-$ difficult to code, large non-linear system

Formulations and solution strategies

Eliminate unkown equilibrium reaction rates by introducing mobile and immobile totals.

$$\phi \partial_t C + \partial_t F + LC = 0 \text{ with } F = \Psi_c(C + F)$$

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

- $+$ easy to program, code reuse
- $-$ not robust, small time steps

Direct subsitution Lichtner et al., Saaltink et al.

- $+$ accurate, robust,
- $-$ difficult to code, large non-linear system

Others

- Nonlinear conjugate gradient (Bouillard, Herbin, Montarnal)
- Elimination technique (Knabner, Kraütle, Hoffmann)

De Dieuleveult, JE, MK (JCP '09)

A global method based on DAE formulation

CC formulation, explicit chemistry

$$\begin{cases} \phi \partial_t C + \partial_t F + LC = 0 \\ H \begin{pmatrix} \log c \\ \log \bar{c} \end{pmatrix} - \begin{pmatrix} C + F \\ W \end{pmatrix} = 0 \\ F - F \begin{pmatrix} \log c \\ \log \bar{c} \end{pmatrix} = 0. \end{cases}$$

- $+$ Explicit Jacobian
- $+$ Chemistry function, no chemical solve
- $-$ Intrusive approach (chemistry not a black box)
- $-$ Precipitation not easy to include (semi-smooth Newton OK)

Coupled system is index 1 DAE

$$K \frac{dy}{dt} + f(y) = 0$$

Use standard DAE software

C. de Dieuleveult (Andra thesis),

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

Discrete non-linear system

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

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

Discrete non-linear system

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

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

Discrete non-linear system

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

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

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

Discrete non-linear system

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

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

Residual computation

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

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

Newton–Krylov method

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)

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

Newton–Krylov method

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)

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

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 $\eta = \gamma \|f(x_k)\|^2 / \|f(x_{k-1})\|^2$ (Kelley, Eisenstat and Walker)
  - Keep superlinear convergence (locally)
  - Avoid oversolving the linear system

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

Preconditioning

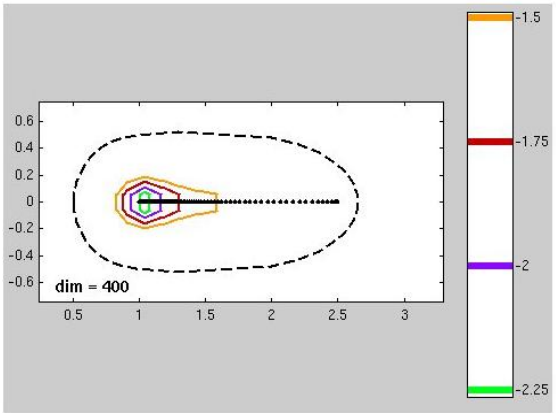
- Essential for good linear performance
- Difficult for matrix free formulation
- Simplified, 1 species model, with explicit sorption (with A. Taakili)

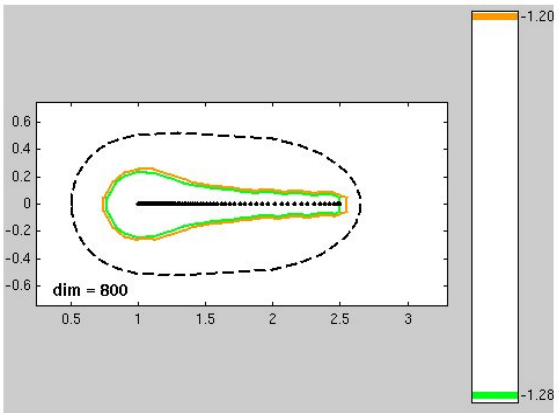
Algebraic elimination of mobile conc. equivalent to Schur complement of block Gauss–Seidel precond.

Preconditioning

- Essential for good linear performance
- Difficult for matrix free formulation
- Simplified, 1 species model, with explicit sorption (with A. Taakili)

Algebraic elimination of mobile conc. equivalent to Schur complement of block Gauss–Seidel precond.
Can show eigenvalues of preconditioned op. bounded away from 0, independent of h, but convergence of GMRES not determined by eigenvalues
Field of values analysis ?





Outline

The Reactive transport benchmark

The setup

- Designed by J. Carrayrrou, M. Kern, P. Knabner
- Concentrate on numerical difficulties : simple geometry, « abstract » chemistry
- 3 levels of chemistry (sorption, equilibrium minerals, kinetics)

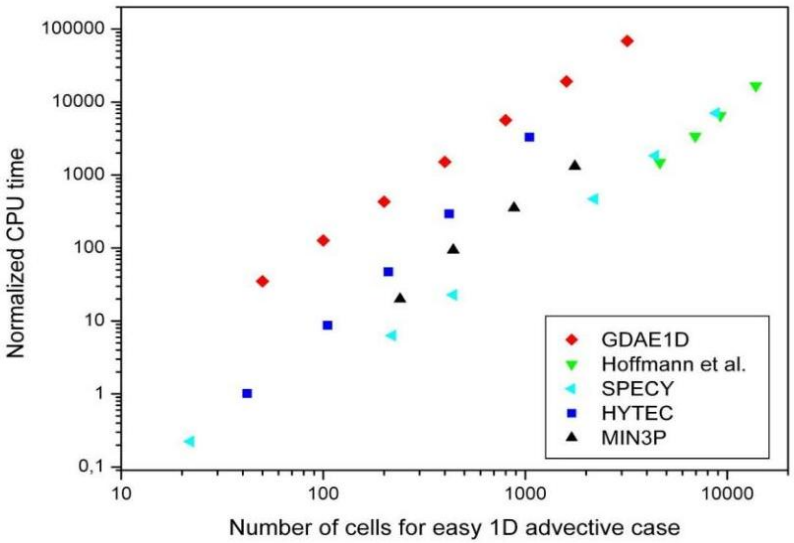
The benchmark

- Results from 6 groups, awards to 4 groups
- International workshop (Ph. Ackerer, Strasbourg, Jan. 2008)
- Special issue in « Computational Geosciences » (ed. Ph. Ackerer), 6 papers + intro + synthesis

1D benchmark results

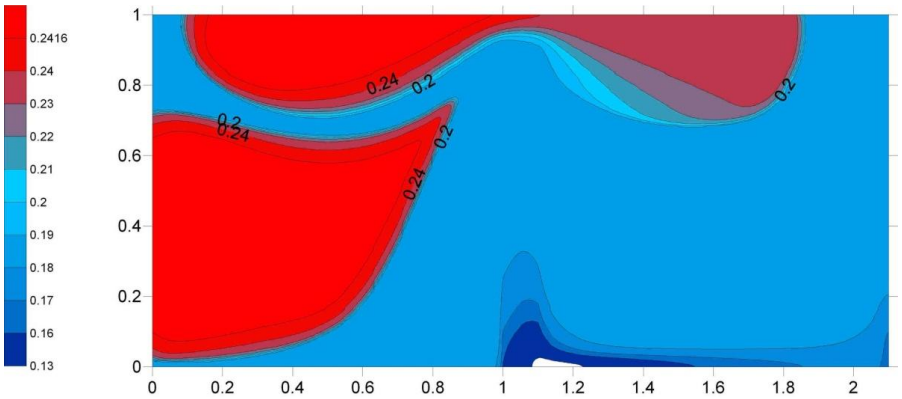
	Loc. of peak	*S* conc.
GDAE1D	0.0175	0.966
NK	0.0167	0.742
Erlangen	0.0167	0.852
Specy	0.0158	0.968
HYTEC	0.0170	0.286
MIN3P	0.0175	0.725
Reference	0.0173	0.985

GDAE1D more accurate, but slower

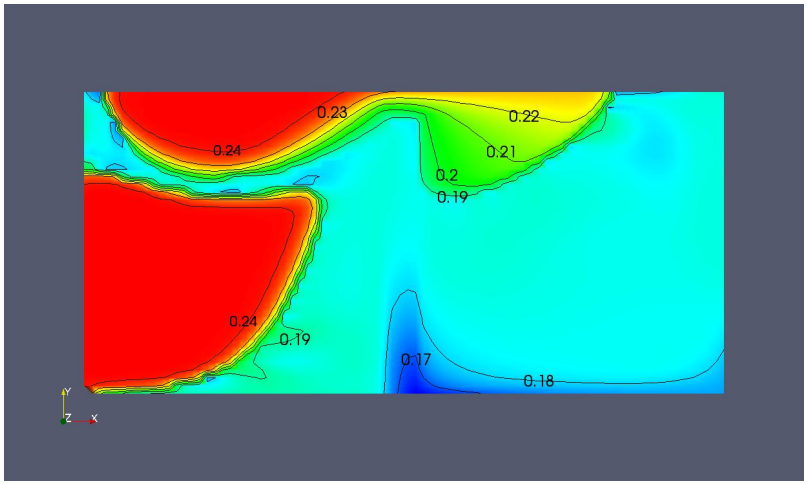


2D benchmark results

One species among 13 at time t=1000



Erlangen result with very fine mesh



GDAE result with coarse mesh
Numerical dispersion due to the coarse mesh but accurate results

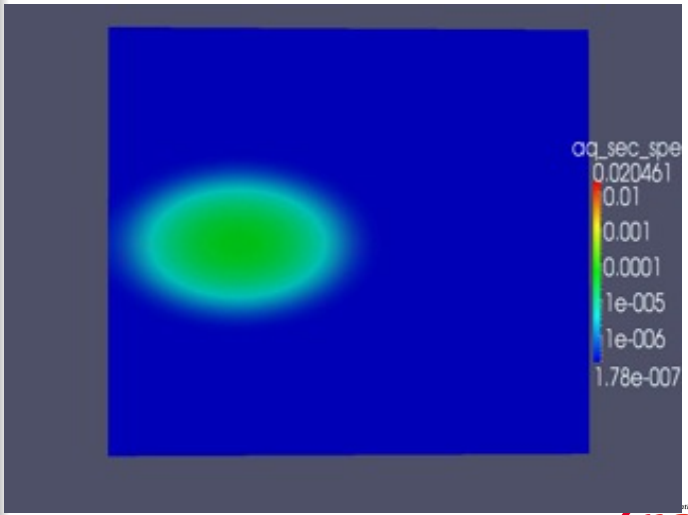
2D Andra test case

Chemical description

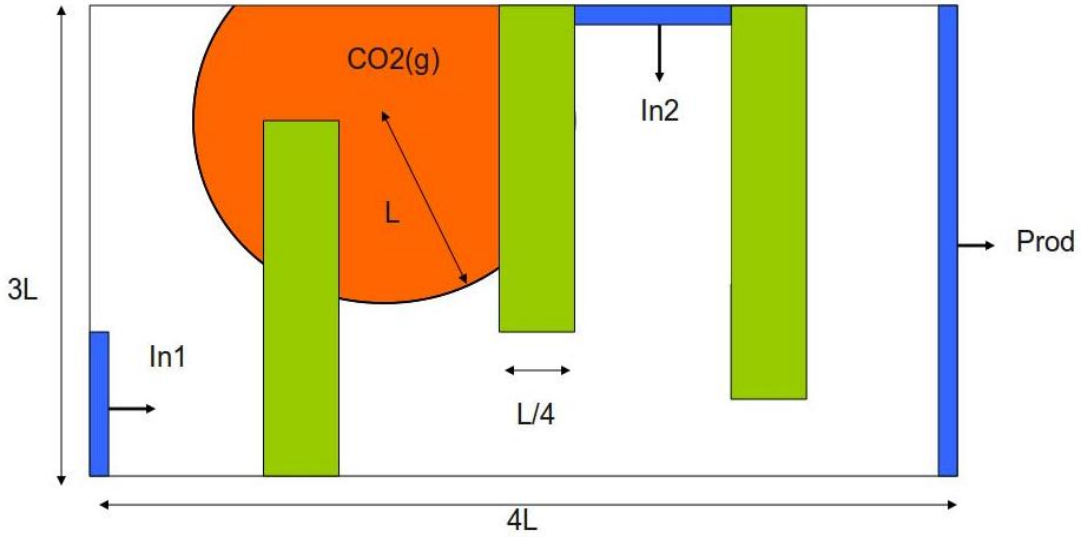
- Injection of alcaline water (NaOH) into a porous medium containing quartz (SiO2)
- Dissolution of quartz : $H_4SiO_4 \rightleftharpoons SiO_2 + 2\,H_2O$
- Aqueous reactions : $H_4SiO_4 \rightleftharpoons H_3SiO_4^- + H^+$, $H_2O \rightleftharpoons H^+ + OH^-$
- Sodium is a tracer

Geometry and transport conditions

- Rectangular domain of size $5\mathrm{m}x3.5\mathrm{m}$
- Injection at time $t = 0$ of NaOH at point $(1, 1.75)$
- Advection ($v = 5.710^{-7}\mathrm{m/s}$) and dispersion
- Duration 30 days



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

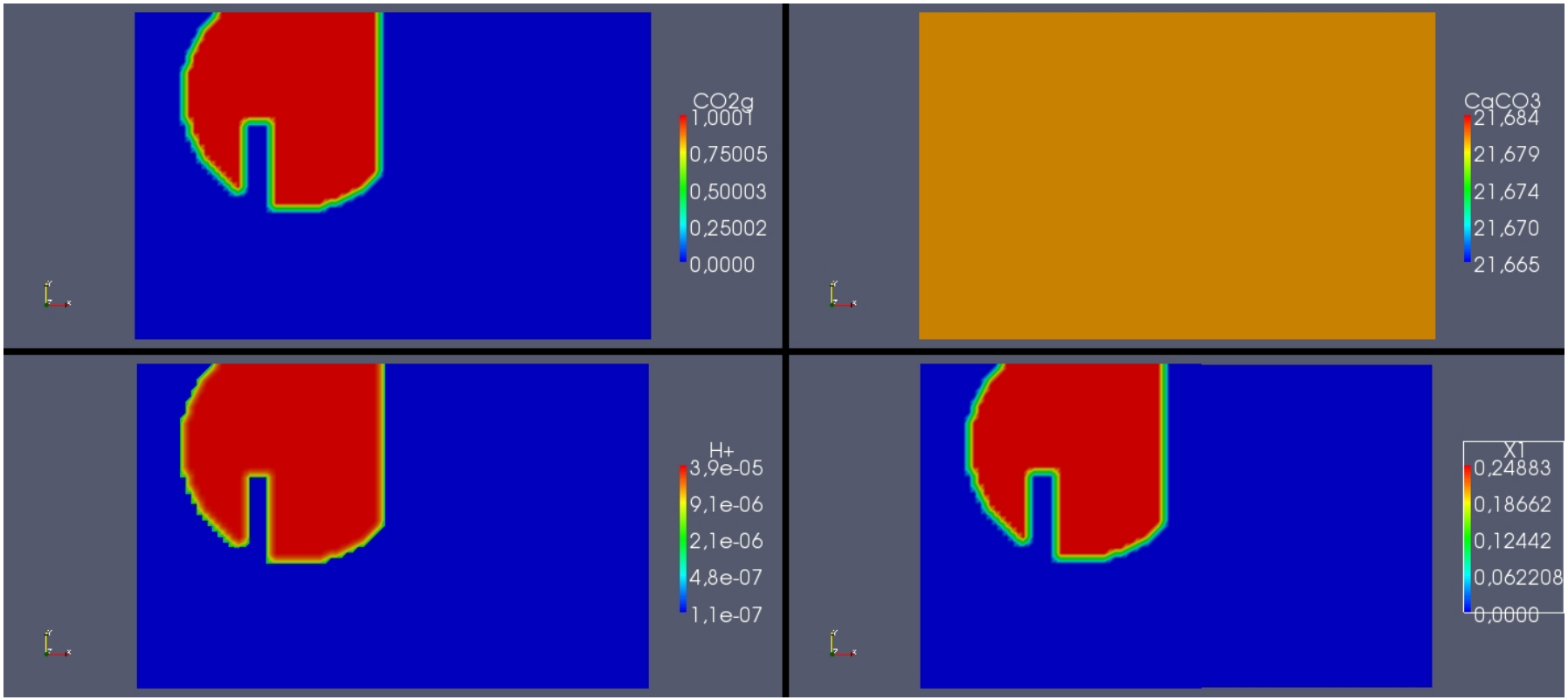


Dissolution of CO_2 in water, dissolution of calcite. Gas assumed immobile (capillary trapping), decouples flow from reactive transport.

Chemical system

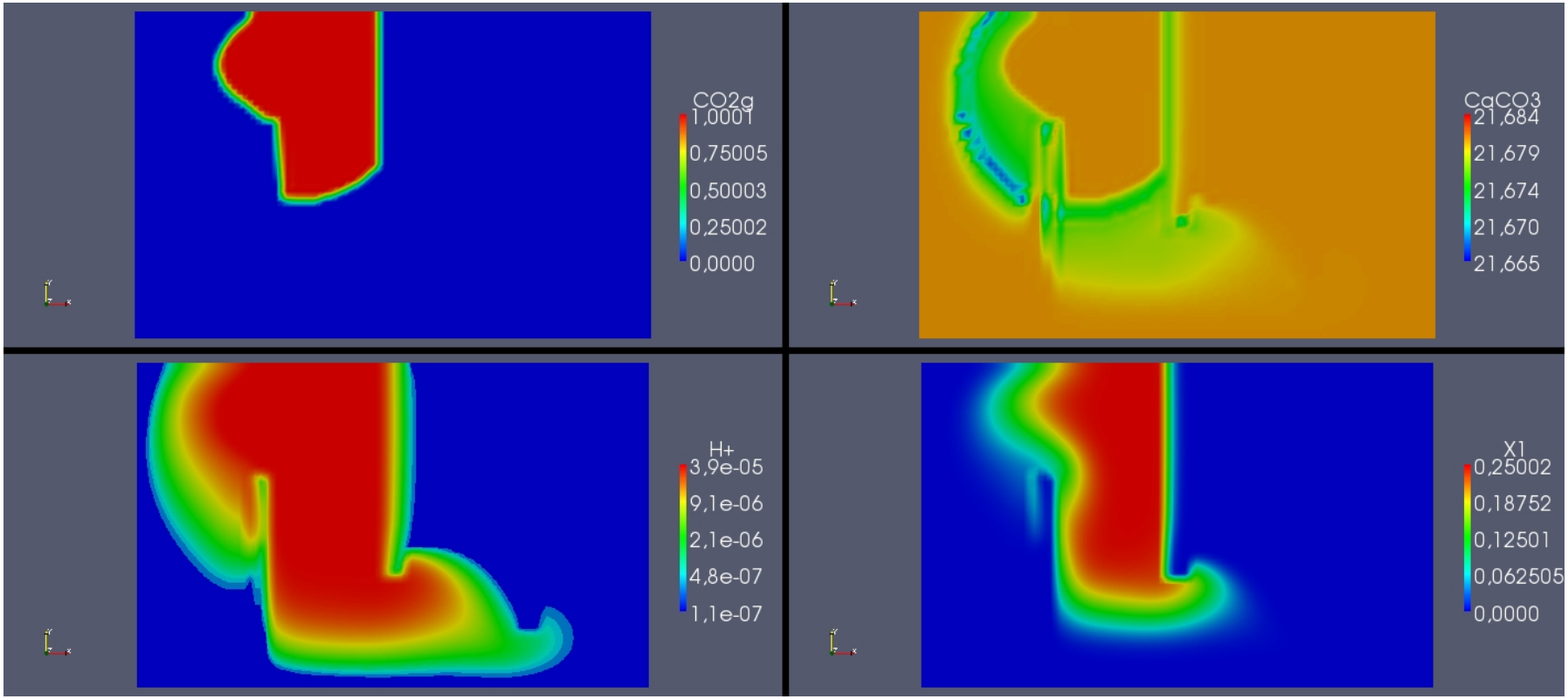
- $H_2O \rightleftharpoons H^+ + OH^-$ water dissociation
- $H_2O + CO_{2\,(aq)} \rightleftharpoons HCO_3^- + H^+$ dissociation of aqueous CO_2
- $CO_{2\,(g)} \rightleftharpoons CO_{2\,(aq)}$ gas dissolution
- $CaCO_3 + H^+ \rightleftharpoons Ca_2^+ + HCO_3^-$ Dissolution of calcite

Evolution of concentrations (Post-doc B. Gueslin)



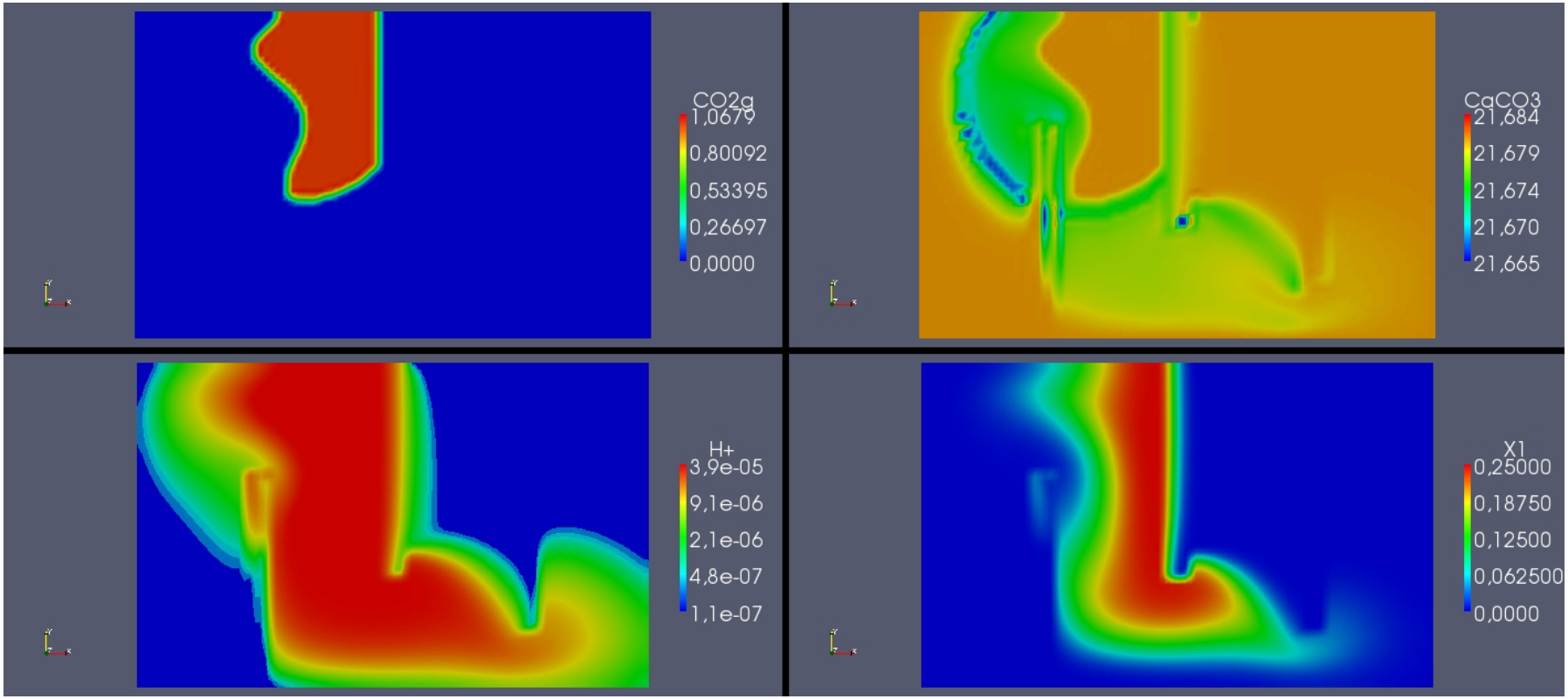
$t = 0$

Evolution of concentrations (Post-doc B. Gueslin)



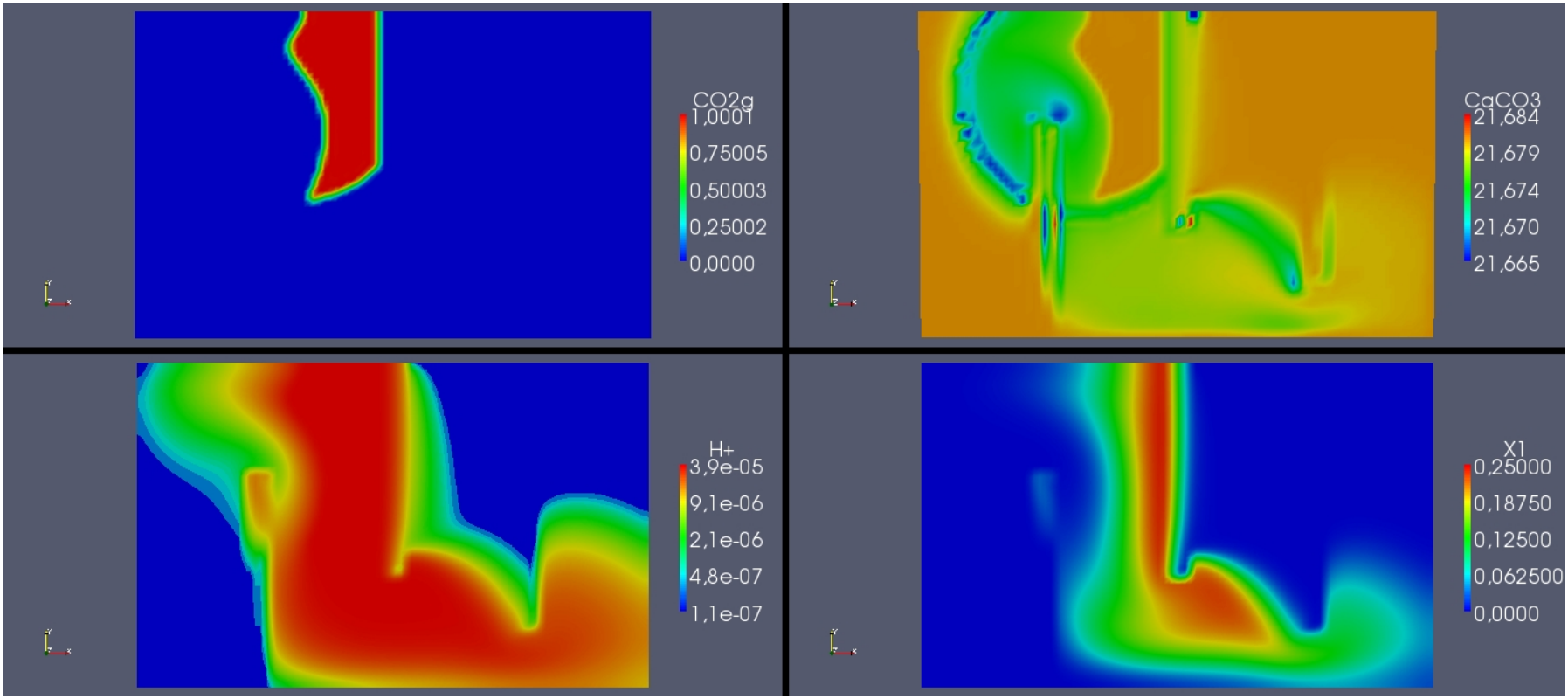
$t = 400$ years

Evolution of concentrations (Post-doc B. Gueslin)



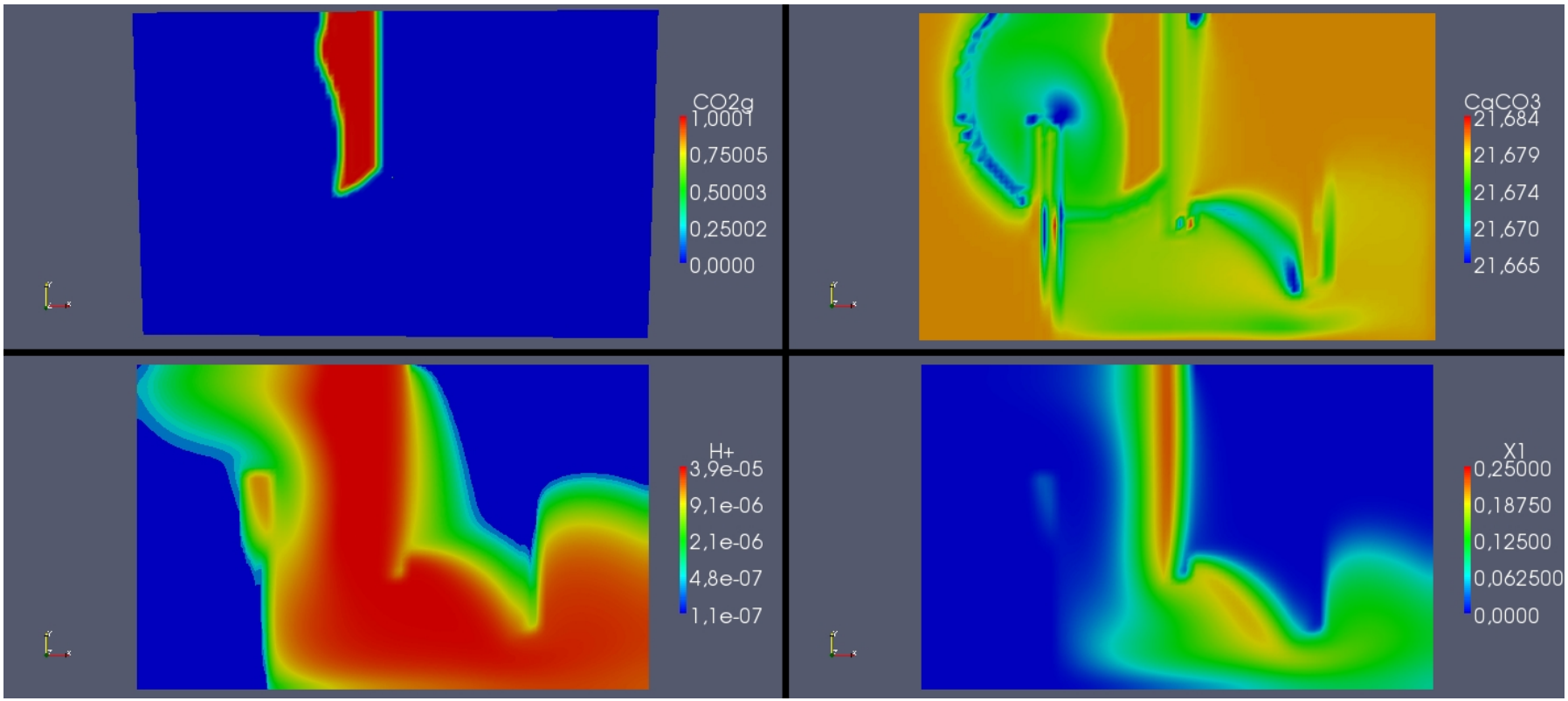
$t = 800$ years

Evolution of concentrations (Post-doc B. Gueslin)



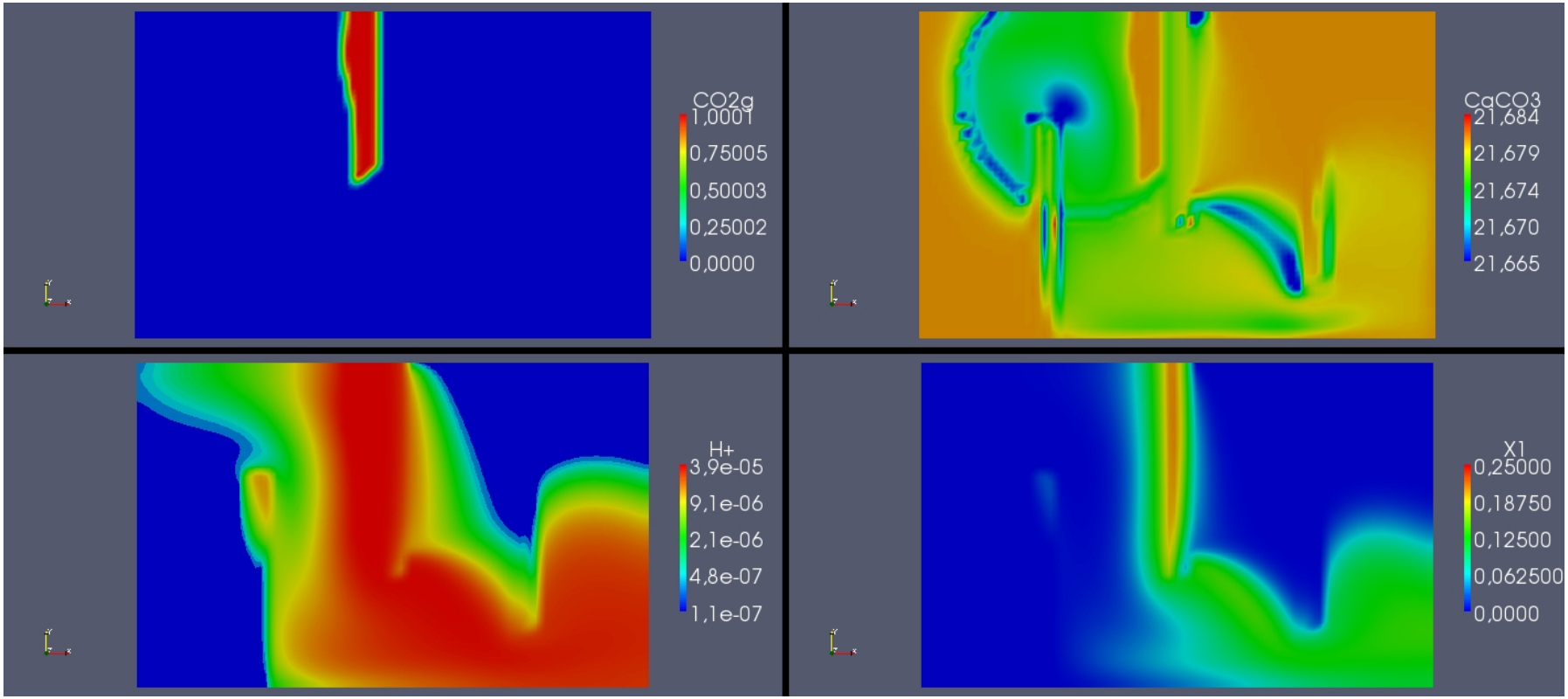
$t = 1200$ years

Evolution of concentrations (Post-doc B. Gueslin)



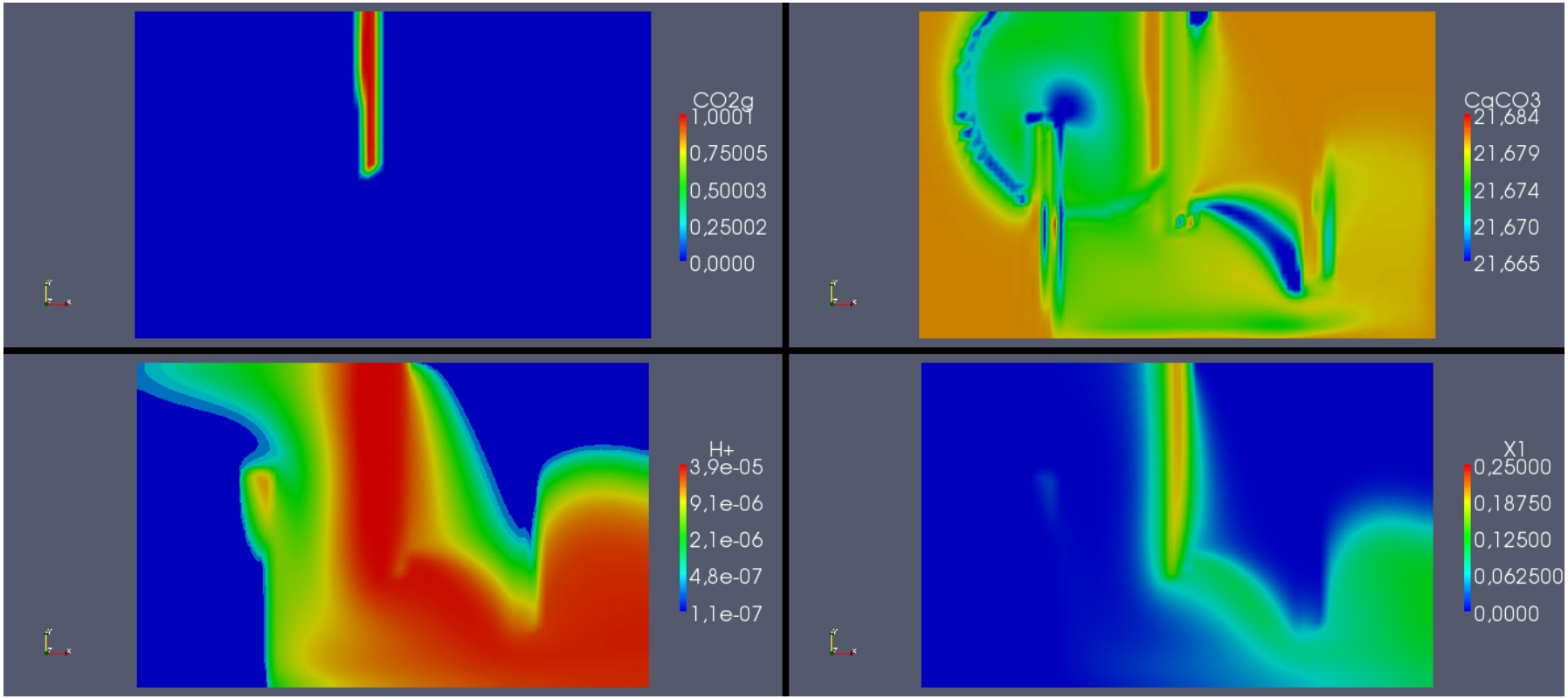
$t = 1600$ years

Evolution of concentrations (Post-doc B. Gueslin)



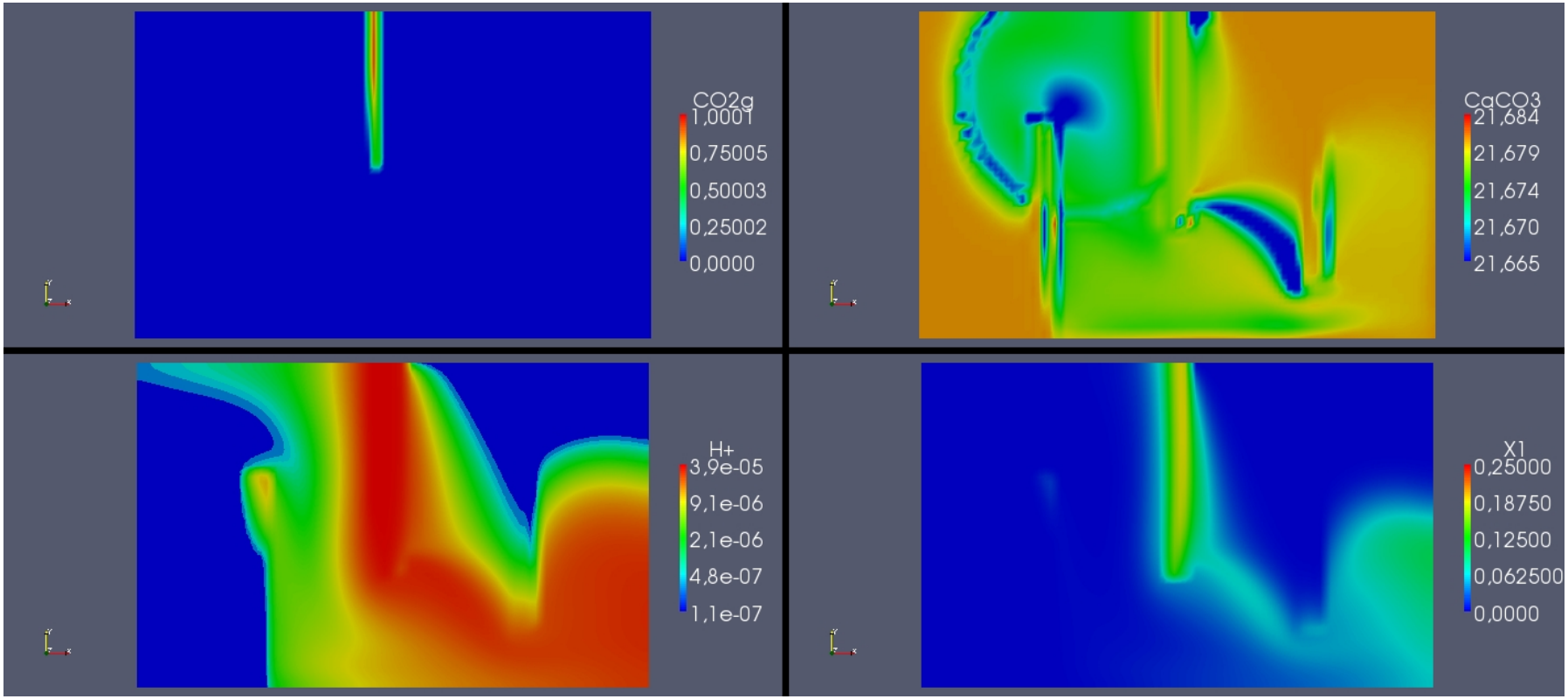
$t = 2000$ years

Evolution of concentrations (Post-doc B. Gueslin)



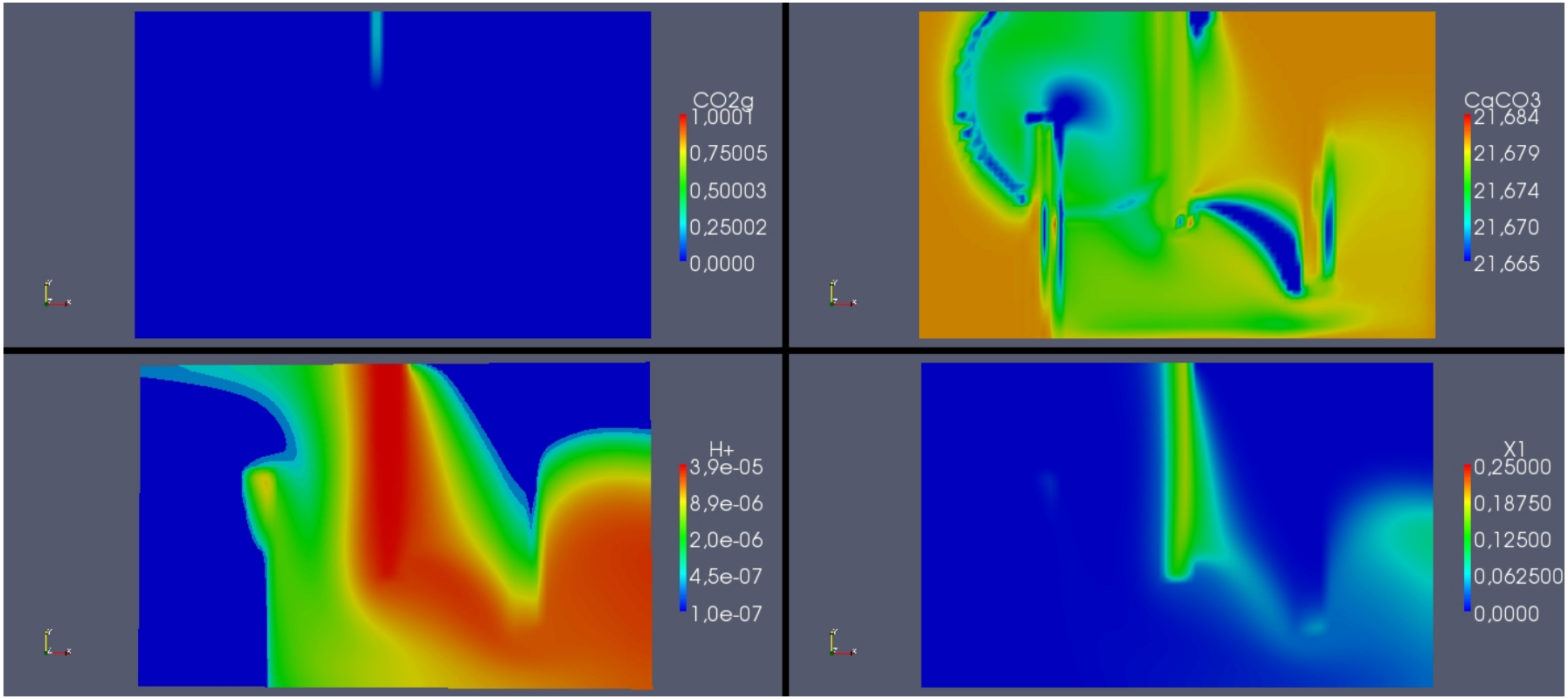
$t = 2400$ years

Evolution of concentrations (Post-doc B. Gueslin)



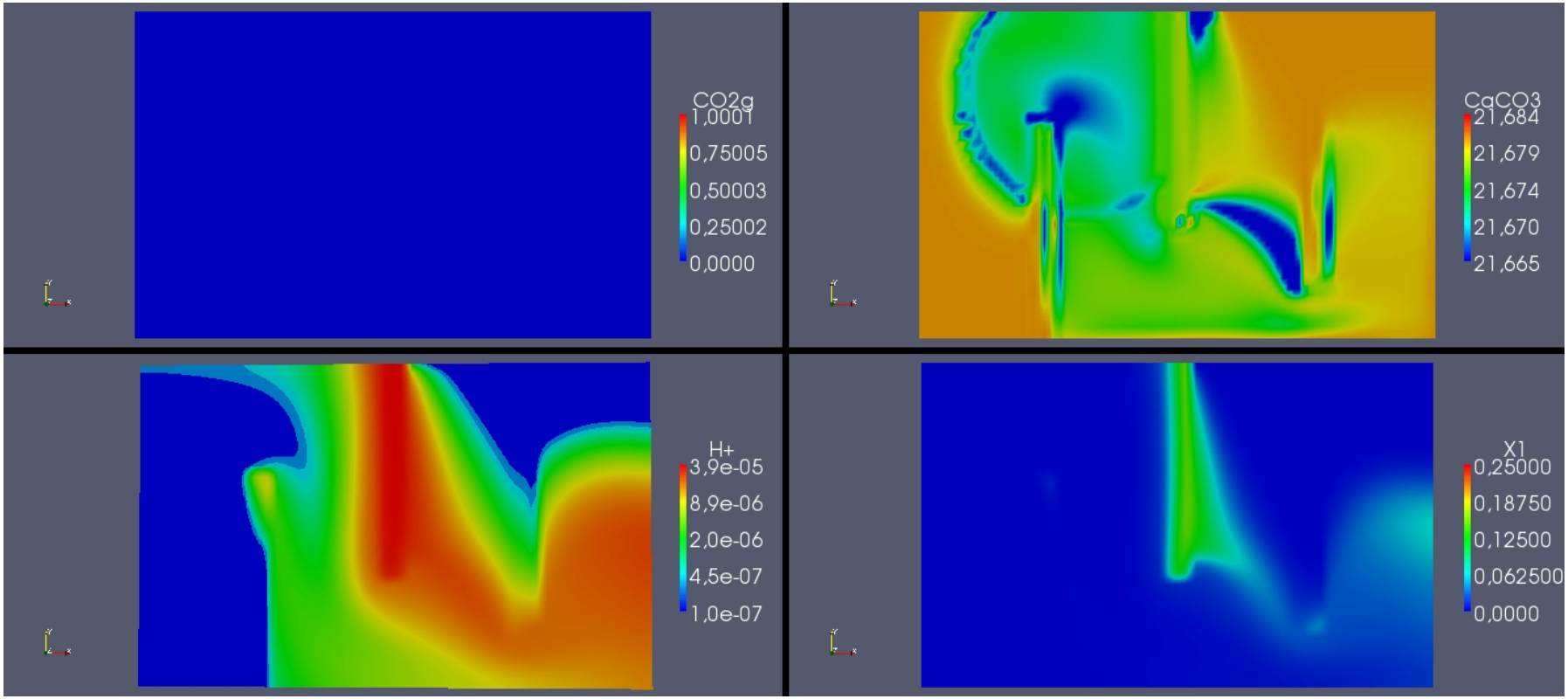
$t = 2800$ years

Evolution of concentrations (Post-doc B. Gueslin)



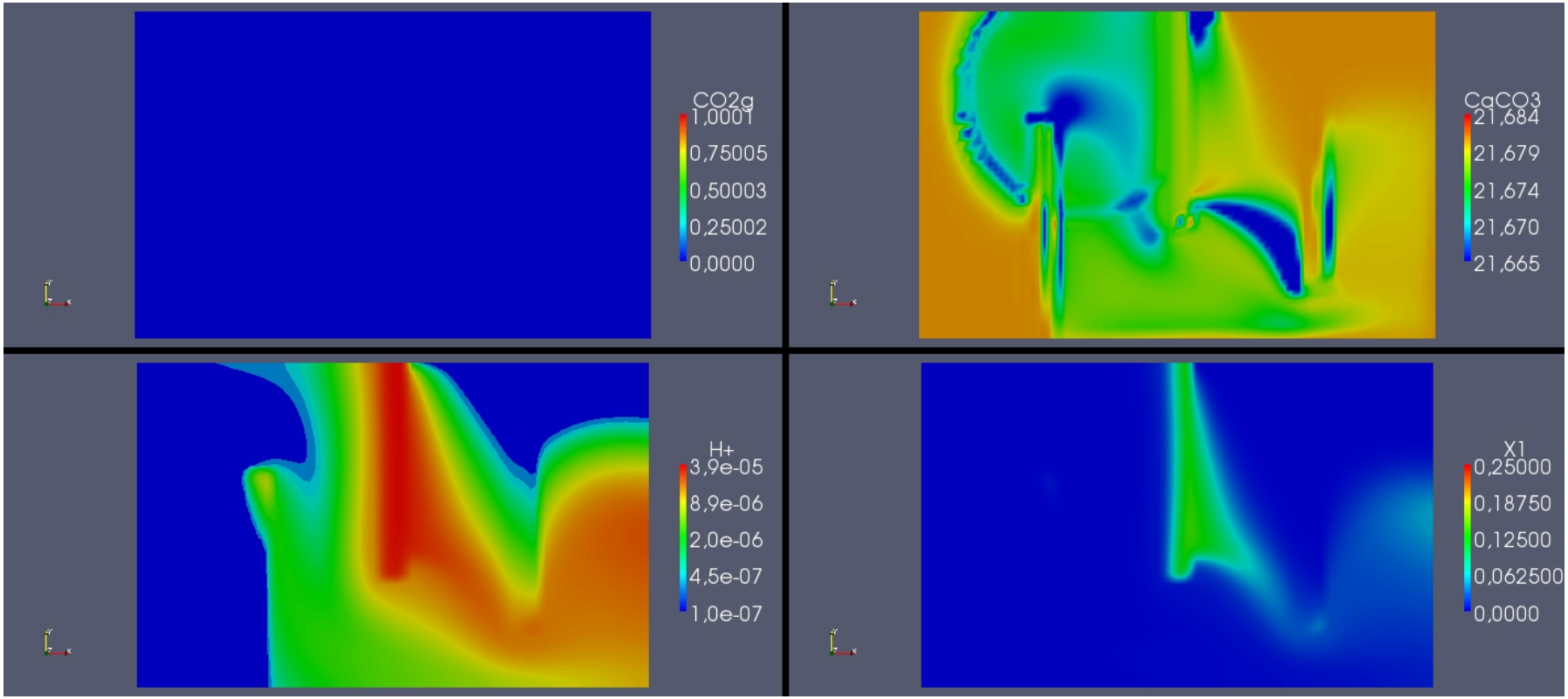
$t = 3200$ years

Evolution of concentrations (Post-doc B. Gueslin)



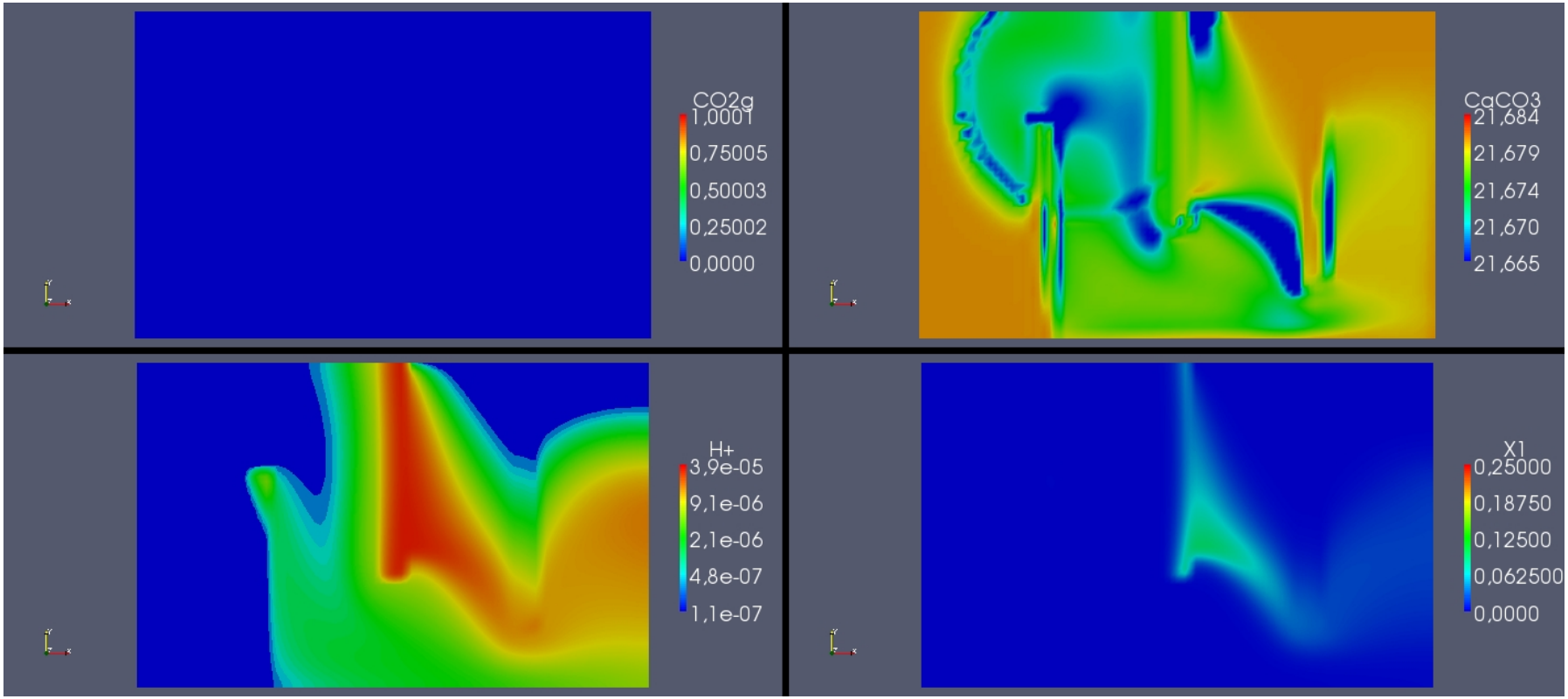
$t = 3600$ years

Evolution of concentrations (Post-doc B. Gueslin)



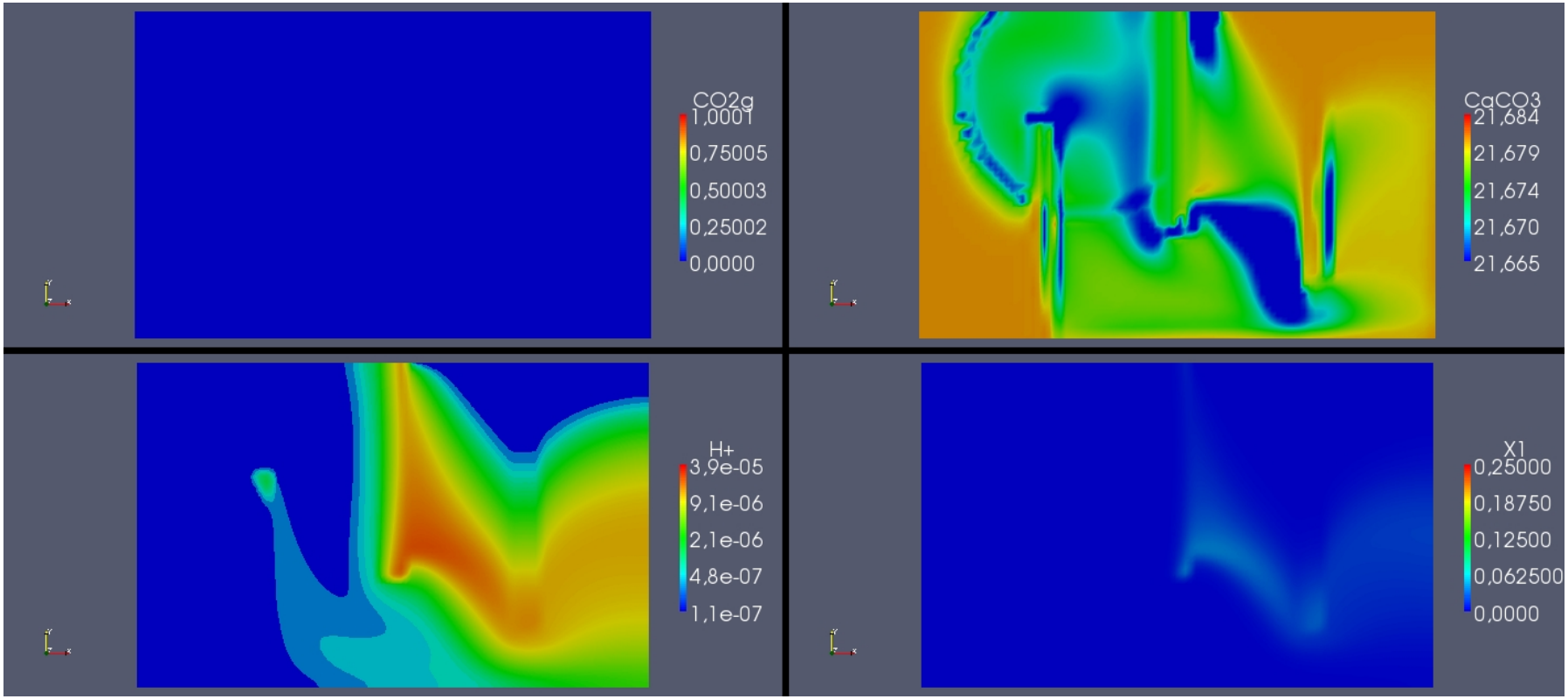
$t = 4000$ years

Evolution of concentrations (Post-doc B. Gueslin)



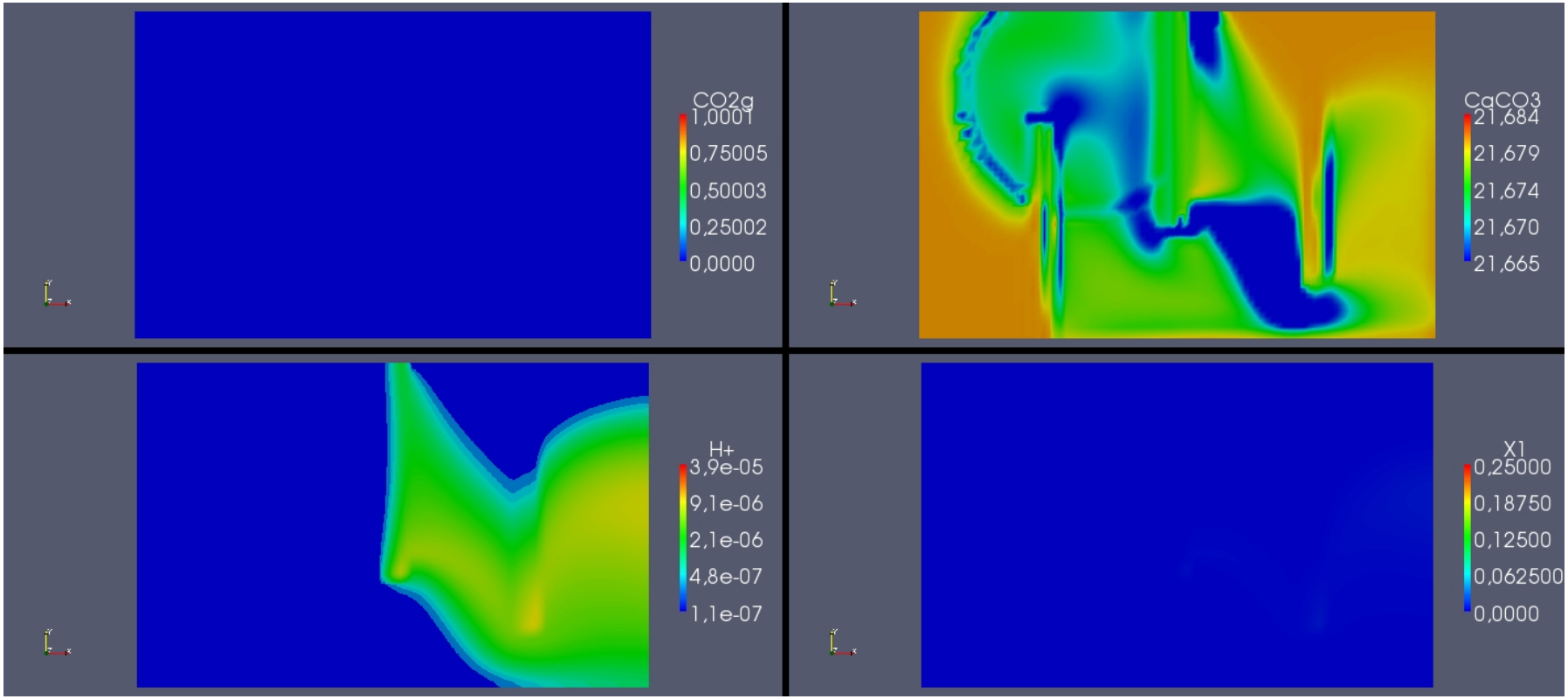
$t = 5000$ years

Evolution of concentrations (Post-doc B. Gueslin)



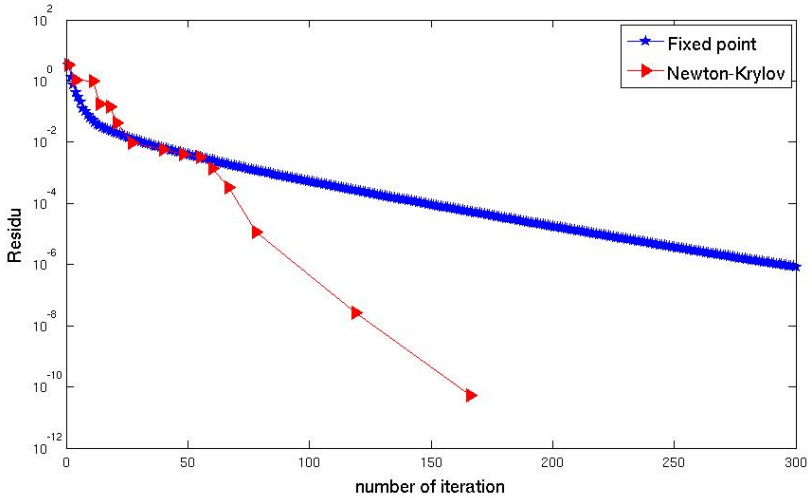
$t = 7000$ years

Evolution of concentrations (Post-doc B. Gueslin)

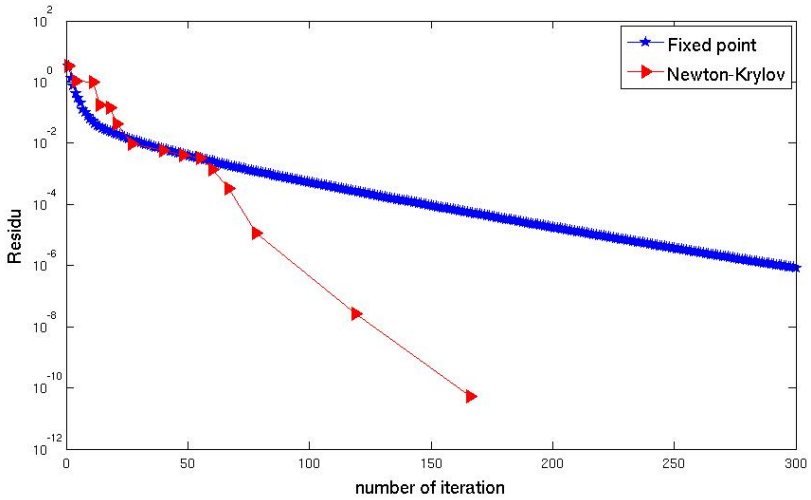


$t = 10000$ years

Newton–Krylov and preconditioner performance



Newton–Krylov and preconditioner performance

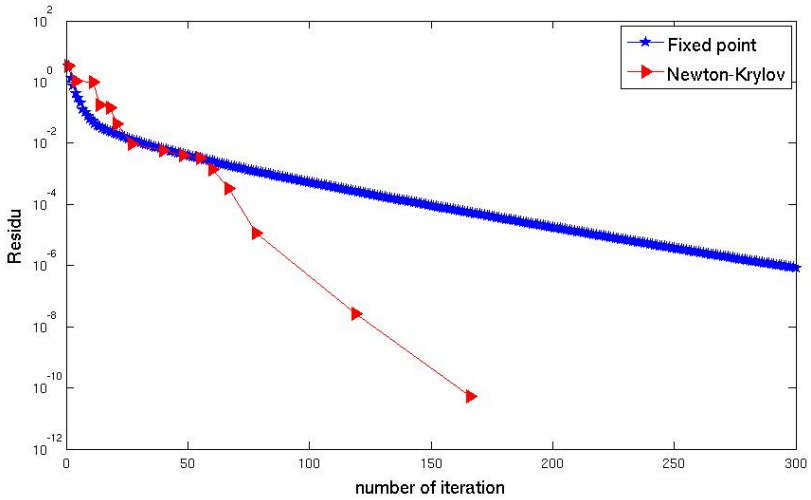


	h		$h/2$		$h/4$		$h/8$	
	NI	LI	NI	LI	NI	LI	NI	LI
No prec.	8	42	8	76	10	105	10	177

Mesh dependance : adaptive forcing term

NI : # nonlinear iters, NLI : total # linear iters.

Newton–Krylov and preconditioner performance

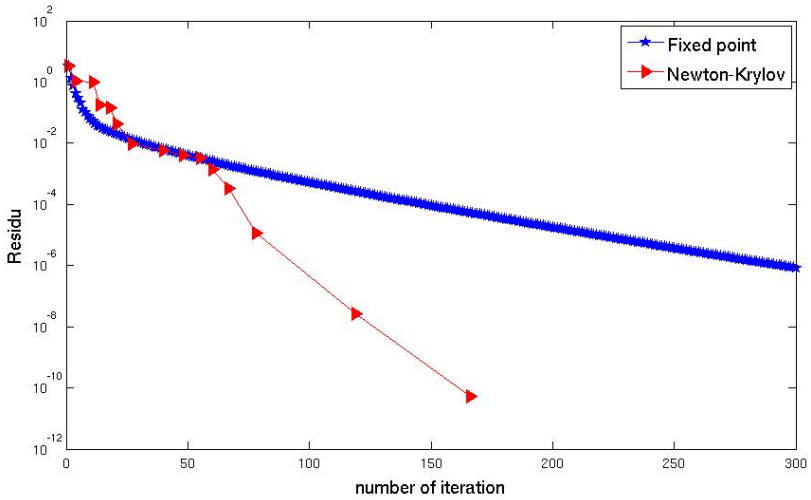


	h		$h/2$		$h/4$		$h/8$	
	NI	LI	NI	LI	NI	LI	NI	LI
No prec.	8	42	8	76	10	105	10	177
BGS	8	23	7	24	7	22	8	25

Mesh dependance : adaptive forcing term

NI : # nonlinear iters, NLI : total # linear iters.

Newton–Krylov and preconditioner performance



	h		$h/2$		$h/4$		$h/8$	
	NI	LI	NI	LI	NI	LI	NI	LI
No prec.	8	42	8	76	10	105	10	177
BGS	8	23	7	24	7	22	8	25
Elimination	5	15	5	15	5	15	5	15

Mesh dependance : adaptive forcing term

NI : # nonlinear iters, NLI : total # linear iters.

Work in progress – future work

Work in progress

- Reduction of CPU time in DAE methods

Work in progress – future work

Work in progress

- Reduction of CPU time in DAE methods
- Parallel software

Work in progress – future work

Work in progress

- Reduction of CPU time in DAE methods
- Parallel software
- Experimental comparison with SNIA method

Work in progress – future work

Work in progress

- Reduction of CPU time in DAE methods
- Parallel software
- Experimental comparison with SNIA method
- Reactive models with kinetics and precipitation-dissolution

Work in progress – future work

Work in progress

- Reduction of CPU time in DAE methods
- Parallel software
- Experimental comparison with SNIA method
- Reactive models with kinetics and precipitation-dissolution

Work in progress – future work

Work in progress

- Reduction of CPU time in DAE methods
- Parallel software
- Experimental comparison with SNIA method
- Reactive models with kinetics and precipitation-dissolution

Future work

- Species appearance / disappearance as complementarity problem

Theses : S. Sabit (JE, Andra), V. Vostrikov (B. Amaziane, MK, Maison de la Simulation)

Work in progress – future work

Work in progress

- Reduction of CPU time in DAE methods
- Parallel software
- Experimental comparison with SNIA method
- Reactive models with kinetics and precipitation-dissolution

Future work

- Species appearance / disappearance as complementarity problem
- Improving the substitution approach

Theses : S. Sabit (JE, Andra), V. Vostrikov (B. Amaziane, MK, Maison de la Simulation)

Work in progress – future work

Work in progress

- Reduction of CPU time in DAE methods
- Parallel software
- Experimental comparison with SNIA method
- Reactive models with kinetics and precipitation-dissolution

Future work

- Species appearance / disappearance as complementarity problem
- Improving the substitution approach
- Exploring mesh refinement strategies

Theses : S. Sabit (JE, Andra), V. Vostrikov (B. Amaziane, MK, Maison de la Simulation)

Work in progress – future work

Work in progress

- Reduction of CPU time in DAE methods
- Parallel software
- Experimental comparison with SNIA method
- Reactive models with kinetics and precipitation-dissolution

Future work

- Species appearance / disappearance as complementarity problem
- Improving the substitution approach
- Exploring mesh refinement strategies
- Two phase flow with chemistry

Theses : S. Sabit (JE, Andra), V. Vostrikov (B. Amaziane, MK, Maison de la Simulation)