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GHGT-12

CO₂ injection in a coal seam – Insights from the European CARBOLAB project with focus on water geochemical monitoring

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Abstract

The CARBOLAB project is funded by the European program RFCS (Research Fund for Coal and Steel). It gathers six partners from Spain, France and Poland. One of the main tasks of this project is to perform in-situ CO₂ injection in a coal seam located at 464 m depth in the Montsacro pit, Asturias, Spain. Injection of CO₂ in coal seams is one of the options for climate change mitigation but it presents lots of uncertainties and technical difficulties. Therefore work is needed to better constraint the processes especially the adsorption of CO₂ and the subsequent desorption of CH₄. This work consists in experimental laboratory work, modelling aspects and real tests. This paper is focused on this last topic and more specifically on the water monitoring aspects. Complementary investigations on the gas phase can be found in other works presented by Lafortune *et al.* in this conference.

In-situ injection of CO₂ has been performed in July 2013 in one selected coal seam. The CO₂ is injected directly in the vein and monitoring boreholes are located at the wall and in the surrounding sedimentary formations. The aim is to get a detailed and representative overview of the consequences of the CO₂ injection. Here the focus is on the geochemical monitoring.

Baseline acquisitions have been performed in 2012 and early 2013. This allowed to define a strategy mainly focused on the monitoring of the saturated zone albeit the site was initially thought to be drier. Acquisitions in the unsaturated zone (free gas phase) have also been done but in a less extent.

Chemical logging of water saturated boreholes (GC1S and GC3N, parallel to the seam) revealed, even at short distance, the existence of different water masses from one side of the coal vein to the other. This is mainly linked to a difference in the amount of dissolved solids in water. During the injection experiment, no noticeable deviations from the baseline values were found for GC1S borehole. At the opposite, the bottom of GC3N experienced strong changes: decrease of more than 2.5 pH units, rise of the electrical conductivity (+29%) and of the alkalinity (+23%).

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Breakthrough in GC3N cannot be timed precisely as work in the gallery was not continuous, but occurred sometime in between the 05 and the 08 of July (the injection started on the 03 July).

The changes on the physico-chemical parameters are linked to changes in concentrations for some major elements and some trace elements. Ca and Mg concentrations were progressively enriched in the water phase whereas species like Na or K remained at stable levels in lien with the geology of the rock formations. Trace elements such as Sr, Ni, Mn, Zn or Ba have experienced rises of their concentrations by factors of 2 to 10. Changes in dissolved gas concentrations were also found for GC3N borehole, the enrichment in CO₂ being prominent (2 orders of magnitude enrichment).

The monitoring of the free gas phase suggests that the main part of the CO₂ has not been injected in the coal seam but rather in the aquifer, on the side of GC3N borehole. Moreover, a strong leakage has been noticed close to the injection borehole wellhead, quantification of the flux with appropriate flux chamber leading to leakage rates of around 20 to 70 l/h.

Carbon isotope ratios may suggest a little influence of the seepage from the wellhead in the atmosphere of the gallery, a depletion of the ratio being noticed at the end of the monitoring period. The free gas phase existing at the top of GC3N was not really enriched in CO₂ (the water phase does) but on the contrary deviations of the carbon ratio from baseline data are obvious. Such statements have been done *e.g.* in Norway during a similar CO₂ injection in rock formations.

Even if the injection has suffered from deviations from the initial plan, it has been possible to detect the injected CO₂ on all the monitored compartments but only on one borehole. Geochemical monitoring methods prove once again their sensitivity and their adaptability to changing conditions.

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Keywords: CO₂ injection pilot; coal seam; water monitoring

1. Introduction

Climate change mitigation is one of the key problematics that geosciences encounter nowadays. Among the options existing for trying to achieve this mitigation, CO₂ storage has been intensively studied during the past two decades. Deep CO₂ storage is a generic term that covers storage in depleted oil or gas fields, storage in deep saline aquifers and storage in unmineable coal seams [1]. This last storage option may have been the less studied option especially when considering practical operations in natural sites, unless this storage option can lead to enhanced coal bed methane (ECBM) production [2, 3] as a result of CO₂ adsorption and subsequent CH₄ desorption. It may be a consequence of the uncertainties and the technical difficulties existing when considering this CO₂ storage option. Coal seams are not all appropriate for CO₂ storage depending on the sorption affinity for CO₂ of the coal, the size, the permeability and the aperture of fractures and the matrix properties of the coal plus, in the case of ECBM, the proportion of CH₄ that could reasonably be desorbed during CO₂ injection [4, 5, 6, 7].

The CARBOLAB project was designed in order to fulfill the gap existing between purely laboratory experiments on core samples and pilot projects at industrial scale [8]. The CARBOLAB project is funded by the European RFCS program (Research Fund for Coal and Steel). It gathers six partners from Spain, France and Poland. Among the tasks defined in the project, one of the main targets of this project was to perform in-situ CO₂ injection in a coal seam located at 464 m depth in the Montsacro pit, Asturias, Spain.

This paper is focused on this last topic and more specifically on the water monitoring aspects. Complementary investigations on the gas phase can be found in other works presented by Lafortune et al. in this conference [9].

2. Settings

One of the main specificities of the CARBOLAB project is that the CO₂ injection was not performed from the surface but from a gallery in a coal mine. Other projects such as RECOPO and MOVECBM in Poland [7, 10],

Yubari pilot in Japan [7, 11] or Canadian and US projects [2, 3, 4] have yet dealt with injection from the surface in burden coal beds. As a consequence of direct injection from a gallery, the quantity of CO₂ that was used during the Spanish experiment (120 kg) was small compared to quantities that have been injected during other operations (more than 10 tons per day for the RECOPOL experiment [10]).

The CARBOLAB experiment is based on the injection of CO₂ in an approx. 1 m wide vertical coal vein. This vein has been equipped with an injection borehole and with four geochemical monitoring boreholes. A schematic view – planar view – of the site is given in Figure 1 and in-situ sampling procedure is illustrated by Figure 2. Other boreholes were dedicated to geophysical measurements. Two geochemical boreholes penetrated the coal vein (geochemical monitoring boreholes in Figure 1) and two others were parallel to the wall rock of the vein (water monitoring boreholes in Figure 1). The two geochemical boreholes penetrating the coal were thought to be dry when drilled but first characterizations after their completions showed that water penetrated into the boreholes, thus leading the gas monitoring to be more difficult. As a consequence, it was decided to focus on the water phase and to monitor water and dissolved gases evolution using the two other boreholes that surrounds the coal vein. One borehole is located northward from the vein (GC3N) and the other southward (GC1S).

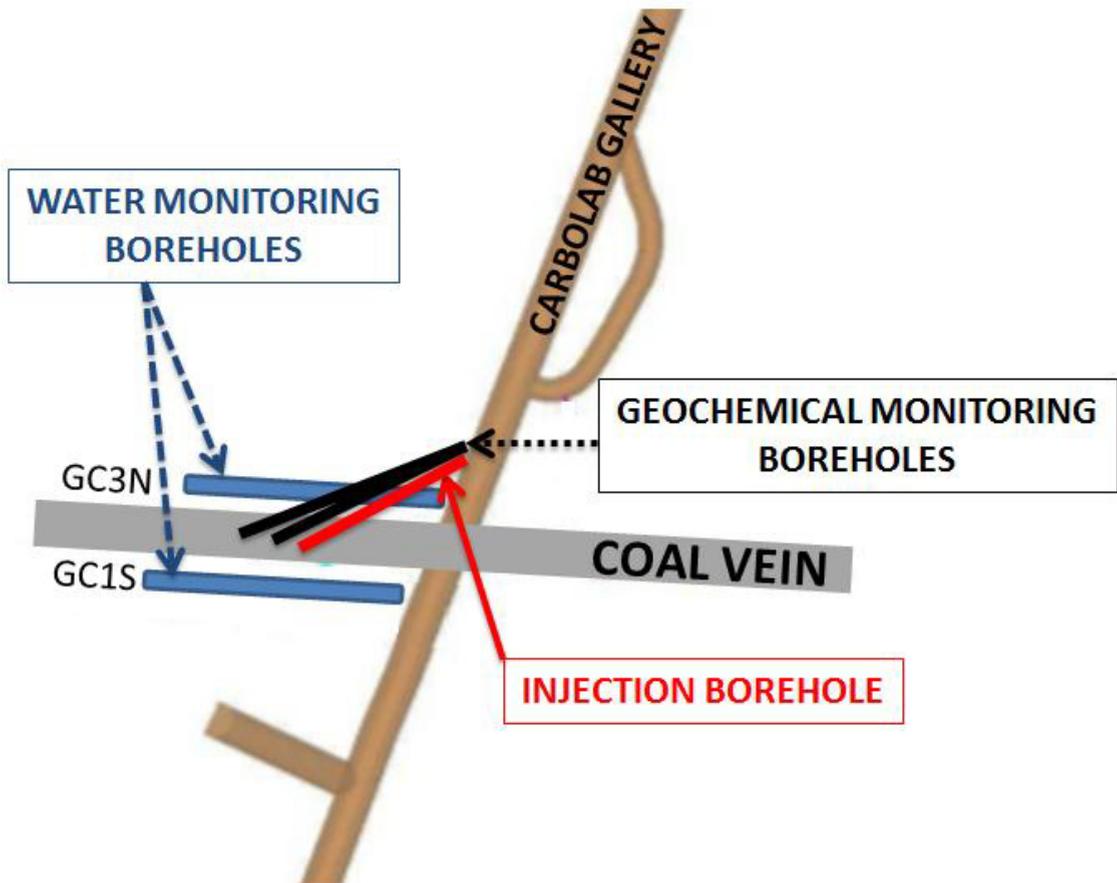


Fig. 1. Schematic view of the CARBOLAB experimental site, 464 m under ground level.



Fig. 2. (a) View of the CARBOLAB gallery with insets focusing on the monitoring technics used; (b) View of a lateral gallery where the material is stored with details on the installations.

Two site investigations were performed in 2012 and in the early 2013 prior to the CO₂ injection that took place in July 2013. These two baseline investigations allowed to determine the drawdown that is consecutive to pumping actions in GC1S and GC3N water monitoring boreholes. This information was used to adapt the monitoring strategy in July 2013 in order to avoid too strong water lowering in the borehole that may induce adverse effects on the experiment. According to the geometry of the system, it was decided to monitor GC1S and GC3N boreholes at 11 m and 21 m of distance from the top of the casing. Taking into account the dipping angle of the boreholes, this corresponds to real depths of respectively 5.5 and 10.5 m under the ground of the gallery. Boreholes are deeper, but their geometry did not allow to use pumping devices having greater investigation depths.

Work performed in the gallery has included the following:

- Water logging using a multi-parameter probe (Itronaut 303) at the beginning of each monitoring session.
- Water pumping at the two above mentioned depths and subsequent measurement of physico-chemical parameters.
- Water sampling and water conditioning in the gallery for subsequent anions and cations analyses.
- Dissolved gas sampling in appropriate evacuated glass flasks for subsequent gas measurements using gas chromatography operating in a dedicated room at the surface.
- Continuous monitoring of water temperature, pH, electrical conductivity, redox potential and dissolved oxygen at 25 m (true depth) in GC1S borehole.

Data and results are discussed in the following section.

3. Results

3.1. Baseline characterization

The two geochemical boreholes GC1S and GC3N were logged in order to determine their characteristics, taking into account that they are cased with screened pipes excepted the upper meters. Chemical profiles as a function of true depth are presented in Figure 3 and show the investigations performed just before the CO₂ injection. These profiles are representative of the measurements performed during baseline acquisitions and the following information can be derived from them:

- The water temperature appears influenced by the gallery in the first two meters. A steeper gradient indeed exists in this shallow water horizon, close to + 25°C per 100 m, whereas the gradient is only +2°C per 100 m for the rest of the borehole (GC1S, Figure 3a).
- The redox potential appears slightly reducing in GC1S borehole whereas a more oxidant character is measured in GC3N borehole. Changes along the depth are similar for both boreholes and a progressive strengthening of the reducing character can be noticed.
- This information is congruent with the dissolved oxygen measurements that show a quick disappearance of the oxygen in the water phase. An anoxic character is not surprising for waters at such depth in geological formations, the slight enrichment near the wellhead of the borehole only suggesting interaction with the air of the gallery.
- The water pH variation with depth is very low in the two boreholes with equilibrium values close to 7.6 for GC1S and 8.0 for GC3N. As the CO₂ injection is believed to induce a lowering of the pH values if the CO₂ has to escape in the water phase [11], then a monitoring of the pH shall be informative during the experiment.
- The electrical conductivity appears to be the most relevant parameter for studying connections between water bodies and possible mixing processes or leakage events in relation to the experiment. Both boreholes have a linear conductivity profile suggesting that the water present in each borehole is homogeneous all along the screened section. As electrical conductivity is a proxy for the content in dissolved elements, GC1S water appears to be twice enriched compared to GC3N water. This also suggests that, in the present case, a near vertical coal vein constitutes a good screen that is able, under steady state conditions, to separate water bodies of different chemistries.

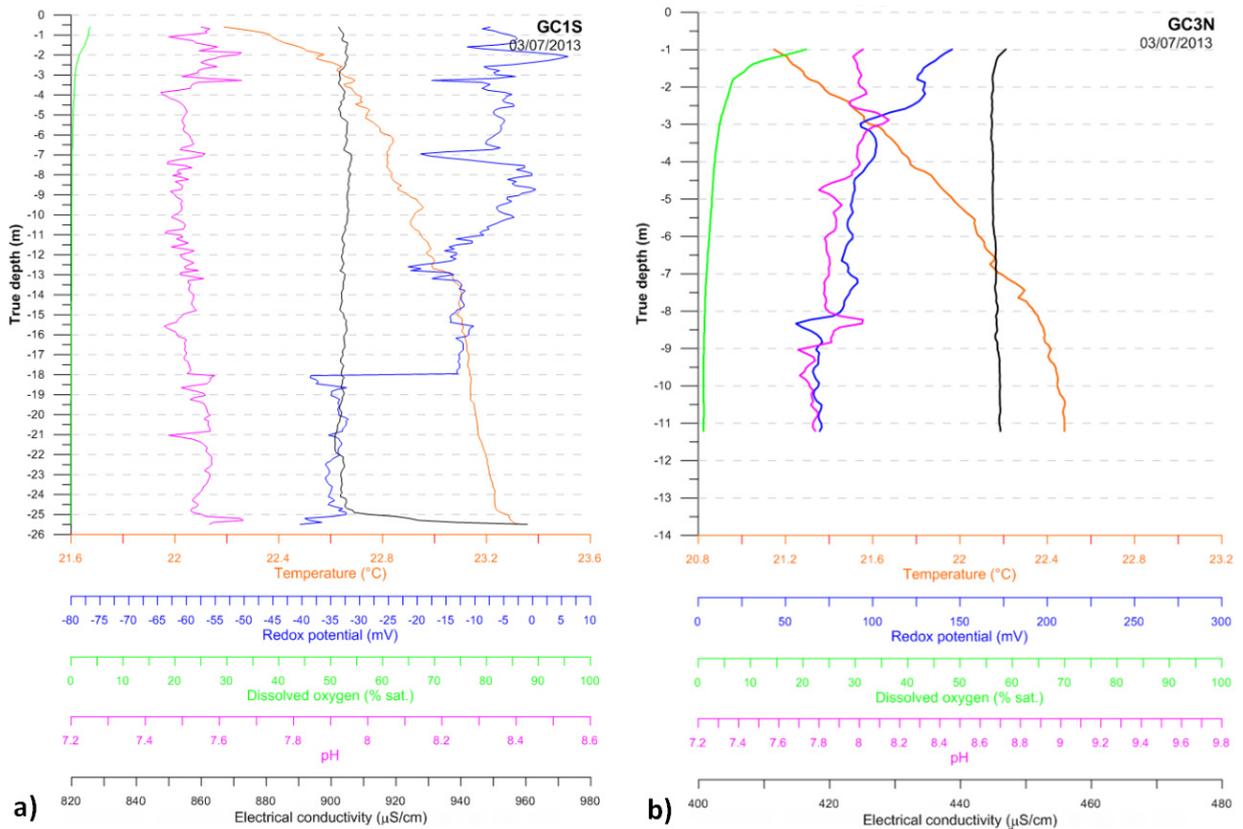


Fig. 3. (a) Temperature, redox potential, dissolved oxygen, pH and electrical conductivity of GC1S borehole prior to CO₂ injection; (b) Temperature, redox potential, dissolved oxygen, pH and electrical conductivity of GC3N borehole prior to CO₂ injection.

Dissolved gases were sampled at 5.5 and 10.5 m true depth in GC1S and at 10.5 m true depth in GC3N, due to progressive lowering of the water table in this second borehole even if care was taken to avoid this phenomenon (see section 2). As the experiment took place in coal environment and consisted in CO₂ injection, focus was on the CH₄ and CO₂ species. Baseline concentrations were close to 4×10^{-3} mol/l for CH₄ and *c.a.* one order of magnitude lesser for CO₂ (*i.e.* $\# 4 \times 10^{-4}$ mol/l).

3.2. Monitoring of the CO₂ injection – underground data acquisitions

GC1S borehole was continuously monitored using an Idronaut 303 probe emplaced at 25 m true depth. Data are not shown as no deviation was stated all along the experiment, especially the pH remained very close to the 7.6 - 7.7 value during the twenty days of deployment. This information is good from an instrumental point of view as it means that sensor drifting did not occur. From an experimental point of view, this suggests that no CO₂ escaped in the water. In other terms, this suggests that the CO₂ remained confined in the coal vein, what was looked for when designing this experiment, or that the CO₂ escaped elsewhere than in the deeper part of GC1S borehole.

This second option is thought to be the most realistic one, as several other datasets pointed out this bias. First, the gas composition monitored in the two geochemical boreholes emplaced directly in the coal vein (Fig. 1) did not significantly change during the CO₂ injection process. Second, probably due to defaults in the injection well completion or to fracture reactivation as a consequence of the well drillings, noticeable leakage was stated and measured in the ground of the gallery close to injection borehole wellhead. Quantification using CO₂ flux chambers gave estimates of tens of liters of pure CO₂ escaping per hour (20 to 70 l/h) thus suggesting that a non-negligible

proportion of the injected gas did not reach the coal vein nor the water bodies surrounding the vein. Indeed, the injection started on the 03rd of July at a 160 l/h injection rate and was still active on the 18th of July at a lower rate (110 to 120 l/h). Third, as highlighted by Figure 4, strong deviations from baseline data were recorded for GC3N borehole.

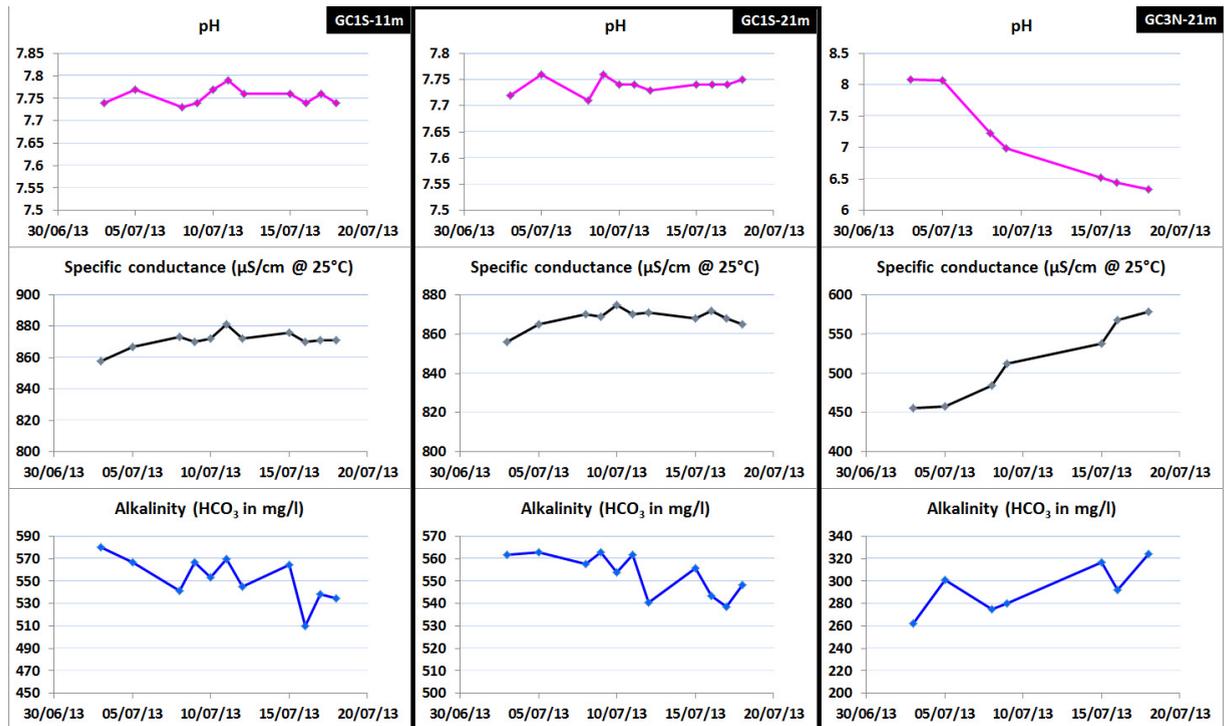


Fig. 4. From left to right, time evolution of water pH, electrical conductivity and alkalinity (bicarbonate content) for GC1S borehole at 11 m (5.5 m true depth) and 21 m (10.5 m true depth) and GC3N borehole at 21 m (10.5 m true depth) during the CO₂ injection.

As suggested by Figure 4, water in GC3N borehole experienced strong deviations during the CO₂ injection:

- The pH value remained stable and similar to that of the baseline during the first two days after the injection began, but a strong decrease then occurred and led to values as low as 6.4 pH units.
- Reversely, the electrical conductivity significantly rose from 450 up to 580 µS/cm.
- Last, the alkalinity, given as the bicarbonate content, did rise too, from 260 to 320 mg/l.

At the opposite, GC1S borehole only experienced small deviations from baseline data, the greater change being associated to a slight decrease of the alkalinity, from 560–580 mg/l down to 530–540 mg/l at the end of the monitoring period. As such, this change does not appear to be linked to the experiment.

These data suggest that the injection of CO₂ was not completely achieved in the coal vein and that the geometry of the geological strata allowed gas migration northward from the injection point in an aquifer but not southward in a distinct aquifer.

3.3. Monitoring of the CO₂ injection – laboratory data

The above mentioned data are supported by complementary investigations performed either on site but at the surface (dissolved gas concentration measurements) or at laboratory (dissolved elements).

Dissolved gas concentrations are reported in Figure 5. Only subtle changes are noticed for GC1S borehole, with slight rises of the CO₂ concentrations (from 7×10^{-4} to 8×10^{-4} mol/l) being associated with slight rises of the CH₄ concentrations (from 5×10^{-3} to 7×10^{-3} mol/l). It is difficult to unilaterally correlate those changes with CO₂ leakage in water but the congruence of both CO₂ and CH₄ rises may nevertheless suggest that the CH₄ rise can be related to desorption processes and then be a consequence of the CO₂ injection.

The behavior of GC3N borehole is clearly different. A progressive dissolved CO₂ enrichment can be noticed from the 07 July and extends until the end of the monitoring period whereas dissolved CH₄ concentrations remained stable. This may surely be attributed to the CO₂ injection that began on the 3rd of July. Nevertheless, as suggested in the previous paragraph, the CO₂ injection in the coal seam may induce the release of CH₄ (2 to 5 moles of CO₂ are needed to desorb 1 mole of CH₄). This is not the case and CH₄ concentrations in GC3N are smaller than concentrations found in GC1S. The CO₂ injection may have not directly been performed inside the vein but rather in surrounding rocks. Or, more probably, and taking into account the slight increases punctually stated in GC1S and the complex geometry of the system, the evolution of dissolved gases may suggest that the CO₂ did not interact with the coal on the northern part of the vein (presence of fractures?) and that the CO₂ did experience some interaction with the coal when migrating southward. The amount of CO₂ southward migrating was nevertheless very low as most of the CO₂ that reached the bottom of the injection well escaped quickly northward in the water body investigated by GC3N borehole.

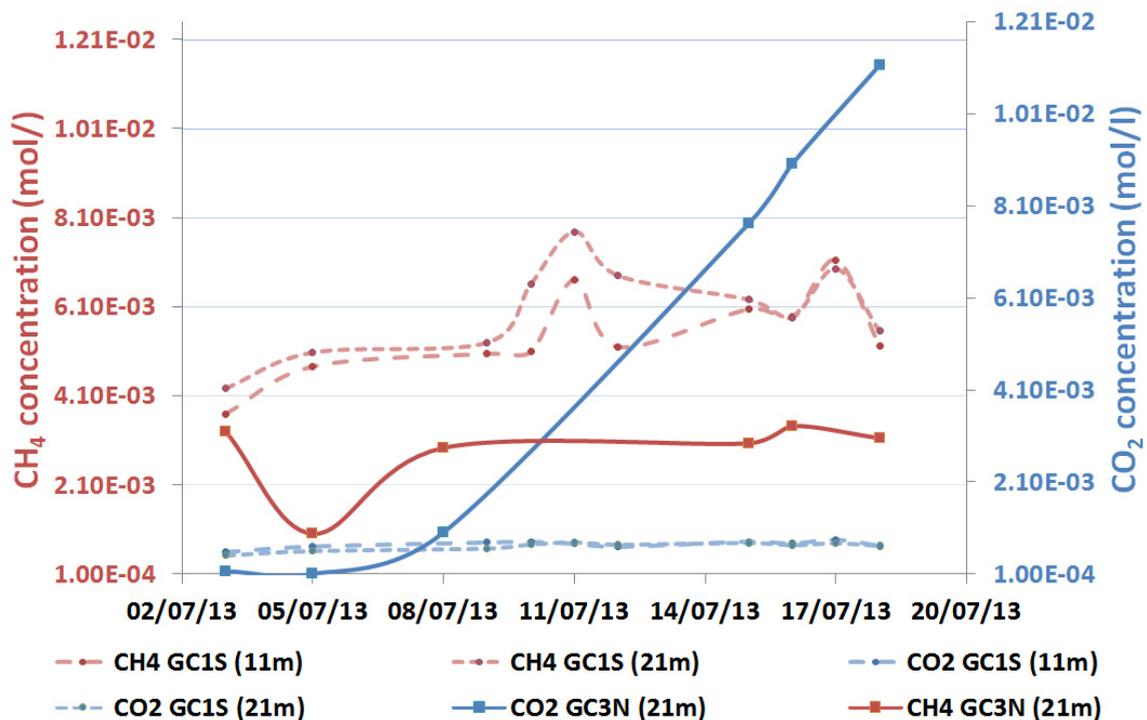


Fig. 5. Time evolution of CH₄ (left axis, in red) and CO₂ (right axis, in blue) concentrations for GC1S (dotted lines) and GC3N (solid line) boreholes.

Chemical analyses are congruent with statements made in the field during the experiment. Two sets of data were acquired: dissolved elements concentrations and stable isotope data on the gas phase.

Concentrations in dissolved elements are reported in Figures 6 and 7 and only for GC3N borehole, as GC1S waters did not experiment strong chemical changes as learned from field acquisitions. GC3N waters show a progressive enrichment in Ca and Mg species whereas other cations (Na, K) remain at nearly the same concentration. For the anions, the only noticeable change is for the bicarbonates, the other species (mainly Cl) remaining at constant – but low – concentrations. This is probably linked to the geology of the formations, with Ca- and Mg- rich mineral phases contributing to enrichment in dissolved elements as a consequence of the pH change linked to the CO₂ injection. Such a statement is quite common in CO₂ injection experiments [e.g. 12].

Major elements are not the only chemical species that experienced concentrations changes. Trace elements, especially trace metals, were also affected by the injection (Figure 7). Elements like Sr, Ni, Mn, Zn or Ba may have experienced rises of their concentrations by factors of 2 to 10 and elements such as Co appeared during the experiment (baseline values were beyond instrumental detection limit). At the opposite, elements like As or Li remained at nearly the same concentration. Last, Al concentrations were found to progressively decrease during the monitoring period. The interaction between two different waters (GC1S is more rich in dissolved elements than GC3N) does not appear to be the main process that can explain those enrichments. For some elements (Al, B, Ba, Li), mixing may have played a role either the decrease in concentration in GC3N (Al) or at the opposite the enrichment (B, Ba, Li). Nevertheless, this process cannot account for the changes stated for Mn, Ni, Sr or Zn. These concentrations changes are preferentially related to trace metal release as a result of mineral dissolution and/or phase interactions linked to the CO₂ injection.

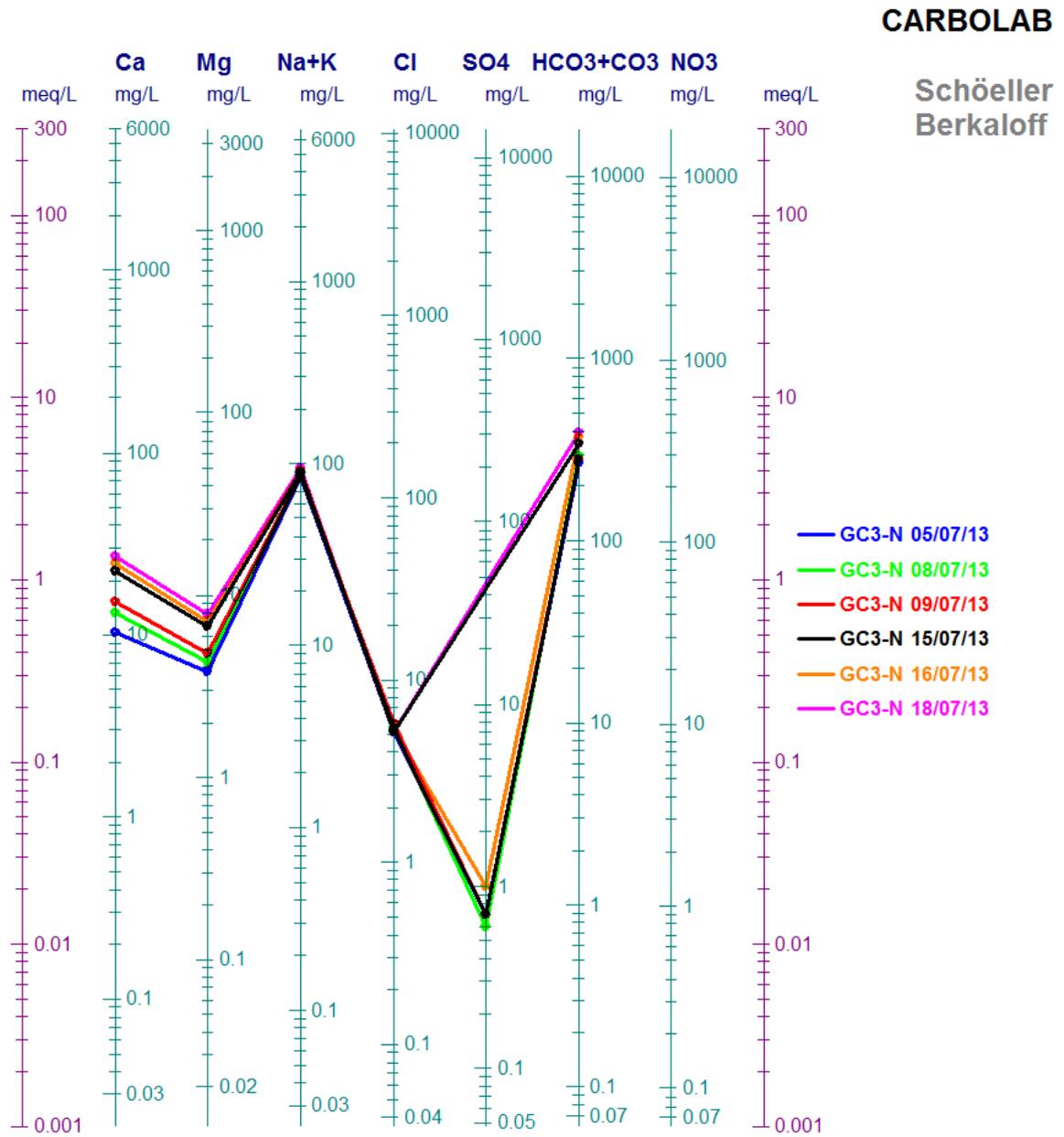


Fig. 6. Schöeller-Berkaloff plot – GC3N waters (sampling depth: 10.5 m true depth); concentrations in mg/l.

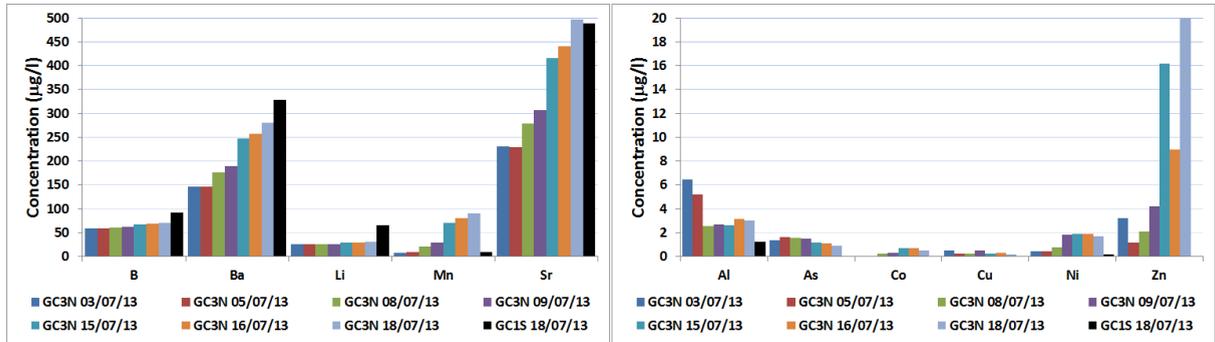


Fig. 7. Trace elements – GC3N waters (sampling depth: 10.5 m true depth); concentrations in µg/l.

Last, isotope data bring complementary information to describe the experiment. Isotope ratios were determined on gas samples. Baseline data only concerned $\delta^{13}\text{C}_{\text{CO}_2}$ but $\delta^{13}\text{C}_{\text{CH}_4}$ measurements were added for the monitoring of the CO_2 injection. These baseline measurements (April 2012 and January 2013) give a good estimate of the system prior to any change. Samples collected in the geochemical monitoring boreholes (free gas phase above the water table level) and in the gallery have a similar $\delta^{13}\text{C}_{\text{CO}_2}$ isotope signature ranging between -10 and -15‰ VPDB independently from the richness in CH_4 (1.4 to 87% vol.) and the CO_2 concentration (0.04 to 0.55 % vol.).

The CO_2 injection led to changes in the pre-injection situation. Data are reported in Figure 8.

First, the leakage of CO_2 noticed near the injection borehole wellhead induced a slight enrichment in CO_2 concentration in the air of the gallery (560 ppm at the end of the monitoring period) and led to a depletion of the carbon isotope ratio (from -13.6 to -16.5‰ VPDB).

Second, geochemical monitoring boreholes (GC1 and GC2) have distinct isotope ratios depending on their relative richness in CO_2 or CH_4 . GC2 borehole was not affected by CO_2 intrusion in the gas phase. This borehole had ratios similar to those measured under baseline conditions (around -10‰ VPDB) and still rich in CH_4 (more than 80% vol.). At the opposite, GC1 borehole was found to be very rich in CO_2 (more than 90% vol.). Its $\delta^{13}\text{C}_{\text{CO}_2}$ ratio was very close to that of the injected CO_2 (-40.7‰ VPDB) thus suggesting that some gas was effectively released in the coal formation, as previously suggested when considering concentrations of gases dissolved in water.

Third, the gas phase that exists in GC3N borehole and that was sampled confirms that the CO_2 gas found as a dissolved phase in the water has a non-natural origin and was originated from the injected CO_2 . Even if the CO_2 concentration in this free gas phase remained low (less than 1% vol.), an imprint on the carbon isotope ratio seems to be present as earlier reported in other geological context [12, 13]. Indeed, as the ratio was close to -32‰, surface processes cannot account for such a change. The interaction with CH_4 does not seem to be realistic in the present case as CH_4 content is only of 3.5%. Thus the best way to deplete the carbon ratio of CO_2 is to equilibrate the gas with the injected CO_2 . Another argument claiming for this process is the ratio determined for the CO_2 collected in the flux chamber (-38‰). This shows that there is little change of the ratio in case of quick escape from the injection point, the imprint in compartments affected by the leakage shall then be strong.

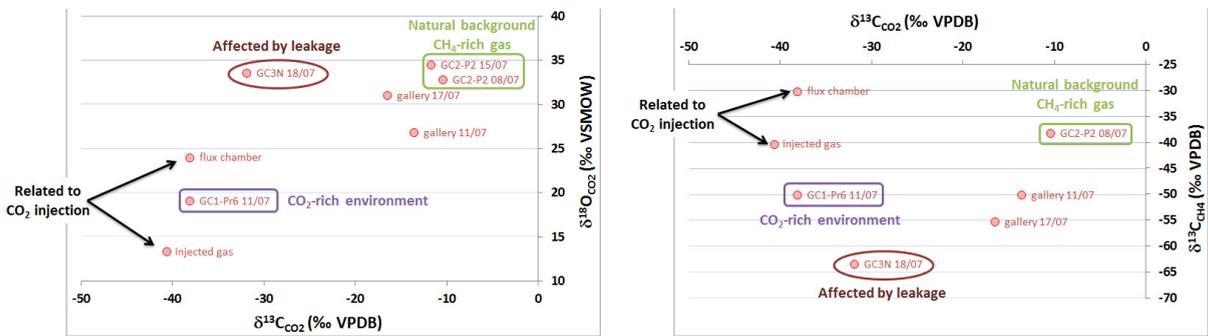


Fig. 8. (a) Carbon and oxygen isotope ratios of the CO₂ gas phase; (b) Carbon isotope ratios of the CO₂ and CH₄ gas phases.

4. Concluding remarks

The CARBOLAB project was designed to strengthen the knowledge of changes, reactions and properties variations associated to CO₂ injection in coal seams. One of its originalities was to perform a CO₂ injection directly at depth in a coal seam emplaced 464 m below ground level, using appropriate injection borehole and related geophysical and geochemical monitoring boreholes. Even if the initial design of the experiment was not fully matched, *i.e.* a non-negligible quantity of the CO₂ directly leaked back into the atmosphere without interacting with the coal, geochemical monitoring methods have allowed to monitor the gas migration during the experiment.

They allowed to detect and to quantify the CO₂ leakage in the gallery.

By using water monitoring boreholes located northward and southward from the coal vein, geochemical methods allowed to determine that only few amounts of CO₂ may have migrated in the coal and then in the southern aquifer (GC1S borehole). At the opposite, gas migration was consequently more developed in the northern direction, but the CO₂ did virtually not interact with the coal as no CH₄ release can be highlighted. Nonetheless, the injection of CO₂ in the northern water body induced changes of the pH thus leading to a rise in dissolved elements concentrations and a rise of the bicarbonate content. These chemical changes are preferentially related to reactions with the rocks surrounding the coal vein rather to reactions with the coal vein itself. Stable isotope ratios of CO₂ and CH₄ changed accordingly to this leakage scheme.

More generally, geochemical methods also allowed to highlight that water bodies existing at depth, especially in the context of near vertical geological formations, are far from homogeneity and can behave distinctly during such an experiment.

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