Experimental and modeling study of dihydrogen co-injection in deep aquifers used as UGS to identify key storage parameters

RINGS

EGATEC2024
The 6th European Gas Technology Conference

Jean Mura[1*], Magali Ranchou-Peyruse[1,2,3], Marion Guignard[2], Marion Ducousso[1,3], Pascale Sénéchal[4], Marie-Pierre Isaure[2], Peter Moonen[4], Guilhem Hoareau[5], Mateus De Souza Buriti[1], Marie Poulain[1,3], Anélia Petit[6], Pierre Chiquet[3,7], Guilhem Caumette[3,8], Pierre Cézac[1,3] and Anthony Ranchou-Peyruse[2,3]

[1] Université de Pau et des Pays de l'Adour, E2S UPPA, LaTEP, Pau, France
[2] Université de Pau et des Pays de l'Adour, E2S UPPA, CNRS, IPREM, Pau, France
[3] Joint Laboratory SEnGA, UPPA-E2S-Teréga, Pau, France
[4] Université de Pau et des Pays de l'Adour, E2S UPPA, CNRS, DMEX, Pau, France
[5] Université de Pau et des Pays de l'Adour, E2S UPPA, CNRS, TOTAL, LFCR, Pau, France
[6] STORENGY – Geosciences Department, Bois-Colombes, France
[7] Teréga – Geosciences Department, Pau, France
[8] Teréga – Environment Department, Pau, France

*Contact: jean.mura@univ-pau.fr

Background / Objectives

- The European Union aims to produce and import 20 million tons of renewable dihydrogen (H_2) by 2030 as part of it decarbonation strategy [1]. The share of H_2 in natural gas is expected to reach 2%. In many countries in Europe, natural gas storages capacities are mainly represented by underground gas storages (UGS) in deep aquifers (around 1000 m deep).

- H_2 behavior in aquifer still remains uncertain because of its potential consumption by autochthonous hydrogenotrophic microorganisms, the site dependency of these phenomena, and the physico-chemistry of the UGS (sulfate, carbonates, temperature, etc.).

- **Pilot experiments and modeling are needed to gather data and extrapolate results to the aquifer scale**
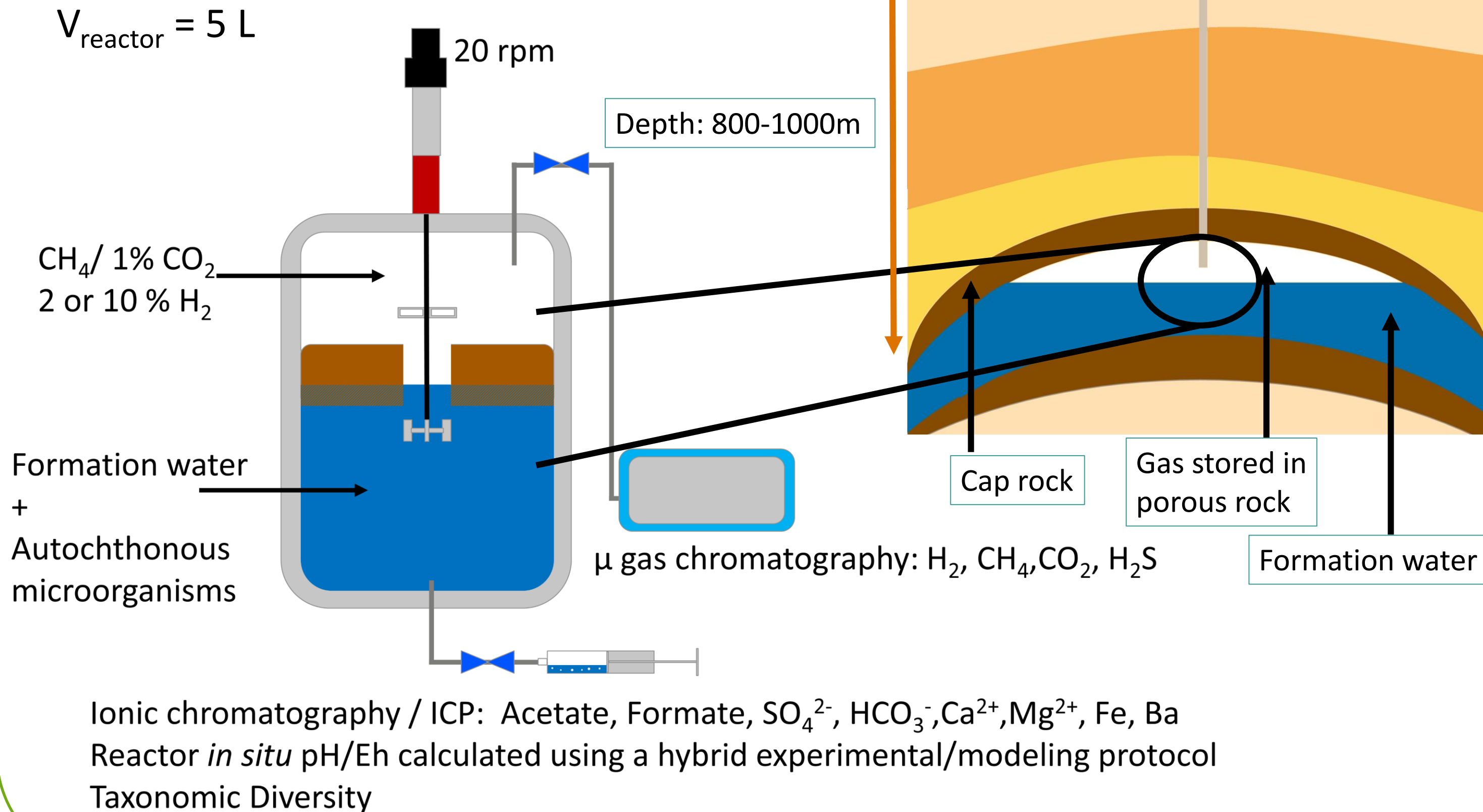
Experimental setup

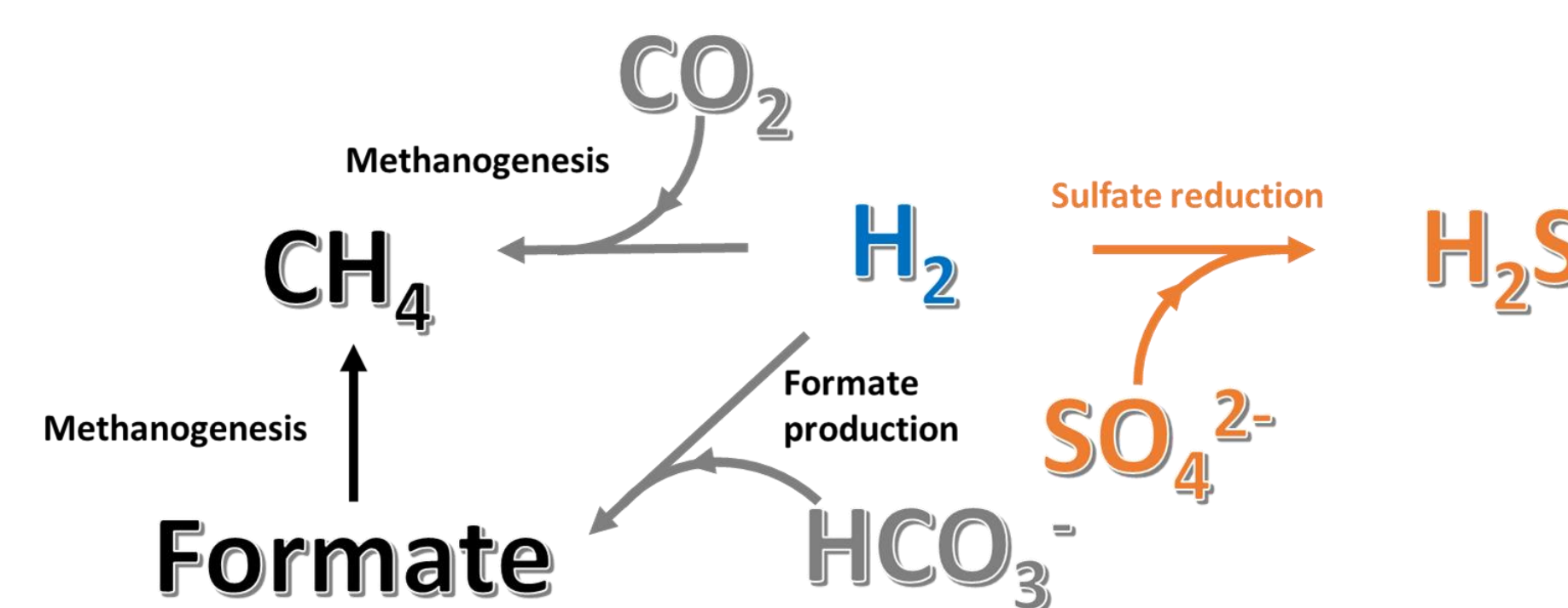- 3 aquifers simulated in a high-pressure reactor [2,3]
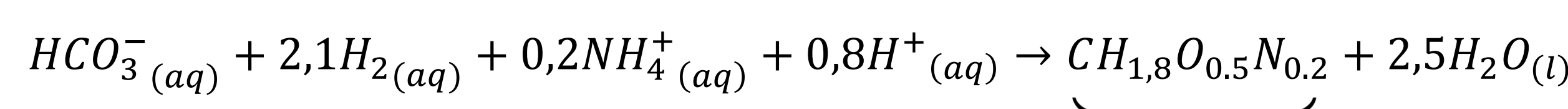
$P \in [85; 95]\ bar$
$T \in [35; 47]\ °C$
$V_{reactor} = 5\ L$

20 rpm

Depth: 800-1000m

CH_4/ 1% CO_2
2 or 10 % H_2

Formation water
+
Autochthonous microorganisms

Cap rock

Gas stored in porous rock

Formation water

μ gas chromatography: H_2, CH_4, CO_2, H_2S

Ionic chromatography / ICP: Acetate, Formate, SO_4^{2-}, HCO_3^-, Ca^{2+}, Mg^{2+}, Fe, Ba
Reactor *in situ* pH/Eh calculated using a hybrid experimental/modeling protocol
Taxonomic Diversity

Modeling approach

- 0D dynamic model of the reactor with PHREEQC v3

CO_2
Methanogenesis
CH_4 H_2 Sulfate reduction H_2S
Methanogenesis   Formate production SO_4^{2-}
Formate HCO_3^-

- Thermoddem database
- Liquid vapor equilibrium (Peng Robinson equation of state)
- Mineral dissolution/precipitation
- Microbial population evolution and respiration rate:
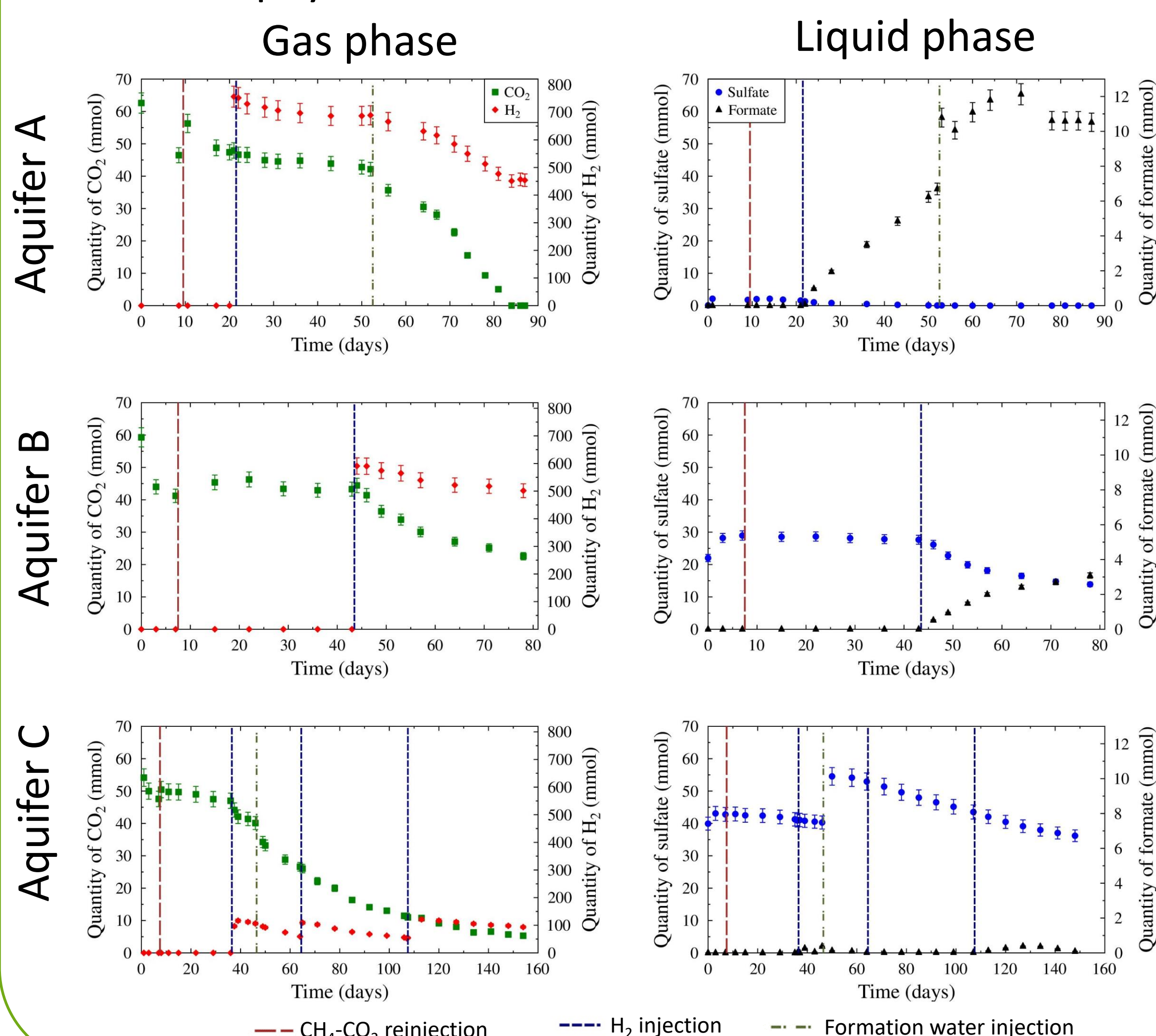 - Biochemical equation of growth:

$$HCO_3^-{}_{(aq)} + 2{,}1H_2{}_{(aq)} + 0{,}2NH_4^+{}_{(aq)} + 0{,}8H^+{}_{(aq)} \rightarrow CH_{1.8}O_{0.5}N_{0.2} + 2{,}5H_2O_{(l)}$$

Theoretical composition of a cell (noted B)

  - Kinetic law of growth and death

$$\frac{dB}{dt} = \mu_{max}F_{inhib}(S, pH, \dots)B - DB$$

With B : Biomass (mol.kg^{-1}), μ_{max}: growth rate in ideal conditions (s^{-1}), F_{inhib}: inhibition factor, S: substrate (mol.kg^{-1}), D: death rate function (s^{-1})

Results

- Main physicochemical results

Gas phase   Liquid phase

Aquifer A

Aquifer B

Aquifer C

— CH_4-CO_2 reinjection   --- H_2 injection   -·- Formation water injection
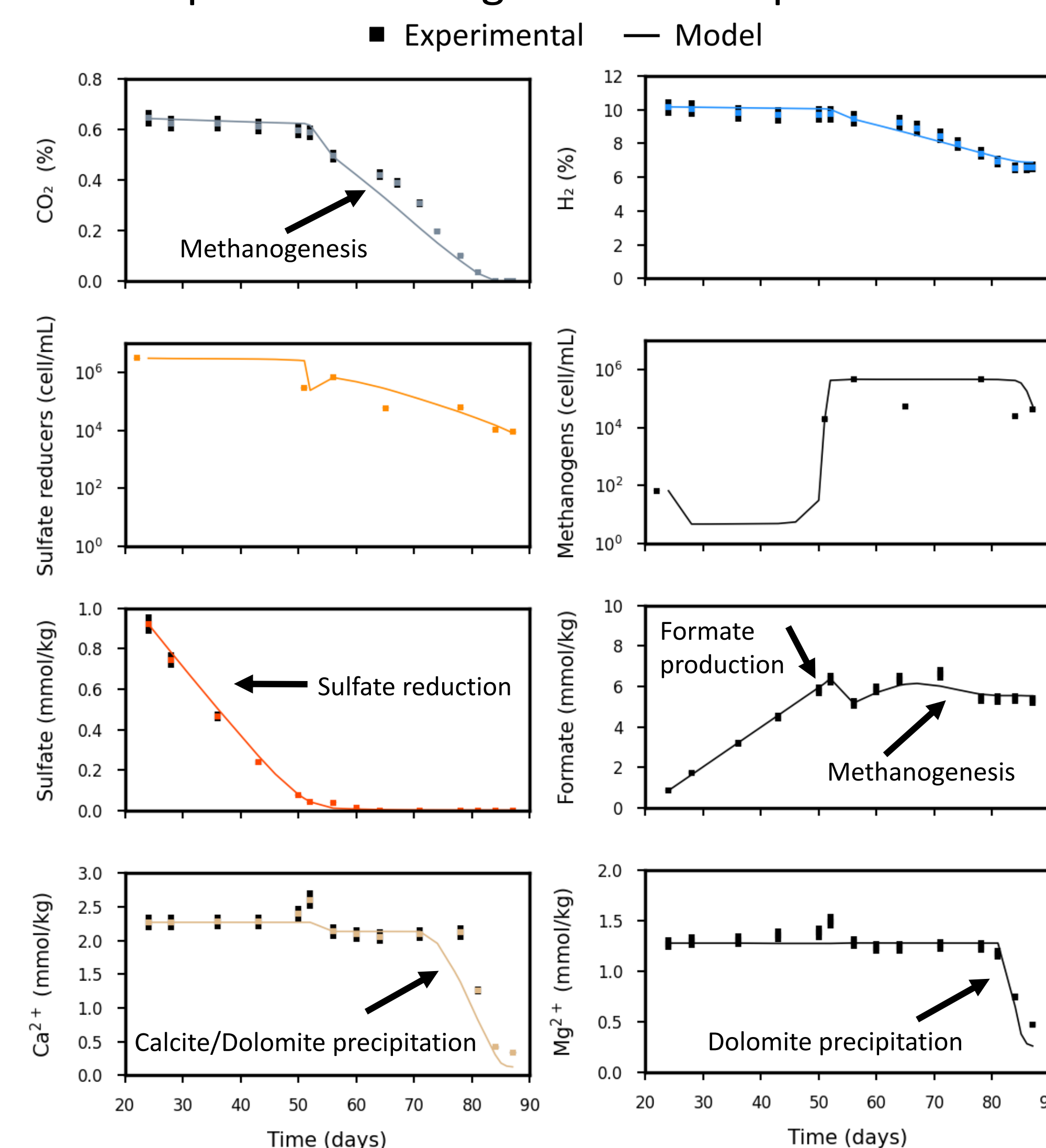
- Sulfate reduction happened in all experiments but didn't always lead to total sulfate consumption.

- When presents, methanogens activity was inhibited during sulfate reduction.

- Inhibition of methanogenesis and formate production by alkaline pH was observed while HCO_3^- and H_2 remained.

- Inhibition of sulfate reduction by lack of nutrients such as nitrogen or phosphorus is hypothesized.

- Attenuation of produced H_2S occurred, probably through iron-sulfide precipitation and adsorption (on the solid and on the reactor walls).

- Global impact on the rock was estimated to be minor, but precipitation/dissolution reactions are still expected.

- Example of modeling results for aquifer A

■ Experimental   — Model

Methanogenesis

Sulfate reduction

Calcite/Dolomite precipitation

Formate production

Methanogenesis

Dolomite precipitation

Conclusion / Perspectives

- **Aquifers with low electron acceptors (especially sulfate), nutrients availability, and pH buffering species (such as CO_2) should be favored to decrease H_2 loss and H_2S production.**

- **H_2 consumption could cease after an initial loss due to inhibition processes or due to the total consumption of electron acceptors.**

- **The model developed in this study should be coupled with large scale experiments to calibrate reservoir simulations in the aim of anticipating the extent of biogeochemical reactions at the aquifer scale.**

References

[1] European Commission. Hydrogen. Available at: https://energy.ec.europa.eu/topics/energy-systems-integration/hydrogen_en (Accessed May 15, 2024); [2] Haddad et al. (2022) Energy Environ. Sci., vol. 15, no. 8, pp. 3400–3415, doi: 10.1039/D2EE00765G; [3] J. Mura *et al. (2024) Int. J. Hydrog. Energy*, vol. 63, pp. 330–345, Apr. 2024, doi: 10.1016/j.ijhydene.2024.02.322.

UNIVERSITÉ DE PAU ET DES PAYS DE L'ADOUR   LaTEP FÉDÉRATION IPRA FR CNRS 2952   IPREM   cnrs   INSTITUT CARNOT ISIFoR   TERĒGA   storengy
Une société de ENGIE