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## CO<sub>2</sub>-Vadose and DEMO-CO<sub>2</sub> projects: two complementary projects about geochemical and geophysical monitoring during CO<sub>2</sub> leakage

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### Abstract

From a natural underground cavity and two locations situated in a well, three CO<sub>2</sub> injections have been designed and performed under controlled conditions in order to study its migration along the carbonate vadose zone and to test geochemical and geophysical techniques. After the understanding of the natural CO<sub>2</sub> dynamics and the establishment of a baseline, some numerical simulations have been performed to help to optimize the monitoring strategy (spatially and temporally). A mixture of CO<sub>2</sub> + noble gases has been injected and the results show that CO<sub>2</sub> subsurface leakage can be anticipated thanks to these inert chemical gases used as tracers (He + Kr). The geochemical and geophysical monitoring approaches are very complementary.

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**Keywords:** CO<sub>2</sub> ; noble gases ; inert gas ; geochemical and geophysical monitoring ; numerical simulations ; carbonate vadose zone

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### 1. Introduction

Capturing CO<sub>2</sub> and injecting it into deep underground geologic formation currently presents great interest, as it is one of the solution susceptible to decrease the accumulation of greenhouse gas into the atmosphere. Carbon capture and storage site (CCS) are subjected to specification relative to security towards population but also toward the environment. As consequences, it has been suggested that a maximum of 0.01 % per year of CO<sub>2</sub> leakage would be the maximum amount of CO<sub>2</sub> tolerated to leak from a CCS site [1, 2]. Therefore geochemical tools have to be developed to monitor and detect CO<sub>2</sub> leakage from the near surface and the shallow unsaturated zone (vadose zone). They must have the potential to precisely detect and quantify leaks, and discriminate natural signal from anthropogenic ones.

Monitoring CO<sub>2</sub> soil gas has been intensively studied for the last years [e.g. 3, 4, 5]. As example, [6] used particularly some gas tracers as precursors of the CO<sub>2</sub> leakage and to identify the origin of CO<sub>2</sub> detected. Those gas tracers are chosen to be chemically inert and with higher diffusion coefficient in order to be displaced faster than CO<sub>2</sub>. It demonstrates the need of having precursors to predict preferential path or heterogeneity of the massive. Indeed, [7, 8] and [9] presented studies with specific isotope signature such as δ<sup>13</sup>CO<sub>2</sub>. [10] presents isotopic tools as ways to determine predominant process during the leakage. They could monitor mixing processes and measure its associated chemical mechanism. However, each approach converges towards the same complex combination of geochemical mechanism occurring in the vadose zone [11]. In order to understand CO<sub>2</sub> dynamics during a potential leakage, it is then important to study different pattern of a release and the different factor susceptible to affect CO<sub>2</sub> mobility.

### 2. Method and/or theory with main results

Release experiment has been performed at Saint Emilion (France) in order to estimate the contribution of parameters in CO<sub>2</sub> dynamic. Thus, [12] injected at 9 m depth a CO<sub>2</sub> - rich gas mixture with associated gas tracers (He and Kr) from a cavity in the limestone and observed differences between the numerical model and field data. It emphasized the difficulty in building geological model relevant to diffuse migration of gases at decameter scale. The definition of the gas plume was successful.

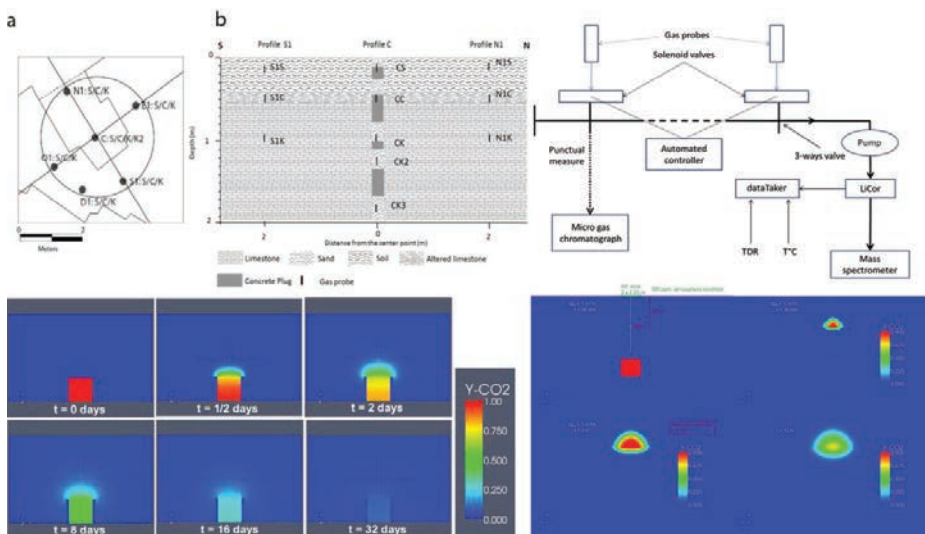


Fig. 1. (top) Map of gas probes implementation on the experimental site and schematic representation of the gas analyses device; (bottom) Numerical simulations of CO<sub>2</sub> leakage from the cavity (left) and from the CK4 point in the well (right).

However, differences in timing and amplitude were noticed, especially for the  $\text{CO}_2$  where the major difference was observed. Indeed, between numerical simulation and experiments, the difference concerning the  $\text{CO}_2$  is equal to 21 days for the peak arrival and 1400% for the maximum concentration observed for the probes near the injection point for example; whereas the differences concerning the He and Kr are not more than 3.7 days for the peak arrival and 600% for the concentration.

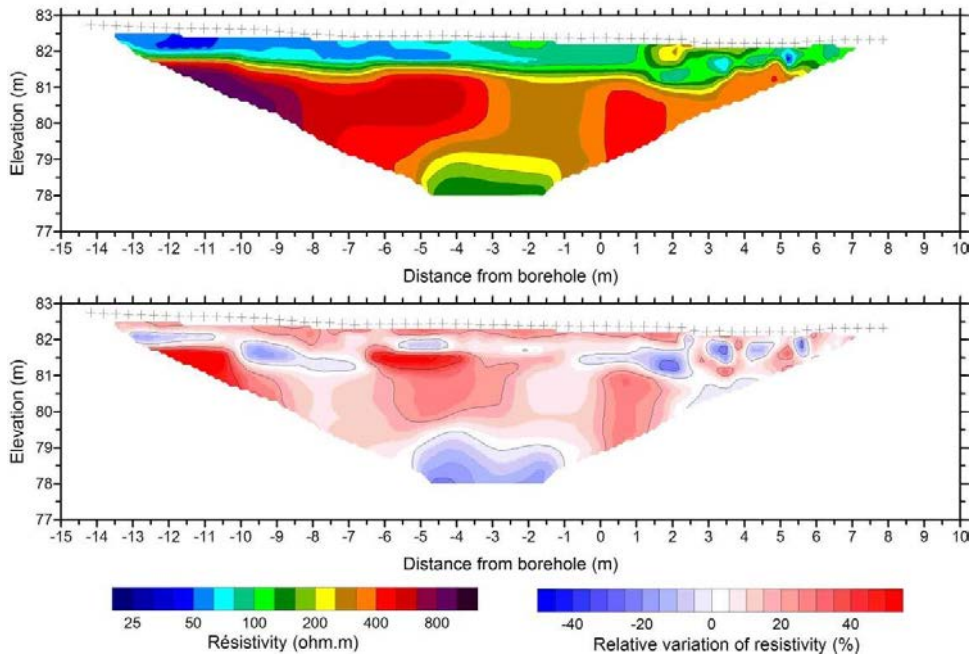


Fig. 2. Evolution of the resistivity traducing  $\text{CO}_2$  concentration variation within the Southwest-Northeast transect of the vadose zone within the limestone.

In the same pilot site, [11] performed a release experiment of enriched  $\text{CO}_2$  mixture with noble gases at 3.7 m depth but with a pressure of 2 bar, through a well. Both diffusive and convective transport were observed. It also showed that difference of peak arrival time of noble gases and  $\text{CO}_2$  could give more information about the dominance of either diffusion or advection processes. In the case of diffusion, displacement would be a function of molecular weight hence it could lead to presence of difference in molecular speed.

Nevertheless, there are few studies on the interaction occurring between the storage formation and the gas phase. [1] explained in their study that depending of the type of release, attenuation efficiency of the unsaturated zone could be drastically different. Higher pressures surrounding the source zone could cause more vertical migration of the  $\text{CO}_2$ . Interactions between  $\text{CO}_2$  and water phase can complicate attenuation dynamic. [6] explain that this is due to the inherent anisotropy of the system, but also to the lack of fundamental understandings on actual rock- $\text{CO}_2$ -water interaction.

### 3. Conclusions

The aim of this study is to understand the attenuation mechanism in the carbonate vadose zone and to estimate the processes of transport and reaction. This work describes the results of a release experiment in the shallow surface of the vadose zone. Then, the results obtained here are compared to a former leakage experiment which was also performed slightly deeper in the limestone with higher injection flow rate [11]. Spatial and temporal variation was continuously monitored thanks to a set of probes placed at different depth. Soil flux, water content and climatic parameters were also regularly measured. This work describes the methods and tools used to continuously monitor the CO<sub>2</sub> plume.

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