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Inducing a CO₂ leak into a shallow aquifer (CO₂FieldLab EUROGIA+ project): Monitoring the CO₂ plume in groundwaters

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Abstract

The CO₂ Field Lab project consisted in an injection of 1.7 tons of CO₂ at a 20 m depth within a shallow aquifer located in fluvio-glacial deposits. Baseline acquisitions, leakage monitoring and post-injection monitoring were performed in the water phase at different depths (5, 10 and 15 m).

Strong deviations of pH, specific conductance or alkalinity were observed and monitored at the horizons affected by the CO₂ intrusion. The complex distribution of waters in the ridge deposits not only induced water/rock interactions but also mixing of saline and fresh waters. This points out the complexity of natural environments and the challenge in identifying CO₂ leakage from deeper-seated reservoirs.

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1. Introduction

Carbon Capture and Storage (CCS) sites are often located at depths greater than 800 m in order to store CO₂ as a supercritical fluid. Overlying aquifers have to be monitored in order to ensure that storage integrity is maintained all along the storage life, including the post-injection phase. This monitoring is of crucial importance [1] but until now few experiments have been done to strengthen the confidence on

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geochemical monitoring methods and their ability to detect changes linked to CO₂ leakage. Apart from numerous studies of natural analogues, most of the recent experiments concerned near surface environments such as the Zero Emission Research and Technology centre [2].

The first phase of the CO₂ Field Lab project is part of these near surface experiments but with a deeper injection depth of 20 m. The project was designed to gather technologies that will enable the detection of possible CO₂ leakage at the earliest possible stage, in view of using these technologies at the industrial scale and of strengthening the confidence people may have into CCS operations. Soil gas and water monitoring were performed but, here, emphasize is on the sole water monitoring.

The first phase of the CO₂ Field Lab project consisted in an injection of 1.7 tons of CO₂ in a shallow aquifer emplaced in fluvio-glacial deposits in September 2011. The site was characterized in 2010 through the drilling of an appraisal well and subsequent geological, geophysical and geochemical investigations [3]. The September 2011 leakage experiment was divided into: i) baseline acquisitions until the start of the injection (on the 7th of September 10:30), ii) leakage monitoring during the 6 days of injection, and iii) post-injection monitoring (until the 19 September).

Deposits of the Svelvik ridge (50 km southwest of Oslo) are heterogeneous glacio-fluvial and glacio-marine deposits from the Holocene ice recession in Southern Norway. Material excavation due to sand exploitation has lowered the ridge level down to few meters above sea level. As a consequence, the water table is at a 1 m depth under ground level. Waters are far from homogeneous as brackish waters, groundwaters and freshwaters can alternate over short distances either laterally or vertically.

The geochemical monitoring of the aquifer compartment is based on boreholes investigating different depths (Fig. 1). Shallower ones were drilled in 2011 in the main leakage area and extend only down to 2 m depth. Wells drilled in 2010 for hydraulic tests (5 m depth) were used for monitoring the water body at a greater geographical scale, as they were all located northward from the leakage site. Last, water was also collected at 5, 10 and 15 m depths in the leakage area thanks to the coupling of sampling ports with electrical cables used for electric tomography (ALERT system from BGS). Four ALERT wells were used for that purpose.

To our knowledge, the CO₂ Field Lab leakage experiment is one of the deepest ever performed. A greater interaction of the injected gas with water or at least a greater path length from the injection point to the surface is then expected when compared to shallower leakage experiments. Duration of the experiment is relatively short as this is dependent from the high permeability of sandy formations that favors gas migration. Amounts of CO₂ injected are comparable to those used, for example, for the ZERT experiment, the difference being a gradual increase in the injection rate for the CO₂ Field Lab experiment (other leakage scenarios using fixed injection rates).

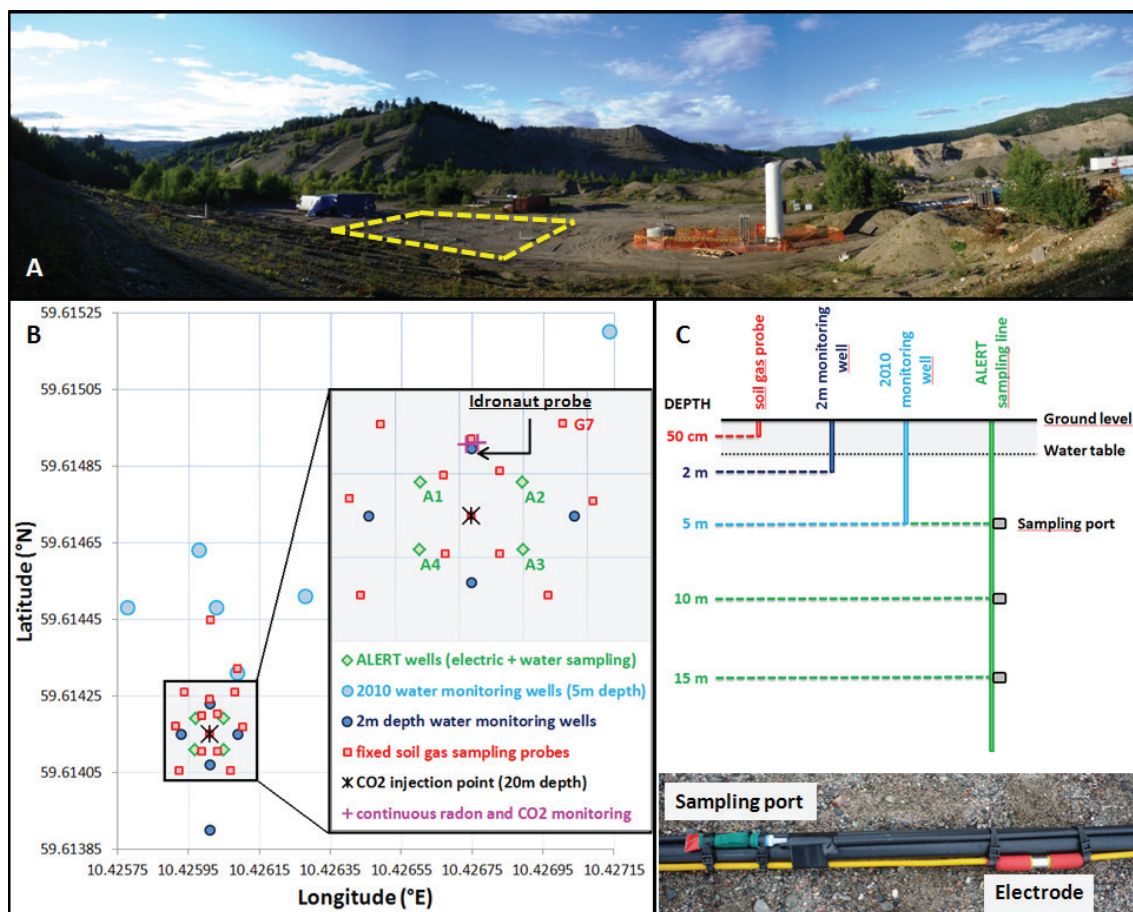


Fig. 1. (A) view of the CO2FieldLab test site; (B) location of the monitoring boreholes; (C) depths investigated for the water monitoring.

2. Results and discussion

2.1. Methods

Geochemical investigations were divided into point monitoring and continuous monitoring. Point monitoring was performed through a low flow pumping, using peristaltic pumps. Direct quantifications of pH, T, redox potential (ORP), specific conductance (EC) and dissolved O₂ (DO) were done in order to determine if sampling was required or not. High frequency sampling concerned alkalinity (expressed as the HCO₃ content) and dissolved ion contents (major and trace elements). At lower frequency, samples were also collected for subsequent isotope analyses ($\delta^{18}\text{O}_{\text{H}_2\text{O}}$, $\delta\text{D}_{\text{H}_2\text{O}}$, $\delta^{13}\text{C}_{\text{Total Dissolved Carbon}}$). Continuous monitoring of pH, T, ORP, EC and DO has been performed in a 2 m depth borehole by using a dedicated multi-parameter probe (Idronaut 303). Further emphasize on chemistry and isotopes may be found in [4].

2.2. Leakage monitoring – point methods

Leakage monitoring, even in near surface environments, may be subject to unwanted events. In the present case, the injection was planned to be performed at 20 m depth. Consequently the CO₂ breakthrough was expected to first occur at the deeper levels (15 m) and to progressively migrate along bedding of geological formations towards the surface. Deviations from this plan were stated 1 day after the injection started with leakage occurring near the injection wellhead, and 2 days later with leakage occurring northward from the main monitoring area (Fig. 1). These leakages were evidenced by soil flux and soil gas measurements. The northward leakage was contemporaneous with CO₂ bubbling in 2010 monitoring wells. The consequence was an adaptation of the monitoring strategy that was formerly focused on the deepest horizons rather than on the shallow ones.

Apart from that bias which highlights the complexity of geological environments even in near surface compartments, and thereof the care that shall prevail for establishing and setting a monitoring strategy for CCS sites, leakage monitoring was successful. The monitoring of physico-chemical parameters allowed registering the following changes, associated to the CO₂ plume migration (Fig. 2):

- pH decrease of a maximum of 2 units;
- Rise of EC by a factor 2 to 3;
- Rise of the alkalinity by a factor as high as 6.
- Changes were also stated for ORP, with the occurrence of less oxidized waters, and for DO, this last parameter being affected by the sampling process.

These intrinsic changes were associated with changes of the dissolved gas contents, the most affected level (10 m depth) being close to saturation with respect to CO₂ during the paroxysm of the leakage.

The monitored area was not homogeneously affected by the leakage in lien with the deviations of the injection from the initial plans. Spatial distribution of geochemical parameters was not only linked to the CO₂ injection but also to the mixing between different types of waters. Fig. 3 illustrates that by comparing pH values, affected by the dilution of CO₂ in water, and the EC values, affected by the desorption of analytes – consequence of the leakage – and also by the presence of waters of different chemistries.

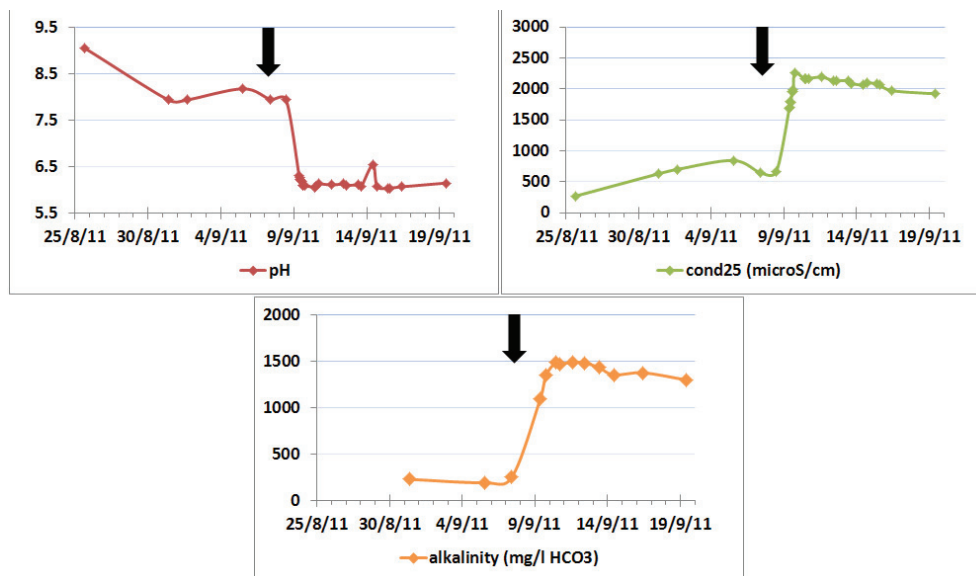


Fig. 2. Changes of pH, specific conductance (cond25) and alkalinity at 10 m depth; the arrow indicate the beginning of the injection.

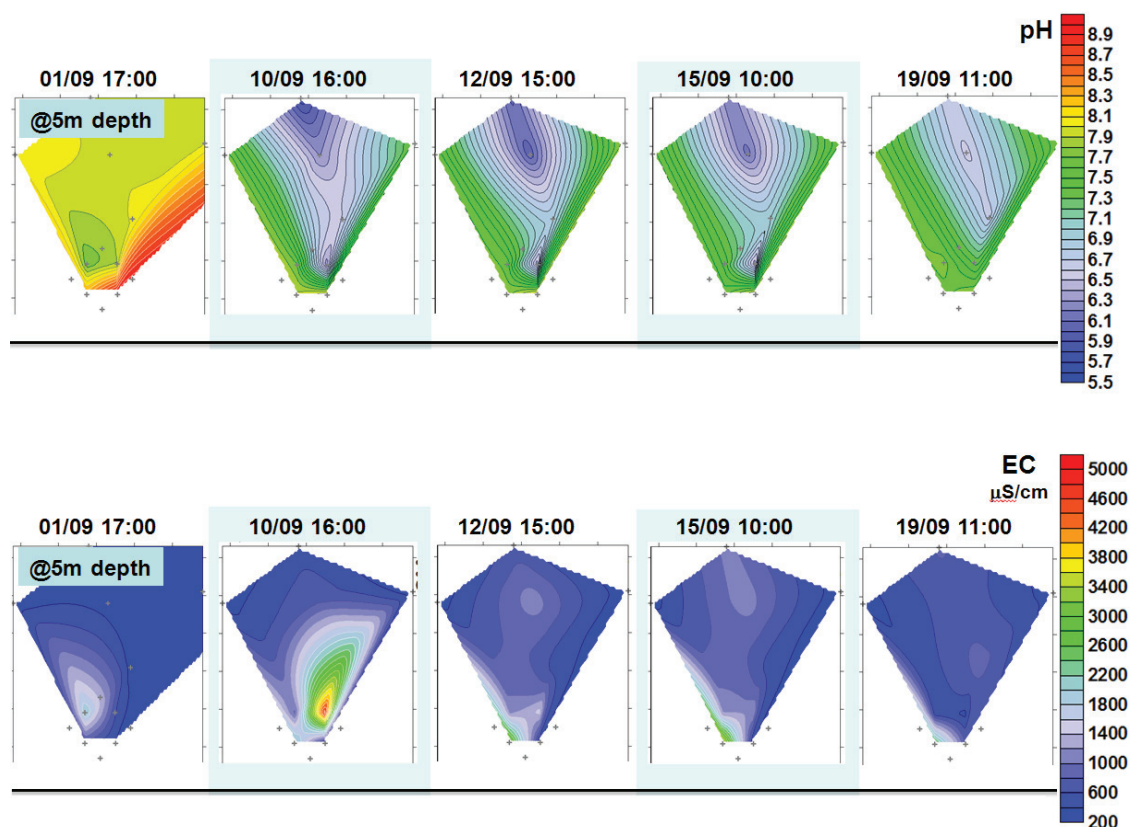


Fig. 3. Top: pH variations at 5 m depth prior to the leakage experiment (01/09) and during the experiment; bottom: variations of the specific conductance (EC) for the same time period; dots refer to location of boreholes as indicated in Fig. 1.

The role of mixing phenomena and changes linked to the presence of CO_2 can be better evaluated using contents of dissolved elements. For example, Cl is a conservative element whose concentration may not change when CO_2 is injected into the aquifer, as Cl contents in the rock matrix are very low. This relation is presented in Fig. 4A for the borehole the most sensible to the leakage event. Two trends are clearly distinguishable:

- The 5 m and 15 m depth levels show hysteresis-like cycles, the rise of HCO_3 concentrations being associated with a decrease, in variable amount, of the Cl concentrations;
- The 10 m depth level, at the opposite, does not experience changes in its Cl contents during the CO_2 leakage.

This suggests that waters located at 10 m depth were very sensitive to the leakage event and poorly sensitive to mixing with other potential end-members. The situation is clearly different for upper and lower horizons, and can be further investigated by referring to Fig. 4B. These levels plot along a mixing line with local seawater and freshwater end-members. There was a progressive dilution of the saline (15 m depth) or brackish (5 m depth) waters as the experiment went on, thus suggesting an intrusion of diluted waters possibly from the 10 m depth level. Upward migration of freshwater from 10 m to 5 m depth may be a consequence of the injection process, but is not a consequence of the dilution of CO_2 in water which would have implied the opposite effect. Similarly, the water movements caused by the

injection, which is thought to have taken place somewhere between 10 and 15 m depth, may also have induced a downward freshwater migration coupled to a slight dilution of the saline waters present at 15 m depth.

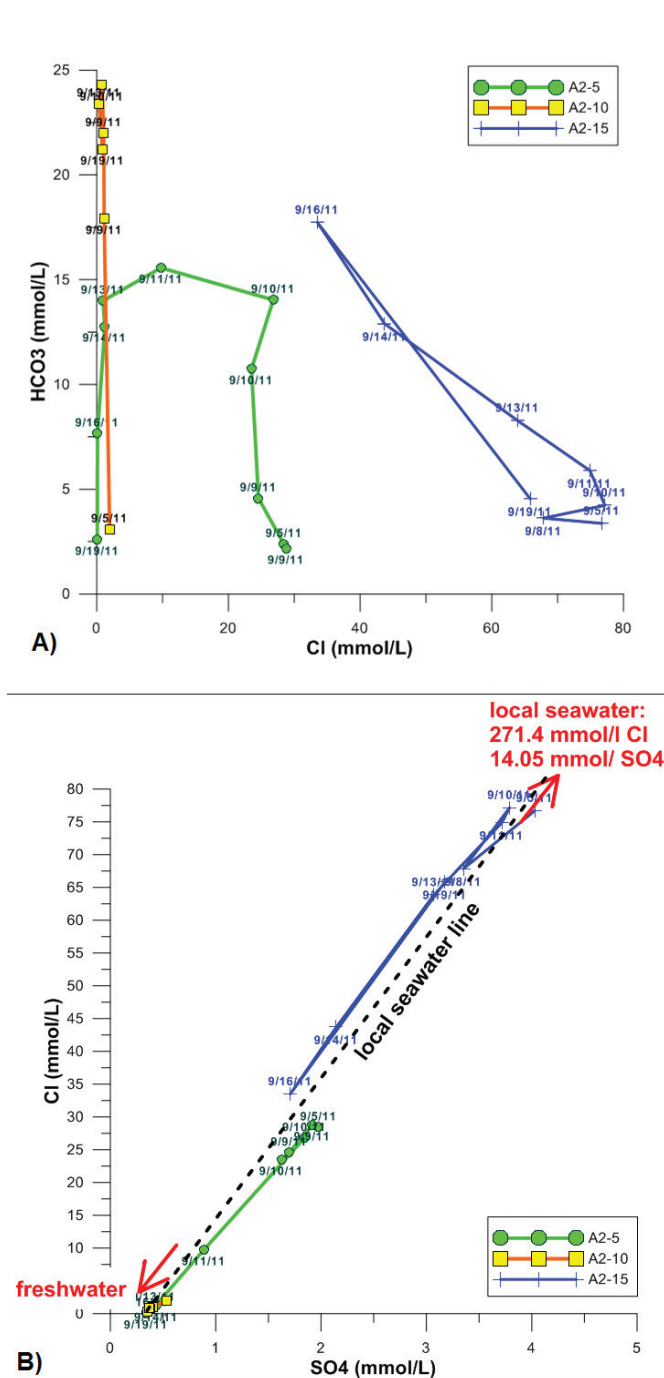


Fig. 4. (A): relation between Cl and HCO_3 contents; (B): relation between Cl and SO_4 .

The relationship between silica and calcium (Fig. 5) further suggests that the 15 m depth level was slightly affected by the leakage, as Ca contents did not vary significantly during the experiment. Saturation indexes, calculated for the 10 m depth level, where changes were mainly caused by the CO₂ intrusion, suggest moreover that carbonate species were more prone to be affected by the dissolution processes. If this had occurred at 15 m depth, then Ca concentrations should have risen much more. Accordingly, the enrichment in silica is rather a consequence of the mixing with freshwaters, enriched in silica, than a consequence of the CO₂ injection.

The situation is different for upper levels (Fig. 5). The 10 m depth level has experienced strong Ca enrichment in relation with the leakage and may also have been affected by the weathering of silicate minerals such as clays or feldspars. Last, the 5 m depth level was preferentially impacted by carbonate weathering that led to the release of calcium in the water.

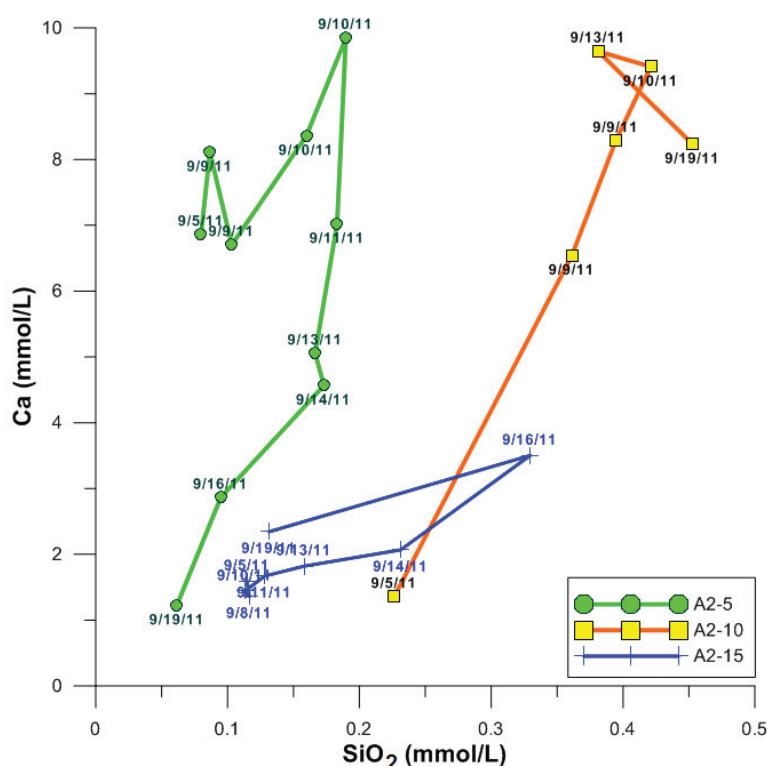


Fig. 5. Relation between Ca and silica contents.

Monitoring of the stable isotopes compositions of oxygen ($\delta^{18}\text{O}$) and hydrogen (δD) comforts the previous conclusions (Fig. 6). There was little scattering of the data for 10 m depth level suggesting the absence of mixing with other waters. When CO₂ is interacting with H₂O, an oxygen isotope equilibrium between the two molecules is expected (i.e. $\delta^{18}\text{O}$ from water and CO₂ will reach isotope equilibrium while δD will not be affected). But here, the CO₂ leakage did not seem to induce a significant shift on the

sole oxygen isotopes as both $\delta^{18}\text{O}$ and δD varied with time along a line parallel to the MWL. Only one sample collected at the paroxysm of the leakage (label 13/9/11 on Fig. 6) showed significant an ^{18}O depletion that may be related to the leakage. The small changes during the experiment may be related to insufficient equilibration time between the CO_2 -water mixture and the hosts' rocks. The situation for 5 m and 15 m depth levels is easier to describe, as the only effect is the mixing between different waters, one end-member being the 10 m depth level. Data from both levels perfectly align along a regression line whose slope is close to the meteoric water line thus defining a local water line rather than changes linked to the experiment.

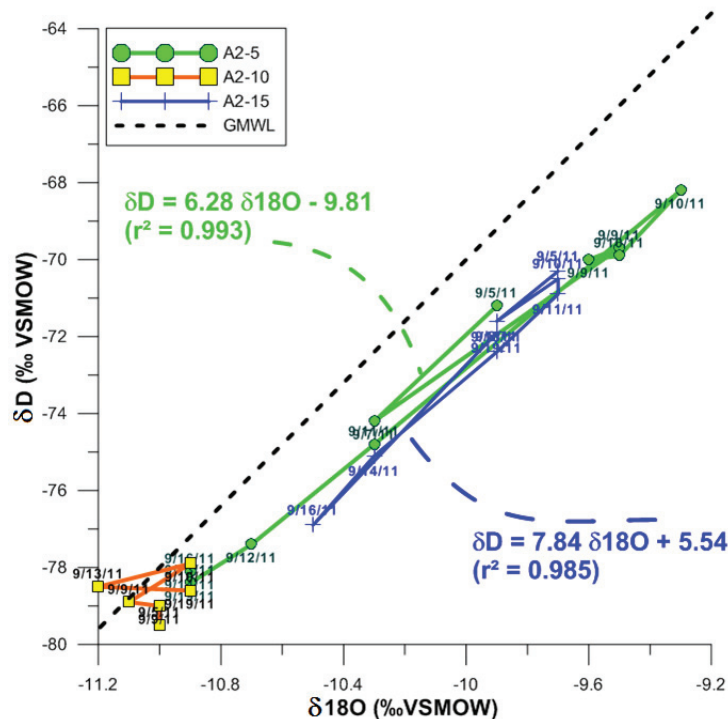


Fig. 6. Oxygen-Hydrogen stable isotopes relation; the Global Meteoric Water Line (GMWL) is indicated for reference.

2.3. Leakage monitoring – continuous methods

Apart from point monitoring, continuous acquisitions were performed at one well located at the northern border of the leakage area (Fig. 1). As the scenario of the leakage differed from the expected one, no chemical changes were monitored at 2 m depth in this borehole. Nevertheless intriguing changes of the physico-chemical parameters were monitored (Fig. 7), as well as changes on the carbon isotope compositions ($\delta^{13}\text{C}$; Fig. 8) measured in the air above the surface at the right of the borehole.

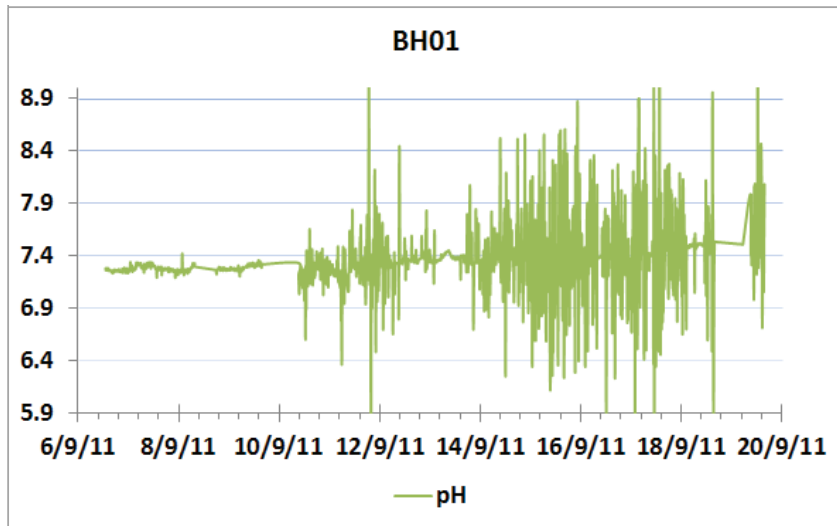


Fig. 7. Variation of the water pH in a continuously monitored borehole (2 m depth).

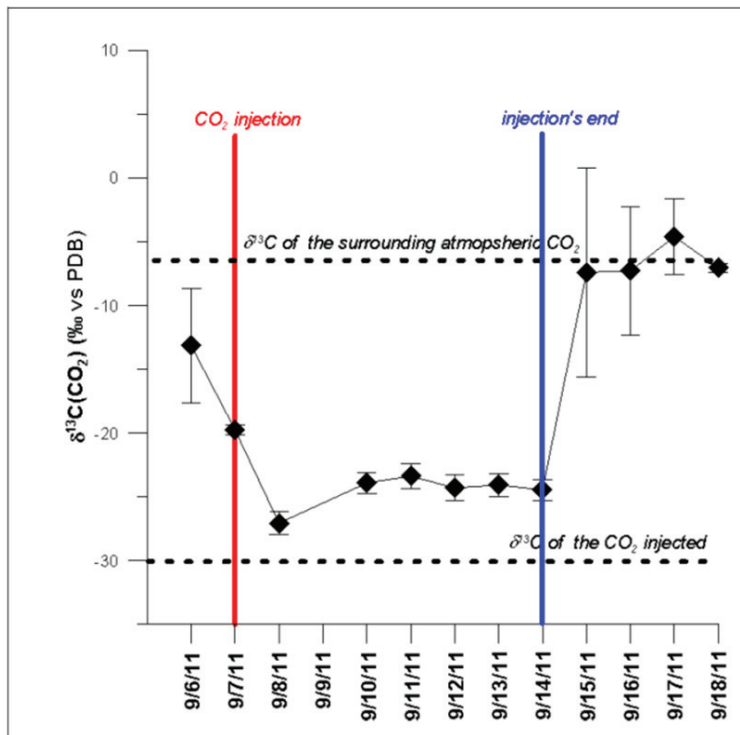


Fig. 8. Variation of the carbon isotope compositions of the CO_2 gas phase at the surface of a continuously monitored borehole.

Even if it was not possible to identify changes linked to a CO₂ breakthrough in this 2 m depth borehole, *e.g.* the pH value remained at the same mean value all along the experiment (Fig. 7), noisy records of pH (also observed for the EC or the temperature) were observed when the leakage started. Similarly, no changes in CO₂ concentrations were observed in this borehole, which remained at near atmospheric concentration, but strong shifts of the corresponding carbon isotope compositions were monitored (Fig. 8). A ¹³C depletion as great as -15‰ was observed right after the injection. This is consistent with a binary mixing between i) the atmospheric CO₂ ($\delta^{13}\text{C}$ measured around -12‰ prior to the injection) and ii) the CO₂ from the injection tank ($\delta^{13}\text{C}=-30\text{‰}$). This low $\delta^{13}\text{C}$ remained as long as CO₂ was injected, and quickly vanished as the injection stopped (*i.e.* $\delta^{13}\text{C}$ are quickly getting back to the atmospheric CO₂ values). Variations of the $\delta^{18}\text{O}$ identify an isotope equilibrium reached between the oxygen of the aquifer water and the oxygen of the CO₂ injected. The origin of such variations remains unclear, as no evidence of CO₂ intrusion can be brought by regular monitoring.

3. Conclusion

A 6-days-long CO₂ injection was performed within heterogeneous fluvio-glacial deposits from a Norwegian fjord. Pre-injection monitoring allowed the characterisation of the environmental conditions prevailing before the experiment. Post-injection monitoring was performed to evaluate the duration of the changes induced by the leakage. Waters were monitored at 4 distinct depths. The consequences of the CO₂ injection were highly variable both in time and in space. The injection of CO₂ not only induced pH or alkalinity changes but also chemical variations that were related to the interaction of the injected CO₂ with the aquifer and partly to the mixing of freshwaters and saline waters that form the saturated horizons of the ridge.

The complex situation described by this short leakage experiment is important for greater CCS projects. First, even if appraisal well drilling and geological, geophysical, hydrological and geochemical investigations were performed prior to the injection, the complexity of natural deposits can be underestimated, thus leading to injection scenarios differing from what was expected. Second, considering storage operations at greater depths, MMV (measurement, monitoring and verification) scenarios must take into account these possible deviations and plan the use of flexible monitoring methods that have investigations radii greater than those initially anticipated.

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