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An experimental versus numerical approach to determine capability of Archie's law to explain impact of evolving porosity on diffusivity

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AN EXPERIMENTAL VERSUS NUMERICAL APPROACH
TO DETERMINE CAPABILITY OF ARCHIE'S LAW TO
EXPLAIN IMPACT OF EVOLVING POROSITY ON
DIFFUSIVITY

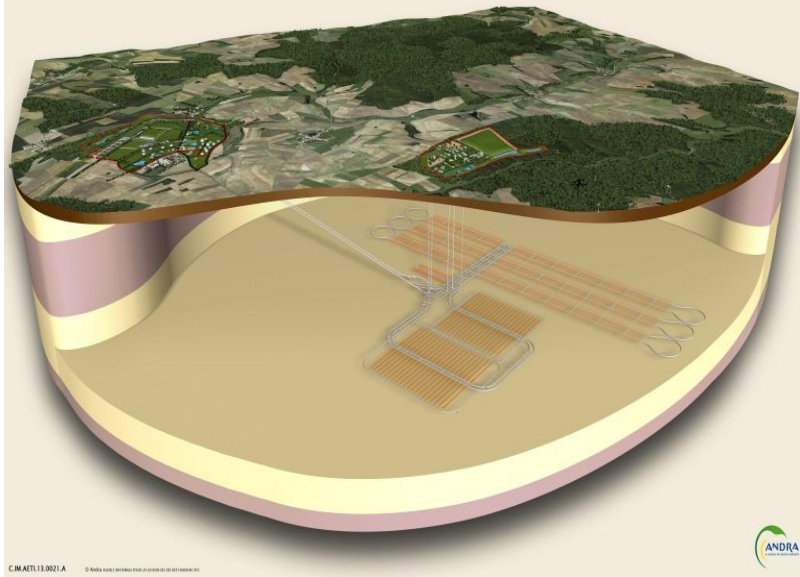
Ashish RAJYAGURU

DPC / SECR/ L3MR

**Directeurs de thèse : Prof. LAGNEAU Vincent & SAVOYE Sébastien
Ecole Doctorale : GNRE– Ecole des Mines de Paris**

Début de thèse : 2 novembre 2015

ILW-HLW radioactive waste disposal facility



Properties of claystones

Very **low permeability**

→ **Diffusion** of ionic species

Negatively surface charged clayey minerals

→ **Sorption** of cationic species and **exclusion** of anionic species

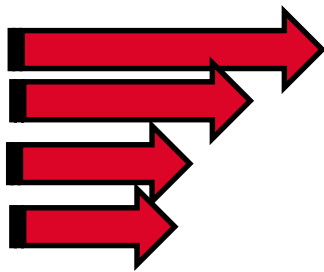
Clay properties essential to delay radionuclide transport into surrounding geosphere

Change in rock containment properties due to mineral perturbation

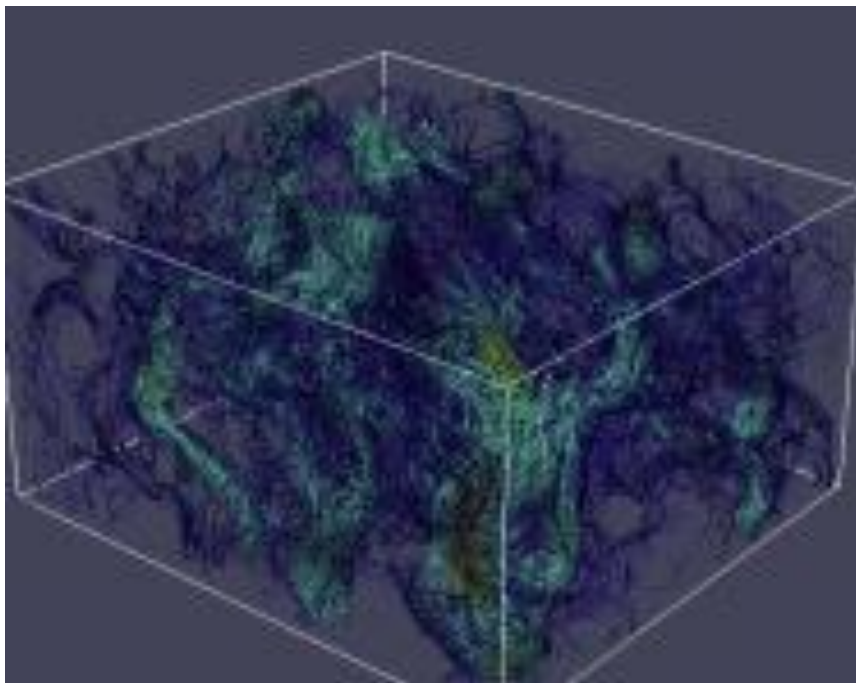
How such chemical impacts observed at lab-scale can be extrapolated for repository space and time scales?



Saline Plume



Porous system¹



Porosity

$$\phi = \left(1 - \frac{\rho_d}{\rho_s} \right)$$

ρ_d (g.cm⁻³) is the dry density
 ρ_s (g.cm⁻³) is the grain density

Mineral perturbation impact on diffusivity

Empirical Archie's law $\frac{D_e}{D_0} = \phi^m$

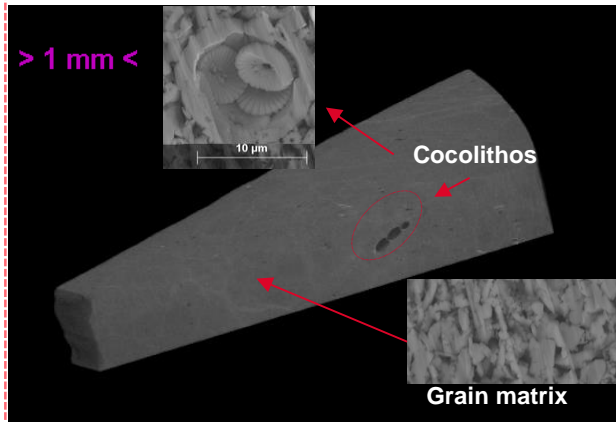
m is the cementation factor (-) (property of intact porous material)
 D_e is the effective diffusion coefficient (m².s⁻¹)
 D_0 is the diffusion coefficient in bulk water (m².s⁻¹)
 ϕ is the porosity (-)

Can empirical Archie's law reproduce mineral perturbation impact on diffusivity for a complex system such as claystones?

1) Gomrod *et al.* 2017

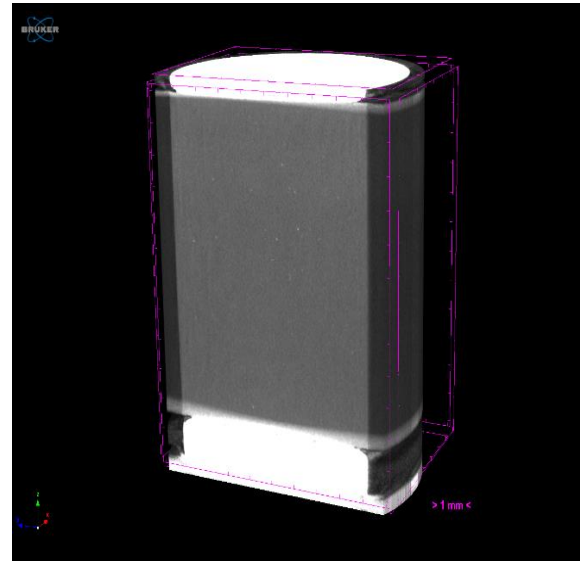
Compacted clay minerals

Micritic chalk

Chalk properties

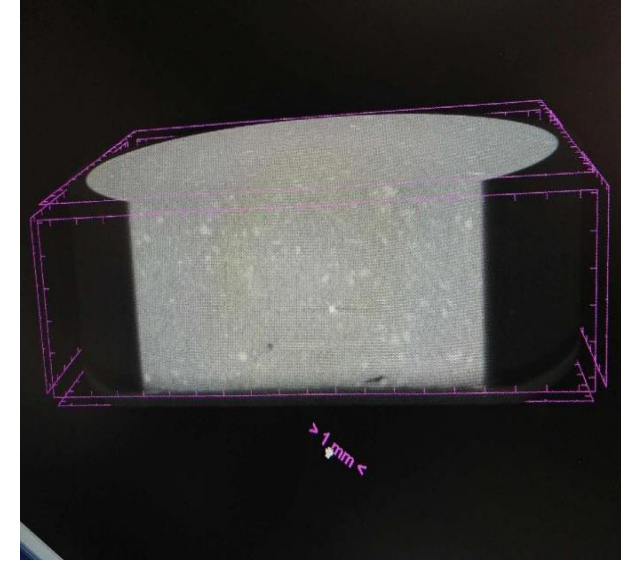
- ❑ 97% calcite matrix
- ❑ Total porosity: 45%
- ❑ No alteration in cations/anions diffusion
- ❑ 2% of coccolithos as macro-porosity

Kaolinite

Properties @ $\rho_d=1.7 \text{ g.cm}^{-3}$

- ❑ Total porosity: 36 %
- ❑ Anionic accessible porosity: 36 %
- ❑ weak negative surface charge (CEC = 38 meq.kg⁻¹)

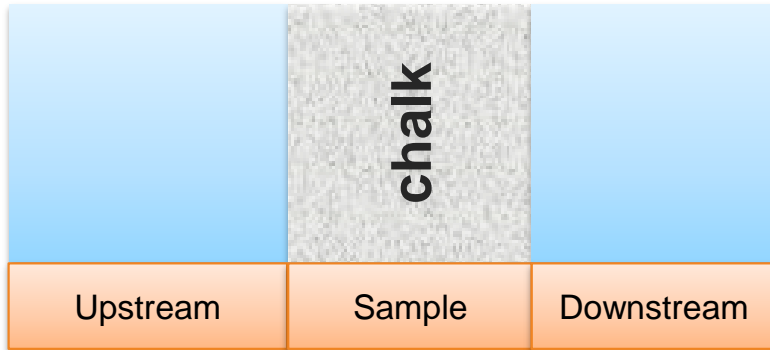
Illite

Properties @ $\rho_d=1.9 \text{ g.cm}^{-3}$

- ❑ Total porosity: 29 %
- ❑ Anion accessible porosity: 19%
- ❑ Negative surface charge (CEC = 225 meq.kg⁻¹)

What is the impact of spatial variability on studied mineral perturbation and subsequently on diffusivity of water tracers?

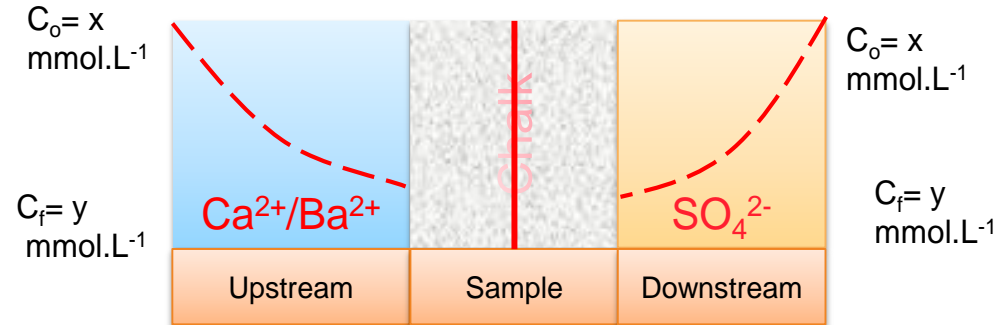
Reactive diffusion setup



Precipitation of two sulfate-alkaline minerals

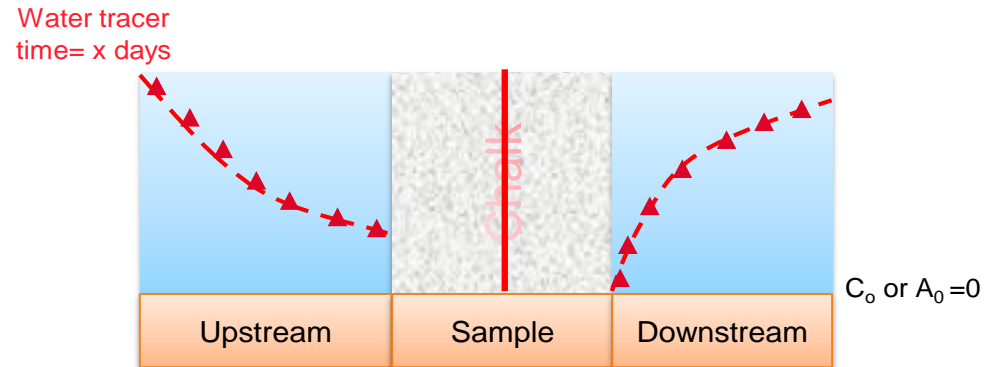
<p>Barite Properties:</p> <ul style="list-style-type: none"> ❖ Sparingly soluble ❖ Slow kinetics of precipitation 	<p>Gypsum Properties:</p> <ul style="list-style-type: none"> ❖ Fairly soluble ❖ Fast kinetics of precipitation
<p>Barite</p>	<p>Gypsum</p>

Precipitation impact on total porosity



- Barite molar volume 74.5 cm³.mol⁻¹
- Gypsum molar volume 52.5 cm³.mol⁻¹

Precipitation impact on diffusivity



RESULTS: PRECIPITATION IMPACT ON TOTAL POROSITY AND DIFFUSIVITY

Precipitation impact on total porosity

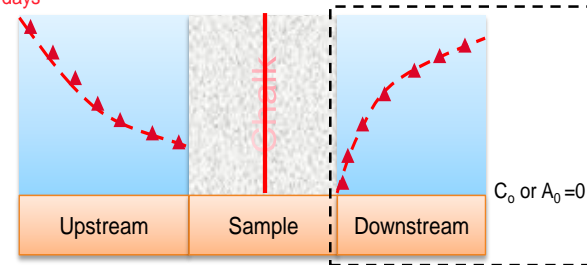
End of Experiments:

- Barium contribution = 2.5 mmol
- Gypsum contribution = 1.5 mmol

Total porosity reduction

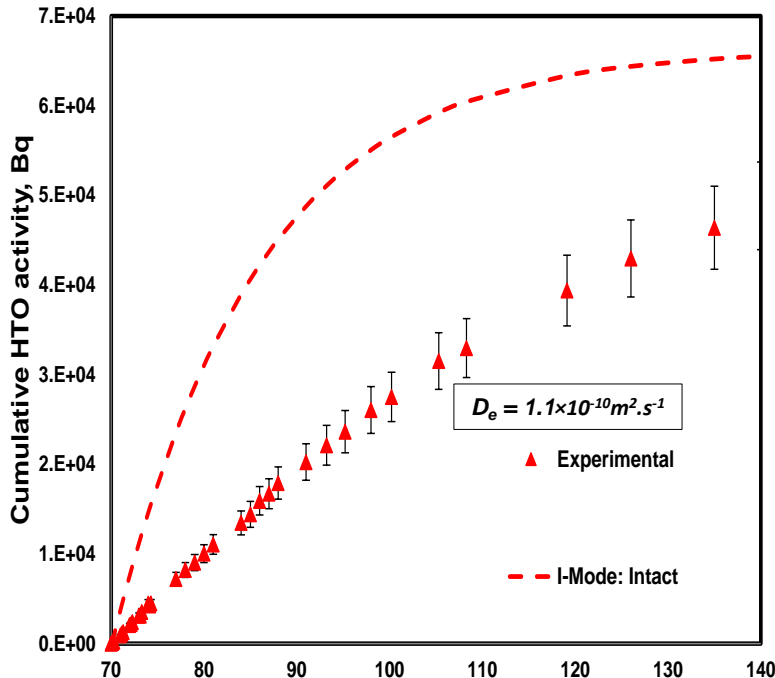
- Barite case = 2.5%
- Gypsum case = 2%

Water tracer
time = x days

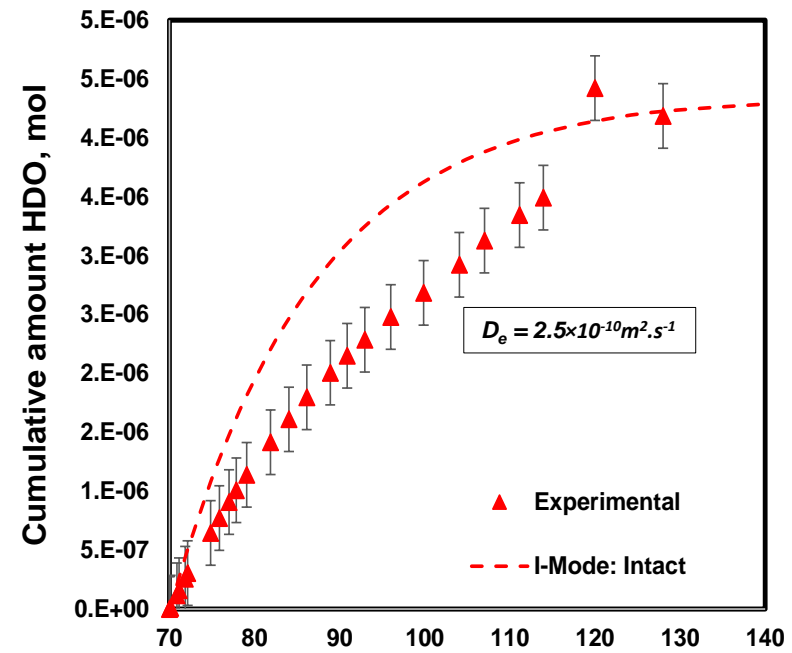


Precipitation impact on diffusivity

Barite: Case: HTO diffusion



Gypsum case: HDO diffusion



Barite & Gypsum led to similar total porosity reduction
Why barite impact more on diffusivity than gypsum?

RESULTS: CAN CODE REPRODUCE PRECIPITATION IMPACT ON TOTAL POROSITY AND DIFFUSIVITY?

Code prediction: Reactant contribution to precipitation

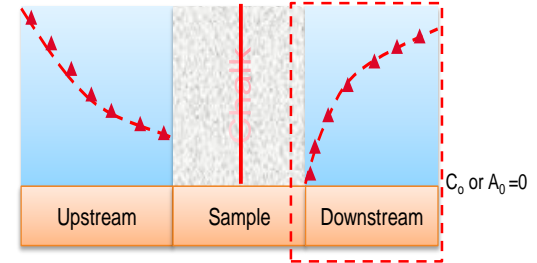
End of Experiments:

- Barium contribution = 2.8 mmol
- Gypsum contribution = 1.5 mmol

Total porosity reduction

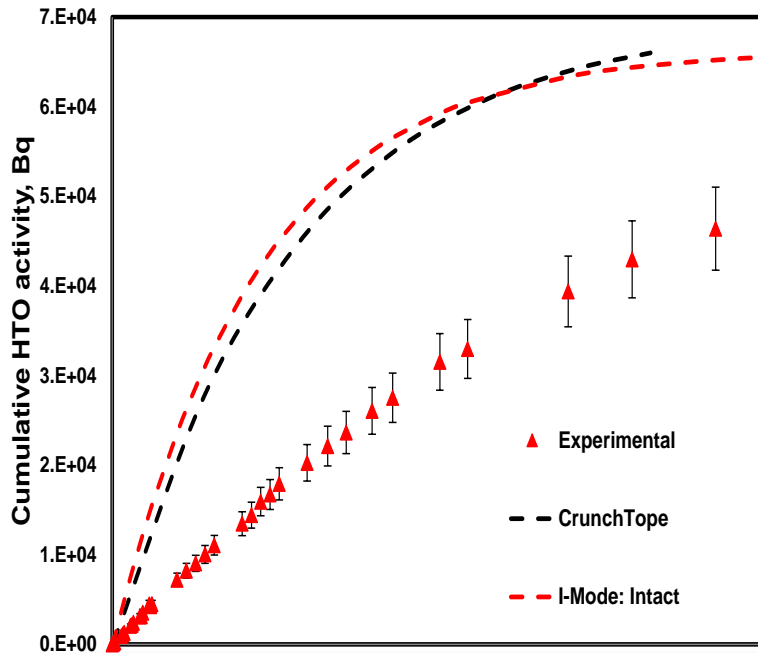
- Barite case ~ 2.6%
- Gypsum case ~ 2%

Water tracer
time= x days

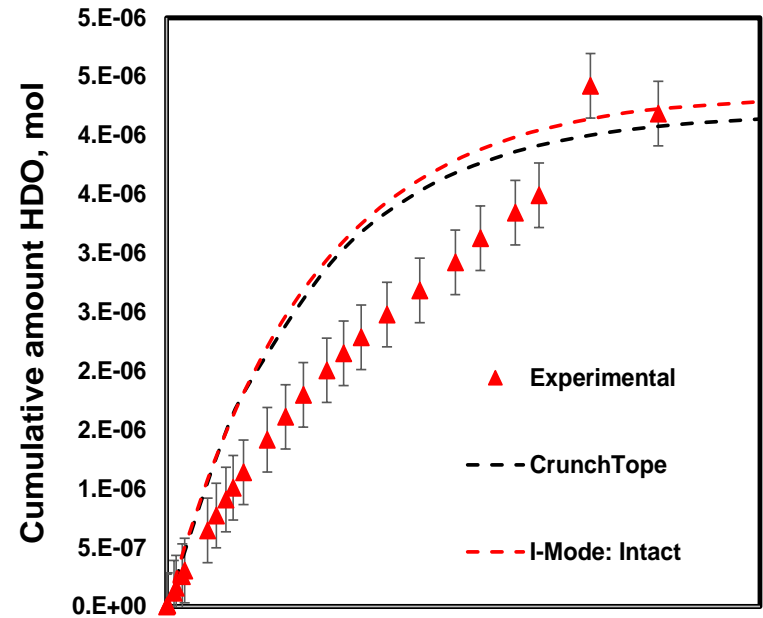


Code prediction: Precipitation impact on diffusivity

Barite: Case: HTO diffusion

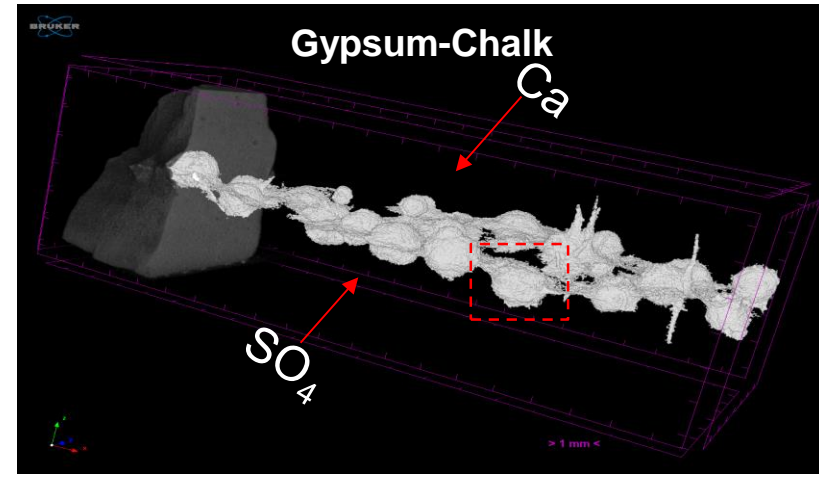
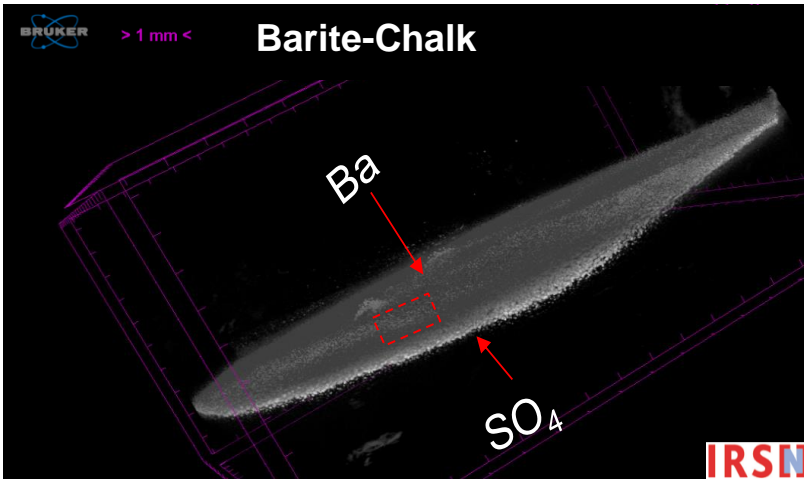


Gypsum case: HDO diffusion



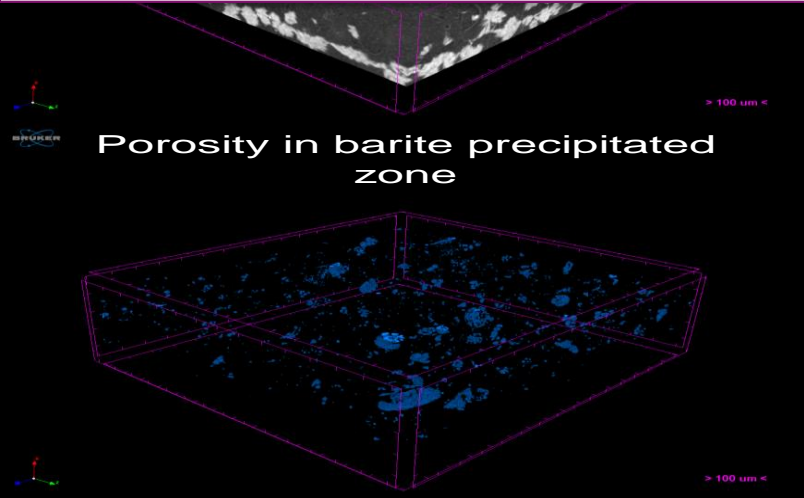
Code well reproduced the porosity reduction due to precipitation
Code under-estimated the precipitation impact on diffusivity

RESULTS: BARITE & GYPSUM EVOLUTION IN CHALK

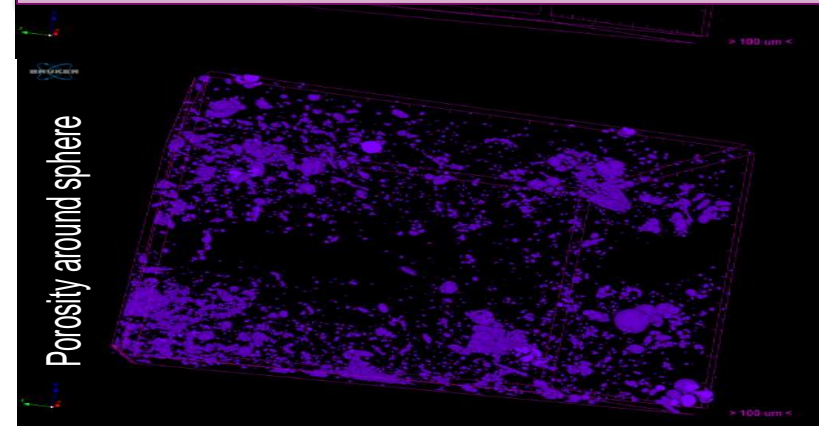


Small selected barite-chalk zone

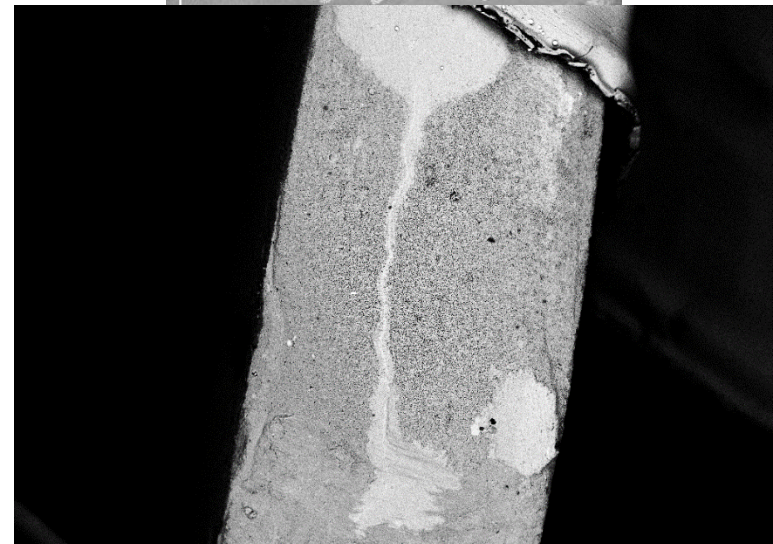
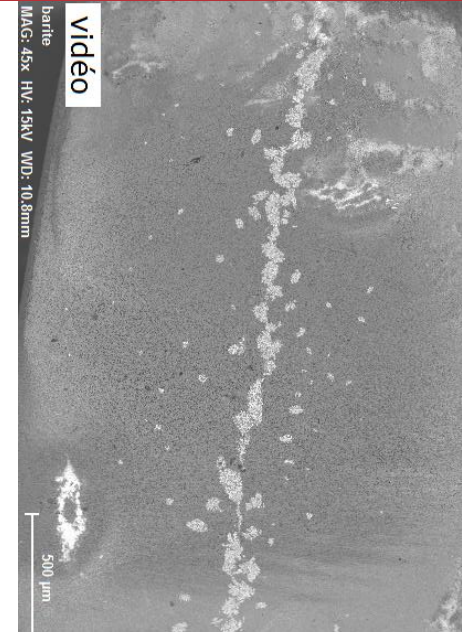
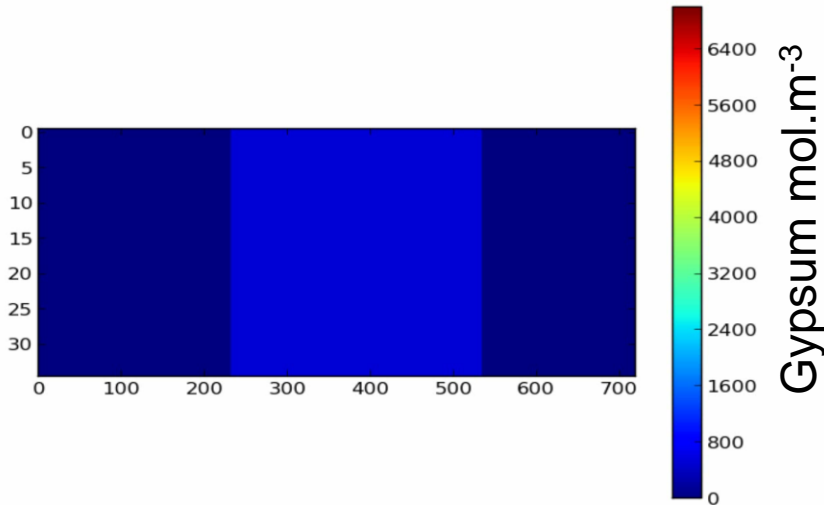
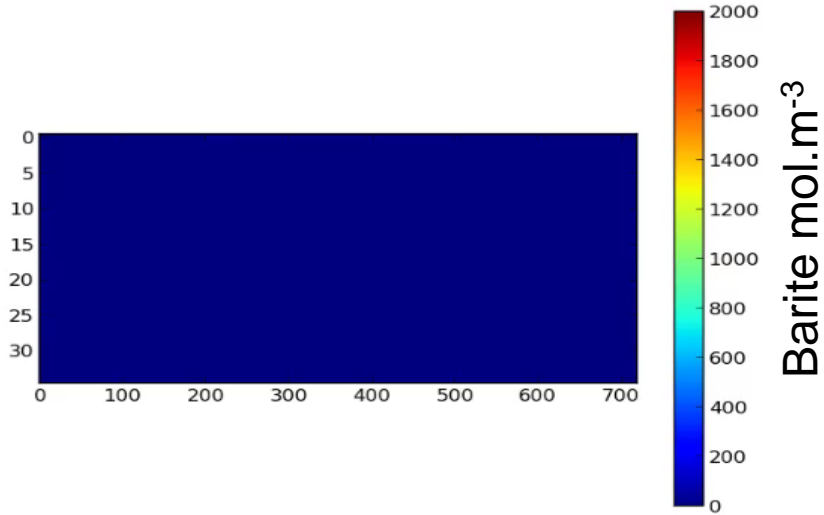
Quasi-continuous barite layer with some remaining unfilled pores
Strong impact of barite on diffusion



The intact chalk matrix around gypsum spheres allowed significant tracer to diffuse
These intact zones lowered the impact of precipitates on diffusion



DISCUSSION: WHAT IS THE IMPACT OF SPATIAL VARIABILITY ON EVOLUTION OF MINERALS?



CONCLUSION

1. Can Empirical Archie's law reproduce mineral perturbation impact on diffusivity for a micritic chalk system?

→ Code properly reproduced macroscopic precipitation impact on chalk porosity

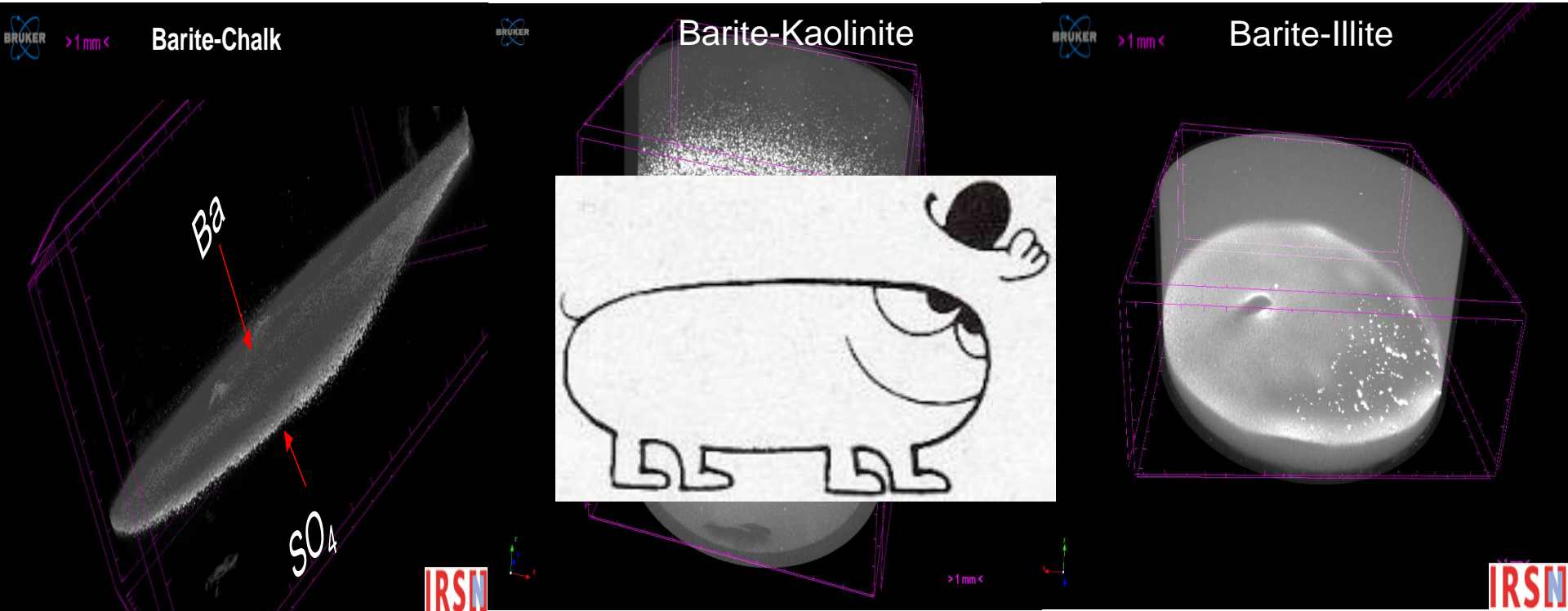
→ Archie's law underestimated the impact on diffusivity

2. What is the impact of spatial variability on studied mineral perturbation and subsequently on diffusivity of water tracers?

→ No significant impact on evolution of barite

→ Significant impact on gypsum evolution ⇔ Isolated spheres = lower impact on diffusion

CLAY RESULTS



Please join me at my
defense in october

Sincere acknowledgements to:

- ❖ SEGARRA Corinne (DPC/SECR/LECBA), VARLET Jérôme (DPC/SEARS/LISL) for SEM image acquisition
- ❖ GRENUT Bernard (DPC/SECR/LECBA) for Mercury intrusion-extrusion porosimetry tests
- ❖ L'HOPITAL Emilie (IRSN Fontenay-aux-Roses) for X-ray µct image acquisition and post-treatment imaging
- ❖ CHARTIER Thomas (IRSN Fontenay-aux-Roses) for development of heterogeneous porosity field



Thank you for your kind attention

Publication

S. Savoye, A. Rajyaguru, N. Macé, S. Lefèvre, G. Spir, J.C. Robinet. How mobile is tritiated water through unsaturated cement-based materials? New insights from two complementary approaches, *Applied Radiation and Isotopes* 2018 139, 98-106.

Publication under revision

A. Rajyaguru, E. L'Hopital, S. Savoye, C. Wittebroodt, O. Bildstein, P. Arnoux, V. Dettileux, I. Fatnassi, P. Gouze, V. Lagneau. Experimental characterization of coupled diffusion reaction mechanisms in low-permeability rocks: A case of two extremities of sulfate alkali minerals. Submitted to *Chemical Geology* the 15th of May 2018

Publication Under preparation

A. Rajyaguru, S. Savoye, C. Wittebroodt, E. L'Hopital, V. Dettileux, V. Lagneau: Barite precipitation in compacted illite & kaolinite: evidence of surface charge on new formed mineral, for *Environment Science & Technology*

A. Rajyaguru, O. Bildstein, S. Savoye, C. Wittebroodt, P. Arnoux, V. Dettileux, V.Lagneau: Numerical characterization of coupled diffusion reaction mechanisms in low-permeable micritic chalk, for *Computers & Geosciences*

International Conference

Oral presentation at the Goldschmidt conference in August 2017, Paris

A. Rajyaguru, S. Savoye, O. Bildstein, C. Wittebroodt, V. Dettileux, P. Arnoux, V. Lagneau: An experimental vs. numerical approach to determine capability of Archie's law to explain impact of evolving porosity on diffusivity

International Workshop

Reactive Transport in the Earth and Environmental Sciences in the 21st Century, October 2017, Amboise, France

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