Tracking and CO2 leakage from deep saline to fresh groundwaters: Development of sensitive monitoring techniques

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Tracking and CO_2 leakage from deep saline to fresh groundwaters: development of sensitive monitoring techniques

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Abstract

In a predictive approach goal and to focus this work on risk assessment related to CCS context, a modeling study of an unfavorable scenario in which CO_2 leakage (willfully provoked) through an abandoned well, from a deep saline aquifer to fresh groundwater was developed. This 3D numerical model handles both fluid-rock geochemical interaction in the storage aquifer and impacted aquifer (code TOUGHREACT). In a second part and in the frame of studying, the use of multi-isotope approaches, like an isotopic monitoring opportunity, is developing in order to detect CO_2 leakage and to record geochemical processes of water quality evolution, induced by the lowering of the pH due to the ingress of CO_2 in the system. The case of study applies to the French Paris Basin.

Keywords: CO_2 geological storage; CO_2 leakage; reactive transport modeling; water quality; isotopic monitoring opprotunities

1. Introduction

The assessment of environmental impacts of carbon dioxide geological storage requires the investigation of the potential CO_2 leakage into fresh groundwater, particularly with respect to protected, high quality, groundwater reserves. The geochemical processes and perturbations associated with a CO_2 gas leak into dilute groundwater are well known, such as precipitation, oxidation and mineral dissolutions. Some of these minerals may contain hazardous constituents which might release contaminant into the groundwater [1] [2] [3].
The illustrated case study is from the French Paris Basin context in which the Jurassic Dogger formation has been used for geothermal purpose since several decades and is now under consideration as a potential target for the French national program of greenhouse gas emission reduction and CO_2 geological storage concept [4].

The Albian aquifer, situated above the Dogger aquifer, is a major strategic drinkable potable water aquifer (www.eau-seine-normandie.fr); the impacts of a CO_2 leak due to potential integrity failure have therefore to be investigated, with a modeling and experimental approach.

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A new 3-year project aims at developing sensitive monitoring techniques in order to detect potential CO_2 leaks and their magnitude as well as their geochemical impacts on the groundwater. In a dimensioning goal, a modeling study of the geochemical impact of a CO_2 intrusion on fresh groundwater during geological storage was also performed and serves as a basis for the development of sensitive monitoring techniques (e.g. isotope tracing). Then, isotopic monitoring opportunities are explored.

2. The reservoir formation

The intracratonic Paris Basin (France) has been a zone site of quasi-continuous subsidence and sedimentation during the Mesozoic era. It is made up of layers of up to 3,000 m of indurated Triassic to Tertiary sediments resting on a basement [5]. The two aquifers targeted in this study belong to a complex multi-layered aquifer system: the Dogger and Albian formations.

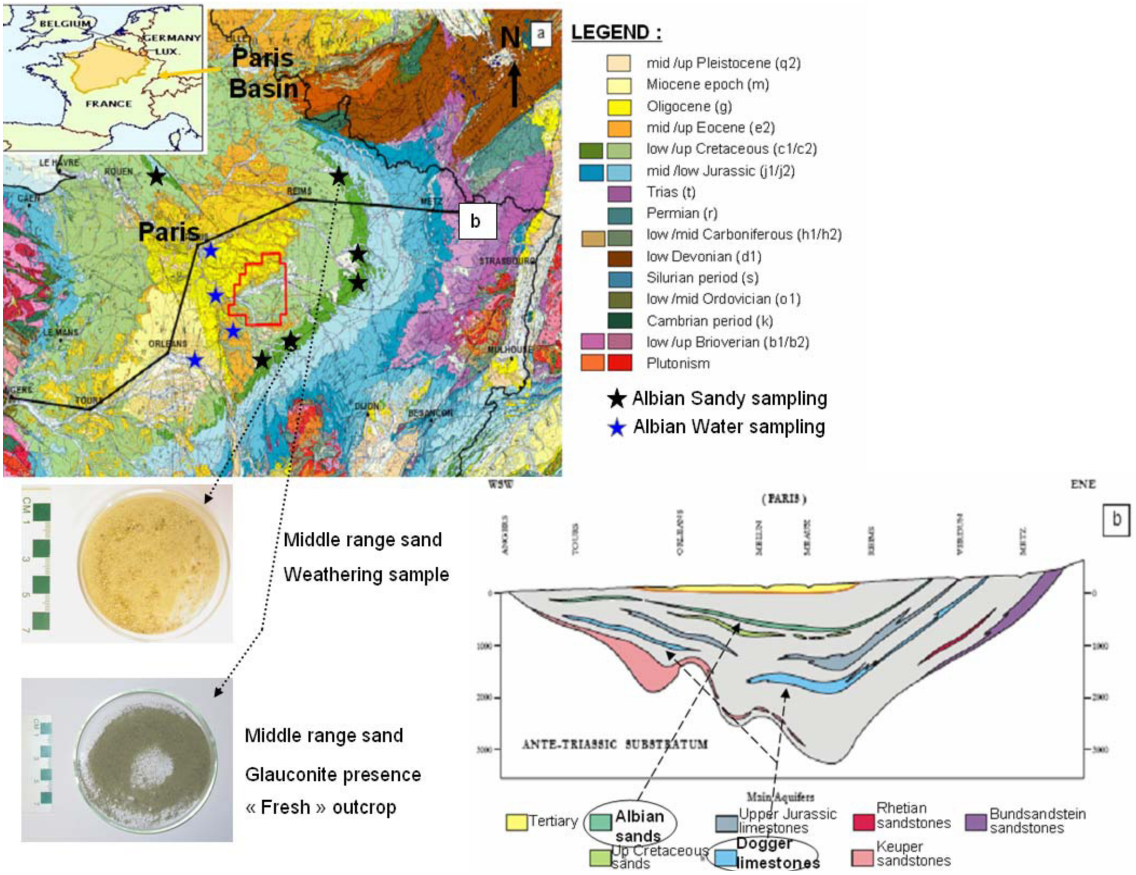

Figure 1 Geological map of the studied area [6] and sampling campaigns of Albian fm

The Dogger fm., investigated for large-scale CO_2 storage concept is mainly composed of carbonates. Variation of groundwater salinity is observed ranging from 5 g/kgw in the Southern up to 35 g/kgw in the Eastern [7]. The confined Albian aquifer, considered as the impacted aquifer in this study, covers an area of about 75,000 km² in the Paris Basin [8]. It is a sandy multi-layered aquifer. The chemical composition of the fresh groundwater in the Albian formation varies through the aquifer, resulting from a complex hydrogeology with multiple recharges and residence times [9]. The Albian groundwater is globally anoxic with high Fe-concentration, a pH of around 7 and low mineral content (around 0.3 to 0.6 g/L).

In order to provide material from the French Paris Basin for the reaction experiments, two sampling campaigns (Figure 1) have been performed to acquire:

- Albian water (from deep bore-holes as fresh water wells, geothermal doublet, piezometer): comparison with literature data show a correct representativeness compared to the center of the basin;
- Albian sands (lower Cretaceous outcrops, quarries, cuttings): the main extension of the Albian outcrops is represented by ring from the S to NE of the Paris Basin and some small outcrops in the lower Seine valley (NW). Samples from the NE outcrops revealed fresher minerals, with presence of glauconite (responsible for the green tint of the sands).

3. Modeling approach

3.1. Definition of a leaking scenario; application to French Paris Basin

The scenario for the case study refers to a potential storage in the Paris Basin. A leakage is simulated from a reservoir in the carbonated Dogger formation, with an impact on the overlying sandstone Albian formation.

The reactive transport code TOUGHREACT [10] was used to assess (i) the multiphase flow evolution during the injection phase including dissolution of the CO_2 in the aqueous phase, (ii) the leakage of the CO_2 from a target reservoir to an overlying formation through a leakage zone approximated by a 1D porous media, and (iii) the mineral dissolutions and precipitations due to CO_2 intrusion in the system. The final objective os the simulation is the evaluation of potential changes in groundwater quality in response to CO_2 leakage from saline aquifer formation, with a focus on acidification process only. In a risk assessment approach, an unfavourable scenario was chosen, in which the CO_2 intrusion into the fresh groundwater is wilfully provoked (Figure 2).

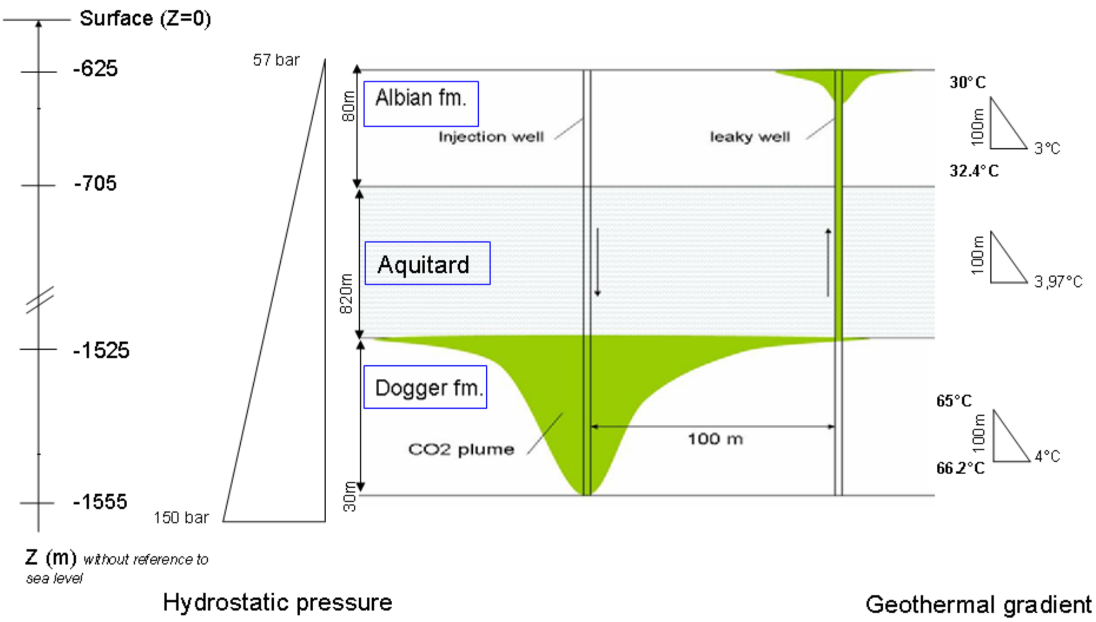

Figure 2 Diagram representing the simulated leakage scenario based [11] with initial pressure and temperature gradient

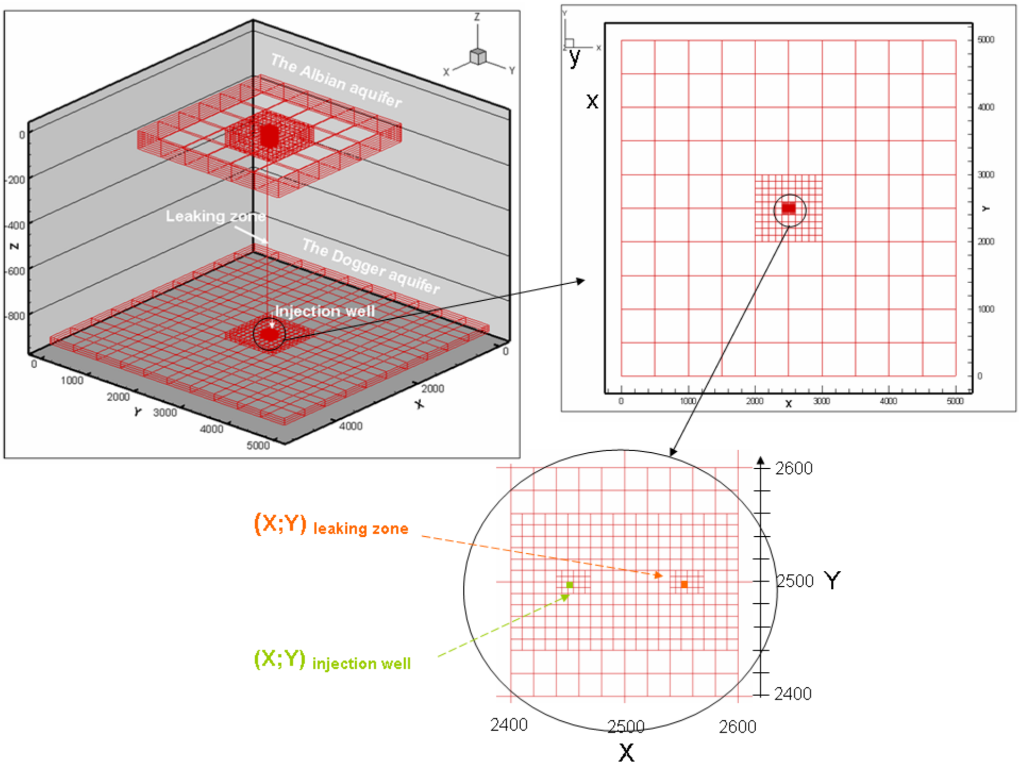

Figure 3 3D mesh representation for the simulation of leakage from the Dogger aquifer to the Albian fm. through an abandoned well. The well is represented as a 1D porous medium (formation rock-cement interface). The grid is locally refined in the Dogger aquifer (top view)

This predictive modelling approach includes a 3D model (Figure 3) with (i) storage saline aquifer, (ii) impacted freshwater in the overlying aquifer and (iii) a leaky abandoned well represented by a 1D porous media as a rough representation of the rock-cement interface. This leaky well is situated 100 m away from the injection well, and allows the CO_2 to reach the overlying formation. This model intends to assess globally the whole migration path of the supercritical CO_2 and the interaction between the fluids and the host rock.

The simulations presented in this study aim at helping the definition of guidelines and selection criteria in the field of environmental risks. Therefore, the examination of the effect of the gas intrusion on the quality changes of the local groundwater as well as a better understanding of the water-rock-gas interactions is of primary importance.

3.2. Impact of CO_2 intrusion on Albian composition

The CO_2 is injected at a rate of 12.7 kg/s (0.4 Mt/yr) in the Dogger. The gas plume in the Dogger formation extends up to 1.5 km from the point of injection after 5 years. The injection builds up the pressure in the Dogger, which drives the migration to the overlying formation. The CO_2 reaches the Albian aquifer, through the leaky well, after 90 days of injection. The upward migration of CO_2 at a rate of 1.1 kg/s is accompanied by fresh water at a rate of 0.03 kg/s at the end of the simulation. The CO_2 injected in a supercritical state in the Dogger reaches the Albian aquifer under a gas state. Major mechanisms such as mineral dissolution in the fresh water are triggered by CO_2 dissolution (regardless of CO_2 sub- or super-critical state).

The principal geochemical process simulated in this study is the acidification of groundwaters (buffered from pH 7.3 to 4.9) due to CO_2 dissolution inducing the dissolution of minerals in the Albian formation and the release of chemical elements (figure 4B). The zone impacted by dissolved CO_2 covers a 0.3 km radial extent, whereas gaseous CO_2 occurs only right above the intrusion point. Significant increases are observed, for the calcium concentration, from 1.3 to 31.6 mmol/kgw, and for the iron concentration (glauconite dissolution) from 1.3 to 2,500 µmol/kgw at the CO_2 intrusion point. The glauconite dissolution (figure 4A) also entails an increase in dissolved silica and aluminium, with resulting precipitation of kaolinite and quartz around the intrusion point. Beyond this zone, the model predicts a similar geochemical reactivity, with one order of magnitude smaller dissolution and precipitation rates.

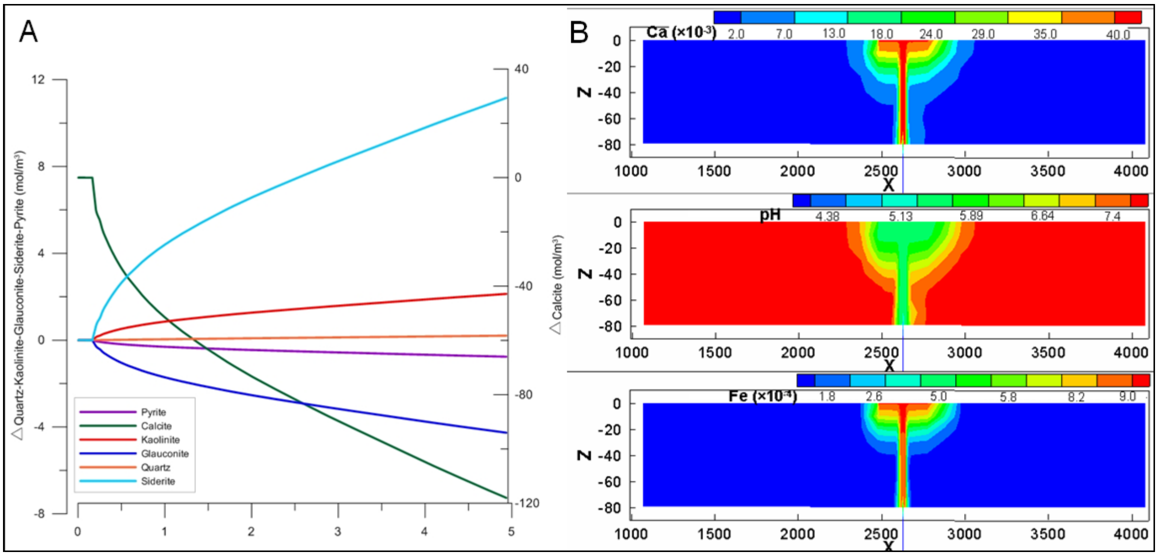

Figure 4 Evolution of mineral phases during CO2 ingress in the Albian formation (at the top of the leaking well) during CO2 leakage (A), aqueous concentrations (Ca, Fe in mol/kgw) and pH in the Albian formation after 5 years (B)

The quality of the model could be increased by taking into account surface reaction (complexation, ion exchange). Finally, the release of trace elements, like lead or arsenic, is of major concern for shallow groundwater chemical integrity: the simulation of their reactivity involves finer chemical description, particularly solid solutions, which were not performed during the study.

4. Isotopic monitoring opportunities

The intrusion of CO_2 into a fresh groundwater, as described previously, seems to be difficult to monitor directly, because it is heavily modified by several reactions (i.e. dissolution, precipitation processes). The use the isotopic signature of several choice elements might be an interesting alternative tool to record geochemical modifications induced by CO_2 intrusion to predictive model.

4.1. How can isotope tools help in deciphering element release/trapping?

Isotopic systems can keep track of reactions associated with small quantities of CO_2 into dilute aquifer which, in case of leakage, would not be detected by others methods. Such isotopic monitoring systems require the application of different types of methods and apparatus. IRMS (Isotope Ratio Mass Spectrometer), CF-IRMS (Continuous flux-Isotope Ratio Mass Spectrometer), TIMS (Thermo-Ionisation Mass Spectrometer) will be used for a large array of isotopic systems. Additionally, other new non classical isotopic systems will be investigated using the new generation isotopic ratio mass spectrometers MC-ICP-MS that combine the ionisation efficiency of inductively coupled plasma with the advantages of multi-collector data acquisition. With this technology, isotopes of many different chemical elements can be analysed with higher precision.

Thus, isotopic fractionation will be investigated during this work. Fractionation is interesting since it is mechanism-dependent: e.g. isotope exchange reactions, physical and chemical processes as kinetic aspect (diffusion etc.) or state changes (adsorption, desorption, dissolution, precipitation etc) [12] [13].

4.2. Isotopic "toolbox"

A laboratory experiment on an Albian representative sample is being developed in order to test these monitoring tools. They involve batch reactions between CO_2 gas and equilibrated Albian water samples in contact with Albian (outcrop) sands. The water-rock-CO_2 interaction and its impact on mineral evolution will be analyzed. Parallel to more classical investigation, isotopic evolution will be closely monitored.

The objective of future work is to develop an isotopic toolbox, which should be able to record different processes of water quality evolution due to CO_2 intrusion. The main focus is to point out suitable isotope systems which can characterize different processes such as:

- Water-rock interaction: lithium, boron isotopes may indicate adsorption/desorption processes [14][15], strontium [16] and carbon isotopes give evidence of carbonate dissolution [17];
- Redox conditions evolution: sulphur [18], selenium, iron and chromium isotopes may indicate redox-active mobility, depending on valence state and redox transformation;
- Secondary processes: calcium isotopes may indicate precipitation reactions;
- CO_2 migration: carbon isotope [17] [19].

These instrumental advances, previously described (4.1), open news research field and favour the use of multi-isotope approaches in isotopic studies as the main interest of hazardous trace elements transport and geochemical evolution of groundwater due to CO_2 intrusion.

5. Conclusion

This work provides a preliminary 3D model which shows simultaneous evaluation and model of element concentration and injection procedure. Indeed, three systems are considered in this numerical approach (i) storage saline aquifer (ii) impact on overlying fresh groundwater aquifer (iii) a leaky abandoned well, represented as a 1D porous column. The main geochemical processes simulated are acidification of groundwater due to CO_2 dissolution, Albian minerals dissolution and precipitation, element release. Particular attention to adsorption and desorption processes via surface complexation constitutes a part of future works. These simulation help constrain the expected range of the impact of a CO_2 intrusion in an overlying aquifer and help in dimensioning laboratory experiments.

Now, we focus on the development of sensitive multi-isotope monitoring in the frame of CCS by testing the feasibility and sensitivity of different isotopic tools at the laboratory scale.

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