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# Evaluation of the geochemical impact of biomethane and natural gas mix injection in sandstone aquifer storage

## sandstone aquifer storage

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### CONTEXT

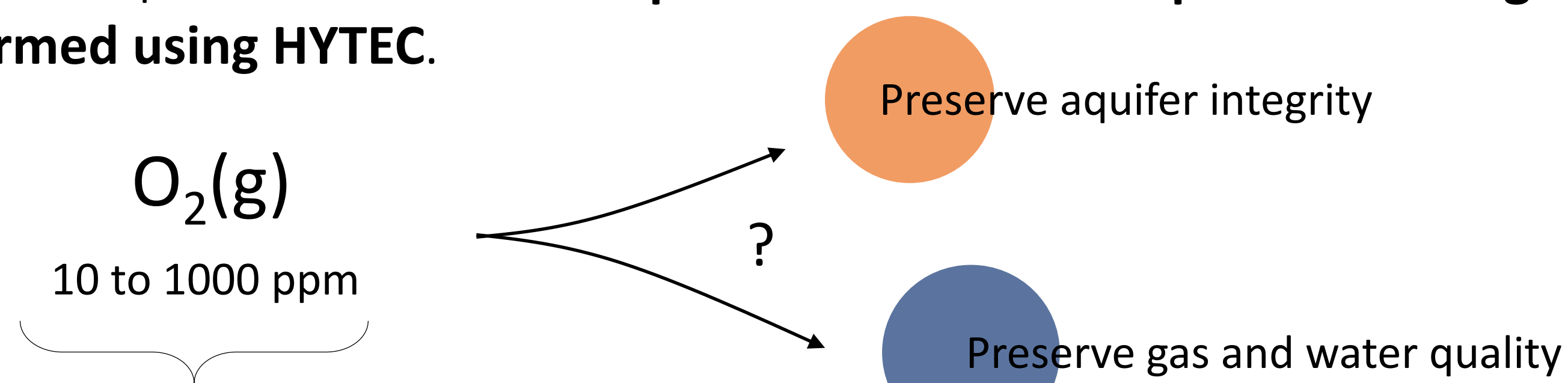
French gas storage capacity mainly relies on **underground gas storage in subsurface aquifers**. In the **REPowerEU plan 2022**, the European commission recently aimed to boost the actual biomethane production of 3 billion m<sup>3</sup> to 35 billion m<sup>3</sup> by 2030 in order to **replace 10% of natural gas (NG) importations**. In short term, biomethane should partly replace NG injected in aquifers.

Until **1%mol** (10 000 ppm) of **O<sub>2</sub>(g)** is measured in biomethane. Thus, **O<sub>2</sub>(g)** could temporarily exceed the 100 ppm French recommendation to preserve grid facilities.

**O<sub>2</sub>(g) injection** may lead to a rapid and important decrease of groundwater pH following pyrite oxidation present in anoxic aquifers triggering important changes in the quality of the stored gas and accelerating the deterioration of the reservoir's injectivity.

### OBJECTIVES

Evaluate the **geochemical impact** of varying oxygen concentration injected in subsurface aquifers. To do so **multiphase reactive transport modeling was performed using HYTEC**.



Injected gas mix (NG + biomethane) also containing **CO<sub>2</sub>, N<sub>2</sub> and H<sub>2</sub>S(g)**

The influence of petrophysical (temperature and pressure) as well as chemical characteristics was studied by modeling gas propagation and reactivity in **two different aquifer storage sites**.

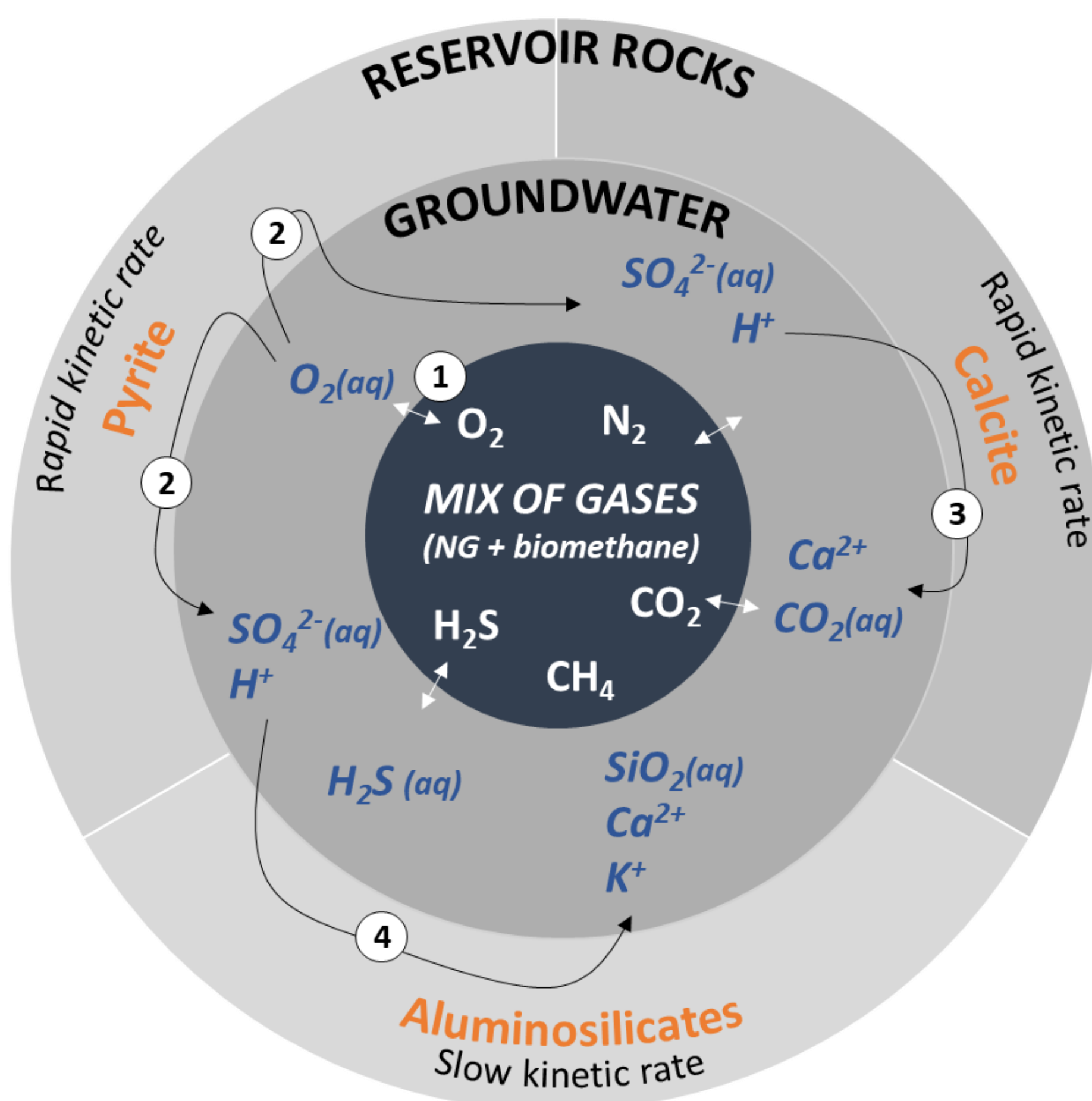
### SITES CHARACTERISTICS

		Mineralogical paragenesis of the reservoirs	
Site 1	Site 2	Minéral (wt. %)	
Feldspathic sandstone with <b>clay cement</b>	Sandstone with <b>calcite cement</b>	Site 1	Site 2 (Sin et al., 2023)
Pressure : <b>110 bars</b>	Pressure : <b>45 bars</b>	<b>Carbonates</b>	
Temperature : <b>55°C</b>	Temperature : <b>30°C</b>	Calcite	1      20
		<b>Alumino-silicates</b>	
		Quartz	63      63
		K-Feldspars	12.5      1
		Albite	9.5      0
		Anorthite	3      0
		Illite	5      10
		Kaolinite	5      5
		<b>Others</b>	
		Pyrite	1      1

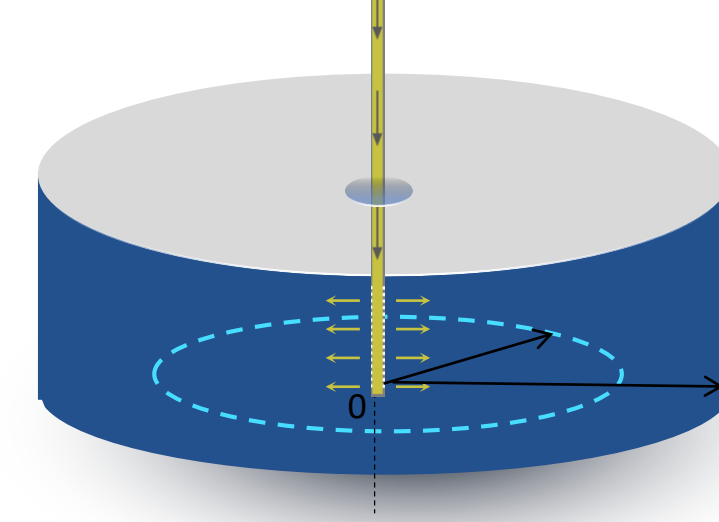
*General petrophysical characteristics*

### METHODOLOGY

Diagram of the modeled reactions following gas injection containing varying proportion of biomethane



GAS INJECTION



CH<sub>4</sub>(g) = 90.9 %  
N<sub>2</sub>(g) = 8 %  
CO<sub>2</sub>(g) = 1 %  
O<sub>2</sub>(g) = 0.001 → 0.1 %  
H<sub>2</sub>S(g) = 0.001 %

1D-radial model used for simulations

**HYTEC** to model fully coupled multiphase reactive transport with gas-water-rocks interactions (Sin et al., 2017)

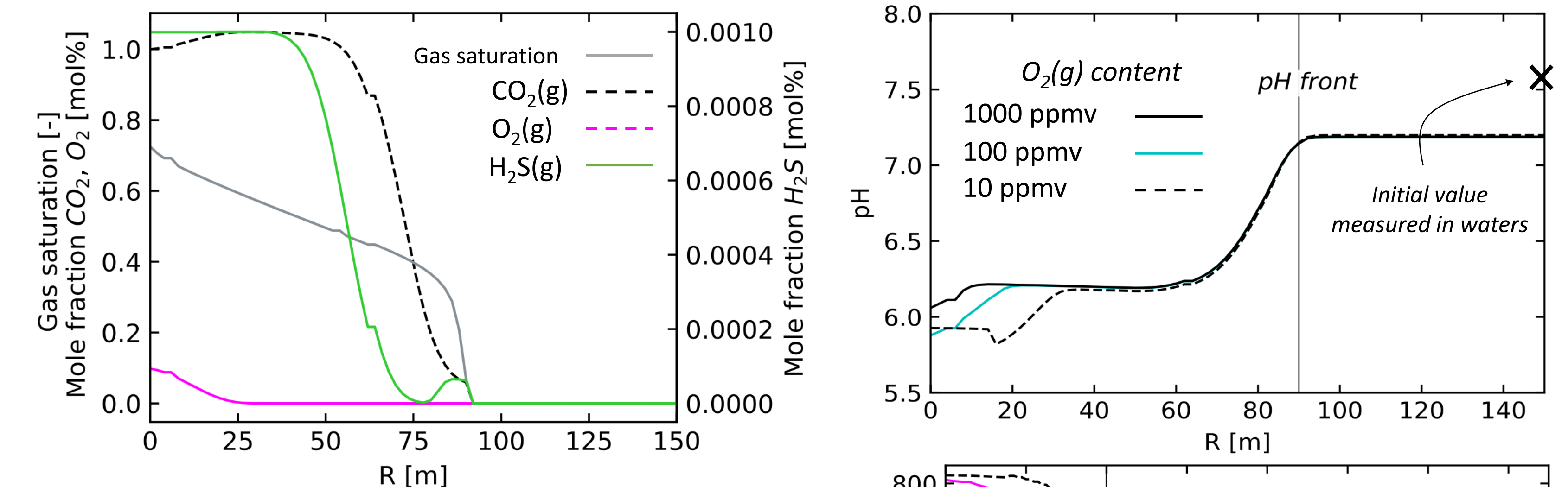
- **Kinetic** dissolution of minerals α Arrhenius law
- **B-dot** model to correct species activity
- Gas-water equilibrium with **Henry's law**, **Peng-Robinson EOS** and **Helgeson** volume correction.

### REFERENCES

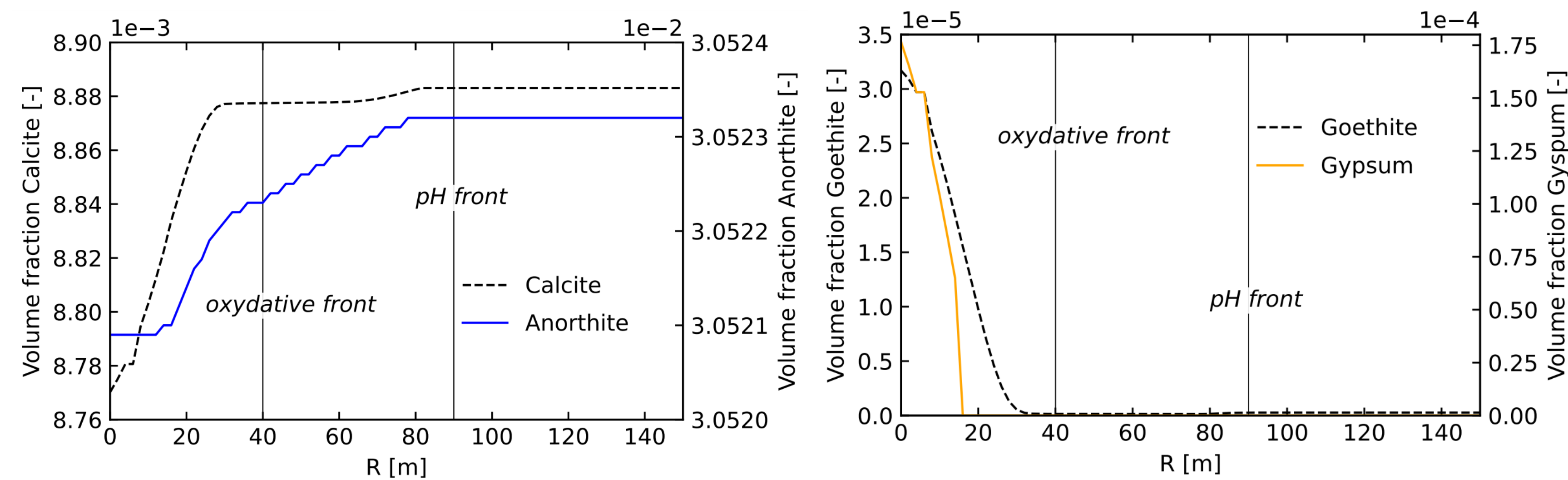
Sin, I., De Windt, L., Banc, C., Goblet, P., & Dequidt, D. (2023). *Science of The Total Environment*, 161657.

Sin, I., Lagneau, V., Corvisier, J. (2017) *Advances in Water Resources*, 100.

### RESULTS OBTAINED ON SITE 1 AFTER 1 YEAR OF INJECTION



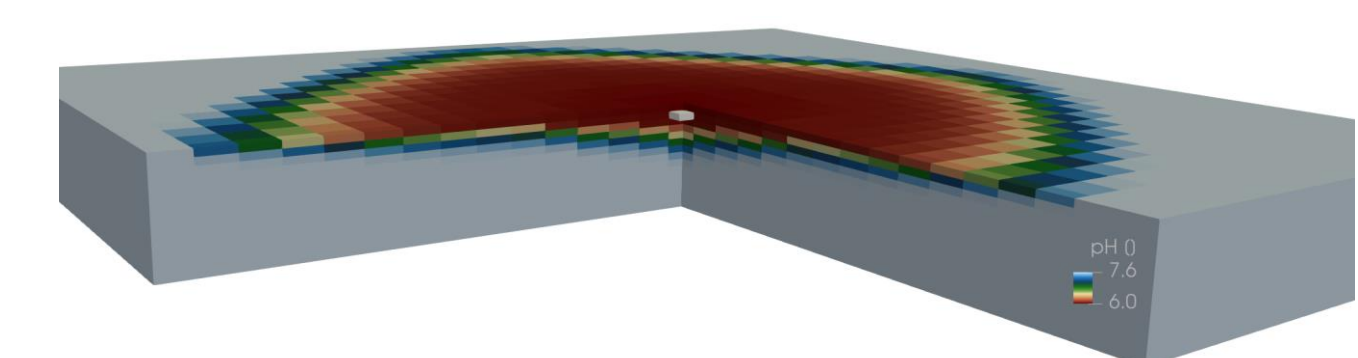
- The **pH front** expands **deeper** into the aquifer compare to oxydative front. Injected oxygen is **consumed by pyrite oxidation** at the vicinity of the injection well.
- The **pH is only slightly affected by oxygen** concentration thanks to aquifer buffering capacity.



- Aluminosilicates minerals and calcite reactivity are sufficient to **limit pH decrease** due to pyrite oxidation.
- Secondary minerals such as gypsum **precipitate** at the vicinity of gas injection well.

### CONCLUSIONS & PERSPECTIVES

- **CO<sub>2</sub>(g)** rather than O<sub>2</sub>(g) content is responsible for decreasing pH inside reservoirs.
- **Aquifers resilience** to oxygen injection is largely **site-dependent**. Oxygen content should be adapted depending on the characteristics of the considered aquifers.
- Despite the local deposition of gypsum in the vicinity of the injection, **no significant impact on the porosity** (<0.01%) of the reservoirs is predicted after one 1 year of constant injection.
- Further work need to be performed to **model injection and withdrawal cycle**.





## Evaluation of the geochemical impact of biomethane and natural gasmix injection in sandstone aquifer storage

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EU is seeking to rapidly replace part of the natural gas usually stored in deep saline aquifers by its renewable analogue, i.e. biomethane. While natural gas is depleted in oxygen, up to 10 000 ppm of O<sub>2</sub> can be measured in biomethane. But to date, the geochemical impact of the injection of a gas mix containing natural gas (thus CH<sub>4</sub> and CO<sub>2</sub>) and biomethane (thus oxygen) in deep saline aquifer has never been studied. The objective of this study is to evaluate the resilience of geological storage to oxygen injection and predict the evolution of the quality of the formation water, the gas plume and the rock formation. To do so, multiple injection scenarios with various gas quality were tested using multiphase reactive transport modeling. The gas mixtures were injected in two different deep saline aquifers in the Paris Basin (France) whose petrophysical and geochemical characteristics were reproduced during the simulation. Site 1 was a felspathic sandstone with clay cement and site 2 was a sandstone with clay-calcareous cement.

One dimensional radial flow model evidenced that in both sandstones, the injection of gas mix induced an acidification of the solution from pH~8 to pH~6. The injected oxygen originating from biomethane was quickly consumed during pyrite oxidation and contributed to the acidification of the formation water close to the injection point. However, the injection of 1% mol fraction of CO<sub>2</sub>(g) contained in the gas mix was the main acidification factor. Model demonstrated that the sandstones pH buffering capacity relied upon three geochemical processes (i) calcite dissolution, (ii) feldspar dissolution and (iii) clays dissolution and sorption capacity. These three pH-buffers were efficient for oxygen contents from 10 to 1000 ppm. Models predicted that the gas mix injection could induce minimal but long-term change in the nature of mineral phases but without significantly impacting the porosity. Overall, the gas quality was preserved in both sandstones. This result was evidenced with the modeling of entire gas injection and withdrawal scenario. CO<sub>2</sub>(g) exsolution, and to a lesser extent H<sub>2</sub>S(g) exsolution, could occur during these cycles. In addition, this study through modeling results concluded that the injection of biomethane did not significantly change gas-water-rocks interactions compared to those of natural gas injection. This study also evidenced that the detailed results were largely site-dependent.

