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## Surface and near surface geochemical surveying of a CO<sub>2</sub> injection pilot: application study to the French Pyrenean foreland (Rousse CCS pilot)

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## Introduction

Many CO<sub>2</sub> Capture and Storage (CCS) actions were undertaken in the last decade throughout the world. One of the key questions to address is how to ensure of the storage integrity. Geological, geophysical and geochemical methods all bring their own input to help getting a better knowledge of a CO<sub>2</sub> storage. Frequent data acquisitions prior to the beginning of the injection are mandatory to evaluate natural variations that exist either at the reservoir level but also at the whole sedimentary pile that overlies this reservoir. From a geochemical point of view, data acquisitions at depth are often restricted to dedicated boreholes but give invaluable results to help in the modelling scenarios. At the opposite, surface or near surface measurements are much easier to perform but their intrinsic spatial heterogeneity requires extensive work to get confidence in the database that results from the baseline measurements.

In that perspective, we present here part of the research work associated to the only onshore CO<sub>2</sub> injection pilot located in France, the TOTAL Rousse pilot (Lescanne *et al.*, 2011). Both baseline acquisitions and the monitoring during the injection phase are presented from a geochemical point of view. These acquisitions consisted in soil gas concentrations and soil gas fluxes measurements at selected sites with quarterly sampling frequency. They were complemented by geochemical surveying of a perched aquifer overlying the storage reservoir through a dedicated monitoring borehole of 85 m depth, located few dozen of meters from the injection well RSE-1. This monitoring included the regular sampling of the aquifer and of the gas species existing above the aquifer water level.

## Methods

As baseline data will serve as reference values all along the injection period but also during the post-operating phase, the use of robust methods is a requirement for CCS applications. Geochemical monitoring of gas species in near-surface environments has been demonstrated as a valuable tool in many applications and may be advantageously combined with water monitoring methods. The approach used here has been earlier deployed by the authors for the monitoring of natural analogues ( Gal *et al.*, 2011; Pokryszka *et al.*, 2010 ; Lafortune *et al.*, 2011).

In the present study the monitored gas species in soils are CO<sub>2</sub>, O<sub>2</sub>, CH<sub>4</sub>, <sup>222</sup>Rn and <sup>4</sup>He. The monitoring of gas flux at the soil/atmosphere interface was realized for CO<sub>2</sub> and CH<sub>4</sub>.

Water monitoring included the determination of water temperature, pH, electrical conductivity, dissolved oxygen content and redox potential. Free gas phase was monitored for CO<sub>2</sub>, O<sub>2</sub>, CH<sub>4</sub> and <sup>222</sup>Rn.

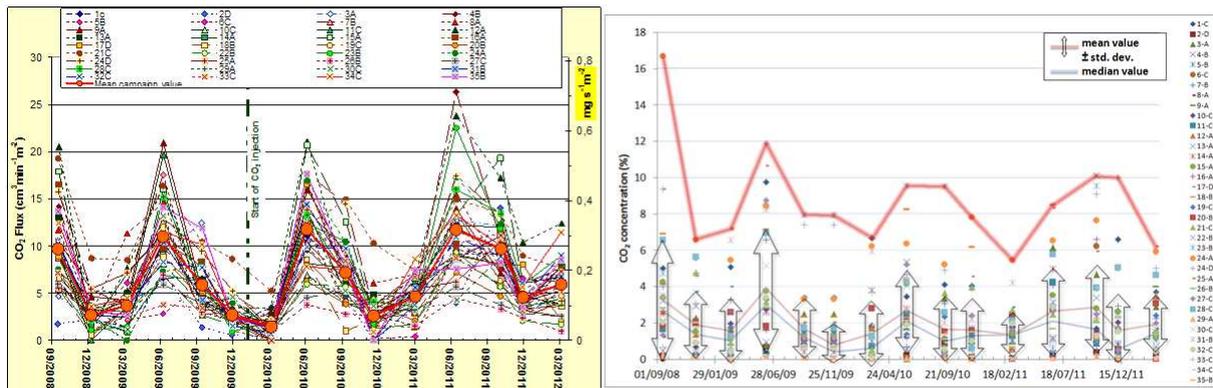
Soil gas data were gathered as point samples on a quarterly basis on 36 sampling stations, one station been equipped with a permanent <sup>222</sup>Rn monitoring system. To integrate the mismatch that may exist between soil gas methods (virtually no restriction in the number of monitoring points) and water monitoring methods (access only through boreholes), borehole monitoring was done at quarterly basis and also in between times. Quarterly measurements allowed to perform logging both in the water phase and in the air phase above water table, whereas continuous monitoring was done at selected depth. Samplings of the water and dissolved gas species were also done each 3 months at 3 to 4 different depths using appropriate nitrogen driven borehole sampler.

## Soil gas monitoring

First site characterisation was done in September 2008 on selected areas. The resulting dataset (112 measurements) was used to select 36 monitoring sites for perennial measurements of soil gas concentrations and fluxes at the soil/atmosphere interface. Five complementary baseline surveys were performed until the beginning of the injection in January 2010. At the stop of the injection in March

2013, a total of 17 distinct campaigns were done. The results of these acquisitions are shown in Figure 1 for the CO<sub>2</sub> specie.

Soil CO<sub>2</sub> fluxes were stated to be highly variable during baseline acquisitions between 0.1 and 21 cm<sup>3</sup>.min<sup>-1</sup>.m<sup>-2</sup>. Except variations linked to the seasonal cycle and local heterogeneities, there are no fundamental changes between baseline conditions and fluxes measured during the injection period. A similar statement can be done for CO<sub>2</sub> gas concentrations, the huge variation range – from near atmospheric concentrations to more than 10% – found during the first site characterization prevailing all along the acquisitions. Extreme values are often higher during summer times as a consequence of enhanced biological activity in soils.



**Figure 1** Evolution through time of: left: soil CO<sub>2</sub> flux at the soil/atmosphere interface; right: soil CO<sub>2</sub> concentration at 1 m depth.

Similar statements can be made using the other gas species. A reverse evolution of the O<sub>2</sub> gas concentrations, or the N<sub>2</sub>/O<sub>2</sub> ratio as determined on selected sampled analysed at lab, suggests that respiration processes in soils are the dominant phenomenon that explains the evolutions of soil gas concentrations and fluxes.

The CH<sub>4</sub> concentrations and fluxes are very low and may be considered as insignificant and only linked to near surface processes if detected (concentration of few ppm at maximum and flux lesser than 0.01 cm<sup>3</sup>.min<sup>-1</sup>.m<sup>-2</sup>). The evolution of the <sup>222</sup>Rn phase, that is in the present case carried by the CO<sub>2</sub> gas phase (Pearson's correlation coefficient > 0.76), is thus similar to that of CO<sub>2</sub> and does not allow to highlight preferential pathways of higher permeability. Last, <sup>4</sup>He concentrations only slightly fluctuate around the mean atmospheric concentration, suggesting that minor local enrichments are not related to deep seated processes.

All the data acquired during the baseline campaigns and subsequent ones range in a similar cluster and do not show any evidence of the influence of gas influx from the deep seated storage reservoir. Only natural surface processes and local heterogeneities (geologic, orographic, pedologic and climatic ones) are involved in their variations during time.

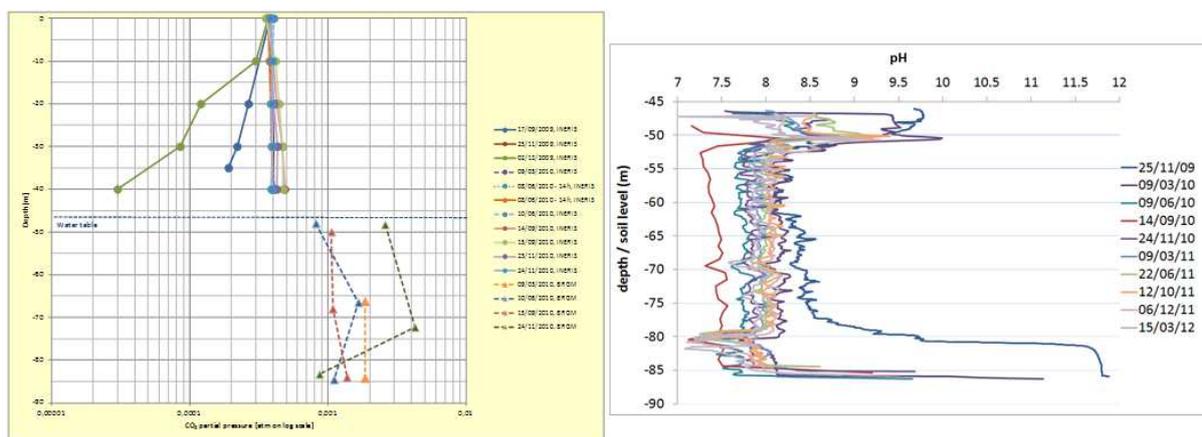
## Aquifer monitoring

A 85 m depth monitoring borehole has been drilled near the RSE-1 injection well of the CCS Rouseau pilot site. This borehole reaches shallow water table at about 45 meters depth. Geochemical monitoring in this borehole has been performed since November 2009 until March 2012. Measurements began before CO<sub>2</sub> injection starts. Aims are to better understand gas exchange dynamics between saturated and unsaturated zones and detect any potential anomalous gas migration towards surface.

Data collected over 4 years show that CO<sub>2</sub> concentration in the gas phase above water table in the monitoring well vary from atmospheric value (~400 ppm) up to 500 ppm (Figure 2). These variations have been understood as alternative phases of water degassing and atmospheric refill. According isotopic analysis, CO<sub>2</sub> degassing from underground water is soil gas dissolved in water during percolation of rainfalls.

The aquifer was investigated using a CTD probe that allowed to periodically investigate the vertical structure of the borehole, that is screened nearly all along the saturated zone. An example of the evolution of the pH is shown in Figure 2, showing a progressive homogenization of the water column along with time as the borehole was not purged prior to be implemented by the instruments. The waters have a Ca-Mg-HCO<sub>3</sub> type that is common in such geological environments and their compositions only slightly varied during time even if the water table level may experiment sudden changes as a consequence of strong rainfall events at the surface.

As the baseline records are shorter than the chronicle existing for soil gas data, getting information on the changes that may affect naturally the aquifer is less straightforward. Nevertheless no significant modification of the elements in solution neither of the vertical physico-chemical structure of the aquifer have been noticed thus suggesting that no event other than external forcing by rainfalls and percolating waters has occurred during the monitoring period. The integrity of the injection borehole in its shallower part is thus demonstrated.



**Figure 2** Monitoring of the surveillance borehole; left: evolution of CO<sub>2</sub> concentration as a function of depth in the 85 m depth monitoring borehole. Water level is around -45m; right: evolution of pH with depth in the aquifer, and comparison with time.

## Conclusions

A panel of different geochemical monitoring tools has been experimented and carried out by BRGM and INERIS at surface and sub-surface of the TOTAL Rouse CCS pilot site. Except variations linked to the seasonal cycle and local heterogeneities, no significant changes have been observed between baseline conditions and geochemical parameters measured during the injection period.

The methods carried out on the Rouse pilot site can be used to design surface and subsurface monitoring plans for other CO<sub>2</sub> storage projects that will be initiated in a future.

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