PROJECT FINAL REPORT

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Executive summary

Funded by the European Commission's 7th Framework Programme, the four-year ULTimateCO_2 project was dedicated to studying the long-term fate of geologically stored CO_2. The goal: to substantially improve our understanding of the long-term effects of CO_2 geological storage. ULTimateCO_2 therefore looked at the main processes and impacts that will occur over the lifetime of a CO_2 storage project and beyond: (i) Trapping mechanisms in the geological reservoir in order to evaluate the evolution of the CO_2 plume (ii) Sealing integrity of wells and caprock in order to evaluate any possible occurrence of leakage, (iii) Regional-scale impacts (e.g. pressurization and fluid displacement) within realistic geological contexts, (iv) Uncertainty assessment tools in order to evaluate the degree of confidence that should be placed in modelling projections. These processes were studied on both local (site/reservoir) and regional (sedimentary basin) scales.

ULTimateCO_2 focused on the long term, i.e. thousands of years, with predictions based on both laboratory experiments and modelling. Real field data were also used to support the studies – not only from suitable reservoir rocks and caprocks, but natural and industrial (oil and gas field) settings or analogues. As with all large construction projects, ULTimateCO_2 also factored in the level of uncertainty associated with long-term predictions, an approach which could be used routinely in CO_2 storage projects.

The main outcomes of the project can be summarized as follows:

- CO_2 injected into a storage site can change its nature over time – for example, by dissolving in water (as in a bottle of sparkling water) or reacting chemically with rocks to produce minerals.
- Modelling of trapping mechanisms in realistic storage scenarios revealed that more than 50% of CO_2 remains in a supercritical form several decades after site closure. However, it is still trapped within the structure, as demonstrated by the vast CO_2 fields that already exist in nature. Studies of natural analogues – and confirmed by numerical predictions – also reveal that mineral trapping is limited in sandstone reservoirs typically used for CO_2 storage. This highlights the importance of having control of CO_2 migration through the combined use of modelling predictions and real-time monitoring once CO_2 is injected into the reservoir. Conformity between modelling and monitoring is more challenging for geochemical processes (such as mineral dissolution due to acidification) for which further research is required.
- ULTimateCO_2 has developed advanced numerical modelling techniques which show the importance of basin-scale models in predicting the long-term evolution of CO_2 – not only at reservoir scale (100s of metres), but at basin scale (100s of kilometres) – in terms of pressure impact. As basin models are not an obligation for operators such models could be undertaken at regional or national level, with the support of government funds, in order to help operators predict the evolution of CO_2 storage after site closure.
- When a typical caprock (e.g. Opalinus Clay) was tested for resistance to various fault rupture mechanisms at the Mont Terri Underground Rock Laboratory, the caprock showed a low risk of failure. These results are specific to experimental conditions and extrapolations to real-life situations should be taken with necessary caution. An improvement in protocols for evaluating the resistance of such geo-materials should therefore be developed in order to validate scientific investigations.
- When the test well at Mont Terri was put in contact with brine acidified with dissolved CO_2 for over a year, the CO_2 had a low impact on well integrity. However, the well had a high sensitivity to pressure and temperature variations, meaning that the 'history' of the well will strongly influence its integrity. Efforts should therefore be made to develop a geophysical tool to

improve the evaluation of abandoned wells which appear to be the main risk for leakage pathways.

- In general, the integrity of caprock and wells showed a tendency to self-heal in the presence of CO_2.
- Evidence from real site operations, such as CO_2 storage or oil and gas production, show that predicting leakage risk from the storage complex is difficult due to the complexity of the underground geology, mineralogy and history of the natural processes involved. Nevertheless, risks identified were considered to be either low or very low.
- In summary, ULTimateCO_2 research confirmed that the impact of long-term CO_2 storage is low – no critical thresholds were reached in terms of pressure, fault reactivation or the development of CO2 flux/flow. While uncertainty should be factored in when looking at long time scales, the migration of the CO_2 plume was limited.

Summary description of project context and objectives

Context

CO_2 emissions must be drastically reduced in order to limit the rise in average global temperature to 2°C – and avoid irreversible climate change. Such a massive reduction can only be achieved using a portfolio of solutions, including energy efficiency, renewables and CO_2 Capture and Storage (CCS) – as confirmed by virtually every global emissions reduction scenario.[1]

In the power sector, CCS will play a critical role in meeting CO_2 reduction targets, while it is the *only* means of significantly reducing CO_2 emissions from industrial processes. It is also the only large-scale technology capable of achieving net negative emissions – for example, via bio-energy with CCS (BECCS). Decarbonizing the energy sector *without* CCS would therefore not only be extremely challenging, but significantly more expensive: CCS is among the lowest-cost options per tonne of CO_2 avoided.

The CCS chain involves capturing CO_2 at source and separating it from other industrial flue gases, transporting it in a compressed form (to take up less space) by ship or pipeline to a suitable storage site (either onshore or offshore), then injecting it via a well into a geological formation – at least 800 m underground. CO_2 geological storage is the final part of the CCS chain.

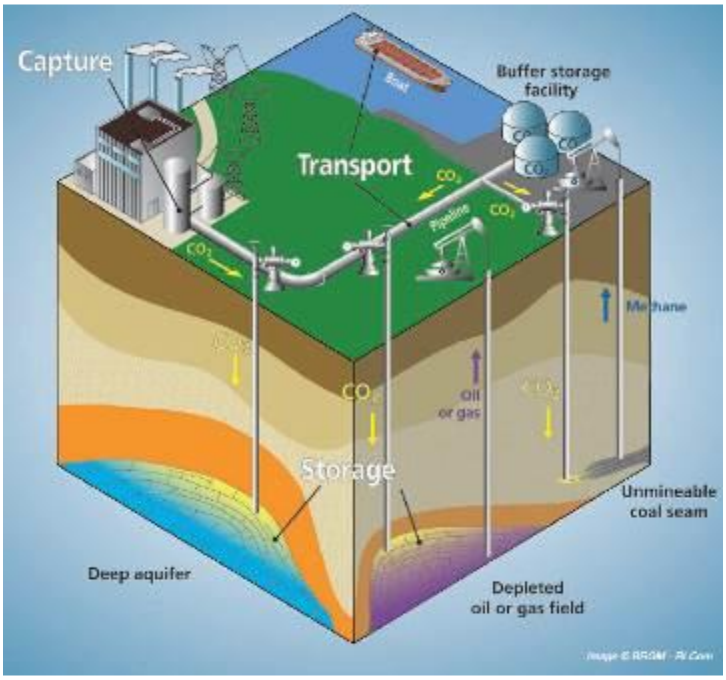

Figure 1: The CCS chain

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[1] Global CCS Institute, 2014 – The Global Status of CCS: 2014; IPCC – Climate Change 2014: Synthesis Report; EU Energy Roadmap 2050, 2012; International Energy Agency – Technology Roadmap: Carbon Capture and Storage 2013.

Objectives

Funded by the European Commission's 7th Framework Programme, the four-year ULTimateCO$_2$ project was dedicated to studying the long-term fate of geologically stored CO$_2$. The goal: to substantially improve our understanding of the long-term effects of CO$_2$ geological storage.

ULTimateCO$_2$ therefore looked at the main processes and impacts that will occur over the lifetime of a CO$_2$ storage project and beyond:

- **Trapping mechanisms** in the geological reservoir in order to evaluate the evolution of the CO$_2$ plume
- Sealing integrity **of wells and caprock** in order to evaluate any possible occurrence of leakage
- **Regional-scale impacts** (e.g. pressurization and fluid displacement) within realistic geological contexts
- **Uncertainty assessment tools** in order to evaluate the degree of confidence that should be placed in modelling projections.

These processes were studied on both local (site/reservoir) and regional (sedimentary basin) scales.

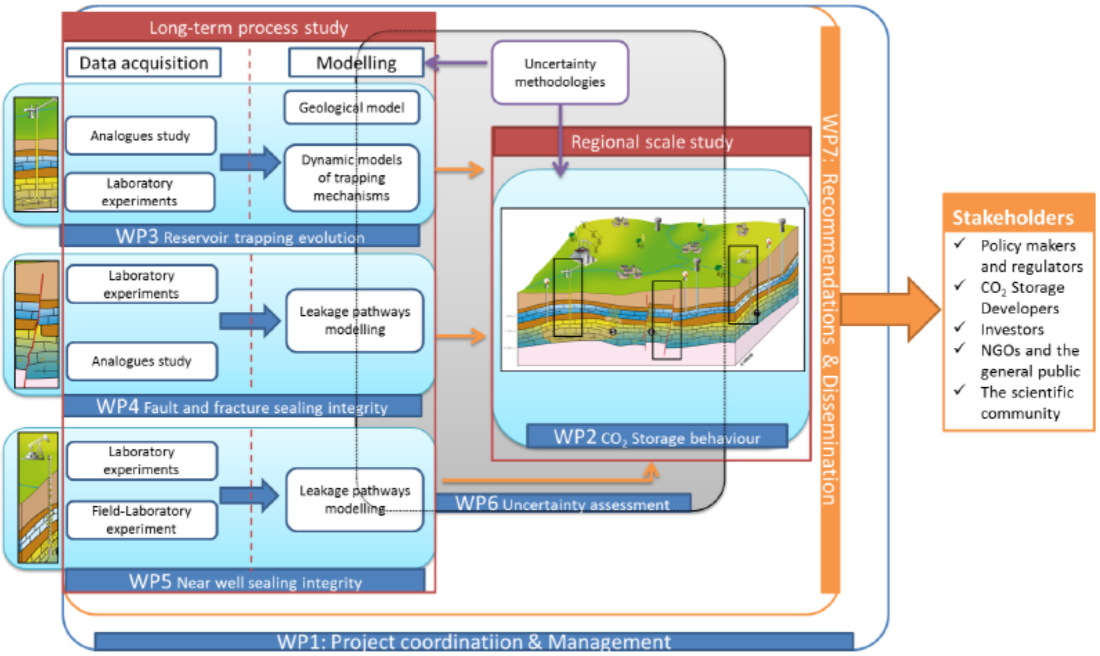

Figure 2: structure of the WPs of ULTimateCO2 project

ULTimateCO$_2$ focused on the long term, i.e. thousands of years, with predictions based on both laboratory experiments and modelling. Real field data were also used to support the studies – not only from suitable reservoir rocks and caprocks, but natural and industrial (oil and gas field) settings or analogues. As with all large construction projects, ULTimateCO$_2$ also factored in the level of uncertainty associated with long-term predictions, an approach which could be used routinely in CO$_2$ storage projects. The structure of the project has been built according to the different phases mentioned above.

Applying ULTimateCO$_2$ findings to a real CO$_2$ storage situation

ULTimateCO$_2$ focused on the post-closure and post-transfer phases of a storage project which in terms of implementation of the CCS Directive, corresponds to the process of transferring responsibility for the storage site from the site operator to the regulator or other Competent Authority or Member State. The transfer of responsibility is the final goal for the operator.

An accurate understanding of the long-term fate of CO_2 is important, especially for the national Competent Authority or regulator who will eventually take over responsibility of the site after CO_2 injection ceases and the storage site is formally closed. Before this transfer can take place, site operators must provide sufficient evidence to satisfy the Competent Authority that there is no detectable leakage and that the injected CO_2 is permanently and securely stored.

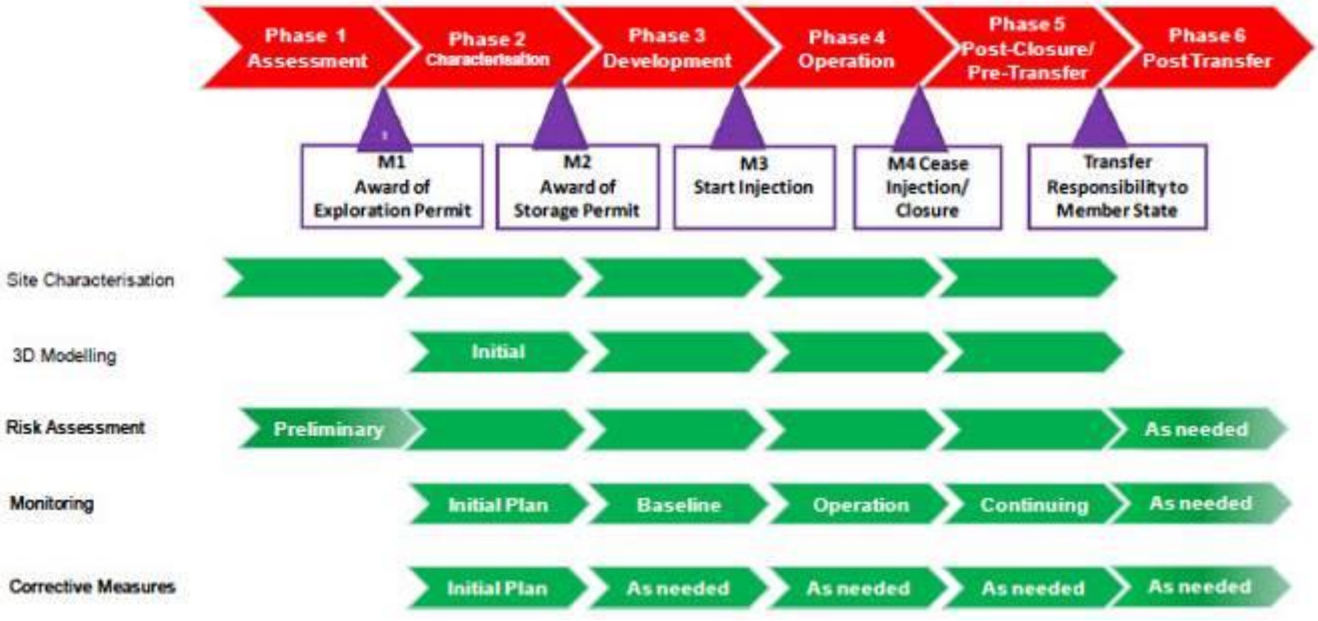

Figure 3: Simple CO2 storage project lifecycle, showing the main stages and permit milestones2

Although ULTimateCO_2 focuses on long-term issues, the conclusions are also relevant to other stages of a storage project lifecycle – from the characterisation and monitoring of the reservoir geology, through to the CO_2 injection, as well as site closure and transfer of responsibility to the Competent Authority.

The geological settings studied in ULTimateCO_2 were the European North Sea (offshore) and the eastern part of the Paris Basin in France (onshore). The reservoir rocks (both industrial and natural analogues) studied were sandstones, which means certain geochemical processes that are specific to sandstone reservoirs might be different in carbonate reservoirs.

ULTimateCO_2 did not focus on either monitoring or risk assessment, but rather on long-term processes in order to avoid duplication with other projects. Nevertheless, its conclusions will contribute to the understanding of long-term risks associated with storage and the robustness of the methodology for designing monitoring plans.

It is also the first time the issue of uncertainty has been assessed in the context of long-term numerical predictions for CO_2 geological storage. Although a general approach has been put forward, it should be noted that was based on a limited number of specific scenarios.

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[4] Implementation of Directive 2009/31/EC on the Geological Storage of Carbon Dioxide. Guidance Document 1: CO_2 Storage Life Cycle Risk Management Framework. European Commission 2011

Three criteria for demonstrating permanent storage

The European Directive on the geological storage of CO_2 (2009/31/EC), known as the 'CCS Directive', has established a legal framework for the geological storage of CO_2. In order to promote coherent implementation of the Directive throughout the EU, four Guidance Documents (GDs) have been produced.

The GDs contain much helpful advice for assessing the long-term fate of geologically stored CO_2 – particularly Guidance Document No. 3 which concerns the transfer of responsibility to the Competent Authority,[3] in line with the CCS Directive (Phases 5 and 6**Erreur ! Source du renvoi introuvable.**).

"The CCS Directive calls for the operator to demonstrate that "all available evidence indicates that the stored CO_2 will be completely and permanently contained". The Directive also suggests that operators can demonstrate permanent containment by meeting at least the three requirements or high-level criteria noted in Article 18(2):

- the conformity of the actual behaviour of the injected CO2 with the modelled behaviour;
- the absence of any detectable leakage;
- that the storage site is evolving towards a situation of long-term stability."

ULTimateCO2 key conclusions are designed to complement the CCS Directive's Guidance Documents in addressing the transfer of responsibility from operators to the Competent Authority.

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[3] Guidance Document No. 3: Criteria for Transfer of Responsibility to the Competent Authority: http://ec.europa.eu/clima/policies/lowcarbon/ccs/implementation/documentation_en.htm

Description of the main S&T results/foregrounds

The following summarizes the main outcomes of the ULTimateCO$_2$ project. Results are presented in line with the project structure.

1. Long-term reservoir trapping evolution (WP3)

Establishing that a storage site is evolving towards long-term stability requires the monitoring of four key parameters before, during and after CO$_2$ injection – as identified by the European Commission:

1. Pressure within the storage complex
2. Movement of the plume
3. Geochemical changes in the storage complex and wells
4. Integrity of materials used in the well throughout its lifetime (construction, operation, closure or abandonment).

Long-term stability is considered to have been achieved when:

- Models are able to predict stability of the CO$_2$ plume within the storage complex, with no expectation of future leakage.
- Values for key monitored parameters are within the range of future stable values predicted by modelling.
- The rate of change in key monitored parameters is small and declining.
- Back-casted values, from modelling historically monitored parameters, are within their respective intervals of uncertainty.
- For a specific storage complex, a parameter may be defined as stable if numerical modelling shows that its value is likely to change by less than a defined percentage over 1,000+ simulated years. It is expected that many parameters will change (often decrease) from their initial value at site closure towards a stable value.

To demonstrate that the storage site is evolving towards long-term stability requires an understanding of the relative contributions of trapping mechanisms over time.

ULTimateCO$_2$ has improved estimates of mineral trapping through new data on:

- The effects of trace components in the CO$_2$ stream
- Methods for selecting data for geochemical simulations
- Evidence of long-term processes in natural analogues
- Trapping stability estimates.

1.1. Improving the estimates of trapping mechanism contributions

Once injected into the storage reservoir, CO$_2$ occupies pore spaces within the rock that are normally filled with native fluids (generally salty waters unfit for human consumption). In its compressed, supercritical state (>31.1°C and >73.8 bar), CO2 is less dense than the rock's native fluids so behaves like a buoyant fluid and migrates upward. A range of trapping mechanisms then contributes towards permanent trapping in the reservoir: Structural, Residual, Dissolution and Mineral (SRDM).

A well-established concept in CO$_2$ storage, trapping diagrams show the contribution each trapping mechanism might be expected to make up to 10,000 years after CO$_2$ injection.

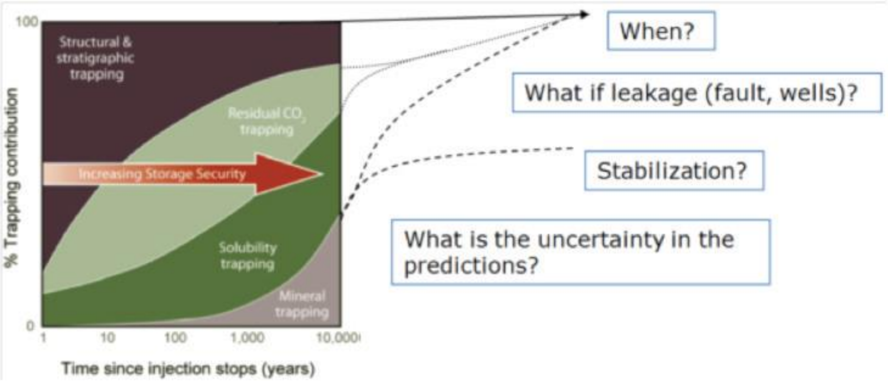


Figure 4: trapping diagram from IPCC 2005

Generally speaking, structural and residual trapping contributions reduce with time, while mineral trapping increases, improving the security of storage.

N.B. Trapping diagrams reflect hypothetical predictions that still need to be confirmed by, or at least compared with, calculations based on site-specific data.

1.2. The effects of trace components in the CO_2stream

Many reservoir sandstones contain limited amounts of calcium or magnesium which may limit their ability to trap CO_2 as secondary carbonate minerals. However, many contain abundant iron, usually as a hematite coating on detrital grain surfaces, and previous modelling has suggested that acid gases co-injected with CO_2 can free this iron (Fe) to form Fe-rich carbonates (e.g. siderite over the long term) – trapping CO_2 in the process. ULTimateCO_2 carried out a study of geochemical reactions between CO_2, gaseous impurities, saline waters (0.5 M NaCl) and sandstone. Experiments lasted around 3 and 9 months and were carried out in low-maintenance batch reactors operating at 140 bar (14 MPa) and 70°C. Acidic impurities (NO2, H_2S, or SO_2) in the CO_2 stream resulted in enhanced dissolution of the sandstone, dominated by the dissolution of carbonates. In all experiments containing impurities, as well as the CO_2-only experiments, the fluid chemistry and mineralogical analysis showed an increase in the reactivity of the sandstone.

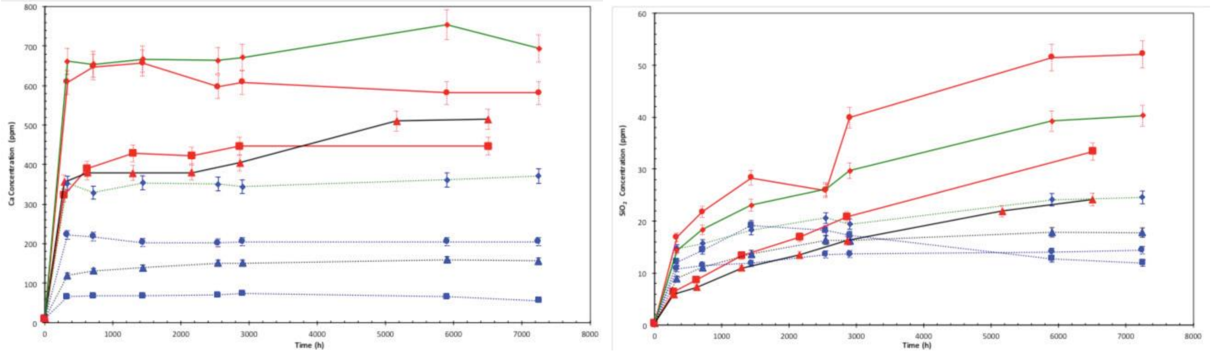


Figure 5: Ca (left) and Si (right) evolution versus time during the impurities experiments in batch

Dissolution took place soon after contact (within hours) with the sandstone and was dominated by changes caused by the acidification of fluids and the dissolution of carbonates that followed. This released calcium (Ca) and other associated elements from the carbonates into the solution.

In conclusion, the co-injection of trace quantities of acidic species (NO_2, H_2S or SO_2), which may be present in captured CO_2, resulted in enhanced dissolution of the sandstone dominated by the dissolution of carbonates. Levels of silicate dissolution were small.

These new laboratory data on the potential impact of trace components in the CO_2 stream were used to calibrate the geochemical model and upscale the long-term geochemical effects of co-injected species from laboratory to field scale with greater accuracy.

1.3. Methods for selecting data for geochemical simulations

In order to identify the best approaches to selecting appropriate data for modelling transport processes, common geochemical simulation software packages were compared and analysed, together with the underlying methods for determining thermodynamic data used. This involved the identification of required parameters and data, including an analysis of dissolution and precipitation reactions. If no measured data were available, knowledge was required on how thermodynamic data were estimated and interpolated, with estimation methods for minerals, gases and aqueous species reviewed. Starting with thermodynamic databases, the different parameters needed for geochemical simulations were compared (log K, activity models, kinetics law, Pitzer equations,…). Starting with thermodynamic databases, the different parameters needed for geochemical simulations were compared. on a critical analysis of the apparent Gibbs energy for Al_3+ in the temperature range up to 150°C and pressure range between 0.1 and 50 MPa, it was concluded that in general the standard Gibbs energy and entropy at reference pressure and temperature are the crucial parameters for predicting the behaviour of an aqueous species at higher temperatures and pressures Several codes have been also compared (PHREEQC, Geochemical Workbench, TOUGHREACT) to evaluate uncertainty associated to the simulation predictions of mineral trapping.

1.4. Evidence of long-term processes in natural analogues

ULTimateCO_2 tested the hypothesis that CO_2 injection may, over the long term, lead to mineral trapping by examining, in great detail, naturally occurring CO_2-rich systems that are directly analogous to sites likely to be used for CO_2 storage. The two analogues selected were the Werkendam Field in The Netherlands and the Brae Area of the UK North Sea because:

- The reservoir rocks had mineralogies comparable to many of those identified as potential storage sites in Europe
- They were known to contain significant amounts of gas with variable CO_2 concentrations
- Nearby sites, experiencing similar diagenetic histories but without CO_2 exposure, allowed the identification of reactions attributable specifically to CO_2
- Samples of reservoir material were available for detailed petrographic and geochemical studies
- Pre-existing data and previous studies enabled the results to be placed in a wider geological context.

Several geochemical data measurements (x-ray, MEB, isotopes, …) have been performed these natural analogues samples to evaluate the origin of the gas, the chemical reactions that occurred and the potential of mineral trapping for such geological formations.

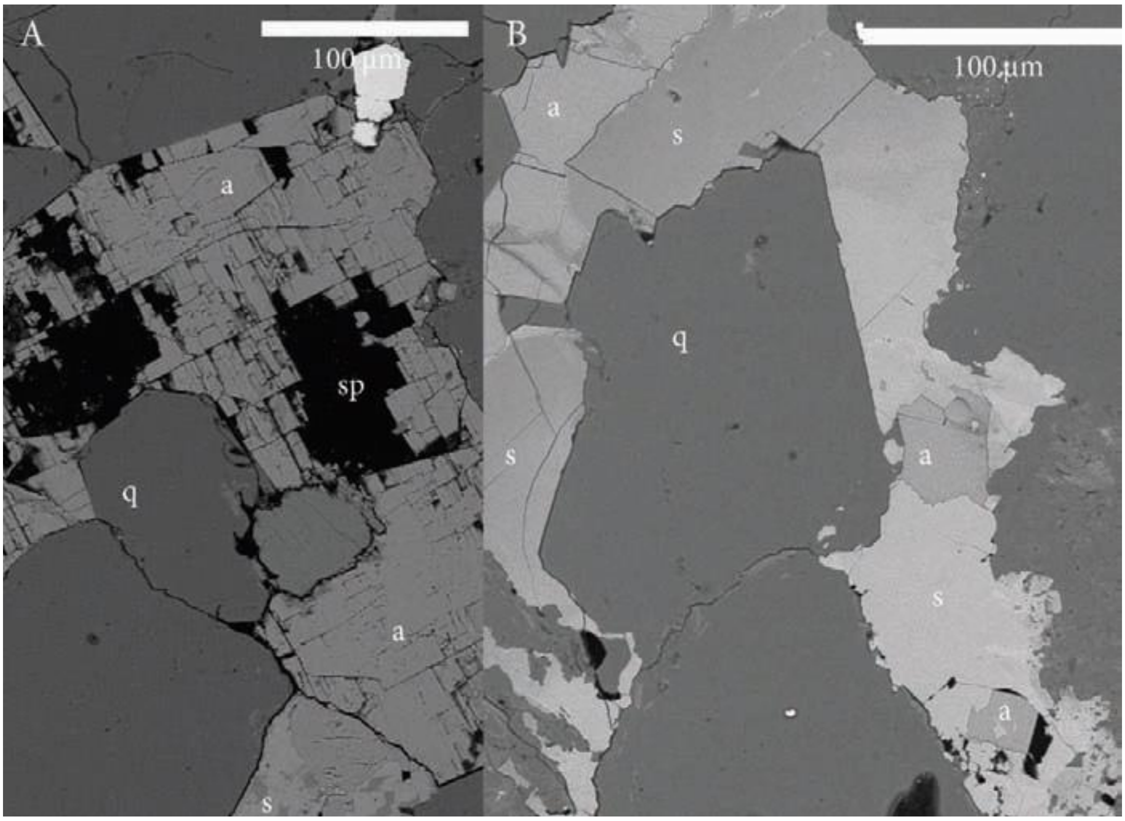


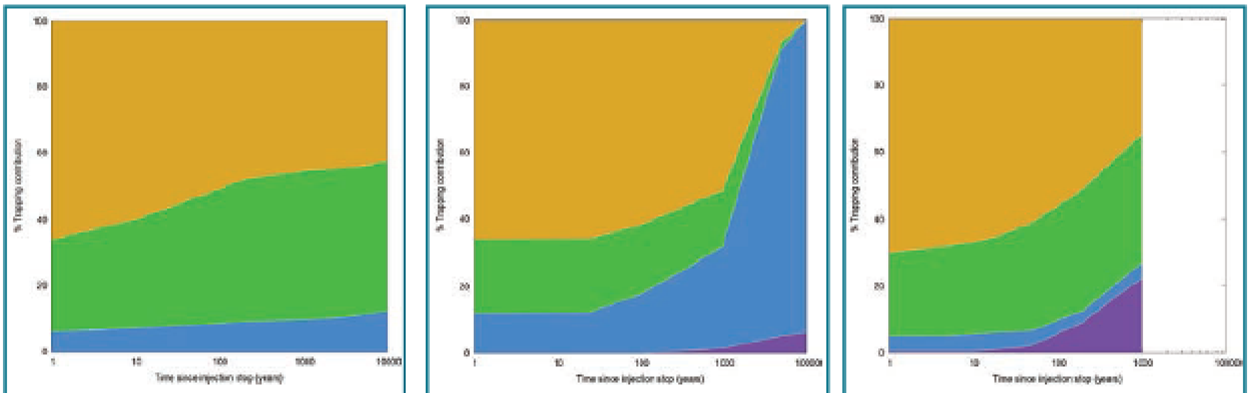
Figure 6: Petrographic images from Werkendam illustrating anhydrite cement that is engulfed by later siderite cement (B). The siderite post-dates the partly dissolved anhydrite cement. Dissolution of anhydrite creates secondary porosity (A).

The presence of CO_2 in the Brae Area of the UK North Sea and in the Werkendam Field in The Netherlands demonstrates that CO_2 can be successfully trapped over 10s of millions of years. In both cases, emplacement of CO_2 has not resulted in complete trapping in new mineral precipitation. This indicates that CO_2 storage in similar reservoirs is unlikely to be able to rely on mineral trapping as a significant contributor to permanent containment. Structural trapping, residual trapping and dissolution into the formation water, and any hydrocarbons if present, will be far more dominant mechanisms in both the short and long term.

Attributing analogue mineral precipitation to CO_2 is challenging. Careful analyses are necessary to provide evidence of new mineral precipitation that can be definitively attributed to exposure to higher CO_2 concentrations. Identifying any response from elevated CO_2 therefore requires the development of multiple lines of evidence, including comparisons with similar systems that have not been exposed to naturally occurring CO_2.

1.5. Trapping stability estimates

In order to fulfil the requirements of the CCS Directive, evidence that the storage site is evolving towards stability must include an evaluation of storage security, i.e. the potential for migration, escape or leakage of CO_2 over the long term <10,000 years. This includes situations where the CO_2 can be migrating slowly, dispersed and dissolved. Very few attempts have been made to achieve full coupling and simulation over the very long time periods required. In order to limit the cell number for large-scale models, large cells are necessary. To compensate for the resulting overestimation of CO_2 dissolution, post-simulation calculations can be applied. In a method developed by Green and Ennis-King, the over-estimation of CO_2 dissolution during injection is estimated by assuming that dissolution only takes place in cells with gas saturation above a certain threshold. ULTimateCO_2 applied this method to the Bunter Sandstone case during the injection period. The over-estimation is rather limited in that case (due to the high salinity, CO_2 dissolution is moderate) and decreases with time. Structural–Residual–Dissolution–Mineral (SRDM) trapping diagrams can be produced presenting a simplified reservoir response to the injection of CO_2. ULTimateCO_2 created a type-model of a closed structure, including some flank migration. Dissolution and diffusion are incorporated and residual trapping is reflected in the hysteresis. A meta-analysis of published results was performed to the sites studied by ULTimateCO_2 and to illustrate the process of generating trapping diagrams. Material (data) was drawn from published trapping charts of all kinds, particularly those that span long (10,000 year) time periods. ULTimateCO_2 data for the GeoLorraine and North Sea Bunter Sandstone cases have been analysed in the same manner and present two very different reservoir responses. This is due to both the different types of structures concerned (sloping aquifer and closed structure) and the simulation tools used. In particular, geochemical trapping was not included in the North Sea Bunter case. In that case, mineral trapping contributes relatively little because the system is highly dynamic and never reaches an equilibrium state that favours large-scale mineralization, and also sandstones, compared to carbonates, are relatively unreactive. ULTimateCO_2 produced trapping diagrams based upon actual modelling results. They show, in all cases, that large quantities of injected CO_2 remains in a supercritical form even after 10,000 years and that, in many cases, mineralization is limited even in the very long term. This important conclusion supports evidence from many natural analogues studies.



- North Sea Bunter case, without mineralization
- Closed domal structure
- A very large proportion of CO_2 remains in gaseous form, either structurally or residually trapped

- Sleipner storage site
- Flat aquifer
- Convection develops over the long term, giving rise to massive dissolution.
- Re-plot of the trapping diagram of Audigane et al., 2007[63]

- GeoLorraine case
- Tilted aquifer
- A large amount of CO_2 remains in supercritical form

Figure 7: Simulations for the Bunter and GeoLorraine case studies up to 1,000 years (without mineralization), showing supercritical CO_2 (orange), residually trapped CO_2 (green), dissolved CO_2 (blue) and mineralized CO_2 (purple).

2. Long term sealing integrity of faulted or fractured caprock (WP4)

As long as supercritical CO_2 is present in the storage site, buoyancy forces tend to drive it upwards. The caprock must therefore keep it contained until it dissolves. This can last thousands of years during which time the caprock will undergo geological processes (e.g. developing tectonic stresses, creating and propagating faults, undergoing pressure changes due to compaction and diagenesis, and slow chemical reactions between acidic CO_2 and the caprock and gouge material). Impurities present in the CO_2 complicate the reactions and may have a significant effect on the processes involved. A careful description of caprock characteristics is required for every potential storage site prior to injection – starting with an assessment of the baseline state. Examples of how this can be achieved are given below, including the study of natural caprock analogues. Laboratory experiments using Opalinus Clay (a typical caprock material) provide a good understanding of flow mechanical- chemical interactions. However, in order to understand long-term processes, numerical modelling exercises are essential.

2.1. Industrial analogues review

ULTimateCO_2 has reviewed published and unpublished data on the likely properties (mineralogy, flow properties and architecture) of faults and fractures in and around potential CO_2 storage sites. Faults in porous, normally consolidated, sedimentary rocks are not generally surrounded by a high permeability damage zone. The mineralogy of potential storage sites is quite limited and relatively unreactive in a closed system (i.e. they are quick to reach equilibrium with the pore fluid after which no further reaction occurs). Key gaps include:

- Information on the structure and flow properties of faults in the caprock in potential storage sites.
- Key mechanical properties of shale-rich caprocks (e.g. the apparent preconsolidation pressure) which would allow an assessment of leakage risk via faults and fractures.

Overall, all leakage mechanisms identified have low or very low risks. The largest risks identified have been leakage along natural fractures within the overburden and via faults formed either naturally or due to human activity.

Recommended future work: There is a distinct lack of data on the structure and flow properties of faults in the caprock to potential storage sites. This is partly due to the lack of core and rapid weathering of such material in outcrop. However, online resources indicate that some good exposures of faults in caprocks could exist (e.g. Mercia mudstone in UK) which could be analysed further. Key mechanical properties of shale-rich caprocks (e.g. the apparent preconsolidation pressure) would allow an assessment to be made of the risk of leakage via faults and fractures.

2.2. Measurements of the impact of stress state and fluid penetration on caprock sealing capacity

ULTimateCO_2 performed experiments in which Opalinus Clay was first subjected to shear stress until it failed to create fractures. The roughness of these new fracture surfaces was then determined. After rehydrating the fracture plane, a range of normal (perpendicular) and shear (parallel) loading conditions were then applied to the fractured caprock and changes to its mechanical properties measured. The following properties were examined:

- Hydraulic or fluid transport properties under different normal loads

- The influence of shear on hydraulic/gaseous transmissivity and the ease with which fluids can migrate through a material
- Clay swelling properties
- Fault reactivation
- Transmissivity and transport mechanism/behaviour for a 2-phase (gas + fluid) flow in intact rock and fault gouge material, i.e. the material along the fault surface that results from wear along the fault.

Experiments on a shale deposited in a marine environment showed that shearing along artificially 'prefabricated' faults is likely to be an effective self-sealing mechanism for water. This aligns with current understanding of the influence of clays and clay smearing in North Sea faults. More data is needed, but these early results are promising, as they point towards a tendency for slip on reactivated faults to encourage self-sealing, rather than increase fluid flow along the fault. Sliding along reactivated faults seems to encourage self-healing. Once sheared, fracture transmissivity was marginally greater for gas compared to water. Shearing caused no change in the gas entry pressure, suggesting that the physics that govern the first movement of gas is not significantly altered. However, the form of pressure response to repeated gas injection is not consistent. This suggests that whilst the formation of gas flow pathways occurs at a reproducible pressure, the distribution of these pathways is much more random, resulting in different fracture transmissivities. The evidence for reduced flow of water is compelling nonetheless. Fault reactivation or failure leads to an initial increase in transmissivity/permeability, but this effect generally decreases with ongoing shear. Evidence has been found that stress history influences the response of caprock to loading. Fracture transmissivity was shown to decrease with increasing normal load in a non-linear fashion. These findings for fractured rock are also consistent with observations made on clay gouge material. On unloading, very little recovery of fracture flow was observed, suggesting that the long-term integrity of faults will partly depend on the stress history and previous burial or exhumation. Stress history influences the long-term integrity of faults (i.e. the likelihood of fluid migration along faults).

ULTimateCO_2 studied fault reactivation at laboratory-scale by locally elevating the pore pressure on a fracture plane. Fractured caprock, representing new fractures, was then examined. Parallel testing on clay gouge, representative of fractures that have accumulated shear was carried out in very similar apparatus. This is undoubtedly the result of contributing factors such as surface roughness and properties of associated asperities present, as well as damage distribution along and off the fracture plane. In the laboratory, the consequence is that for a close-to-critically stressed fractured caprock, a greater degree of instability was observed compared to tests conducted on gouge. How well any quantified observations may upscale to field conditions is unclear and emphasizes the complexity of upscaling simplified laboratory tests to much larger damage zones at field scale. Fractured caprock shows more unstable behaviour than fault gouge indicating that fault maturity (i.e. the amount of previous slip accumulated) is important for controlling fault slip stability. As with testing where a shear load is applied to the sample, where slip occurs in Opalinus Clay as a result of reactivation, the results indicate a strong tendency towards reduced transmissivity. All experiments using Opalinus Clay samples indicate that slip generally encourages fault sealing – a response that appears to be permanent, at least on laboratory timescales (i.e. days). Below, a more detailed presentation of the experiments is given.

2.3. Elastic and ultrasonic properties of faulted host rock

For this work, a retarded acid solution (which is only activated at specific temperatures) was injected. The main challenge, caused by low permeability, was to attain contact between the acid solution and the reactive minerals. Three methods were tested to achieve this, while maintaining integrity of the sample:

1. Applying triaxial loading without reaching failure
2. Drying and re-saturating samples to generate microcracks (drying alone did not cause cracks in the caprock to develop)
3. Cycles of freezing

➢ *Indentation tests*

Indentation experiments were performed using an indentation test arrangement which measures the hardness of rock to deformation. Indentation experiments are useful for deriving static elastic parameters and seeing whether this is affected by the preserved state of samples.

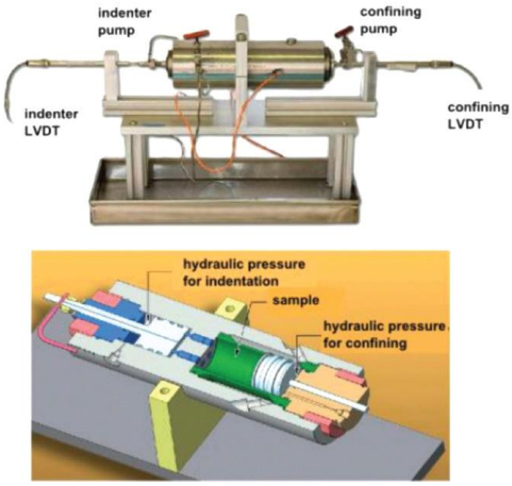


Figure 8: Experimental apparatus used for indentation tests

Two preserved states were used: samples with 5.4% water saturation ('preserved samples') and those with 1.4% water saturation ('unpreserved samples'). Different confining pressure states were tested (5, 10 and 15 MPa). ULTimateCO2 obtained a very stable Poisson's ratio (ν), between 0.17 and 0.18. The Young modulus (E) is also fairly constant, around 4.5 GPa +/- 10%, consistent with other mechanical tests on Opalinus Clay samples. The cohesion parameter differs between the unpreserved and preserved Opalinus Clay samples: the unpreserved have a cohesion of c.5 MPa, whereas the preserved have a cohesion of c.3 MPa (no such observation was made on the internal friction angle). Finally, the Opalinus Clay samples appear to be more brittle at preserved confining pressure states.

➢ *Ultrasonic velocity and cyclic loading*

The speed at which ultrasound (P- and S-waves) travels through caprock indicates whether cracks are present: slower velocities generally indicate more cracks. After the drying procedure described above, a P-wave was not detected and S-wave velocities had the same value as without drying. Freezing, on the other hand, caused a preferential crack network to develop in the direction of the bedding; axial P-wave and S-wave velocities increased, while ultrasound velocities parallel to the bedding plane decreased. Ultrasonic velocities were also measured as a function of increasing axial stress. An increase in S-wave velocity by c.5% between 10 MPa and 37 MPa of axial loading was observed, probably due to a strengthening of the interfaces between shale layers. A classical triaxial press was used to derive the shear and compression resistance of the rock. The result: a Poisson's ratio of c.0.1 that is roughly constant for all samples, but relatively low according to literature data. Young's moduli were also fairly constant for dry samples or those saturated with brine: 4-6 GPa. However, the sample saturated with an acid (to represent contact with CO_2) showed a Young's modulus of c.2.3 GPa. This suggests that the acid solution could reduce the stiffness of the clay. However, this procedure is difficult to use and numerous manipulations could create mechanical damage.

2.4. The impact of CO_2 and CO_2-brine on the mechanical and transport properties of caprock

This series of experiments was set up to study:

1. The effect of normal stress, shear displacement and changes in mineralogy on the fluid transport properties of gouge-filled faults (Effect 1)
2. The effect of changes in mineral composition and temperature on the frictional behaviour of gouge (Effect 2)

The aim in both cases was to clarify how shear displacement and mineralogical changes caused by interactions between the CO_2, fluid and caprock impact fault friction, slip stability and the potential for fault reactivation and permeability. To determine the transport properties (Effect 1), a new permeability set-up was developed that allowed for measurements across and along simulated faults of Opalinus Clay gouge during active shearing, in a direct shear configuration. This aimed to clarify how shear displacement, and changes in mineralogy due to CO_2, influence fault friction, slip stability and the potential for fault reactivation and permeability.

➢ *Effect of normal stress and shear displacement on gouge permeability*

The permeability of 'simulated' clay-rich gouges decreased as normal stress and shear displacement increased – independent of the direction of flow. The decrease in permeability was greater after dynamic shear than after a period of hold, the latter simulating a period of no slip between fault movement. Permeability was anisotropic (shows a directional dependence), with across-fault permeability an order of magnitude lower than along-fault permeability. This is presumably due the development of an internal fault-parallel foliation (i.e. minerals aligned in a planar fashion, homogeneously distributed throughout the rock volume) that allows easier flow parallel to the fault than perpendicular to it.

Increasing normal stress and shear displacement decreased fault gouge permeability. The decrease in permeability in the first 2-3 mm of shear displacement accounted for the majority of the overall reduction, with additional shear displacement causing relatively little additional reduction in porosity or permeability. Other studies suggest that some clays (i.e. smectites) may swell, causing an additional drop in permeability by as much as an order of magnitude when CO_2 is used as a pore fluid. This may result in self-healing. High fluxes of dry CO_2, on the other hand, cause clay minerals to shrink and could result in leakage. This means that the mechanical and transport properties data obtained in these short-term experiments should be valid for the long term. Thus clay-rich, and especially smectite-bearing faults and fractures, show a tendency to self-heal due to clay swelling in the presence of CO_2, provided there is no desiccation by dry CO_2. Geochemical modelling and experiments showed that for typical caprock material (i.e. clay-quartz-carbonate), chemical reaction with CO_2 had only minor effects on the mineralogy and mechanical and transport properties, compared with the natural variability observed in the materials studied and with clay-rich caprocks in general.

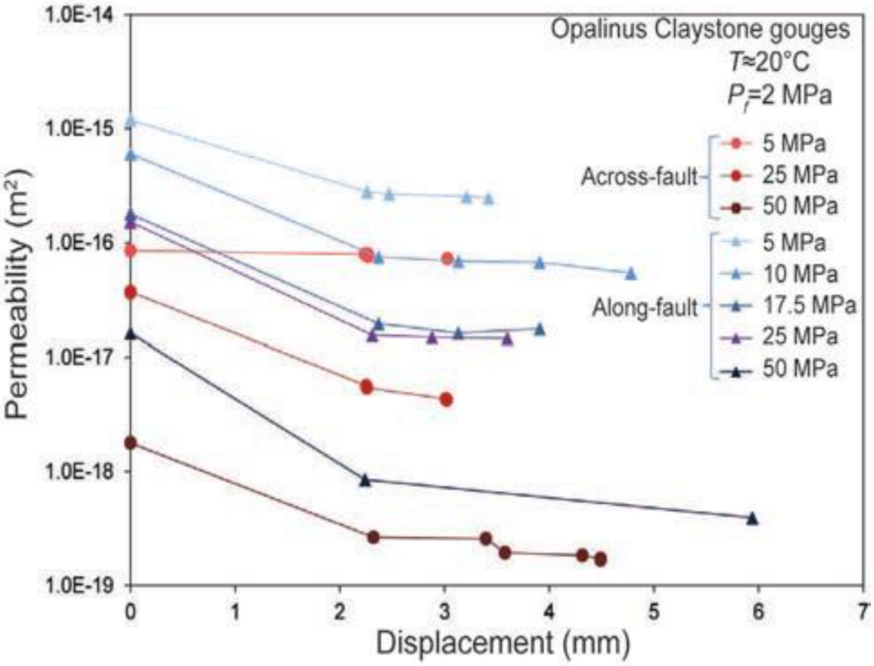


Figure 9: Fault permeability as a function of displacement for simulated clay-rich fault gouge prepared from crushed Opalinus Clay without leaching (see text). The permeametry experiments were performed at room temperature and a pore fluid pressure, Pf, of 2 MPa (argon gas).

➢ *The effects of changes in mineralogical composition and temperature on the frictional behaviour of Opalinus Clay gouge*

How the frictional strength of fault gouge responds to changes in mineralogical composition and temperature after CO_2 injection (Effect 2 above) was addressed in similar tests to those used earlier to examine normal stress, shear and mineralogical changes (Effect 1), but with varying amounts of calcite also added to the Opalinus Clay gouge. The temperatures were 20, 100 and 150˚C. This aimed to account for the presence of relatively large amounts of carbonate, either because it was originally present in the caprock, or because it has been deposited through reactions with CO_2 in the presence of a divalent cation source (i.e. Ca2+). ULTimateCO2 found that frictional strength increased as the calcite content in the samples increased. The effects of temperature (since temperature increases with depth) on the potential for faults to slip (slip stability) were also studied by recording the conditions under which unstable (stick-slip or microseismic) behaviour was observed. Stick-slip events (i.e. the laboratory equivalent of earthquakes) were observed only at high carbonate content (c.80-90%) and temperature (100 and 150˚C); a carbonate-dominated clay-bearing fault at c.4 km depth may therefore exhibit unstable slip behaviour.

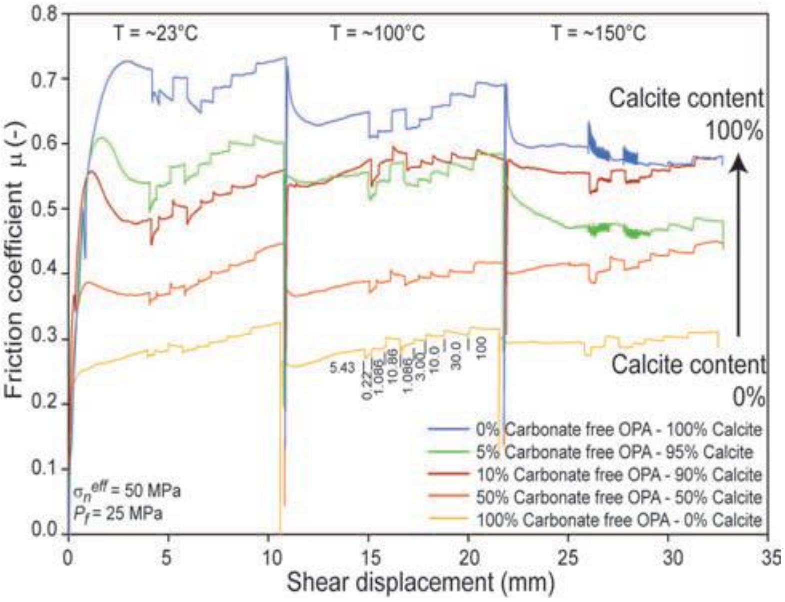


Figure 10: Evolution of the friction coefficient with shear displacement for the experiments on different Opalinus Clay plus added calcite mixtures. The velocity steps employed are indicated (in µm/s) and marked with vertical 'ticks' above the 100% (untreated) Opalinus gouge curve.

ULTimateCO_2 has found that frictional strength tends to increase with increasing carbonate concentrations. It also found that faults will generally not be prone to unstable stick-slip behaviour except at high carbonate contents. However, in these systems, fault strength is relatively high and reactivation more difficult. Microphysical modelling suggests that this unstable slip results from a competition between shear-induced processes increasing and decreasing the porosity, involving stress-driven dissolution and re-precipitation (pressure solution) of calcite. The model provides rough quantitative constraints on the slip rates at which faults will be stable rather than unstable, and permeable rather than impermeable, as well as providing a method for calculating the timescales on which faults reactivated by CO_2 storage are likely to heal by pressure solution mechanisms. However, upscaling simplified laboratory tests to much larger damage zones at field scale is highly complex and should be treated with caution.

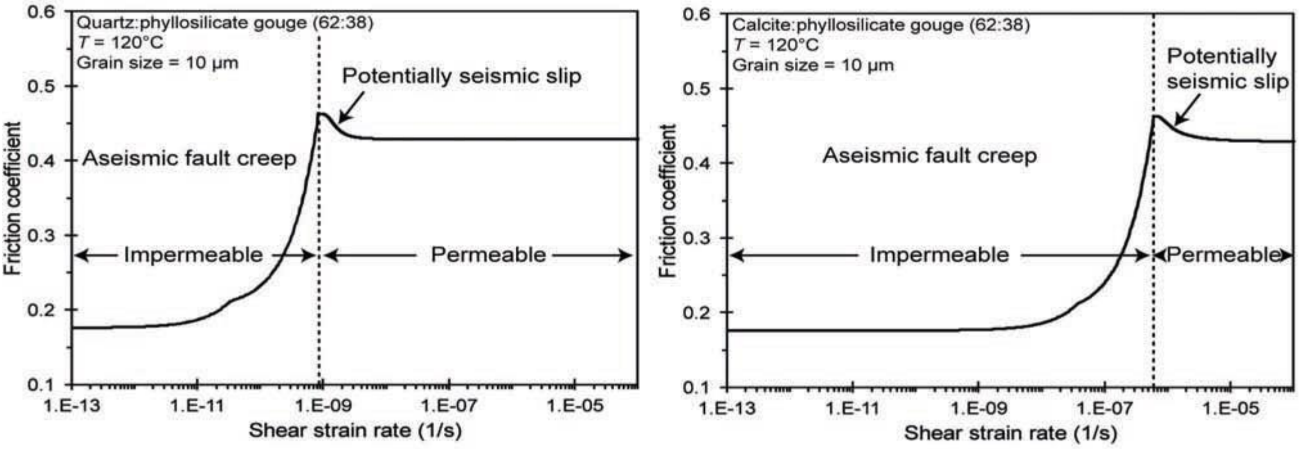


Figure 11: Preliminary model predictions for phyllosilicate-quartz (left) vs. phyllosilicate-calcite (right) fault gouges, showing the sliding velocities at which the behaviour is expected to be seismic vs. aseismic and permeable vs. impermeable. The calculations were carried out for a temperature of 23°C, an effective normal stress of 50 MPa and a gouge layer thickness of 0.5 mm.
The microphysical model used was developed at Utrecht University and modified to account for the

diffusion controlled pressure solution (right), as opposed to the reaction controlled pressure solution originally used (left).

2.5. Modelling to assess whether chemical reactions affect the mechanical integrity of caprock

In ULTimateCO_2, modelling methods were developed that account for coupled hydraulic-mechanical chemical processes, addressing the problem from a mechanical perspective. This was applied in order to evaluate the role of chemistry on the mechanical integrity and flow properties of fractured caprock. ULTimateCO_2 studied the combined effect of chemical and mechanical processes on the long-term stability of caprocks (intact or fractured) and identified important processes in space and time, for example the reactive kinetics that affect the degradation or healing of altered caprocks. ULTimateCO_2 found that, under specific conditions, the CO_2-induced increase in porosity can make the caprock material more collapsible, which may favour the onset of ductile deformation, which in turn may lead to a decrease in permeability, thereby limiting leakage.

➢ *Application to Opalinus Clay case study*

Reactive transport models and mechanical models were coupled in order to predict chemical reactions of pure, or impure, CO_2 along a flow path. Mechanical models examined shear reactivation of a clayey fault gouge due to an increase in pore pressure resulting from porosity/permeability degradation induced by pure or impure CO_2-rock reactions.

The coupled model was then applied to two situations in which a gouge fault may be reactivated by CO_2 through: a) changes in porosity/permeability and b) fluid intrusion. Shear reactivation of a clayey fault gouge due to an increase in pore pressure resulting from porosity/permeability degradation induced by dissolved pure or impure CO_2-rock reactions was modelled. The coupled chemicaltransport-mechanical model addresses the short- and long-term effects of pure or impure CO_2 on water-rock interactions and, consequently, the mechanical stability of faulted caprock. It is based on iterative steps between the chemical reactions between fluid and rock modelled by PHREEQC™ and the flow modelled by MRST.31 Using the calculated pressures produced, mechanical stability is assessed using simple slip tendency analysis to indicate the reactivation potential of the fault. The model performance indicates that the presence of pure CO_2 results in such low porosity increases that it has no distinguishable effect on the caprock mechanics.

ULTimateCO_2 went on to examine the effect that trace components in the CO_2 stream may have on caprock mechanics. By following chemical reactions between water and rock in the presence of CO2 that contained SO_2 as an impurity, SO_2 was found to have caused minor additional increases in porosity, and thus a minor effect on the mechanical strength/stability of the caprock. This may make the structure of the fault/fracture more likely to collapse and effectively heal itself, which will limit leakage. On the other hand this may still affect the mechanical stability of the fault, even though the model indicates mechanical stability is secure. Therefore the purity of the CO_2 stream may still be an important factor in ensuring integrity of the caprock in the long-term. Numerical coupled transport-chemical models were also used to simulate CO_2 intrusion into a fractured caprock and the matrix surrounding it. The fractured caprock was idealized here by inner high permeability/porosity pathways surrounded by outer damage zones of lower permeability and porosity. The outputs of these simulations (mineral fraction, porosity changes, gas saturation, pore pressure) were then converted into mechanical changes.

Three main processes were considered:

- The effect of capillary pressure (suction) on the material strength
- The effect of the mineral fraction and porosity evolution
- Alteration of the poroelastic properties.

Depending on the assumptions for damage zone properties (in particular, its initial damage state and over-consolidation ratio) and magnitude of the degradation, the composite structure showed a tendency to ductile deformation. Contrary to brittle deformation, which is characterized by the formation of distinct shear failure surfaces (i.e. an increase in permeability corresponding to a worst case in terms of risk of leakage), ductile behaviour is characterized by a more distributed deformation making the fractures contract (i.e. leading to reduced permeability with increasing shear deformation) which should help limit the magnitude of the leakage. This demonstrates the need to consider the over-consolidation ratio, an important factor when screening rock formations as suitable caprocks for CO_2 storage. In the mechanical processes and storage scenarios examined, chemical reactions between water and rock in the presence of CO_2 that contained SO_2 as an impurity resulted in only a minor increase in porosity and thus a minor effect on the mechanical strength/stability of the caprock. The results also suggested that impure CO_2 (resident reservoir fluid + CO_2) may make the structure of the fault/fracture more likely to collapse and effectively heal itself, which would limit any leakage. Overall, long-term geochemical effects, and other effects on the hydro-mechanical behaviour and transport properties of fractured or faulted caprock during CO_2 storage, are unlikely to increase the occurrence of leakage. Coupled modelling is a powerful tool for studying effects and interactions between flow, mechanics and chemistry in order to understand leakage in terms of flow through a fractured caprock. However, it requires specialist knowledge of:

1. The chemical system:
    a. What reactions occur?
    b. How do these reactions change as pressure and temperature change?
    c. How do different chemical compositions of fluid and rock affect flow?
2. The mechanical system:
    a. Do faults exist?
    b. Is stress close to failure?
    c. Is the rock brittle or ductile?
    d. How do rock properties change on failure or collapse?
    e. What impact does the chemical composition of fluid or rock have?

ULTimateCO_2 advocates a phased approach to building and using coupled models, beginning simply (e.g. 1D flow, limited number of chemical species, etc) and building up the complexity gradually towards a more representative, complex model of a real storage site.

2.6. Studying natural sandstone analogues representative of CO_2 storage sites

Natural analogues can provide insights into long-term geological processes. CO_2-brine-rock interactions in the reservoir may have been active for 10s of thousands of years – timescales not reproducible in laboratory experiments. Natural CO_2 fields can be found both on- and offshore (e.g. Werkendam, onshore near Rotterdam, the Netherlands; Brae and Miller Fields, offshore in the North Sea, the UK). CO_2 emplacement is commonly related to (late) volcanic exhalations and may be present in a supercritical form (gas cap), or dissolved in the reservoir brine. The latter is the case for many mineral water provinces in central Europe. Fault outcrops, in formations representative of CO_2 storage sites, can also be used to parameterize fracture network characteristics needed to investigate the potential for leakage. Such fracture network studies can be undertaken on onshore outcrops, or using exposed walls in underground mines and tunnels. Typical caprocks such as shales and mudstones are strongly affected by weathering under surface conditions which make them unsuitable for outcrop studies. Exposed fracture networks in sandstones may be used; for shales, underground outcrops, such as mine shafts and tunnels, are also an option. However, due to the mechanical instability of these rocks, such cavities are commonly covered by shotcrete or support beams.

Figure 12: Field operation of the terrestrial laser scanning (TLS) system, February 2014

N.B. In order to translate observations from natural analogues into numerical flow and transport models, some simplifications are required due to the complexity of natural systems which cannot always be captured in numerical simulations.

➢ *Natural CO_2 fields: showing the effect of exposure to CO_2*

In studying natural CO_2 analogue fields, it is pertinent to compare rock samples that have been exposed to CO_2 (e.g. drill cores or outcrop samples) with samples from the same geological formation which were not exposed to CO_2. During exposure, CO_2 or other trace gases (e.g. such methane, H_2S) react with the primary minerals present in the formation, potentially altering its mineral content, porosity and permeability. Mineral trapping of CO_2 by Mg-, Fe- or Ca-rich carbonates and chemical alteration of the overlying caprock are important processes in ensuring the long-term security of CO_2 geological storage. It should be borne in mind that comparison of rocks from the same stratigraphical unit, either unexposed or CO_2-reacted, is key to evaluating the effect of CO_2-exposure on mineralogical composition, though natural (lateral) variability should be taken into account. The mineralogical and geochemical comparison of rock samples was performed on drilled cores from two sites, some 60 km apart: Gemmingen and Bad Teinach, in south-western Germany. The site at Bad Teinach was selected because of its proximity to a well-known area of deep crustal CO_2-exhalations, resulting in numerous utilized mineral springs. Exposure to moving CO_2-rich fluids and trace gases would therefore be expected. Samples from Gemmingen served as the unreactedcounterpart. Thin sections from rock samples obtained from the Bad Teinach site show strongly corroded feldspar grains through the rock and elevated carbonate contents in individual samples. Both sites showed red (oxidized) and green (bleached, reduced) zones, occurring in close proximity. This bleaching could be attributed to the presence of CO_2 and associated trace gases. However, the green colouring is not associated with a depletion in iron mass, but indicates a change from iron oxides (red) into greenish FeTi-oxides and diagenetic illite clay (green). CO_2 exposure did not cause these diagenetic changes. Furthermore, the natural geochemical variability between and within the two sites is fairly high, with the porosity of the green samples commonly slightly lower than that of the red samples. Overall, it is not possible to relate mineralogical differences between green and red cores to different levels of exposure.

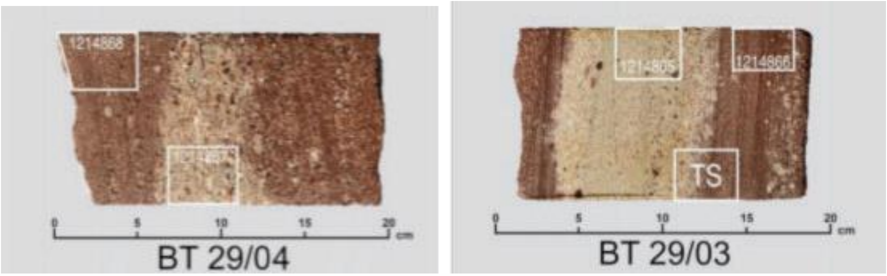


Figure 13: Example of thin sections from the site near Bad Teinach

➢ *Using natural analogues to evaluate long-term sealing capacity*

In order to obtain an accurate impression of fracture properties, a sandstone outcrop near Rastatt in south-western Germany was selected. Despite somewhat different mechanical properties, the sandstones of the outcrop were affected by the same tectonic stresses as neighbouring shales which are the type of caprock that would be considered for CO_2 storage. The fractures outcropping in this sandstone quarry are located in a CO_2-rich mineral water province in southwestern Germany, near Rastatt. Important fracture network characteristics measured include spatial orientation, spacing, aperture, interconnectivity and roughness of the sandstone fractures. These measurements were included in a geological geometry model, capable of describing the movement of fluids through the subsurface. N.B. Geometrical models of fracture distribution are necessarily a simplification of reality due to the multitude of parameters and their natural variability.

➢ *Modelling parameters*

- Spatial orientation and spacing (length, distance) of fractures were investigated in a sandstone quarry in the Rastatt region in south-western Germany along several transects. The field data were translated into a digital geological model which is available for numerical modelling of flow and transport processes in a fractured formation.
- Interconnectivity of fractures, important for fluid flow and transport, were obtained from a statistical assessment of fracture length and orientation.
- The geometry model contained simplified assumptions on the fracture aperture.
- An aperture: a transport-relevant parameter required to model fluid flow in fractures correctly using a ‘Cubic Law’, often used to correlate fracture aperture and permeability. Here, the aperture sizes followed a normal distribution, something commonly recorded in literature for fractured rock sites. However, the measured aperture values cannot be transferred directly into a numerical flow and transport model, as they were obtained from a surface outcrop. In a real-life situation, normal stress acting on the fractures and fault tend to lead to fracture closure, or at least a reduction in fracture aperture. Any model therefore needs to include a depth-aperture function which can be obtained from literature.
- Surface roughness of fractures: a transport-relevant parameter is also required to model fluid flow in fractures correctly using the Cubic Law; high surface roughness negatively affects the fluid flow due to resistance, but can help keep fractures open, at least partially, against a

confining pressure. Roughness is a cumbersome parameter as it is a function of the area of measurement and thus dependent on the scale being considered. This can probably be related to the fractal dimension of roughness, i.e. to the different scales of roughness-creating features in the rock (e.g. fine crystal precipitates, bulk rock grains, micro and macro cleavages). Modellers who wish to include fracture roughness therefore need to define a size-roughness function, similar to that commonly undertaken for dispersion.

3. Long term near well sealing integrity (WP5)

The integrity of wells is very important to long-term CO_2 storage as they can be direct pathways towards sensitive stake (shallow potable water aquifers, surface). The initial bounding quality between interfaces of well elements (clay–cement–casing) is recognized as a key factor affecting a well integrity. During the lifetime of a well, its integrity can also be modified by various constraints due to pressure and temperature changes, and chemical interactions – especially contact with CO_2.

ULTimateCO_2 built a full-scale section of a well into a typical caprock (Opalinus Clay) at the Mont Terri Underground Laboratory in Switzerland. The well was exposed to CO_2-rich water with temperature and pressure conditions changed to imitate typical site operations. Well integrity was then monitored for changes in permeability and geochemistry.

Key chemical reactions affecting the long-term containment capability of a casing-cement-caprock system, in terms of the physical and geochemical properties of its materials, were then identified. Finally, key chemical reactions potentially modifying the long-term water quality of shallow groundwater resources affected by CO_2 intrusion from a storage site were identified.

3.1. Long-term sealing properties of the wellbore

An experimental study was carried out to follow the evolution of well integrity over time in response to changes in well conditions – similar to those to which an abandoned well might be subjected during its lifetime and after its abandonment. This particularly focused on the consequences of changes in pressure, temperature and fluids in contact with the well. The well assessment was made in situ at shallow depth and in a caprock-like formation (Opalinus Clay at Mont Terri. The innovativeness of this approach was that observations were made continuously at field scale, allowing all the components of a well-close environment to be integrated, using classical geometry and materials for a realistic well, but with significantly more detailed instrumentation than is usually in place at a genuinely active or abandoned well. This enabled access to parameters usually difficult to measure, e.g. hydraulic conductivity, control of pressure and temperature conditions, and the geochemical context. The experimental protocol included the quantification of the well's hydraulic properties and how these evolve over time, the sampling of fluids (pore waters) to follow changes in their geochemical composition and the over-coring of the system to inspect changes (notably mineralogical) in the well-close environment.

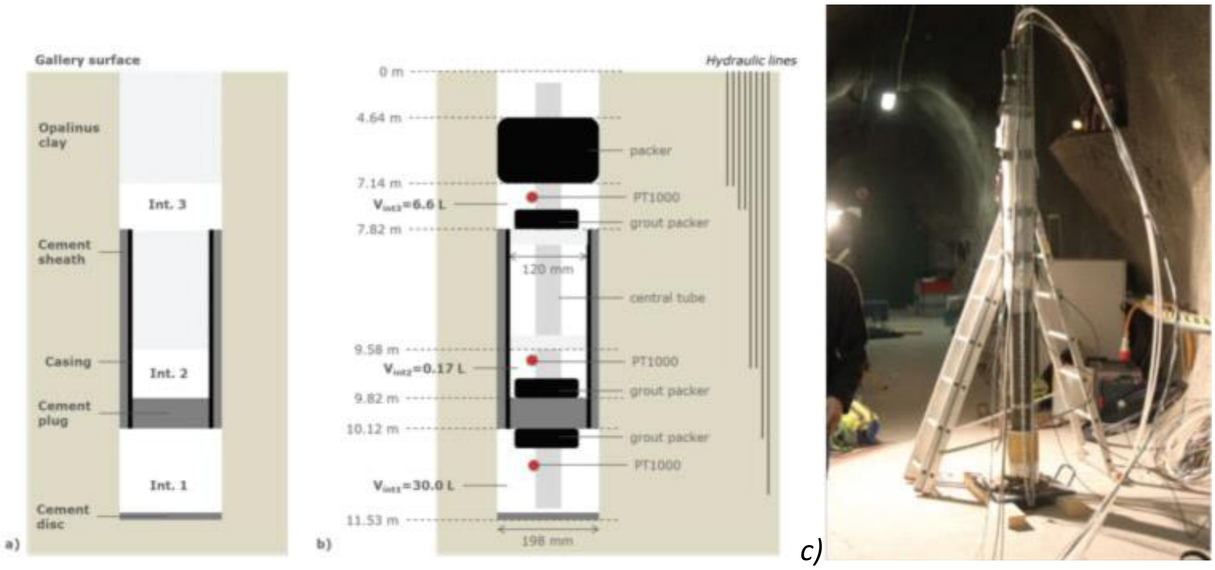


Figure 14: a) Concept of the experimentation showing the observation intervals (not to scale). b) Technical layout of the completion; only the hydraulic lines used for pressure monitoring, fluid extraction and injection are represented (not to scale) after Manceau et al., 2015.32 c). Experimental set up at the Mont Terri Laboratory

The results confirmed the ability of the selected design and observation scale to estimate, comprehensively, the evolution of well integrity over time, flow characteristics along the well-system and the reasons for the observed evolution:

1. The initial effective permeability of the well was higher (c.20mD) than the cement's or caprock's
2. own intrinsic permeability. This suggested that preferential flow pathways existed at the very beginning of the experiment which was later confirmed by following the geochemical evolution of the fluid and the observation that well integrity depends on pressure. This means that effective well permeability would be partly, possibly entirely, due to the flow through the interfaces between the caprock and cement annulus – reinforcing the significance of the cementing process during well construction.
3. Well integrity or effective well permeability can be influenced significantly by changes in temperature and pressure. Increasing temperature decreased effective well permeability drastically – by three orders of magnitude. Significantly increasing pressure at the well bottom increased effective permeability by more than two orders of magnitude. This indicated that operations-induced stresses could influence well integrity as much as the cementing process. These effects (abrupt changes in pressure) were significantly diminished after dissolved CO_2 was injected into the system. This could be due to reactions with carbonated brine at the interfaces that limit flow and improve well integrity, even with marked pressure changes. Consumption of dissolved CO2, as it flowed through the well, was also noticed which would confirm the occurrence of such geochemical reactions.

N.B. The pressure and temperature variations studied here (10 bar and 28 bar, and 17°C and 52°C) represent only a limited range of subsurface conditions.

The caprock formation, as well as the drilling and cementing processes, are specific to this experiment and cannot be more widely generalized. Any upscaling of these observations should therefore be considered with caution.

In parallel to the Mont Terri experiment, autoclave experiments of one-month duration were performed in the laboratory in order to evaluate the consequences of the interactions of the well elements (Opalinus Clay and Class G cement stone) with CO_2-enriched Opalinus Clay pore water under P/T conditions close to the Mont Terri experiment (20 bar total pressure and 30°C), and also under more relevant CO_2-storage conditions (100 bar total pressure and 40°C). The conclusions were as follows:

- Opalinus Clay caprock shows a dissolution of carbonates, especially calcite. Considering the near-well zone of a regular CO2 storage well, this could be an issue especially at the caprock/cement stone interface as carbonate degradation could create pathways along the interface aggravating the sealing properties.
- Class G cement stone shows carbonation, but the effects of this process have to be considered carefully. On one hand, the carbonation process is associated with a decrease in the total pore volume of the cement stone, which in turn can decrease its permeability and therefore improve sealing efficiency.

On the other hand, the volume increase could lead to crack formation inside the cement stone, which can increase its permeability and therefore worsen its sealing efficiency. A further negative effect is the decrease in the pH resulting from the carbonation. The pH decrease would result in a loss of corrosion control, in this context with regard to the casing at the cement stone/casing interface. In the long-term, such loss of corrosion control could lead to casing failure and leakage.

- Class G cement stone showed dissolution of Mg^{2+} and SO_4^{2-} containing phases – most probably brucite and gypsum – which are not initial phases of the cement stone, but precipitate when the Opalinus Clay pore water is contact with the cement stone, before the injection of CO_2. The precipitation process is ambivalent with regard to sealing efficiency as on the one hand, due to the associated volume increase, clogging of pores in the cement stone and/or at associated interfaces is possible, which would result in a permeability decrease and therefore improvement in sealing efficiency. On the other hand, the volume increase could lead to crack formation inside the cement stone which would have a negative effect on its sealing efficiency.
- The fact that the precipitate completely dissolved after CO_2 injection is of general advantage to the cement stone sealing efficiency, as the precipitate is available for additional CO_2 consumption, compared to the initial cement stone before contact with Opalinus Clay pore water. However, this advantage is only relevant if no cracks form during precipitation.
- Class G cement stone showed a dissolution of calcium silicate. This process can be assumed to be irrelevant for sealing efficiency as calcium silicate is a relic of the initial raw cement before mixing with water. As long as water/cement (w/c) ratios are 0.4 or above for the preparation of the paste, and the curing time of the cement paste is long enough (which can be assumed on-site), there should be little or no calcium silicate due to the formation of the cement stone phases (calcium silicate hydrate (CSH) and portlandite).

The reactions listed above occurred unexceptionally for both P/T combinations. This means that Mont Terri experiment conditions can be qualitatively compared to regular CO2 storage conditions.

The quantity of reactions occurring, in particular carbonate dissolution for Opalinus Clay and the carbonation process for the Class G cement, varied between Mont Terri conditions and regular CO_2 storage conditions due to pressure/temperature (P/T) differences due to CO_2 solubility. Under regular CO_2 storage conditions, the experiments showed an increase in these reactions.

3.2. Modelling long-term geochemical processes in wells and aquifers exposed to CO_2

As already demonstrated for caprocks, numerical modelling is a powerful method for studying coupled physical and chemical processes associated with leakage and can be used to predict medium- to long-term risks of leakage from near-well environments and contamination of shallow aquifers.

Simplified geochemical and reactive transport models were therefore established: data from experiments performed by ULTimateCO_2 and direct measurements were fed into numerical models which were applied to a CO_2 storage site. Where no information was available for essential parameters, values were taken from published papers. These parameters included the chemical composition of pore-waters, the mineralogical composition and phase composition of rocks or solid engineered materials and their porosity, permeability, capillary pressure and relative permeability.

Since the systems in question are, by their nature, poorly known, these models were not used to make absolute predictions for the medium and long term, but focused on identifying key phenomena and processes that might be expected in the case of well leakage.

As shown above, leakage from wells is most likely to occur when a poorly cemented casing/hole annulus allows pathways to form that then allow fluid to flow along the caprock-cement and/or cement-casing interfaces, rather than through the cement itself. Unless external stresses cause fractures or pathways to form within the cement's structure, fluid is unlikely to migrate through the pore spaces of intact cement.

However, changes in downhole conditions during the service of the well (casing pressure and temperature, casing displacement, changes in pore pressure/temperature during reservoir production) can cause radial stress cracks to form in the cement sheath, which can potentially lead to a loss of the annular isolation of the well. Low-pH fluids in the storage reservoir have the potential to cause chemical alteration of the cement and modify any micro-fracture networks that have developed around the well.

The manner in which near-well materials react to contact with CO_2-rich fluids is site-specific. It depends on the geochemical properties of the fluid, gases, and solid well components participating in any reaction with CO_2. The degradation of the primary phases of cement and caprock may lead to large-scale precipitation of carbonates and affect their porosities. A decrease in porosity is potentially beneficial in terms of the containment capacity of the storage reservoir, whereas an increase in porosity is a concern as it may lead to an increased risk of leakage.

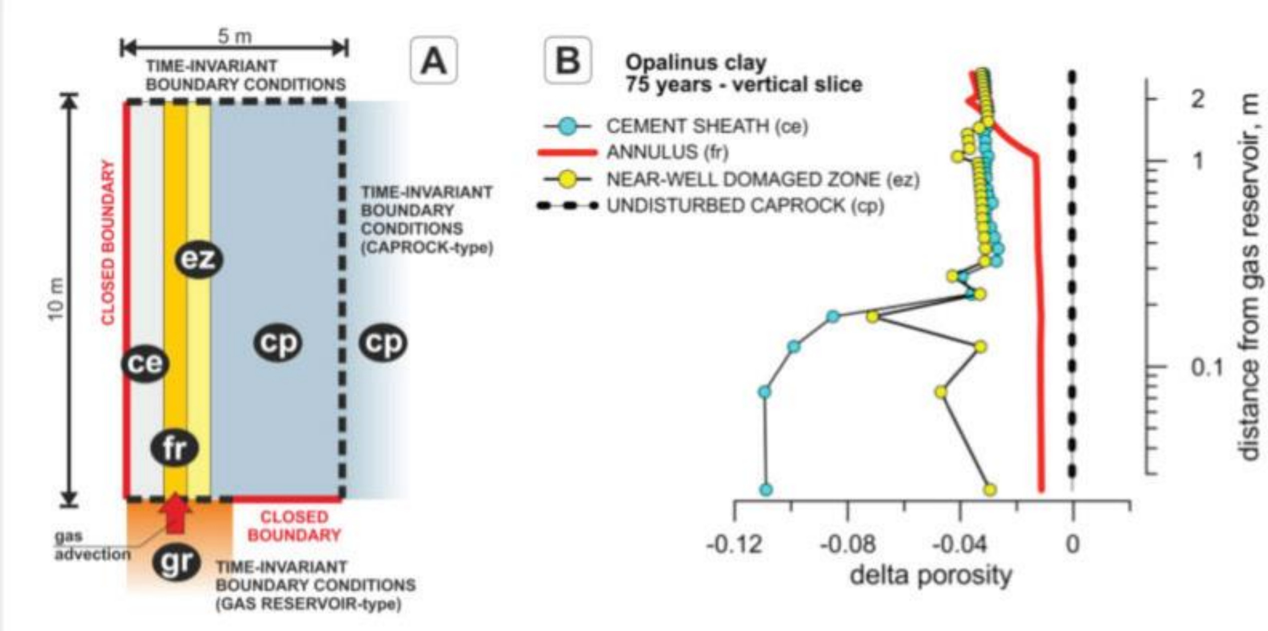


Figure 15: The near-well environment (A) can be modelled by considering (i) a gaseous reservoir ('gr') at the bottom of the system; (ii) a cement sheath around the casing of the well ('ce'); (iii) a damaged zone of the caprock ('ez'); (iv) a highly conductive annulus ('fr') at the interface between the cement and the damaged zone; (v) a large volume of 'pristine' caprock ('cp') far from the axis of the well. At the interface between Opalinus Clay and an ordinary Portland class G cement (B), porosity is predicted to decrease after gas-water-rock interactions driven by CO_2 intrusion through the annulus and the damaged zone (75 years simulation time).

The containment capacity of sealing materials also depends on additional non-geochemical factors that are not taken into account by the current generation of reactive transport models. The formation of fractures and the degradation of the mechanical properties of well construction materials are practically unknown. For this, a new generation of simulators is needed that considers full coupling between geochemistry and geomechanics. Reliable analysis of the geochemical processes controlling the integrity in the near-well environment and long-term containment of CO_2 is possible over limited time-scales (i.e. decades). Extrapolation over longer time-scales is still highly uncertain due to the lack of information on how these cement materials evolve in the long term.

Leaking CO_2 has the potential to mobilize chemicals that would otherwise be stable, and non-hazardous, in shallow aquifers and groundwater systems. This is because the high CO_2 content and reactivity of CO_2 lowers the pH of fluids and can cause solid minerals to dissolve into the water. However, the vulnerability of aquifers to CO_2 intrusion depends strictly on specific local hydrogeological, geochemical, and mineralogical conditions. The identification of any minerals containing potentially hazardous trace elements that may leach into groundwater is of paramount importance. The concentration and extent of any resulting contamination depends on a number of physical-chemical parameters of the aquifer, including the CO_2 intrusion rate and the mechanism of release of the contaminant. Site-specific information is therefore required for a reliable, semi-quantitative assessment of the pollution risk to potable supplies.

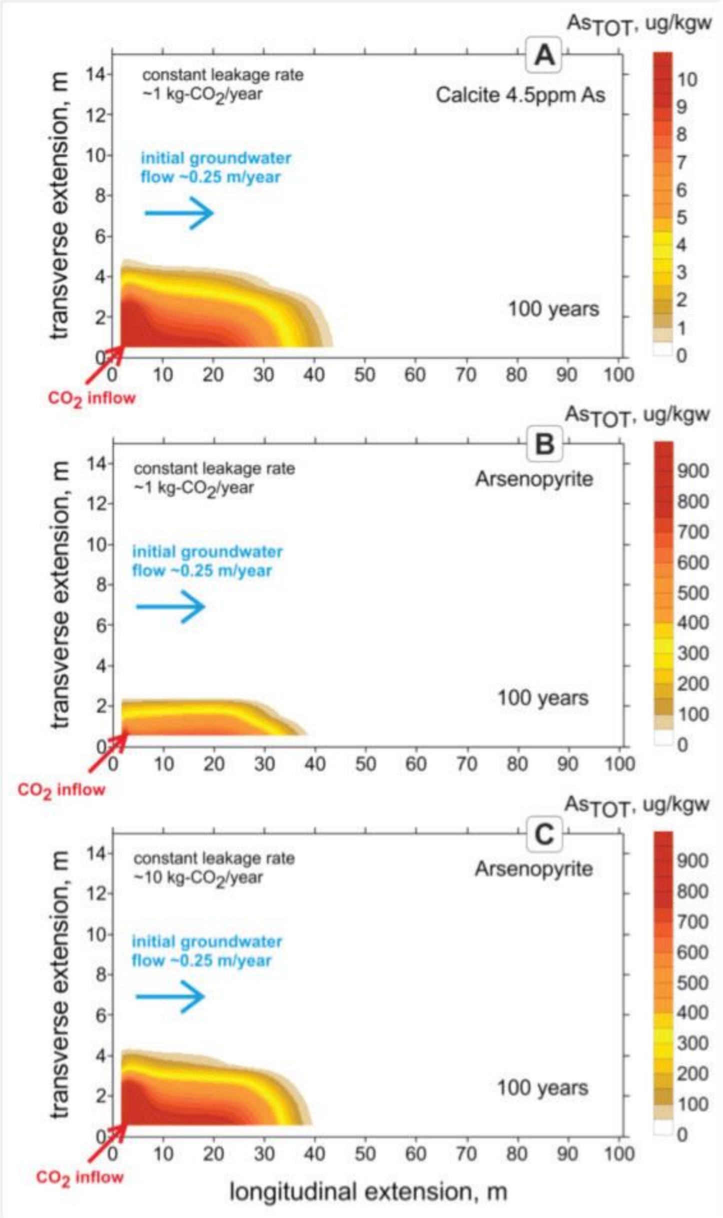


Figure 16: Spatial variations of aqueous arsenic concentration (micrograms per kgw) after 100 years in a horizontal slice of the Buntsandstein aquifer, Lorraine Region, NE France (top view), with the assumption of constant leakage rate (leakage point at 0,0 coordinates) and constant groundwater flow. The effects of different primary mineral as sources (arsenian calcite, A, vs. arsenopyrite, B) and CO_2 leakage rates (1 kg-CO_2/year, B, vs. 10 kg-CO_2/year, C) are shown.

4. Storage behaviour at basin scale (WP2)

In order to assess the long-term fate of injected CO2, 3D numerical models are usually built for the reservoir only, taking into account the formation's natural spatial variability and associated petrophysical properties. These models are used to simulate the evolution of the injected CO2 and induced modifications to the reservoir pressure due to injection. Previous storage models have usually been spatially restricted, with assumed boundary conditions either limited to the area surrounding the injection point or using analytical formulation to take into account the parts not included in the model.

In many cases, the numerical flow model assumes a hydrostatic equilibrium. However, this type of initialization does not take into account regional groundwater flow or irregularities in the pressure field that may result from the geological history. Having accurate initial conditions is therefore key to predicting the behaviour and impact of CO_2 injection.

ULTimateCO_2 combined two basin-scale 3D numerical models:

- Basin history simulation model: to define the present-day groundwater flow, geology and salinity field by simulating its history through time (how sediments deposited, eroded, compacted under overlaying sediment weight, or deformed over time; how the formations were progressively filled with water and how this water evolves by becoming more saline). The model therefore computes the evolution of pressure, temperature and salinity over geological timescales (millions of years) due to the variable geometry of the basin. This results in a present-day basin state that may not be in equilibrium with hydrostatic conditions due to past geological events.
- Flow model: to model CO_2 injection and its impact on pressure and flow. Present-day pressure and salinity fields resulting from the basin simulation model are then used to set the initial and boundary conditions of the flow model.

Thus a basin simulator computes present-day pressure, temperature and salinity fields, consistent with the basin's geological history. A reservoir simulator simulates the injection and long-term fate of CO_2 in a reservoir (with its initial conditions derived from the basin simulator results).

The combined use of these two models enables:

- A more accurate assessment of the initial conditions before CO_2 injection is simulated – particularly for pressure, temperature and salinity.
- An understanding of the impact of the pressure pulse on brine displacement induced by CO_2 injection and any possible change in the hydrodynamic regime at basin scale.

Basin-reservoir coupling allows the initial hydrodynamics and pressure singularities of a reservoir to be taken into account – leading to more accurate predictions for CO_2 storage.

4.1. The Paris Basin case study

The Paris Basin is a 600 km-wide sedimentary basin in the north of France that offers the potential for CO_2 storage. It is composed of Triassic to Tertiary sediments that reach a maximum thickness of 3 km in the central part of the basin. A 2 million-cell 3D model was created for basin and injection simulations. Water in contact with the salt layer (cells in white in the figure) becomes very saline and then, depending on the basin evolution, the brine migrates over time, resulting in today's salinity field (0 My).

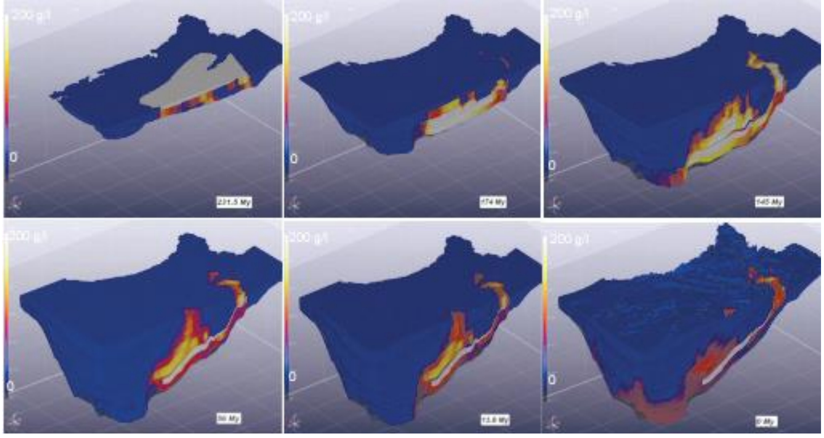


Figure 17: The modelling of the Paris Basin and the evolution of salinity simulated through geological time (million years, My). 3D view of the simulated salinity field through time (231.5 My, 174 My, 143 My, 56 My, 12.8 My, today (0 My)). Color scales range from 0 g/l (deep blue) to 200 g/l (white).

Applying the basin and flow models is a two stage process:

➢ *Stage 1 – Basin modelling*

Basin modelling has two steps:

1. The first step reconstructs the evolution of each layer – from present-day geometry back to that during sedimentation. This process is called 'backstripping' and consists of progressively 'peeling-off', or removing, each sedimentary layer. The model then computes the decompaction resulting from this peeling off. The effects of subsidence, deposition and erosion are also 'undone' to lead back to the original geometry at the time each layer was deposited.
2. The second step goes forward in time – from sedimentation to the present-day state, using the geometry defined in the previous stage. By simulating the basin filling over time, the model computes the evolution of petrophysical properties (e.g. permeability and porosity) and of pressure and water salinity. The 3D fluid flow simulator simultaneously solves several equations up to the present day: conservation of mass and momentum for solids and fluids, plus salinity transport by advection, compaction, energy conservation, the generalized Darcy Law for the three-phase flow and, if necessary, the maturation and generation of hydrocarbons. For validation purposes, results are then compared against estimations of temperature and salinity fields in existing literature and previous modelling work.

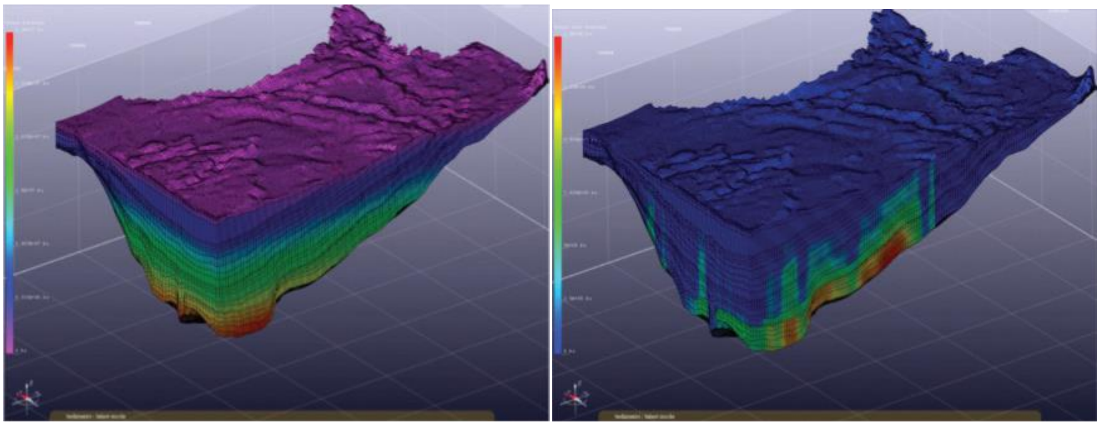


Figure 18: Improving the assessment of initial pressure conditions: initial pressure based on the basin model (left) and the difference between the hydrostatic and basin model pressure (right). Color scales range from 0 to 32 MPa and 1.5 MPa, respectively (1 MPa = 10 bar).

➢ *Stage 2 – The flow model*

The results of the basin simulation in Stage 1 are then used to initialize a flow model at basin scale before the simulation of CO_2 injection. In providing accurate initial conditions, basin-scale modelling results also allow an accurate prediction of the impact of CO_2 injection – especially on pressure and flow.

➢ *Simulating CO_2 injection*

The simulated scenario corresponds to an injection of 200 Mt of CO_2 over 50 years. The CO_2 plume extension remains very low: only 20 km^2 at the end of the injection; the injected fluid also remains confined in the Triassic layers. The maximum overpressure value is 143 bar (i.e. 54% of the initial pressure value at the same depth). In this model, the zone impacted by CO_2 transport is very localized and the resolution used for the basin model (cell size: 2 km) implies that the shape of the plume is relatively coarse.

4.2. Adaptive Mesh Refinement significantly improves modelling resolution

The results of CO_2 injection are strongly influenced by the grid size used for the basin history simulation.

A possible solution for achieving a higher resolution for the CO_2 plume – and avoiding long processing times – would be to extract a sub-grid corresponding to the reservoir scale and use pressure boundary conditions to simulate the formation extension beyond the modelled area. However, the localization of the boundary condition may impact on the simulation result if not accurately selected. Another method involves using a static Local Grid Refinement (LGR). The main difficulty is to define the size of the refined grid in order to properly account for the size of CO_2 plume (before starting the simulation): an iterative process is often needed and the final number of grid blocks could still be very high. An alternative solution involves refining only the grid blocks in contact with the CO_2 to achieve fine CO_2 flow modelling, leaving the rest of the model at basin scale in order to simulate the pressure effects. This gives refined modelling results while controlling the total number of grid blocks and simulation time. An Adaptive Mesh Refinement (AMR) prototype has already been developed to address this and was applied by ULTimateCO_2 to simulate CO_2 injection in the Paris Basin model illustrated above. The same pressure temperature- salinity calculation performed by the basin history simulation was used to initialize the model. The maximum level of refinement allowed the basin grid size to be sub-divided by a factor of 16 within the CO_2 plume during the injection simulation.

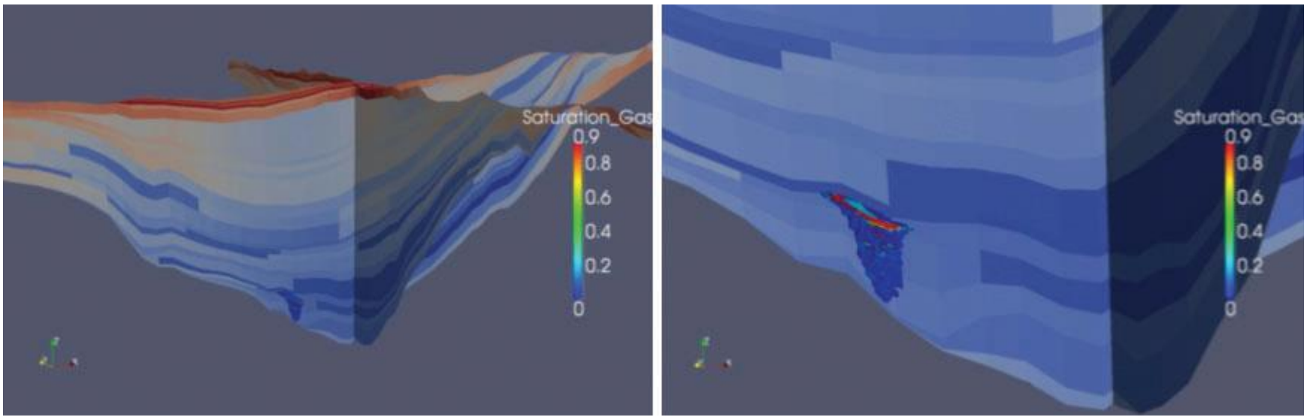


Figure 19: CO2 gas saturation after 1,000 years by using an AMR technique (left – basin-scale view, right – reservoir-scale view)

When comparing CO_2 injection simulations performed on a basin-scale grid size with simulations performed with AMR techniques, results show that the displacement of the CO_2 plume and associated trapping mechanisms are simulated more accurately, while reducing computing times compared to traditional techniques.

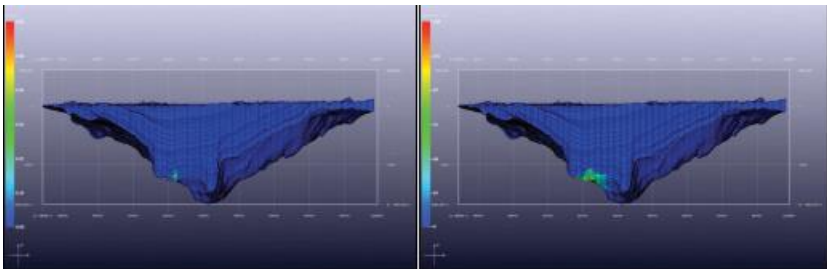


Figure 20: CO_2 saturation after 1,000 years (left) and overpressure after 50 years (right)

4.3. Improving the simulation of CO_2 dissolution for large-scale domains

Of the trapping mechanisms described in the ULTimateCO_2 example in Section 2.4, dissolution of the CO_2 plume is a slow process occurring at pore scale that is difficult to simulate effectively in reservoir- or formation-scale models. Once accumulated below the caprock, the CO_2 dissolves only at the interface between the gas and liquid phases over time. Initially, part of the brine in contact with the CO_2 will quickly become saturated and dissolution limited by the very slow molecular diffusion of CO_2 in the brine. This is known as the purely diffusive phase. However, because the CO_2-saturated brine is denser than the underlying indigenous brine, the brine-CO_2 contact zone gradually becomes unstable. After the purely diffusive phase, a complex convection process starts where the CO_2-saturated water sinks and the CO_2-free water rises.

This 'convection' accelerates the global dissolution of CO_2, i.e. the bulk transfer of CO_2 from supercritical form to the aqueous phase. Once CO_2-saturated brine reaches the bottom of the reservoir, CO_2 transfer progressively decreases and very slow molecular diffusion again drives the final homogenization until equilibrium is reached and CO_2 transfer stops.

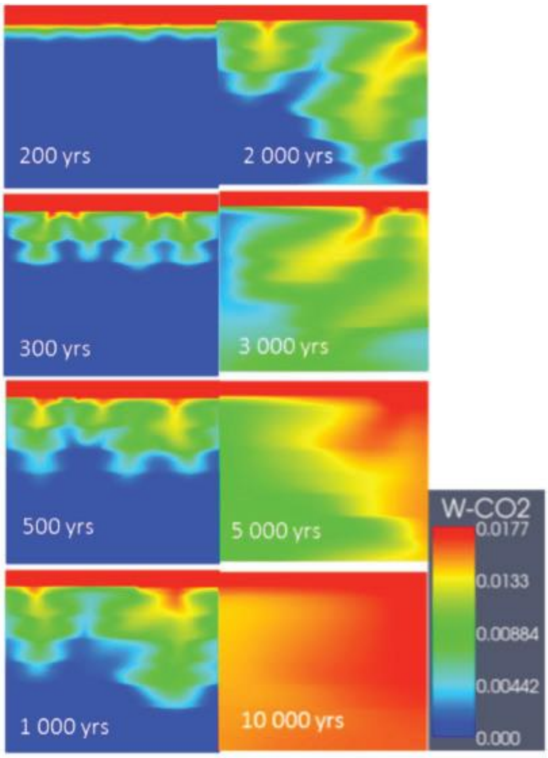


Figure 21: CO2 molar concentration maps at different timescales for a layered permeability map at 200, 500, 1,000, 1,500, 2,000, 3,000, 5,000 and 10,000 years

This whole process of gravitational instability due to CO_2 dissolution in water depends on the diffusion rate, formation permeability and heterogeneity distribution. In a strongly-layered sequence, the convection process is retarded compared to a homogeneous formation. A fine model grid is therefore needed to reflect the diffusion and convection process correctly in the flow simulation. In full field models, it is necessary to use a coarser grid for computational reasons and design an upscaling strategy.

The goal of an upscaling method of gravitational instability is to represent the phenomena described above, that happen at pore scale, in a coarse grid. In a coarse grid, only a purely diffusive regime can be simulated: the second (convection) and third (homogenization) phases are not represented due to the large size of the cells.

➢ *Upscaling using the pseudo-diffusion method*

In order to obtain similar values of dissolved CO2 at intermediate timescales (between 200 and 2,000 years), the upscaling pseudo-diffusion process is based on the calculation of a pseudo-diffusion coefficient for coarse grid size that is larger than the native fine-scale diffusion coefficient. This means accepting an error at very short timescales (for small values of dissolved CO_2) and verifying that the final trend extrapolated from the upscaled model is consistent with the trend observed for the refined model. The pseudo coefficient is calculated by creating two vertical models: a coarse model with cells corresponding to the large-scale model and a refined model representative of the vertical heterogeneity of the reservoir. The global transfer of CO_2 between the gas cap and the reservoir water over time is used to calibrate the pseudo-diffusion coefficient.

The diffusion process is proportional to the square root of time, i.e. the global quantity (tons per cubic metre) of dissolved CO_2 is plotted against the square root of time (days). The pseudo-diffusion coefficient is derived from the average slope of the global CO_2 transfer in the refined model (red line), so that the global transfer in the coarse model (green line) fits best. Such a calibration of the pseudo-diffusion coefficient will be required according to the coarse model discretization (the interval applied) and the heterogeneity of the reservoir (to date only upscaling in the horizontal direction has been investigated).

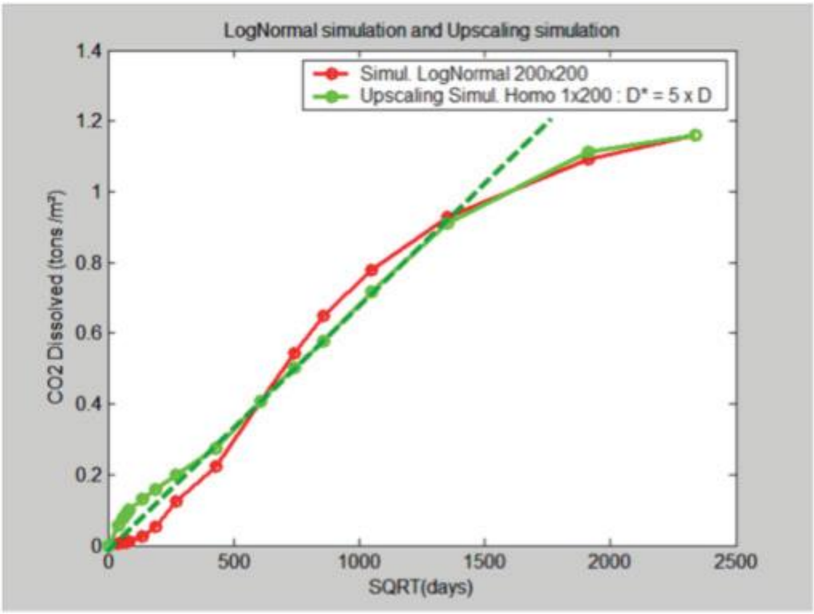


Figure 22: Comparison of the total amount of dissolved CO_2 in the refined model (red) and the upscaled model after pseudocoefficient calibration (D = 5 x D initial here)*

Using a detailed reservoir model and incorporating heterogeneities as explicit elements, or via an upscaling/averaging strategy, will significantly improve the resulting pseudo-diffusion coefficient.

➢ *Application to the GeoLorraine case study*

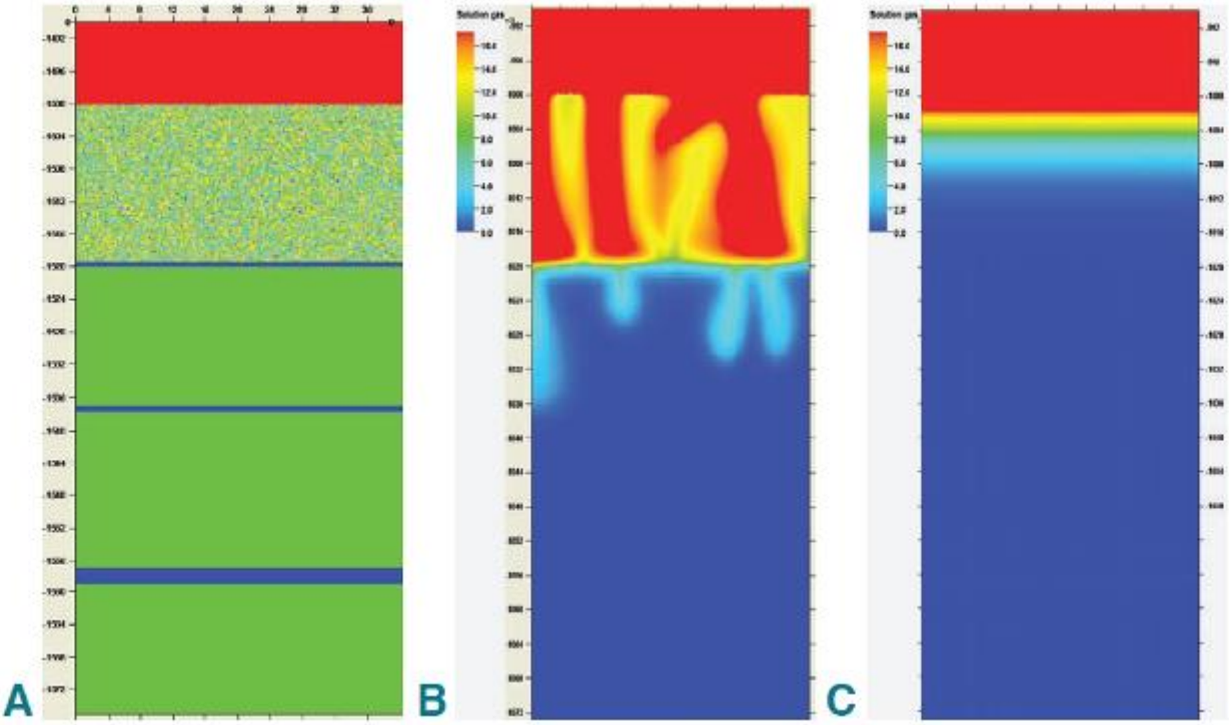


Figure 23: A) Schematic illustration of the simulation model for the layered case with three shale layers at 20 m intervals (Model 1). The permeability perturbation is visible in the upper 20 m reservoir. The zone above the CO_2-brine contact with full CO_2 dense-phase saturation is assigned a high permeability to aid uniform distribution. B) Dissolved CO_2 after 100 years in Model 1 with 20 m thick reservoir layers separated by thin, continuous shales with 1 mD permeability, showing convection in the uppermost layer. C) Dissolved CO_2 after 100 years in Model 2 with homogeneous 20 mD upscaled permeability.

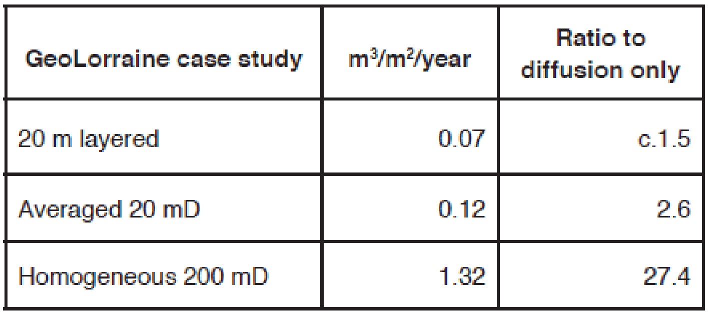
This upscaling process was incorporated into the GeoLorraine case study, testing different assumptions. Three cases for the refined model were investigated (Figure 23), corresponding to different decisions on the modelling of thin, low permeability layers:

Model 1 Layered reservoir with explicit modelling of thin, continuous shales

Model 2 Homogeneous reservoir with harmonic averaged vertical permeability (20mD) in a fine grid

Model 3 Homogeneous high-permeability (200mD) reservoir ignoring thin sealing shales

- The transport of the CO2 downwards across the CO2-brine contact was monitored and plotted for the three cases as the cumulative of m3/m2 (Figure 3-8). This illustrates the very different time evolution of the dissolution transfer for the three different models.
- An approximation of the transport rate can be deduced by estimating a linear trend for segments with a near-linear trend. This is achieved by selecting an interval on the transport curves free from initial growth and prior to late-stage interference with the bottom boundary of the model.
- The estimated amounts of global CO2 transfer and the resulting estimated transfer rates caused by late-stage convection are compared and shows ratio to diffusion only scenario ranging from 1.5 to 27.4..



GeoLorraine case study	m^3/m^2/year	Ratio to diffusion only
20 m layered	0.07	c.1.5
Averaged 20 mD	0.12	2.6
Homogeneous 200 mD	1.32	27.4

Table 1: Rates and ratios of transport

Ignoring the low-permeability shales inside the reservoir and using a homogeneous model can lead to an overestimation of the amount of convection transport by a factor of 20. It is therefore key to incorporate realistic heterogeneities in the reservoir model when simulating instability and convection processes over the long term. Even if the shale layers are considered in a permeability averaging scheme, the transport rate is still slightly overestimated.

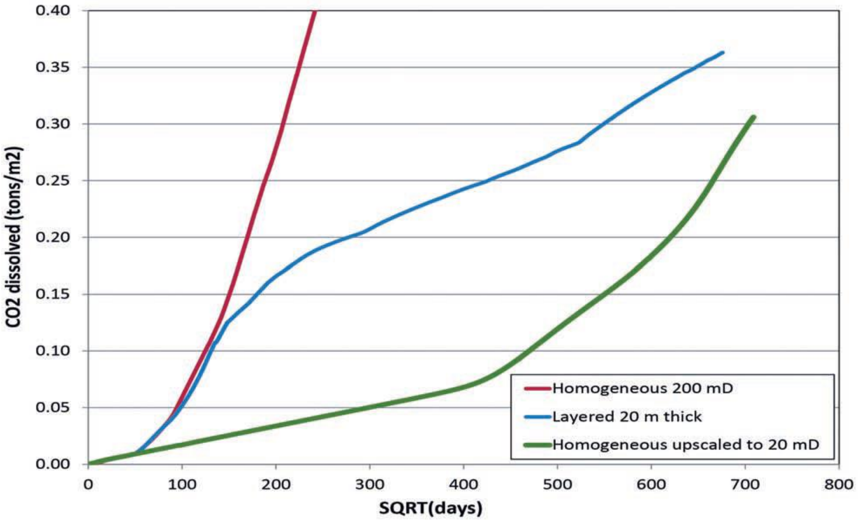


Figure 24: Comparison of cumulative transport across the CO2-brine contact in the three model versions of the GeoLorraine case study over 1,200 years. Curves for layered model with seals (Model 1: blue), for a homogeneous low-permeability model (Model 2: green) and homogeneous high-permeability model (Model 3: red).

5. Uncertainty assessment for long term predictions (WP6)

Modelling data used to predict the evolution of a storage site can vary considerably from site to site depending on the availability of data. It is therefore critical to integrate an appropriate method for assessing uncertainty in order to enhance confidence in modelling predictions. There is potential to adapt uncertainty assessment methods developed for other applications (e.g. civil engineering) to CO_2 storage, provided any similarities/differences between these subsurface applications are duly considered. ULTimateCO_2 has combined an uncertainty assessment with CO_2 storage modelling – the first time such a methodology has been used in this context. It assesses typical factors known to contribute to uncertainty – in particular, the kinetics of major reactions between fluids and rocks, how a multi-phase (CO_2-water) flow model with heterogeneous properties behaves and the coupling of mechanical and geochemical processes (e.g. the behaviour of faulted systems in the storage aquifer that have been chemically altered by the presence of CO_2 and trace components dissolved in the brine).

Uncertainty assessments are broken down into three steps:

Step A: Define the model, or sequence of models, used to assess key geological phenomena

The model may only be known through point-wise evaluations for each input vector due to time/cost constraints which limit the number of times a model can be run, or because neither an analytical nor a numerical representation of the model is known. Defining which parameters are considered deterministic (i.e. known) and which are uncertain is therefore key. Defining what type of dependencies exists between the input parameters can also drive the choice and scope of the assessment. a decision criterion should be established to drive the iterative process of uncertainty assessment (e.g. the probability of a percentage of CO_2 being trapped via solution and mineral reaction).

Step B: Classify the uncertainty

Typically, this means making a distinction between aleatory and epistemic uncertainty. Aleatory uncertainty arises from the natural variability of phenomena under consideration, while epistemic uncertainty refers to a lack of knowledge. Modern uncertainty theories were conceived with this in mind.

Step C: Characterize the model's uncertainty response using modern uncertainty theories

The choice of framework used depends on the nature of the model, the decision criterion and the mathematical constructs on which the uncertainty of the input parameters is expressed. This evolves during the lifetime of a storage site – as more information is gathered, the amount of epistemic uncertainty is reduced. Some input parameters become less uncertain and can be represented by more informative mathematical constructs. This calls for a revision of the uncertainty propagation in order to update the impact on the model.

5.1. Quantifying uncertainty: examples of application

➢ *The role of chemistry: caprock minerals*

The uncertainty workflow developed by ULTimateCO$_2$ was applied to the long-term evolution of caprock near an injection well. The numerical modelling was based on experimental data in order to assess the most influential minerals and their kinetic parameters (the amount and kinetic properties (i.e. reactive surface) of the minerals are highly uncertain – see section 5.2.3). A simple screening method, varying one parameter at a time (Morris method) was used to minimize the number of simulations for 29 uncertain parameters. However, several interactions were identified which requires an analysis of the variance (i.e. a different screening method (Sobol Sensitivity Indices).

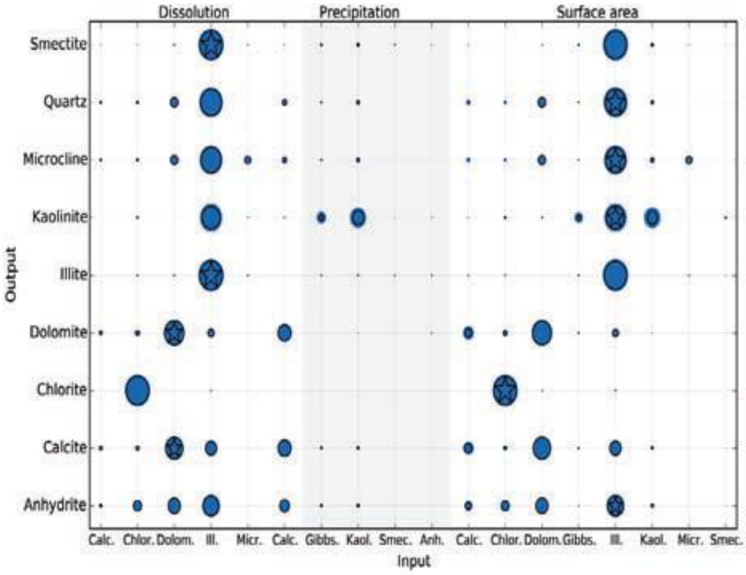


Figure 25: Sobol indices of the amount of minerals (weight ratio). Areas of the symbols are proportional to the magnitude of the quantities.

The results show a strong influence of two inputs – illite kinetic parameters (reactive surface area and dissolution rate) on the amount of illite (output). A surrogate model was then built to represent the experiment and the entire uncertainty domain. The model mean was in good agreement with the observed data.

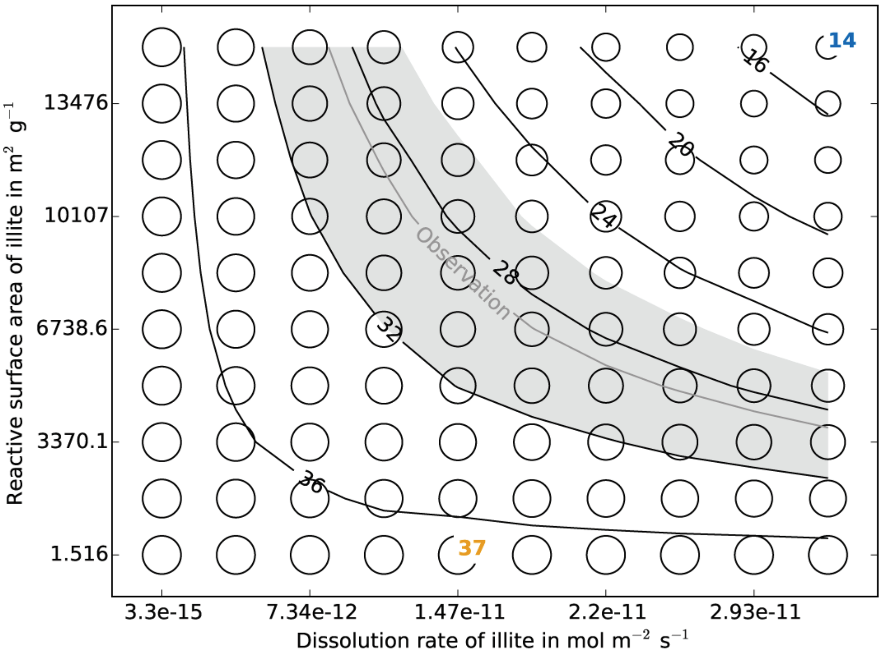


Figure 26: Final weight ratio of illite as a function of dissolution rate and reactive surface area.

➢ *The role of reservoir properties: subseismic faults*

Some features of geological storage may induce or enhance CO2 migration beyond its expected primary storage. One such feature is linked to small faults which may enable migration between different geological horizons. These are beyond the detection level of current seismic resolution and thus may not be characterized prior to CO_2 injection tests. The uncertainty workflow developed in ULTimateCO_2 was used to estimate the probability of failure of geological storage (i.e. the probability of migration of CO_2 from the storage formation to secondary storage within the storage complex). Following an initial local sensitivity analysis to identify the most influential uncertainties from the 18 uncertain input variables, a second sensitivity analysis was performed based on the four uncertainties that were shown to most influence the mechanical integrity of the base of the sub-seismic blind fault (Sobol Sensitivity Indices). A surrogate model was developed based on a limited set of simulations of the full model. This allowed the probability of rare events (associated with low probabilities) to be computed in a reasonable time. In order to estimate low probabilities, a considerable amount of simulations is required. Expected mean behaviour remains in the safe domain.

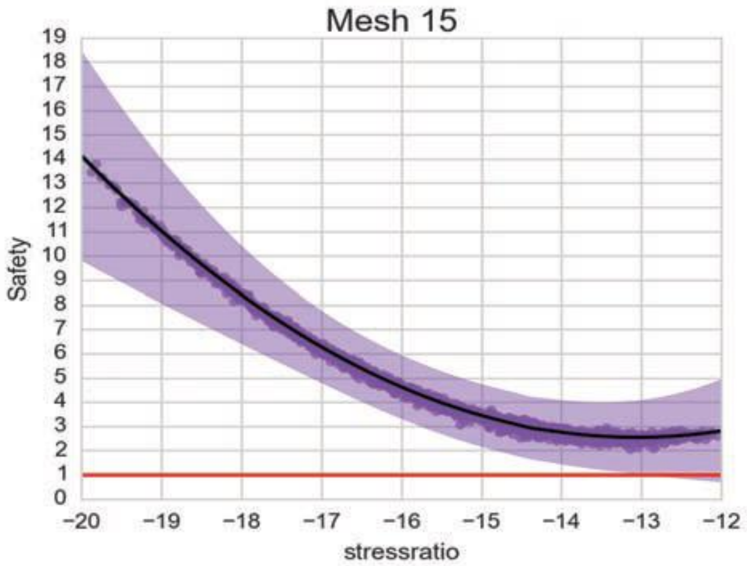


Figure 27: Estimation of mechanical failure at the base of the subseismic blind fault. The orange line indicates mechanical failure of the base of the fault. The black dot indicates the base case.

Due to uncertainty, the unsafe domain (mechanical failure) may be reached at the upper bound of the uncertain domain. The failure probability was estimated to be lower than 10-3 for this case. A confirmation run with the detailed numerical model at the upper bound of the uncertain domain confirmed that the mean behaviour was in the safe domain. The uncertainty of the surrogate model was thus reduced, which further decreased the failure probability below 10-6. In short, mechanical failure of the base of the sub-seismic blind fault was very unlikely.

➢ *Trapping diagrams: a focus on residual trapping*

Fluid flow modelling is challenging given the uncertainties of subsurface formation intrinsic properties (parameter uncertainty), but also modelling choices/assumptions for representing and numerically implementing processes occurring when CO_2 displaces the native brine (model uncertainty). Sensitivity analysis is therefore needed to identify the group of factors which most contribute to uncertainties in the predictions. ULTimateCO_2 proposes an approach for assessing the ranking of uncertainty sources with regard to the behaviour of supercritical CO_2 post-injection: a variance-based global sensitivity analysis on three output parameters which characterize the location and quantity of supercritical CO2. The use of ACOSSO-type advanced meta-modelling techniques circumvents two key difficulties: 1. The large number of computationally-intensive, reservoir-scale flow simulations 2. The differing nature of uncertainties, whether linked to parameters (continuous variables) or modelling assumptions (scenario-like variables). The feasibility of this approach is demonstrated using a potential storage site in the Paris Basin for which the amount, nature and quality of data available and associated uncertainties are representative of those of a storage project at the post-screening stage. Special attention was paid to comparing the results of the sensitivity analysis with the physical interpretation of the processes.

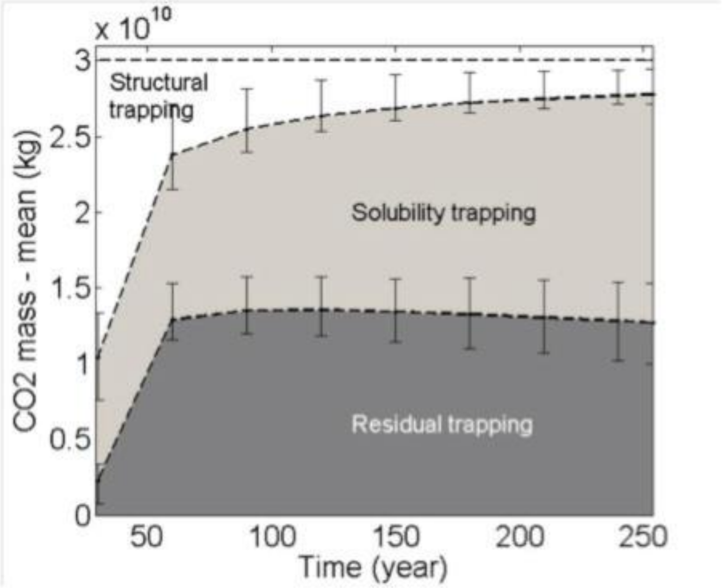


Figure 28: Evolution of the proportion of trapping mechanisms in the total CO_2 *mass computed from the mean of 300 simulations after injection ends (i.e. at 30 years). The error bars correspond to the 25th and 75th percentiles.*

The potential impact (including the socio-economic impact and the wider societal implications of the project so far) and the main dissemination activities and exploitation of results

ULTimateCO$_2$ intended impacts and dissemination objectives

The original intended impacts of the ULTimateCO$_2$ project were to:

- Improve scientific knowledge on the long-term fate of geologically stored CO$_2$
- Increase confidence by tackling the uncertainty associated with long-term CO$_2$ storage
- Draw up guidelines on the long-term processes of geologically stored CO$_2$ so that these can be better considered by CO$_2$ storage regulations

This was with the aim of providing the necessary insights to support the characterisation and assessment of potential storage sites and their surrounding areas as required in the Directive on geological storage, thus facilitating the large-scale deployment of CCS.

Based on these intended impacts, and bearing in mind the overall aim of helping to make CCS an acceptable climate change mitigation measure, the ULTimateCO$_2$ dissemination objectives were to:

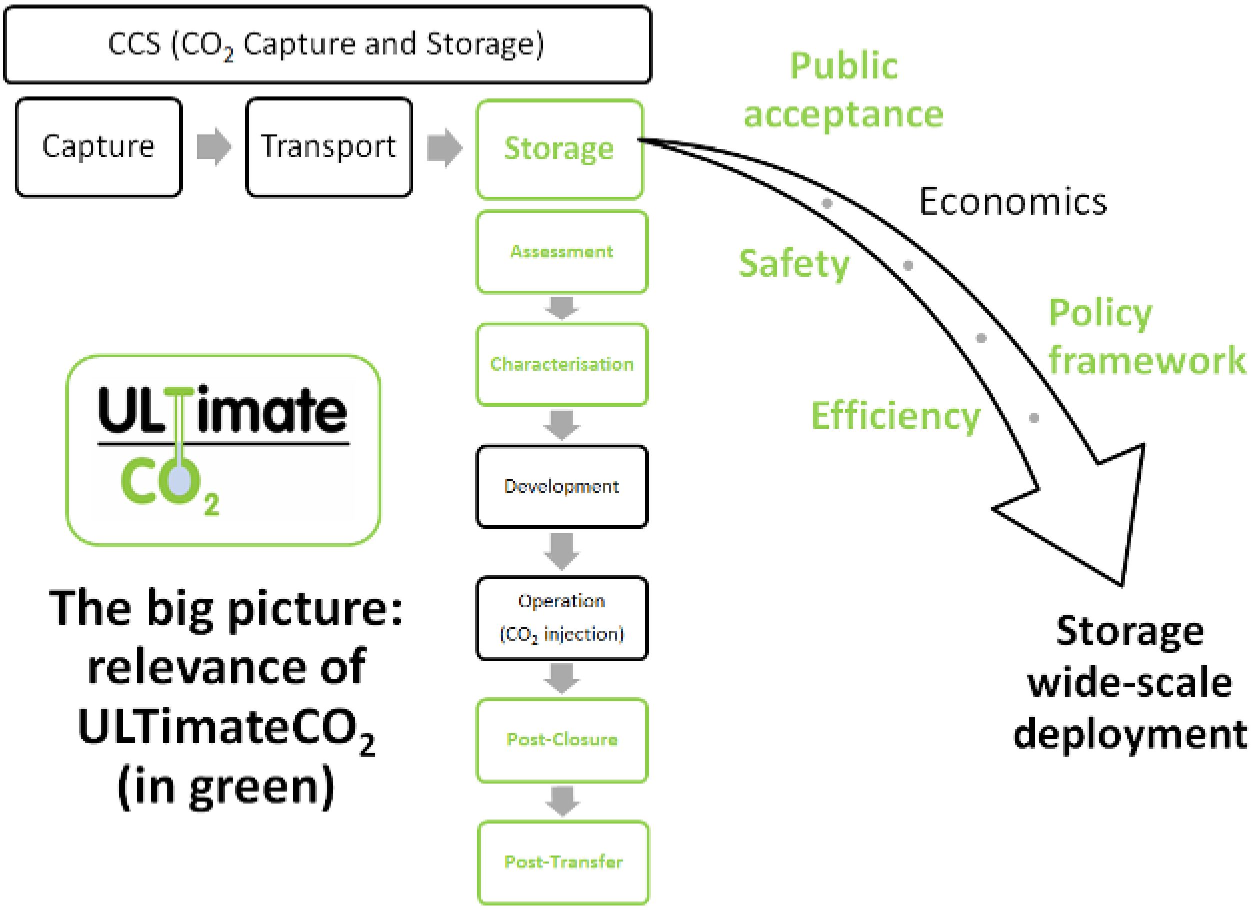
- attract interest in the project and give visibility to the research results
- disseminate project results to four stakeholder groups (see below) and increase their understanding of i) the efficiency and security of CO$_2$ geological storage, and ii) the long-term evolution of processes
- transfer, in an adapted form so they can be exploited and generate follow up, our improved scientific knowledge gained through activity in the technical workpackages (2,3,4,5 and 6) on i) the specific processes that may affect the long-term fate of geologically stored CO$_2$ and ii) which tools and methods are pertinent for predicting long-term storage site performance
- based on this improved knowledge, produce key conclusions (guidelines) in the aim of i) reducing the uncertainties associated with long-term CO$_2$ geological storage ii) raising confidence in the safety of geological storage of CO$_2$ in the long term and iii) helping enable future operators to successfully (or otherwise) demonstrate permanent containment as required by the EU Storage Directive by providing guidelines on the key considerations and most appropriate approaches to evaluate the long-term, post-abandonment behavior of storage sites
- help support the fulfilment of the EC CCS Directive (2009/31/EC) by providing complementary information on the long-term aspects of CO$_2$ storage and by addressing the transfer of responsibility from operators to the Competent Authority

Potential impacts and societal implications

The big picture

ULTimateCO$_2$ has addressed CO$_2$ geological storage, more specifically how the CO$_2$ plume and storage complex will evolve over the long-term (100s to 1000s of years). This issue is just one part of a larger context: CCS, or CO$_2$ Capture and Storage, which is a potential bridging technology in combating the negative climatic effects of carbon dioxide. CCS is highly pertinent to society in general, and to the major challenge of our necessary transition from fossil fuels to a greener sustainable energy.

The world is waking up to the gigantic challenge of climate change and our need to drastically cut CO_2 emissions in order to limit the rise in average global temperature to 1.5°-2°C, as reflected by the Paris Agreement representing a consensus of 196 parties following negotiations at the UNFCCC COP21 climate conference. Global experts agree that CCS is a critical technology to combat climate change and that decarbonizing the energy sector without CCS would therefore not only be extremely challenging, but significantly more expensive: CCS is among the lowest-cost options per tonne of avoided CO_2.



However, various challenges are holding back what we so evidently need: large-scale deployment from 2020-2030 at hundreds of sites, and the value drivers are currently not sufficient to overcome these. Much knowledge and expertise has already been gained through EU research programmes to support commercial deployment: criteria for selecting appropriate storage sites, methods and tools for site characterization, modelling, monitoring, control of risks and environmental impacts, assessment of storage capacity. ULTimateCO_2 fits into this context and has helped fill the knowledge gap concerning CO_2 geological storage in the long-term. Innovative scientific experiences have been proposed to evaluate specific long term processes related to efficiency and security of CO2 storage. Real field data and natural CO_2 analogues have also been used to ensure a pertinent study in direct link with geological context under consideration today for pilot projects in Europe. More specifically, ULTimateCO_2 has provided new and improved insight into the issue of long-term processes, area traditionally not very well understood but important for being able to assure safe and permanent storage in the long term.

Impacts benefitting a range of stakeholders

The results have been, and will continue to be disseminated to key target groups in order to enhance their understanding of the long-term security and efficiency of CO_2 geological storage. The main aim was to transform the scientific outcomes of the project into digestible information to be taken up and made use of by the four identified target audiences:

- Scientific community
- EU Policy makers and regulators
- CO_2 storage developers / operators (industry)
- Environmental NGOs

Indirectly, it is also expected that the general public may also benefit from the research undertaken by ULTimateCO_2.

The foreground of the ULTimateCO_2 project is relevant to both the upstream part of the CCS chain for future site selection and licensing, and the downstream part for safe decommissioning after site closure and transfer of responsibility**.**

The identified impacts of ULTimateCO_2 include:

➢ *Improved scientific knowledge on the long-term fate of geologically stored CO_2.*

- A better understanding of the specific long-term processes involved to help define what to look for during site characterization, but also what to avoid, i.e. identification of any processes that might prevent permanent containment (potential leakage pathways, fluid mixing, etc.).
- Help develop and enhance monitoring programmes
- Help benefit the process of handing over the responsibility of a storage site by making it possible to state that a site is 'secure'

➢ *Increased confidence (or reduced uncertainty) that CO2 can be completely and permanently contained.*

- Help answer questions, backed up by scientific facts, on the security and efficiency of long-term CO_2 geological storage
- Contribute to raising the profile of the CCS technology as a climate-change solution and to resolving one of the major current issues holding back large-scale deployment: social acceptance of CCS

➢ *Key conclusions and guidance for CCS stakeholders*

- The basic scientific facts and generic lessons learned will form the basis of key conclusions
- Provide sound criteria on which a site can be defined as secure: vital for transferring responsibility of a storage site from operators to the Competent Authorities
- Improved clarity and robustness of CO_2 storage regulations and thus support for the EU CCS Directive. ULTimateCO_2's main dissemination activities and exploitation of results

An address book tailored to ULTimateCO$_2$'s stakeholders

Before considering in detail examples of the main dissemination actions carried out during ULTimateCO$_2$, it is worth mentioning the ULTimateCO$_2$ address book. There is little point in having good results if they are not disseminated into the appropriate hands. Therefore, a comprehensive stakeholder address book was created under Excel in the early stages of the project by calling upon the experience of all partners and identifying key players in the CO$_2$ storage landscape, essentially in Europe, but also internationally. This is a transverse tool that formed the backbone to many of the dissemination actions. It was purposefully designed around the four ULTimateCO$_2$ target audiences with separate sheets per target group (policy makers and regulators (171 entries), CO$_2$ storage developers and industry (148 entries), NGOs (54 entries), and the scientific community (265 entries). It was a live document that was continuously fed and updated (638 addresses by the end of the project) and it served as the central tool for rapidly and effectively disseminating project outcomes.

Overview

The full list of dissemination actions is available in the Use and Dissemination of Foreground table (section 4.2 of the Final Report delivered to the EC).

ULTimateCO$_2$ dissemination activities have included: a re-designed project Website that was regularly updated; 1 project leaflet; 3 newsletters; 2 press releases; 2 public reports: one early on in the project giving the state-of-the-art, and one at the end summarising the learnings and key conclusions; annual consortium meetings and/or workshops; visibility at major scientific meetings and conferences through oral (21) and poster (14) presentations in 11 countries, and organization / chairing of conference sessions (GHGT11, Kyoto, Japan), presentations given at conferences in 11 countries: Australia, Austria, Denmark, France, Germany, Italy, Japan, Korea, Norway, Switzerland, USA; 12 publications (4 peer-reviewed and 8 proceedings) in scientific journals, with a further 9 publications expected (including 1 accepted and 2 submitted); participation in the COP-21 climate conference in Paris end 2016.

Outreach: raising visibility of ULTimateCO$_2$ and CO$_2$ geological storage in general

➢ *Website*

www.ultimateco2.eu

Laboratory experiments to better understand fluid-rock interactions, chemical reactivity, effects of other substances injected along with the CO2 (impurities).

A re-designed website was brought online in June 2014 and has been updated regularly with news and research highlights since. It is aimed at a wide audience, ranging from the general public curious about CO$_2$ storage (the introductory sections are written to be accessible by an alert reader but not necessarily with previous knowledge of CO$_2$ storage), to the scientific community (Workpackage descriptions, research highlights and the publications page with links to posters articles and presentations given at conferences). Visitors have access to the project content and the consortium, relevant reports, papers and presentations from project partners. From the homepage, visitors can access two 'non-scientific' pages explaining i) what the project is about and ii) the Mont Terri well experiment, plus a links page with general information on CCS. The ULTimateCO$_2$ website will be maintained and updated with any relevant news and publications until end 2018. After this time, the main content will be transferred to a page on the Coordinator's BRGM website.

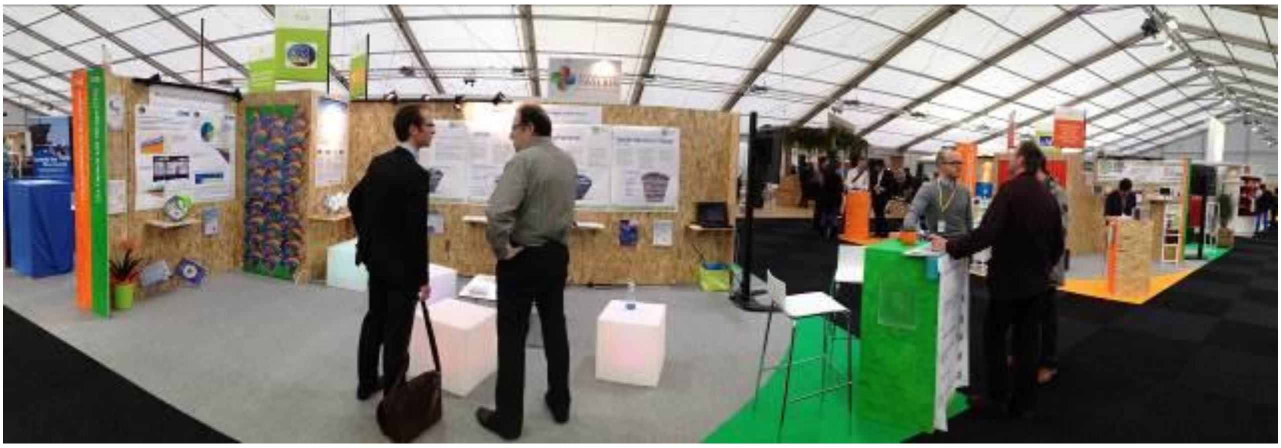
The website was used as a tool to disseminate various other actions, including the reports, press releases, project leaflet, and the main oral and poster presentations given by partners.

➢ *Participation in the UNFCCC COP21*

Through relationships with CO$_2$GeoNet, ULTimateCO$_2$ was given the opportunity to display material on two stands at the COP-21 climate conference in Paris from 30 November to 11 December 2015:

- a 18m^2 stand presenting CCS in the Climate Generations Areas accessible to the public,

- a small booth in the negotiations Blue Zone with restricted access.



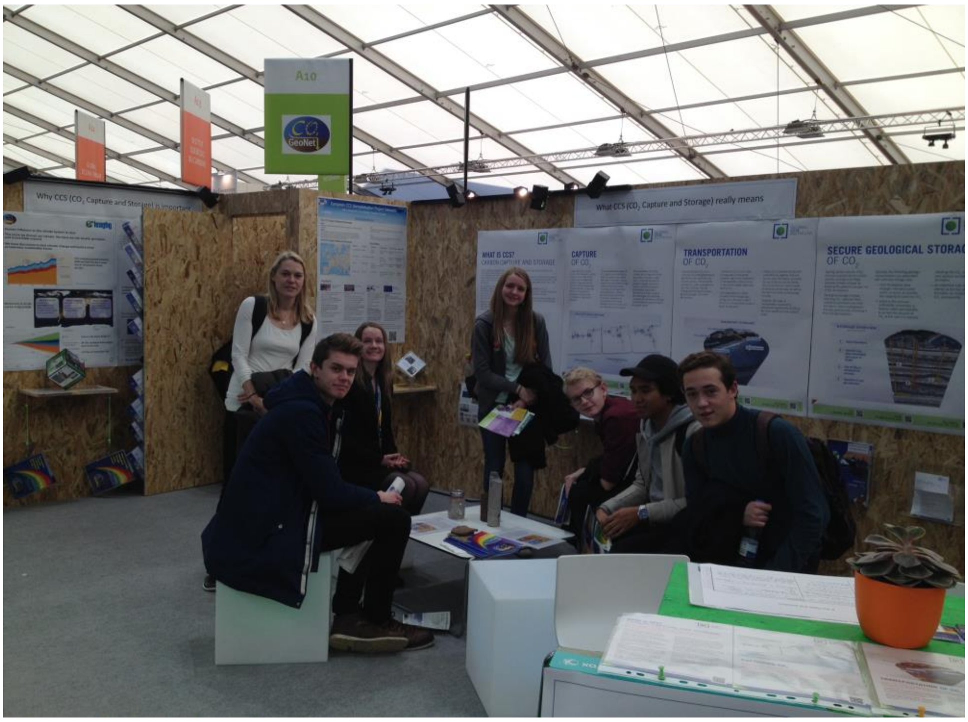
CCS stand in the public Climate Generations Areas

The stand in the public Climate Generations Areas was designed to raise the awareness of CCS in general, and materials were aimed at the general public / a non-scientific audience. Posters showed the various stages of the CO_2 Capture-Transport-Storage chain, and the role that CCS needs to play in our future energy transition, backed up by scenarios with and without CCS. The stand proved very successful with many curious visitors from all walks of life and interesting discussions over the two weeks.

ULTimateCO_2 material on display:

- two double-sided A4 flyers: one giving a general introduction to the project, aims and consortium, and the other announcing the forthcoming key conclusions Report;
- an eye-catching rotating cube displaying 6 themes: 1) ULTimateCO2 contact details and QR flash code; the multidisciplinary nature of the research with 2) 3D modelling, 3) laboratory work and 4) fieldwork; 5) the innovative well-integrity experiment at the Mont Terri Underground Rock Laboratory, and 6) the forthcoming key conclusions report.

ULTimateCO_2 rotating cube (farthest right) on the public Climate Generations Areas stand



Danish students in full debate at the CCS public Climate Generations Areas stand

Spreading the word to the scientific community

Under the main project aim of improving scientific knowledge (which in turn can also help increase confidence in the CCS technology), dissemination actions towards the scientific community were planned including publications in scientific journals and participation at high-level international conferences, to enable others working in CO_2 storage to benefit from the advances.

➢ *Scientific publications*

A total of 12 publications were published during the project, 4 in peer-reviewed scientific journals (Mathematical Geosciences, Geophysical Journal International, Water Resources Research, ARMA) and 8 others as proceedings (Energy Procedia GHGT11 and 12).

At least a further 9 are expected in the forthcoming year(s): Applied Geochemistry, Computational Geosciences, International Journal of Greenhouse Gas Control, Journal of Geophysical Research or Journal of Structural Geology, Energy Procedia (GHGT-13).

➢ *Participation at scientific conferences*

The results from ULTimateCO_2 were presented at various conferences internationally:

- 21 Oral presentations
- 14 Posters
- 11 countries: Australia, Austria, Denmark, France, Germany, Italy, Japan, Korea, Norway, Switzerland, USA

Some examples of high-level international conferences:

- 2012-2015: CO2GeoNet Open Forum, Venice, Italy
- 2012: GHGT-11, Kyoto, Japan
- 2014: European Geosciences Union in Vienna, Austria
- 2014: Goldschmidt, California, USA
- 2014: IEAGHG Monitoring & Modelling Combined Network Meeting, W Virginia, USA
- 2014: AGU, San Francisco, USA
- 2014: GHGT-12, Texas, USA
- 2015: 14th annual DOE-NETL conference on CCUS, Pittsburg, USA
- 2016: GHGT-13, Lausane, Switzerland

ULTimateCO2 Final meeting, IFPEN, Paris

The conferences were a good opportunity to interact with other projects and CCS initiatives. Some specific examples include:

- Participation in a workshop Brainstorming Day, together with other EU-projects, on the long-term effects of CO_2, in Trondheim, Norway (4-6 June 2013). Interaction with other EU FP7 projects: PANACEA (main organizer), CO2CARE, CARBFIX and MUSTANG.
- Participation in the Panacea project (http://panacea-co2.org) final meeting in Paris (19 Dec 2014). ULTimateCO_2 gave a presentation and sat on the discussion panel.
- The CO_2GeoNet Open Forum for each year of the project duration. This was an excellent opportunity to meet with other researchers and CCS stakeholders. In 2012, ULtimateCO_2 was present in a special session dedicated to 'Major research results and plans from European projects' and gathering the projects MUSTANG, SiteChar, COMET, CO_2ReMoVe, CO_2FieldLab, RISCS, ECO_2, CO_2CARE, PANACEA and ULTimateCO_2.
- Participation to the Canadian North American Wellbore Integrity Workshop in Denver, October 16 – 17, 2013.

➢ *Interaction with international high-level experts*

In addition, a team of pertinent international high-level experts were closely linked to the project via the ULTimateCO_2 Advisory Board. They played an active role in the annual consortium meetings and/or workshops, and offered steering advice on the project research, and particularly on how best to formulate and target the final report on key conclusions from the project.

Eight active members with varied profiles and viewpoints:

- CSLF link through PTRC, Canada, and Laurence Berkeley National Laboratory, USA
- research link through PTRC, Canada, and their Aquistore injection pilot
- international CCS initiatives' link through the IEAGHG and the GCCSI
- Regulatory Authorities' link through the French government department (DGEC Directorate General Energy and Climate)
- public outreach link through the NGO ZERO, Norway
- industry link through a private consultant with 30 years' experience with Statoil

Following an invitation from the EC scientific officer to attend two high-level international workshops on CCS, the coordinator benefitted from the opportunity to interact with other coordinators of worldwide CCS projects. The meetings were designed to enrich relationships between overseas and Europe and enhance collaboration for future research proposals.

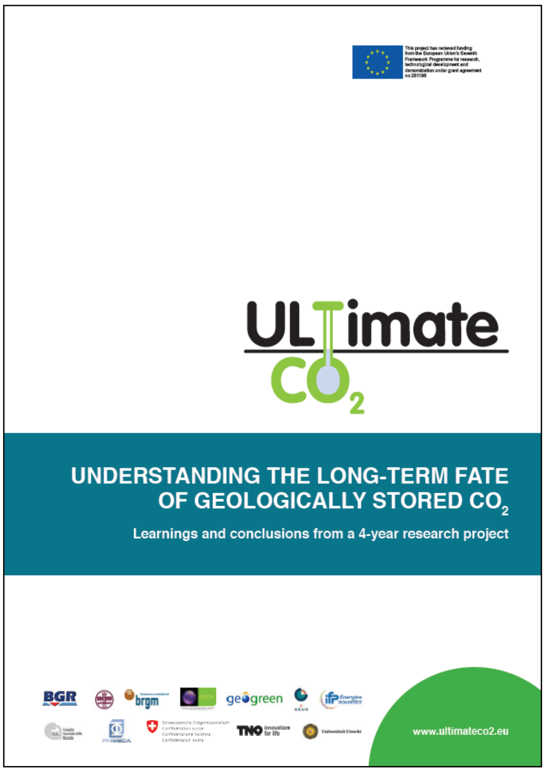
- 2014: EU-Australia Cooperation on CO2 capture & storage, Melbourne and Sydney, Australia
- 2015: Korean-EU CCS workshop, Jeju Island, Korea

Learnings and conclusions from a 4-year research project

➢ *Report*

Basic scientific facts and generic lessons learned from four years of collaborative research form the basis for the public report released just after the end of the project (January 2016).

The aim is to provide improved technical criteria for establishing the conditions under which CO_2 can be permanently contained in the long term, which is vital for robust clear regulations in the context of transfer of responsibility of a CO_2 storage site after injection has ceased and the site has been closed. This transfer will take place between CO_2 storage operators and the Competent Authority of the Member State, hence the relevance of this report for regulators & policy makers and CO_2 storage operators because they both need clear criteria to successfully (or otherwise) demonstrate permanent containment, as required by the EU Storage Directive.



This project has recieved funding from the European Union's Seventh Framework Programme for research, technological development and demonstration under grant agreement no 281196

ULTimate CO_2

UNDERSTANDING THE LONG-TERM FATE OF GEOLOGICALLY STORED CO_2

Learnings and conclusions from a 4-year research project

www.ultimateco2.eu

Although the report is written with operators and regulators in mind, it is also expected to be useful to national authorities, NGOs and the scientific research community, and also of benefit to the general public as the information will help to answer key questions such as:

- What happens after CO2 is stored?
- How will the CO2 change or evolve with time?
- Will the CO2 leak?
- Are we technically able to provide the long-term predictions needed?

➢ *Supporting the fulfilment of the CCS Directive*

A sound regulatory framework is essential to large-scale deployment of CO_2 geological storage: the European Directive on the geological storage of CO_2 (2009/31/EC), also known as the 'CCS Directive', has established this legal framework. In order to promote coherent implementation of the Directive throughout the EU, Guidance Documents (GDs) have been produced and GD3, which concerns the transfer of responsibility to the Competent Authority, is particularly relevant to the stage that ULTimateCO_2 focuses on.

The CCS Directive calls for the operator to demonstrate that "all available evidence indicates that the stored CO_2 will be completely and permanently contained". The Directive also suggests that operators can demonstrate permanent containment by meeting at least the three requirements or high-level criteria noted in Article 18(2):

1. the conformity of the actual behaviour of the injected CO_2 with the modelled behaviour;
2. the absence of any detectable leakage;
3. that the storage site is evolving towards a situation of long-term stability."

The outcomes of ULTimateCO_2 provided in the abovementioned report are hinged around these three criteria and designed as such to complement the CCS Directive's Guidance Documents in addressing the transfer of responsibility from operators to the Competent Authority. In terms of influence of the ULTimateCO_2 findings with respect to the stages in the lifetime of a commercial-scale CO_2 storage site, the findings are essentially aimed at the post site closure stage (transfer of responsibility to the Competent Authority), although they could also be of use at other stages: design and setting up - characterisation and monitoring of the reservoir geology - CO_2 injection.

Wrap up

In short, the results of ULTimate CO_2 have provided:

- Enhanced confidence in long-term predictions for CO_2 geological storage
- Better assurance of a permanent and safe storage of CO_2
- Knowledge on improved well design and sealing properties, thereby reducing the risk of leakage
- Better identification of any long-term risk of leakage via the caprock
- Enhanced efficiency of numerical simulation tools used to make long-term predictions for CO_2 behaviour
- Assistance to operators and regulators to complete the transfer of responsibility phase of a CO_2 storage site