



CarbFix Project no. 281348



CarbFix final report

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Contents

1	Final publishable summary report	2
1.1	Executive summary	2
1.2	A summary description of project context and objectives	4
1.2.1	WP1: Project management and planning.....	4
1.2.2	WP2: Injection technology	5
1.2.3	WP3: Surface and subsurface monitoring.....	6
1.2.4	WP4: Laboratory experiments	6
1.2.5	WP5: Improvement of thermodynamic and kinetic databases for CO ₂ -rich fluids.....	7
1.2.6	WP6: Geochemical and hydrological modelling.....	7
1.2.7	WP7: Dissemination and exploitation	7
1.3	A description of the main S&T results/foregrounds	9
1.3.1	Work Package 1: Project management	9
1.3.2	Work Package 2: Injection technology	9
1.3.3	Work Package 3: Surface and subsurface monitoring.....	15
1.3.4	Work package 4: Laboratory experiments of fluid-host rock and fluid-cap rock interaction	21
1.3.5	Work Package 5: Improvement of thermodynamic and kinetic databases for CO ₂ -rich fluids	29
1.3.6	Work package 6: Geochemical and hydrological modelling	33
1.4	The potential impact and the main dissemination activities and exploitation of results (not exceeding 10 pages)	46
1.4.1	Potential impact	46
1.4.2	Main dissemination activities and exploitation of results.....	47
1.5	The address of the project public website, if applicable as well as contact details.....	53

1 Final publishable summary report

1.1 Executive summary

Members of the CarbFix project have over the past three years developed the technology and expertise to capture, transport and geologically store CO₂ as carbonate minerals through in-situ carbonation in the subsurface. This knowledge has furthermore been demonstrated at the pilot scale at Hellisheidi power plant, SW-Iceland, where a pilot gas separation station, pipes for transport and injection and monitoring infrastructure was successfully built and operated.

CarbFix differs from many current carbon storage projects in several important aspects. First, it aims to provide a complete carbon capture, transport and storage (CCS) solution at a single operating power-plant. Second, in contrast to most projects, CarbFix aims at storing carbon by accelerating the transformation of CO₂ into stable carbonate minerals (e.g. calcite) as rapidly as possible. Because calcite is stable over millions of years, once CO₂ is transformed into carbonate minerals, there is little need for further monitoring. One can 'walk away' from the storage site.

To accelerate this transformation, CarbFix developed and adopted a unique approach. Most subsurface carbon storage projects until now have injected supercritical CO₂ into large sedimentary basins (Figure 1 a). Recent work has suggested that the transformation of CO₂ to carbonate minerals in such systems takes tens of thousands of years or more, if it occurs at all (Figure 2). This is because of the slow reactivity of silicate minerals in sedimentary rocks and the lack of the calcium, magnesium, and iron that are necessary for making carbonate minerals. The alternative method developed by CarbFix is to dissolve the CO₂ in water prior to or during injection into a basalt hosted reservoir to make it much more reactive with the basaltic host rock (Figure 1 b).

Injection of carbon dioxide into basaltic rocks has several advantages. First, approximately 25 weight percent of basalt is made of calcium, magnesium, and iron oxides. Second, basaltic rocks are far more reactive than sedimentary silicate rocks, meaning that the metals contained in basalts are more readily available to combine with injected CO₂ to form carbonate minerals. Third, basaltic rocks are abundant on the Earth's surface with about 10% of the continents and much of the ocean floor comprised of basalt. These advantages suggest that basalt carbonation could be an important carbon storage solution for the future.

In this project, we have demonstrated that:

- Solubility trapping of CO₂ occurs immediately
- Injected carbon is trapped in minerals within a few years using the CarbFix injection method in basalt hosted storage reservoirs (Figure 1 b)

These results suggest that the CarbFix method can change the time scale of mineral carbon trapping considerably.

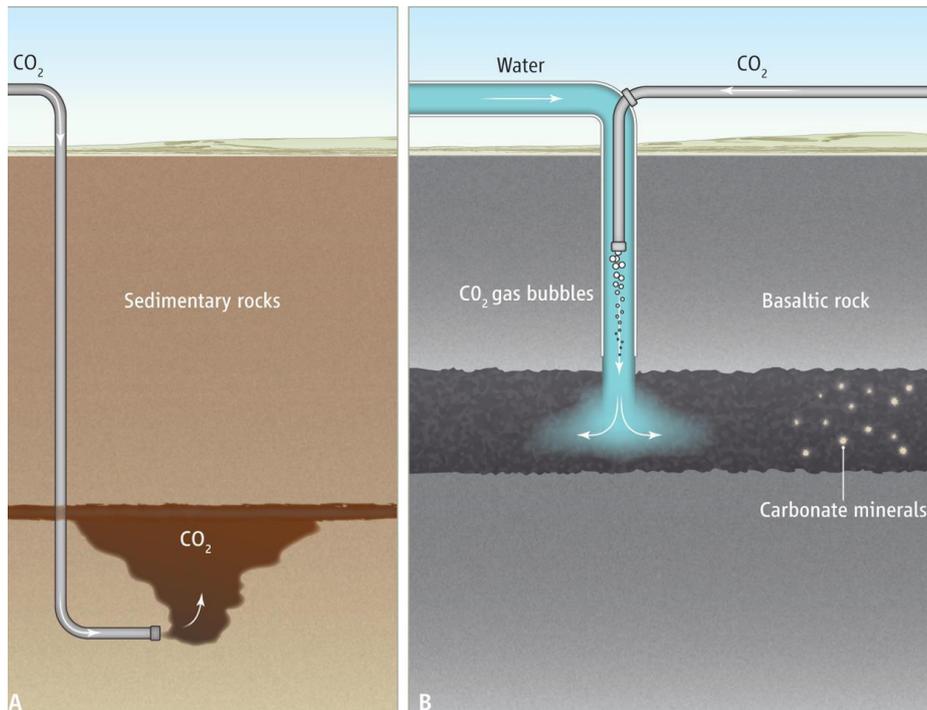


Figure 1: Carbon storage (a) as supercritical CO₂ in large sedimentary basins, where cap rock is necessary and (b) the CarbFix approach, CO₂ dissolved in water, injected into basaltic rocks, where cap rock is not required because dissolved CO₂ is not buoyant so it does not migrate back to the surface. Solubility trapping occurs immediately and most of the carbon is trapped permanently in minerals within few years. Figure from Gislason and Oelkers (2014) Carbon Storage in Basalt. *Science* 344, 373-374.

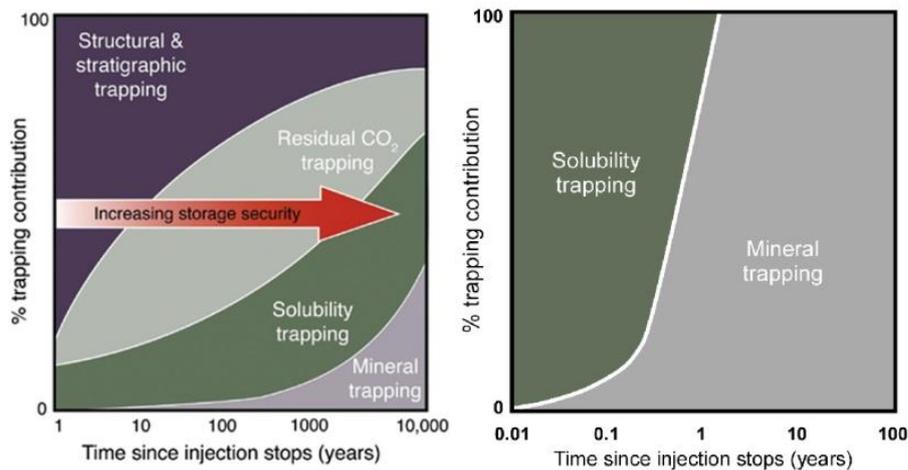


Figure 2: Schematic illustration comparing the trapping mechanisms followed by left) traditional geological carbon storage in sedimentary basins proceeds by injecting pure CO₂ into porous sedimentary rocks where CO₂ is trapped below an impermeable cap rock (structural trapping). Eventually some of this CO₂ becomes stuck in small pores, limiting its mobility (residual trapping). Over time, CO₂ dissolves in the formation water (solubility trapping). Some of this dissolved CO₂ reacts to form stable carbonate minerals (mineral trapping). As the system progresses from structural to mineral trapping, the CO₂ immobilised and storage is more secure, but carbonate mineral formation by this process can take thousands of years or more, and right) carbon storage using the CarbFix method. Solubility trapping occurs immediately and carbon is trapped in minerals within few years. Left figure from IPCC (2005) IPCC Special report on Carbon Dioxide Capture and Storage. Right figure from Matter et al. (2014) in preparation.

1.2 A summary description of project context and objectives

CarbFix was from its inception been a combined industrial/academic research program, consisting of a CO₂ injection pilot test, laboratory based experiments, study of natural analogues, predictive model development, numerical modelling and model validation. The overall goals of CarbFix have been:

- 1) To increase measurably our understanding of the long-term fate of CO₂ injected into the subsurface
- 2) To develop new technology to facilitate safe and permanent geologic carbon storage
- 3) To publicise the results of this research, allowing them to be applied internationally.

The final results of this project are anticipated to contribute towards the understanding of the long term fate of CO₂ when co-injected with water into the subsurface. The injection method developed and tested by this project minimizes pressure build-up in reservoirs as water used for co-injection is sourced from the reservoir itself. In addition the method has been demonstrated to have the following main benefits:

- 1) Solubility trapping is achieved within a few minutes of injection hence minimizing the risk of buoyant CO₂ floating to the surface.
- 2) The potential environmental impacts associated with disposing of reservoir brines that are often pumped from the reservoirs to prevent pressure increase as supercritical CO₂ injected is limited.
- 3) Between 80-90% mineralization of injected carbon is trapped only 1 year from injection.
- 4) Due to increased storage security, the method may be used as a demonstration to promote carbon storage as a safe method of reducing CO₂ concentrations in the atmosphere.

Over the past three years the CarbFix consortium has worked on interlinked tasks, milestones and deliverables in seven different work packages with the objective of achieving the goals listed above. The Pert chart shown on Figure 3 shows the main tasks in each of the work packages. These are explained in more detail in the following subsections.

1.2.1 WP1: Project management and planning

WP1 was led by OR. One deliverable was handed in in WP1 and one milestone was appointed to the work package. The main objectives of the work package were:

- 1) To plan, organise, maintain and review all consortium and participant activities.
- 2) To include the necessary activities to ensure coordination and planning of the project with the EU and the rest of the consortium.
- 1) To insure the smooth, efficient, and successful running of the CarbFix project.
- 2) To submit all required reports in a timely manner.
- 3) To manage all financial aspects of CarbFix. To coordinate the other work packages so that all milestones are met.
- 4) To manage intellectual property between CarbFix members.

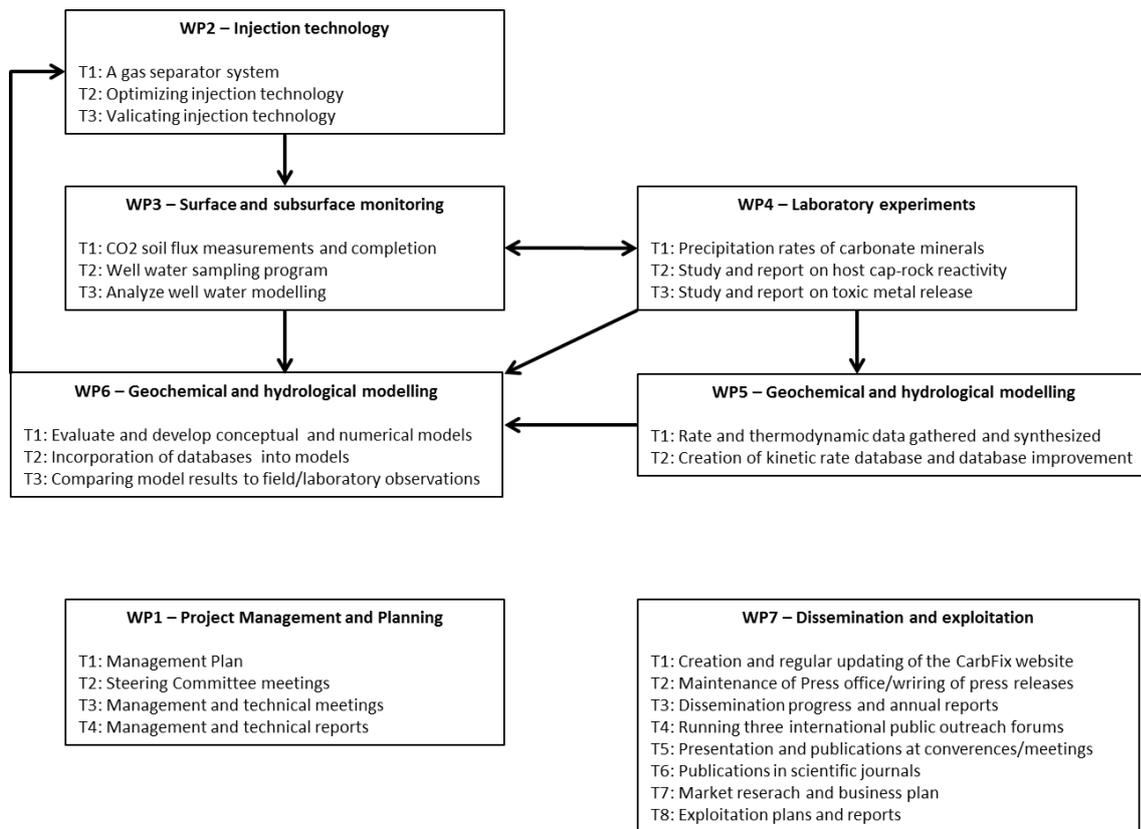


Figure 3: Pert chart showing main tasks in work packages in CarbFix and how they are interrelated.

The following tasks were carried out to meet these goals:

- 1) Upon initiation of the project, the Project Coordinator constructed a Project Management Plan, which was maintained to ensure consistency with project schedule and activities.
- 2) Regular meetings were held in the projects steering committee, as well as management and technical meetings.
- 3) The Project Coordinator and Technical Manager wrote and submitted all management and technical reports.

1.2.2 WP2: Injection technology

WP2 was led by OR but CNRS and UI also participated in the work package. Three deliverables were handed in in WP3 and three milestones were associated with the work package. The main objectives of the work package were:

- 1) To determine the optimal gas composition for CO₂ injection.
- 2) To optimize industrial methods to separate this gas from the exhaust of power plants.
- 3) To develop new technology favouring the dissolution of this gas into the aqueous formation or injection fluids.
- 4) To generate well monitoring tools that allow assessment of the success of the injection process.

The following tasks were carried out to meet these goals:

- 1) A gas separator system was created, tested and optimized to generate gas to be injected at the Hellisheidi site.
- 2) Design and optimization of injection technology that promotes the dissolution of the injected gas with an aqueous phase during its injection.
- 3) Validating the success of this injection technology to aid in the dissolution of the injected gas through a series of injection well monitoring techniques.

1.2.3 WP3: Surface and subsurface monitoring

WP3 was led by UI but OR, CNRS and UCPH also participated in the work package. Four deliverables and one milestone were associated with the work package. The main objectives of the work package were:

- 1) To design, implement, and demonstrate a coherent surface and subsurface monitoring plan for the Hellisheidi carbon injection site.
- 2) To create data from laboratory experiments that assist with assessing the long-term viability of carbon storage sites and validate hydrogeochemical models in WP6.

The following tasks were carried out to meet these goals:

- 1) Perform regular CO₂ soil flux measurements to check for CO₂ leakage to the surface.
- 2) Design and implementation of a well water sampling program. This task included the optimization of techniques to sample fluids at elevated pressures, some of which may exsolve during depressurization that occurs by bringing samples back to the surface.
- 3) Chemically analyse the solutions that have been sampled, and their interpretation including solution modelling and mass balance for tracers.

1.2.4 WP4: Laboratory experiments

WP4 was led by UCPH but CNRS and UI also participated in the work package. Three deliverables and one milestone were associated with the work package. The main objective of WP4 was to perform key experimental to improve significantly our ability to accurately predict the long-term fate of injected CO₂. Many of the parameters required for the accurate modelling of the long-term fate of CO₂ injected into the subsurface, and in particular the rates and distribution of precipitated phases were unknown prior to the start of the CarbFix project.

The following tasks were carried out to meet this goal:

- 1) Measurement of the precipitation rates of carbonate minerals including calcite and magnesite leading to development of a robust set of equations to describe these processes as a function of temperature, CO₂ pressure and the availability of suitable nucleation sites.
- 2) Acquisition of host and cap-rocks from Hellisheidi and Compostilla sites and reaction of these samples at temperature and CO₂ pressure conditions that are predicted by model calculations to be present during CO₂ injection efforts.

- 3) Further efforts on the Hellisheidi and Compostilla samples have been performed to assess if the reactions between rocks and CO₂-rich fluids in the subsurface might liberate toxic metals to the environment.

1.2.5 WP5: Improvement of thermodynamic and kinetic databases for CO₂-rich fluids

WP5 was led by CNRS but A21 also participated in the work package. Three objectives and one milestone were associated with the work package. The main objectives of WP5 were:

- 1) To update the existing thermodynamic databases.
- 2) To create a kinetic database that will allow for the accurate prediction of fluid-rock interaction during carbon storage efforts.

The following tasks were carried out to meet this goal:

- 1) Gathering and synthesizing rate and thermodynamic data obtained in the literature.
- 2) Evaluating these data to generate a consistent improved database for these properties with emphasis on those mineral sand fluids most likely involved in carbon storage systems.

1.2.6 WP6: Geochemical and hydrological modelling

WP6 was led by A21 and OR. Five deliverables and two milestones were associated with the work package. The main objectives of WP6 were:

- 1) To use existing geochemical and hydrological codes to model and predict the behaviour of fluid transport, injected CO₂ and reactivity at the Hellisheidi and Compostilla sites.
- 2) To use these comparisons to validate and improve these models and thereby optimize short- and long-term management of the Hellisheidi and Compostilla sites.
- 3) To identify the secondary minerals that will compete with CO₂ for dissolved cations.

The following tasks were carried out to meet this goal:

- 1) Evaluating and updating conceptual and numerical models
- 2) Incorporation of the improved databases generated from WP5 into existing geochemical and hydrological modelling codes and as there is cause for, setup of primary rock, alteration minerals and fluid chemistry will be revised
- 3) Comparing the results of model calculations with field and laboratory observations to assess the quality of and to validate model calculations

1.2.7 WP7: Dissemination and exploitation

WP7 was led by OR and CNRS. Ten deliverables and two milestones were associated with the work package. The main objectives of WP7 were:

- 1) To ensure that the results and advances obtained by CarbFix are adequately reported to the stakeholders including government decision makers, international scientists, interest groups, industrial companies, the media, and the general public.
- 2) To define the exploitation route for the results of the project and to manage intellectual property between CarbFix members.

The following tasks were carried out to meet this goal:

- 1) Creation and regular updating of the CarbFix website.
- 2) Maintenance of Press office/writing of Press releases.
- 3) Writing and submission of annual dissemination reports.
- 4) Running international public outreach forums.
- 5) Presentations at international conferences and specialist meetings and publication of scientific papers.
- 6) Market research and development of a business plan in order to create the foundation for exploitation results.
- 7) Presenting exploitation plans and results.

1.3 A description of the main S&T results/foregrounds

The work in CarbFix was divided into seven interlinked work packages. The following subchapters describe the main results obtained in each package and the work done to achieve the results.

1.3.1 Work Package 1: Project management

One of the first tasks in CarbFix was to develop a Project Management Plan. The plan contained the description of partners' roles, project procedures, structure and interaction of work packages (Pert chart), project schedule (Gantt chart), meeting schedule, milestone and deliverable schedule, and financial reporting schedule. The first version of the Project Management Plan was delivered Oct. 31 2011 (deliverable D1.1). The plan has been maintained since to ensure consistency with project schedule and activities. Overall, the consortium kept track with original tasks, milestones and deliverables in the project.

The main achievements of the consortium as a whole are:

1. Successful injection of pure CO₂ into the shallower reservoir in Hellisheidi between Jan-Mar 2014. Injection infrastructure worked without problems according to design. Solubility trapping was confirmed to occur within minutes within the injection well resulting in a single phase fluid entering the storage formation.
2. Injection of CO₂-H₂S gas mixture (75% CO₂, 25% H₂S) into the shallower reservoir in Hellisheidi (400-800 m depth) was carried out in summer of 2012 and injection into deep reservoir (> 1000 m depth) in Sept-Dec. The injection ran smoothly in both reservoirs but microbial activity in the shallower reservoir caused clogging problems. No such problems occurred within the deep reservoir where conditions are more reduced and microbiota not present.
3. Impressive and promising monitoring results indicate between 80-90% of injected carbon to be mineralized within a year from injection. The results show how efficient the CarbFix injection method is, as mineralization in geologic carbon storage is generally considered to take thousands to tens of thousands of years.
4. We have developed much needed thermodynamic and kinetic databases describing reactions of importance for CO₂ sequestration. These are of great importance for increasing the accuracy of numerical simulations.
5. Complex 3D reactive transport models developed of Hellisheidi and Los Páramos injection sites. Predictive simulations indicate CO₂ mineral storage in basalts to be a viable option for reducing anthropogenic CO₂ emissions.
6. We followed the ambitious dissemination and exploitation activities that we set out to follow at the project's kick off meeting. The consortium has given over 70 talks and presentations at international conferences and about 30 papers have already been published, several of those in high impact journals. In addition, we have developed a web-based interactive educational material on CCS and CarbFix for children and were highly involved with four international public outreach forums.

1.3.2 Work Package 2: Injection technology

The following deliverables explained in detail the work carried out within WP2 over the course of the project:

- D2.1 Report on optimal gas compositions for CO₂ injection
- D2.2 Development of gas mixing techniques favouring gas dissolution down-hole
- D2.3 New and improved well sampling techniques

The main results obtained within this work package include:

1. The injection system has been proven successful in co-injecting CO₂ and water to the subsurface for prolonged period of time.
2. Subsurface monitoring technologies (bailer sampling, WP3) and downhole camera have validated a complete dissolution of CO₂ in water prior to entering the target reservoir. Failure of complete gas dissolution in the injection system may be rapidly detected by gas detectors in the injection well headspace.

These are explained in more detail in the following subsections.

1.3.2.1 Injection System

Designing and optimizing injection technology that promotes the dissolution of the injected gas with an aqueous phase was one of the major tasks in WP-2. A detailed description of the injection system can be found in deliverable report 2.2 and in scientific paper Sigfússon et al. (2014), accepted for publication in International Journal of Greenhouse Gas Control pending revisions.

At the Hellisheidi CCS site, the availability of ample water supplies makes it possible to co-inject sufficient water to dissolve CO₂ during the injection process. The dissolution of CO₂ is accelerated by increasing CO₂ pressure and CO₂/water interface area. At Hellisheidi these factors are made use of by injecting water and CO₂ together to a depth of 300-350 m, where the injected gas is added to the water using a 'bubbler' that creates small gas bubbles. We have demonstrated that all injected CO₂ is dissolved prior to its infiltration into the target aquifer at a depth of 520 m. The dissolution of the gases during the injection promotes necessary water rock reactions to trap CO₂ into minerals in the formation receiving the CO₂ loaded waters as well as greatly increasing the storage security.

Two types of gases, pure CO₂ and a gas mixture from Hellisheidi Power Plant, have been injected. The injection system was essentially the same in both injections except for the part that provides the gas for injection. In the pure CO₂ injection carried out in January – March 2012 commercial CO₂ was stored in a pressurized tank but in the gas mixture injection in the 2012 a gas separation station provided gas for the injection.

The main components of the injection system at the Hellisheidi CCS site are the following:

1. Gas supply system comprising either of
 - a. A gas storage tank for the pure CO₂ gas (phase 1)
 - b. A gas separation station to treat the geothermal gas prior to injection (phase 2). This step essentially involves separating the poorly soluble geothermal gases H₂, N₂, CH₄, Ar and O₂ from the more soluble sour gases CO₂ and H₂S.
2. A pipe to transport the gases 3.3 km at 25-30 bars towards the shallower injection site.
3. A pipe to transport the gases 3 km at 8 bars towards the deeper injection site.

4. Injection well HN-02 at the shallower injection site where the gases are dissolved in water at 300-350 m inside the well.
5. Gas mixing facilities where gases and water were mixed on surface at 8 bars at the deeper injection site.
6. Injection well HE-08 at the deeper injection site.
7. Water supply well HN-01 at the shallower injection site which provides water to dissolve the gases during injection.
8. Tracer mixing system in the gas separation station and in facilities near both injection wells.

Gas injections carried out in the project are as follows:

Pure CO₂ injected into the shallower storage reservoir in Hellisheidi (400-800 m depth):

- 1.1 tons in March 2011
- 174.6 tons January – March 2012

Gas mixture (75% CO₂, 24% H₂S and 1% H₂) from Hellisheidi Power Plant injected into the shallower storage reservoir in Hellisheidi (400-800 m depth):

- 60 tons June – August 2012
- 3.5 tons January 2013

Gas mixture (64% CO₂, 35% H₂S, 1% H₂) from Hellisheidi Power Plant injected into the deeper storage reservoir in Hellisheidi (>800 m depth):

- 100 tons September – December 2012

The injection of pure CO₂ in January to March 2012 was highly successful. Gas flow into the injection well is shown in Figure 4. After initial start-up problems caused by clogging in the sparger at the end of the gas pipe 300-350 m in the injection well, the gas flow into the well was relatively stable. The clogging in the sparger is evident in Figure 4 **Error! Reference source not found.** from the repeated sharp decrease in gas flow rate from the beginning of injection on January 17th until 2nd of February. The purpose the sparger is to secure that the gas bubbles leaving the gas pipe are small to speed up the dissolution process. Unfortunately it also acts as a filter at the end of the gas pipe. To prevent clogging the average diameter of the holes in the sparger was increased from 0.01 mm to 1 mm. After this increase gas flow was stable. Detailed characterization of the solids causing the sparger clogging was carried out. Variations in the gas flow rate from 2nd of February and onward are caused by pressure changes in the CO₂ storage tank.

Gas mixture injection started in June 2012 (Figure 5). 1-2 % of the gas from Hellisheidi power plant was diverted to the gas separation station where it was separated into soluble and non-soluble gases and subsequently the soluble gases (CO₂ and H₂S) were injected. For the first time in the CarbFix project a complete CCS cycle was achieved. Geothermal gases coming from the production field of Hellisheidi Power Plant were captured and re-injected back into the ground. At the time of injection of the gas mixture the gas separation station was providing gas for injection into both storage reservoirs in Hellisheidi (400-800 m depth and >800 m depth). The flow rate from the station was

however not high enough leaving both injections starved for gas. Decision was then made to postpone injection into shallower system until gas injection into the deeper system was finished. In total around 60 tons gas were injected into the shallower system in the summer of 2012. About 100 tons of gas was injected into the deeper system between September and December 2012.

In January 2013 it became evident that road construction work in the fall of 2012 had damaged the gas pipe transporting the gas from the gas separation station to the injection site. The pipe was seriously damaged and repeated failures on pipe followed. When the gas pipe had been fixed and injection was ready to resume a rising water level in the injection well was observed. Rising water level at constant flow rate of water means decreasing transmissivity of the well. Water level measurements showed an increase in the water level of the injection well after the injection of gas mixture stopped in August 2012. Injectivity tests showed that the injectivity index had decreased from 2.0 kg/s/bar to 0.2 kg/s/bar. The transmissivity of the well was regained via airlift pumping of the well, during which we learned that microbial activity had caused the clogging of the well by linking together small Fe-S particles in a gel-like precipitation.

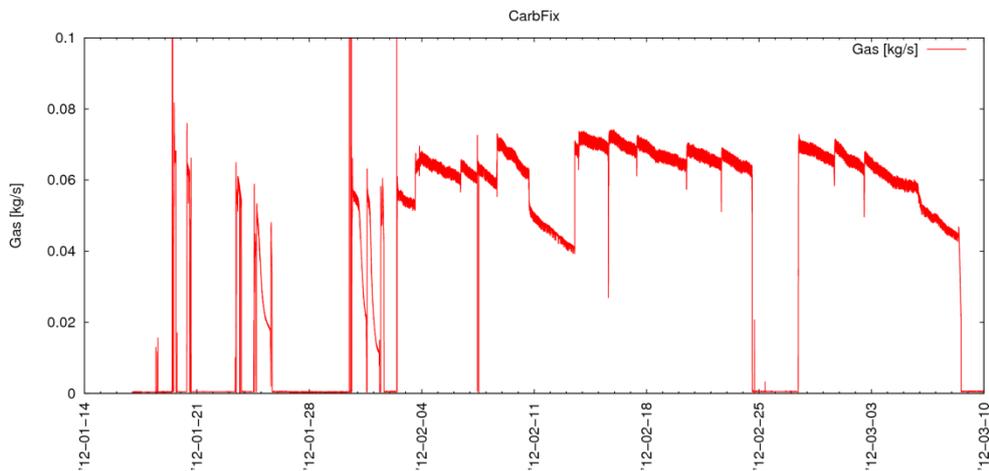


Figure 4: Gas flow rate into HN-2 during the pure CO₂ injection from January until March 2012.

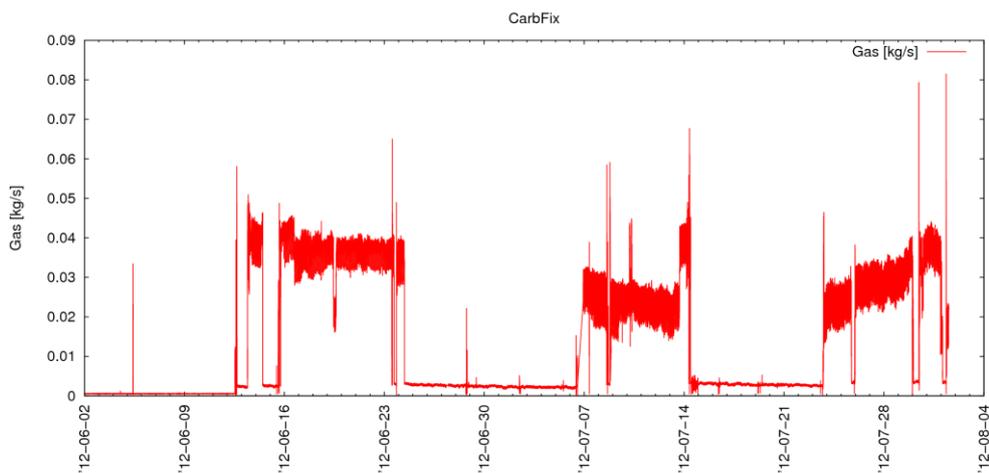


Figure 5: Gas flow rate into HN-2 during the gas mixture injection from June until August 2012.

1.3.2.2 Validating injection technology

One of the main tasks in WP-2 was to validate the injection technology. Validating the injection technology was done in three ways. 1) Direct camera observation in the injection well. 2) Down hole sampling of the injected fluid. 3) Monitoring the CO₂ and H₂S concentration in the headspace of the injection well.

Direct camera observation

Verification of the complete dissolution of CO₂ during its injection was performed by digital downhole camera (CCV Engineering & Manufacturing Inc., WC1750 slimline waterwell inspection camera). Images were taken during pure CO₂ injection on 16 March 2011 at depths from 90 to 550 m. These images, such as presented in Figure 6 **Error! Reference source not found.** show the well to be void of gas bubbles consistent with the complete dissolution of CO₂. Mass balance calculations indicate that the average residence time of the CO₂-H₂O fluid in the well from the sparger to a depth of 540 m was 4 minutes and 30 seconds. Solubility trapping was therefore achieved in less than five minutes.



Figure 6: Image taken at a depth of 481.9 m in the injection well at 16:06 16 March 2011. The camera enters the injection well through the service pipe. At this time the CO₂ and H₂O inlet flows were 27.1 and 750 g/s, respectively. No CO₂ bubbles are visible consistent with the complete dissolution of CO₂ during its injection.

Downhole sampling

A major effort in WP-2 was to develop and test well sampling techniques to validate the injection technology. This was done by *in situ* sampling of the downhole fluids and measurements of *in situ* pH and total dissolved inorganic carbon. New sampling technique was developed and tested in the CarbFix project and these were detailed in deliverable 2.3.

The downhole sampler was extensively tested during the pure CO₂ injection in January to March 2012. In total 11 fluids samples were obtained from a depth of 530 m. Each of these fluid samples was analyzed for total dissolved inorganic carbon and 6 of the 12 were measured for *in-situ* pH. As shown in Figure 7, in each case the dissolved inorganic carbon concentration of the sample fluid was 0.82±2% mol/kg and the fluid pH was 3.84±0.1 confirming the complete dissolution of the CO₂ during

its injection. The very good match between theoretical dissolved inorganic carbon concentrations calculated from the flow rates of CO₂ and water means that the bailer accurately reproduced the average amount of DIC entering the aquifer over the entire injection period. These findings altogether clearly support the operational capability of the bailer and sampling line for the specifics of the CarbFix injection site.

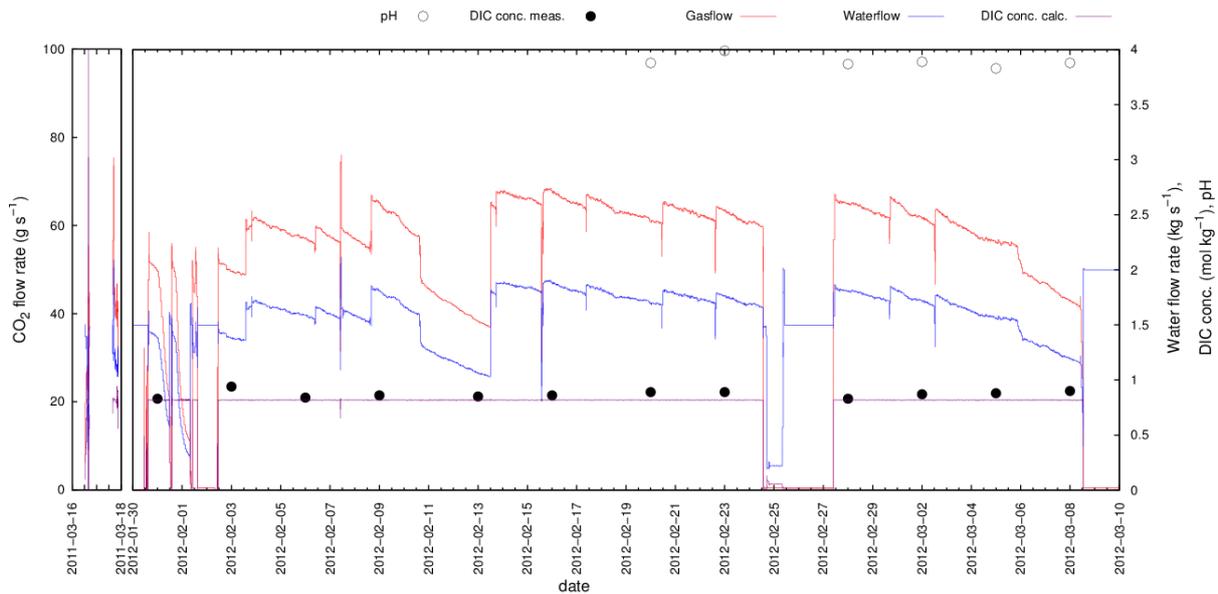


Figure 7: Temporal variation of CO₂ (red curve) and water (blue curve) flow rates of the injected fluid, calculated dissolved CO₂ concentration, and measured fluid pH (open circles) and dissolved CO₂ concentration (filled circles) at the CARBIFIX injection site in Hellisheidi (Iceland) from 15 March to 17 March 2011 and 28 January to 11 March 2012. Figure from Sigfússon et al (2014) accepted for publication in Int J Greenh Gas Control pending revisions.

Concentration of CO₂ and H₂S in headspace of HN-2

Carbon dioxide and hydrogen sulphide levels in atmosphere are measured with infrared technology (Crowcon Xgard) and by electrochemistry (Crowcon Xgard) respectively and the value recorded every minute to a Campbell CR 1000. Monitoring started in June 2012. Initially the headspace was sampled at 70 meter depth, pumped towards the sensors prior to being returned to the headspace near the wellhead. Now the headspace is sampled near the wellhead due to increased water table in the injection well.

Elevated concentrations were measured when the mass ratio of water to gas was 24 instead of 42 leading to insufficient dissolution of gases that consequently rose through the water column and were detected almost immediately (Figure 8). Therefore, installations of gas detectors in the well headspace are useful tool monitor the method effectiveness.

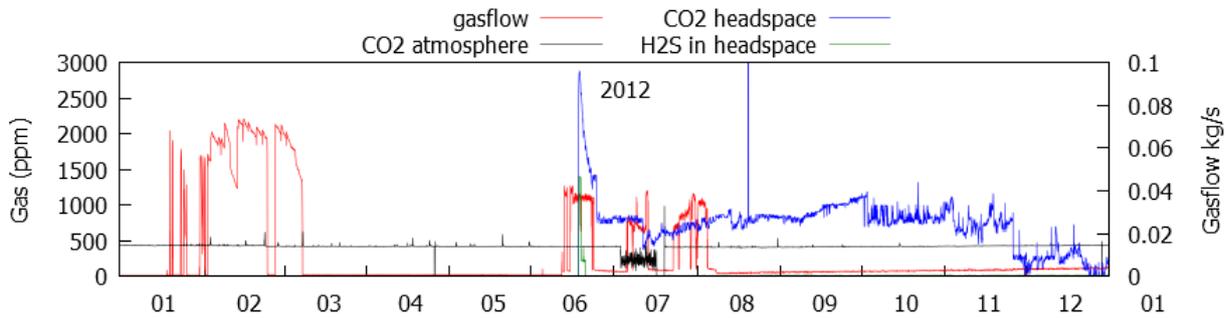


Figure 8: Concentration of CO₂ and H₂S in the HN-2 headspace during the year 2012. Elevated concentrations were measured when the mass ratio of water to gas was 24 instead of 42 leading to insufficient dissolution of gases that consequently rose through the water column and were detected almost immediately.

1.3.3 Work Package 3: Surface and subsurface monitoring

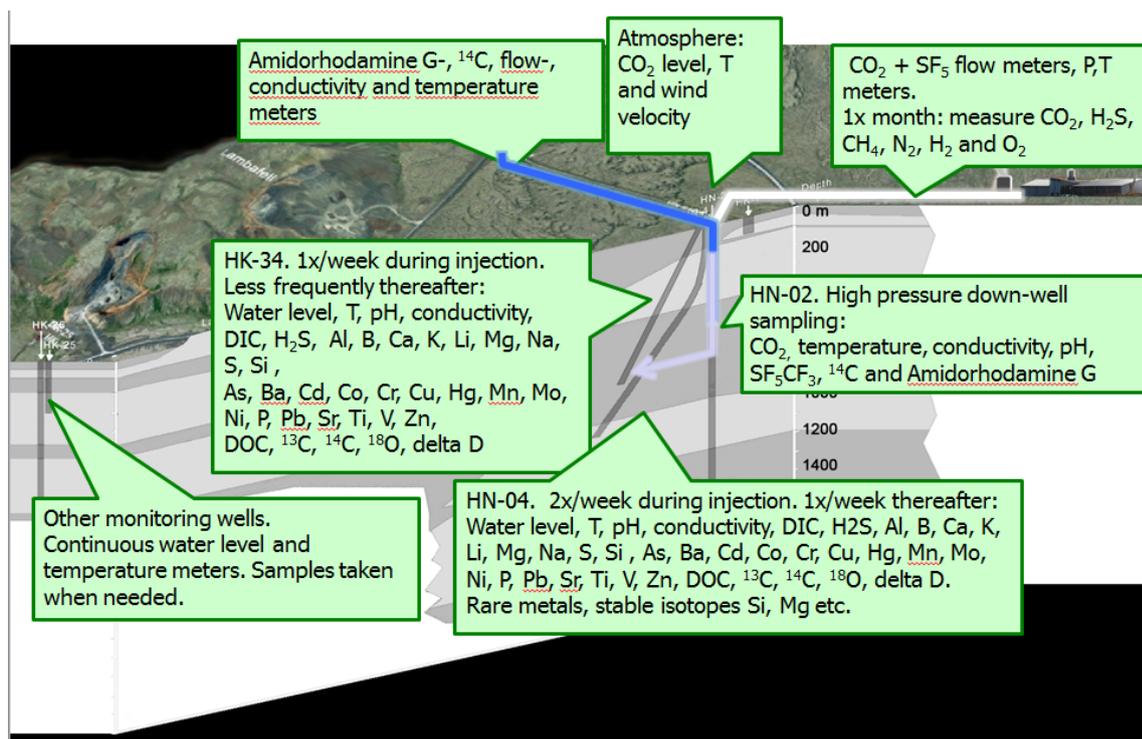


Figure 9: A summary of monitoring in the various wells, pipes and air at the Hellisheidi injection site. In addition the CO₂ soil flux is monitored on selected sites as described below.

Public acceptance requires that subsurface CO₂ storage sites are adequately monitored to assure the safety of CO₂ storage efforts. The goal of this work package was to design, implement, and demonstrate a coherent surface and subsurface monitoring plan for the Hellisheidi carbon injection site. Information obtained by the WP is essential to assess the long-term viability of carbon storage

sites, and has e.g. been used by CarbFix to 1) measure the rate of mineralisation of the injected CO₂ and H₂S, 2) measure the mobility of toxic metals, 3) assess CO₂ leakage, 4) identify potential addition experiments to be run by WP-4, and 5) validate hydro-geochemical models in WP-6. Subsurface monitoring is summarized in Figure 9.

The following deliverables explain in detail the work carried out within WP3 over the course of the project:

- D3.1) Assessment of CO₂ leakage: The CO₂ leakage at injection has been assessed with the aid of CO₂ measurements in the atmosphere, wellhead and by CO₂ soil flux measurements [month 15].
- D3.2) Data acquired to validate reactive transport modelling codes: Waters from monitoring wells sampled and analysed to validate reactive transport modelling codes [month 24].
- D3.3) Soil CO₂ flux measurements [month 36].
- D3.4) -Development of novel injection system, high pressure sampling tool, high pressure sampling lines to bring the samples to ambient pressure, and tracer monitoring tools [month 36].

The main results obtained within WP3 are:

1. Analysis of dissolved inorganic carbon (DIC), S, conservative tracers and the reactive tracer ¹⁴C concentrations from monitoring wells showed that 90% of the injected carbon and sulphur were mineralized within a year. No leak from the reservoir has been detected by surface monitoring.
2. Analysis of major and trace dissolved constituents has shown that dissolution of basalt and secondary minerals within the basalt raises the pH of the pure CO₂ charged injected water, from about 3.8 to 6.5 during injection and above 9 after injection, as the water travelled from the injection well to the monitoring well. The distance between the wells is 125 m at 520 m depth and the first arrival of the CO₂ charged water was after 17 days. Simultaneously, the concentration of divalent cations increases in the CO₂ charged waters as it emerged in the monitoring well. These waters were saturated with Fe-carbonate, during injection and calcite after injection, which sequestered more than 90% of the injected carbon within a year after. The pump in the monitoring well broke down during the monitoring period after the gas injections. The cause were carbonates, the pump was full of calcite. The concentrations of several metals that can pose threat to humans and biota, increase following the CO₂ charged waters rock interactions. However, the concentrations are all well below the allowable limits for drinking water.

These results are explained in more detail in the following subsections.

1.3.3.1 Injection of tracers

The injection of a number of novel geochemical tracers together with the CO₂, including the radioactive tracer ¹⁴C that can be used to determine directly the movement of the dissolved gas, and validate the extent of mineralization in the subsurface. The injection of gases into the shallower injection site was carried out in two main phases as described above:

- a. During the period of January to March 2012, 175 tons of pure commercially bought CO₂ were injected together with ¹⁴C reactive tracer and SF₆ conservative tracer.
- b. During the period of June to August 2012, 65 tons of a CO₂:H₂S:H₂ mixture (75:24:1 vol%) were injected together with ¹⁴C reactive tracer and the conservative tracers SF₅-CF₃ and amidorhodamine G.

All tracers were injected in a fixed ratio to the CO₂ injected. The conservative tracers allow for quantification of the dilution of the injected mixtures into the reservoirs and hence can be used to subtract the dilution factor from the signal obtained by the reactive tracer ¹⁴C. As a consequence the analysis of the isotopic ratio in monitoring waters compared to isotopic ratio of injection mixtures gives an indication of carbon mineral storage in the subsurface. Furthermore, a comparison of “expected” and “measured” dissolved carbon and sulphur concentrations in the monitoring wells can underscore the mineral fixation of these gases. Detailed results describing co-interpretation of tracer monitoring data will be published in a paper that is currently in the final stages of preparation, intended for publication in a high impact journal (Matter et al. (2014) in preparation).

1.3.3.2 Design and implementation of a well sampling program

This task included the optimization of techniques to sample fluids at elevated pressures, some of which may ex-solve during depressurization that occurs by bringing these samples back to the surface. A bailer sampler was designed, tested in the laboratory and in the field before and during the injections. The original design is shown in Figure 10 a) as published in Alfredsson et al. (2011) was modified after several tests as shown in Figure 10 b) when a vacuum chamber was added to the bailer greatly improving its performance (Alfredson et al. (2014) in review). This improved bailer was used during the injection of CO₂ in 2012 as described in discussion for WP2 and in a paper in accepted for publication in International Journal of Greenhouse Gas Control pending revisions (Sigfússon et al. 2014).

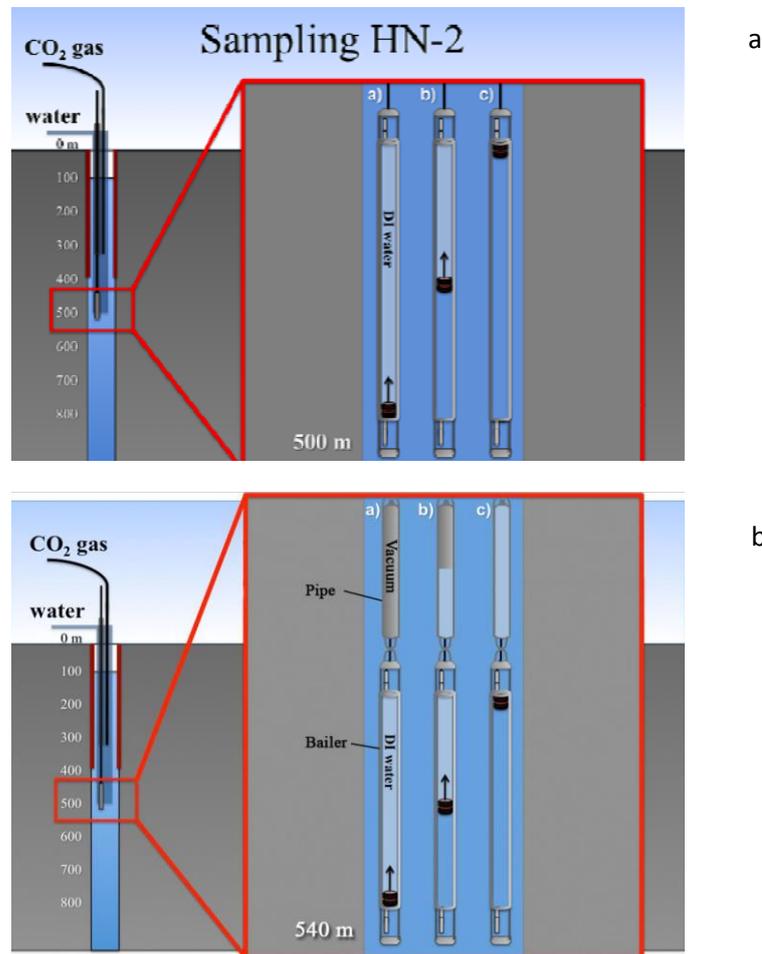


Figure 10: a) An example of downhole sampling at 500 m depth in the CO₂ injection well HN2. The bailer is lowered through a sampling pipe down to about 500 m depth. The CO₂ gas is injected into the down-flowing water stream at about 350 m. When the bailer reaches 500 m depth, a) the bailer opens and the CO₂ charged water flows into the sampling end of the bailer, moving the piston upwards and forces the DI-water out through the upper end via a tubing extending all the way to the surface. b) The sampling is half complete after ~1 hour and c) sampling is finished in about two hours' time (Alfredsson et al. 2011). b) An example of downhole sampling at 500 m depth in the CO₂ injection well HN-2. The bailer is lowered through a sampling pipe down to about 500 m depth. The CO₂ gas mixes with the injected water at about 350 m. When the bailer reaches 500 m depth, a) the bailer opens and the CO₂ charged water flow into the sampling end of the bailer, moving the piston upwards and forces the DI-water out through the upper end into the vacuum chamber. b) The sampling is half complete after ~1 hour and c) sampling is finished in about two hours' time (Alfredsson et al. 2014 in review).

1.3.3.3 Chemical analysis of the sampled solutions

This task focused on chemical analyses of the solutions that have been sampled, and their interpretation including solution modelling and mass balance for tracers. All major and trace elements concentrations in waters and rocks, sampled from all the CarbFix wells at the Hellisheidi injection site (Figure 9), from 2008 to 2010, have been analysed, interpreted and published (Alfredsson et al. (2013) *Int J Greenh Gas Control*, 12, 399–418).

All major and trace dissolved constituents have been monitored from the injection in January 2012 to the end of September 2014. Some isotope ratios and some tracer concentrations are left to be

measured. The frequency of sampling was twice a week during injections to once every week in the following months and recently once every other week in HN4, the first monitoring well as shown in Figure 9. This covers both the pure CO₂ injection in January-March 2012 and the gas mixture injection in June-August 2012. The pump broke down in August 2013 and then again in the summer of 2014, causing two gaps in the data collection. The pump failure was caused by massive calcite (CaCO₃) precipitates within and on the outside of the pump. We were thus “too successful” in mineralising the injected carbon. Detailed results on chemical analysis and subsequent geochemical modelling of sampled solutions will be described in a scientific paper that is the final stages of preparation (Snaebjornsdottir et al. 2014 in preparation).

Most of the tracers and all major and trace dissolved constituents have been measured in the first monitoring well HN-4 before, during and after injection up to September 2014. The pH of the water in the first monitoring well, HN-4 dropped from about 9.5 to about 6.5 as the injected water arrived in the well. It remained low weeks after injection was terminated, and then slowly rose towards the pre-injection value and then dropped in similar fashion during the gas mixture injection. Many other dissolved constituents shows similar, but reverse trend. The concentrations of Mg, Ca and Fe²⁺ increases once the “plume” arrives, and reaches maximum at the lowest pH and then decline towards the pre-injection value after injection was stopped. The concentration of metals that can pose threat to humans and biota were below the allowable limits for drinking water before, during and after injection in the first monitoring well as shown in Table 1.

Table 1: The maximum measured concentration of metals, that can pose threat to humans and biota, in the first monitoring well HN-4, and the maximum allowed concentration according to drinking water guidelines. The concentrations are highest during the injections.

Element	World Health Organization	European Union	Pure CO ₂ injection	CO ₂ -H ₂ S injection	Analytical Method
	(<i>μmol/l</i>)	(<i>μmol/l</i>)	(<i>μmol/l</i>)	(<i>μmol/l</i>)	
Arsenic	0.13	0.13	0.013	0.01	ICP-SFMS
Barium	5.10		0.003	0.003	ICP-SFMS
Boron	222	92	2.43	2.18	ICP-AES
Cadmium	0.027	0.044	0.0001	0.0001	ICP-SFMS
Chromium	0.96	0.96	0.001	0.013	ICP-SFMS
Copper	31.5	31.5	0.004	0.004	ICP-SFMS
Fluoride	79.0	79.0	31.0	21.0	IC-2000
Lead	0.048	0.048	0.001	0.002	ICP-SFMS
Mercury	0.030	0.0050	0.0001	0.0005	AFS
Nickel	1.19	0.34	0.024	0.013	ICP-SFMS
Manganese	*	*	0.78	0.74	ICP-OES
Iron	*	*	36.0	10.3	ICP-AES
Aluminium	**	*	2.59	2.58	ICP-SFMS

*Not of health concern at levels found in drinking-water. May affect acceptability of drinking-water **A health-based value of 33.4 *μmole/l* could be derived, but this value exceeds practicable levels based on optimization of the coagulation process in drinking-water plants using aluminium-based coagulant.

1.3.3.4 CO₂ flux measurements

This task was amended to large extent as experience has been gathered during gas injections. Surface monitoring is carried out continuously for CO₂ in atmosphere (see Figure 11 and Figure 12) and the injection well head space (CO₂ and H₂S, see discussion in relation to WP2). Continuous data are available on-line to the operators of Carbfix at the passport protected site gogn.vista.is.

No indications of gas leak have been detected throughout the duration of the project with either CO₂ analysis in the atmosphere or by the aid of flux chambers (see discussion in deliverable D3.1). The CO₂ flux from the surface was carried out according to monitoring plan that was updated during the injection of pure CO₂ during January to March 2012. There, the CO₂ flux through the surface was monitored regularly (Figure 13) without any CO₂ being detected.

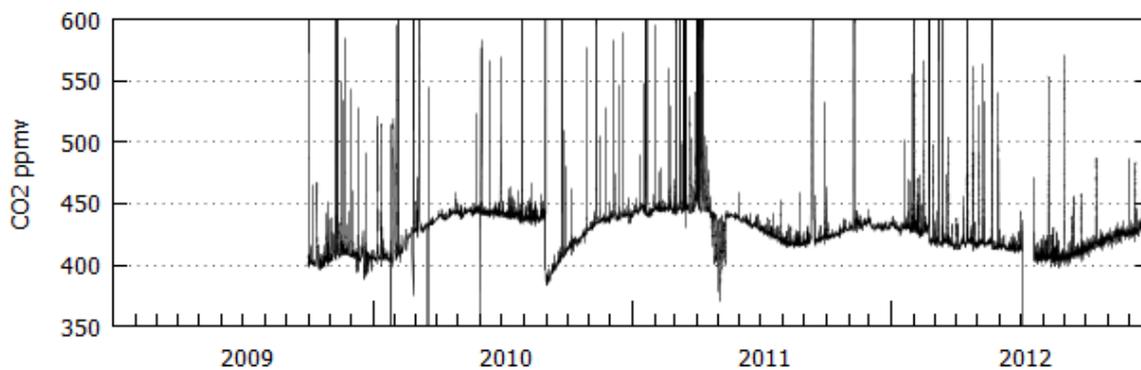


Figure 11: Carbon dioxide concentration at the CarbFix injection site. Spikes in the concentration levels are mainly associated with vehicle traffic in the injection area during sampling, maintenance and setup of equipment.

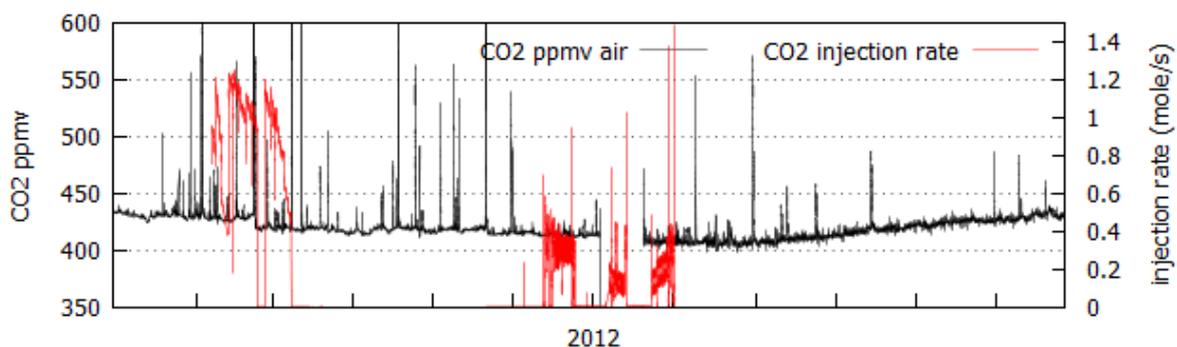


Figure 12: Concentration of CO₂ in atmosphere at the injection site during the year 2012. Elevated levels of CO₂ were never observed for more than 1-6 hours during site visits.

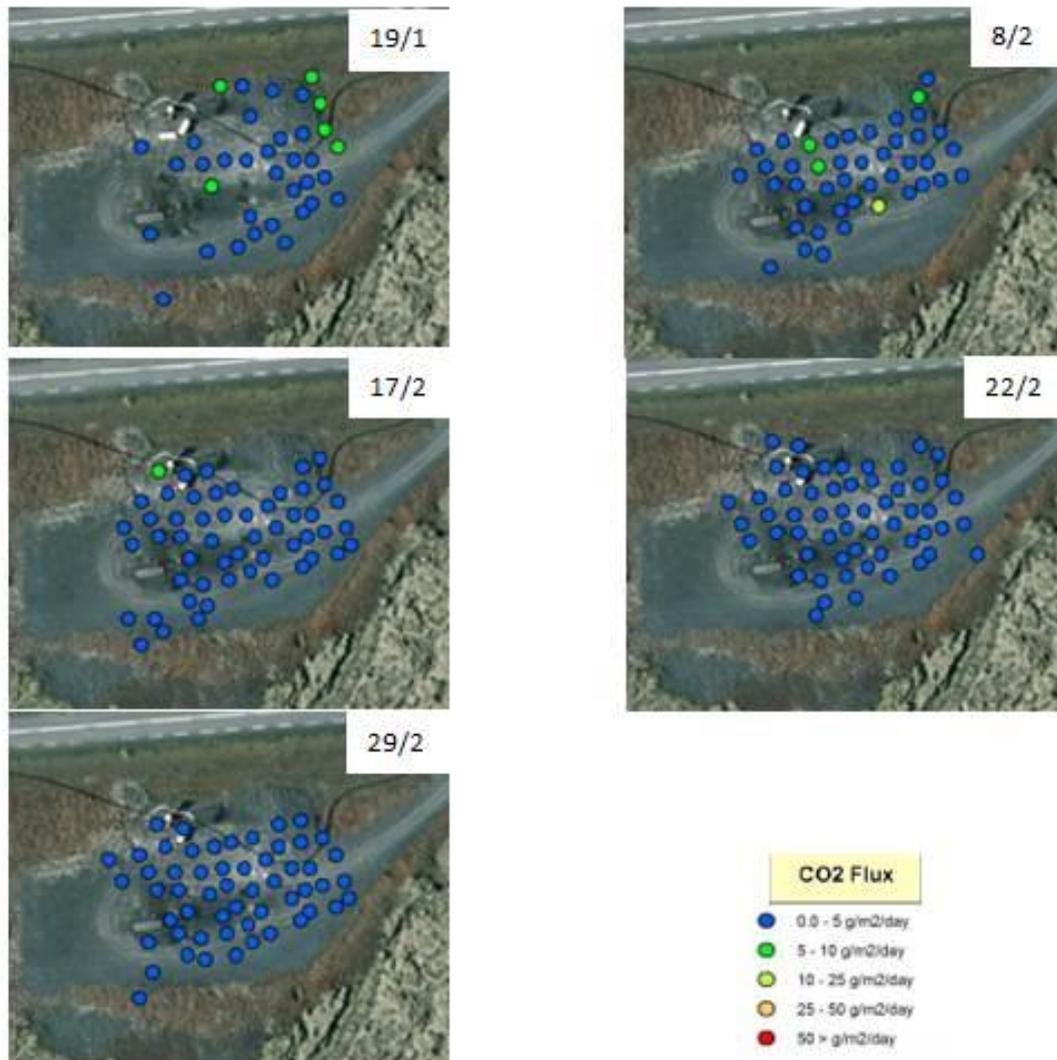


Figure 13: CO₂ flux prior and during the injection of pure CO₂ in January to March 2012. No indication of leak was detected.

1.3.4 Work package 4: Laboratory experiments of fluid-host rock and fluid-cap rock interaction

Work package 4 focussed on experimental investigations of 1) carbonate precipitation rates, 2) host rock and cap-rock reactivity during interaction with dissolved CO₂, and 3) release of toxic elements during the interaction. CarbFix has made substantial progress and a summary of the major results in each of these areas is provided below.

Although carbonate precipitation has been studied extensively for decades, research performed within CarbFix has significantly improved our understanding of carbonate precipitation rates and mechanisms as well as of heterogeneous nucleation of calcite on basaltic minerals and glass.

Studies on host rock reactivity showed that divalent cations are released during interaction between basalt and CO₂-bearing solutions, providing the first step for carbonation. At lower pH, Fe and Mg is

released faster than Ca, suggesting that Fe- and Mg-carbonates are likely to form in the initial stages of carbonation. Consistent with these results, characterisation of basalt after reaction showed that the surfaces became poorer in divalent cations. Importantly, results show that the dissolution of the common basaltic mineral diopside is unaffected by calcite surface coating. Experiments with forsterite, however, show that interaction between this mineral and CO₂ may lead to formation of a passivating layer, at least at higher temperature and pressure.

Experiments using a high pressure, flow-through column reactor, show that the initial interaction between basalt and CO₂-bearing water releases only four elements in concentrations above the drinking water limit (Al, Fe, Mn and Cr). In their trivalent state, these elements form insoluble oxides and hydroxides at circum neutral pH, suggesting that their concentration will decrease significantly upon further buffering of pH by basalt dissolution. However, in an oxic environment Cr(III) may oxidise to Cr(VI)O₄²⁻, which is toxic and much more mobile. The effect of pH buffering by basalt dissolution and the effect of redox reactions will be tested in the batch experiments with the Hellisheidi host rock.

The following deliverables explain in detail the work carried out within WP4 over the course of the project:

- D4.1) Measurement of carbonate precipitation rates in the laboratory: Carbonate mineral precipitation rates as a function of temperature and pressure, required by WP-5. [month 12]
- D4.2) Report on host and cap-rock reactivity: Host and cap-rock reactivity experiments to validate and improve geochemical models. [month 36]
- D4.3) Report on toxic metal release: [month 36]

The main results obtained in WP4 are:

1. Experimental studies on the precipitation rate of magnesite (MgCO₃), calcite (CaCO₃) and siderite (FeCO₃) has provided understanding on formation rates and mechanism as well as on heterogeneous nucleation of calcite on basaltic minerals and glass.
2. Studies on host rock reactivity showed that divalent cations are released during interaction between basalt and CO₂-bearing solutions, providing the first step for carbonation.
3. At lower pH, Fe and Mg are released faster than Ca, suggesting that Fe- and Mg-carbonates are likely to form in the initial stages of carbonation. Consistent with these results, characterisation of basalt after reaction showed that the surfaces became poorer in divalent cations.
4. The dissolution of the common basaltic mineral diopside is unaffected by calcite surface coating.
5. Experiments with forsterite, however, show that interaction between this mineral and CO₂ may lead to formation of a passivating layer, at least at higher temperature and pressure.
6. Experiments using a high pressure, flow-through column reactor, show that the initial interaction between basalt and CO₂-bearing water releases only four elements in concentrations above the drinking water limit (Al, Fe, Mn and Cr). In their trivalent state, these elements form insoluble oxides and hydroxides at circum neutral pH, suggesting that their concentration will decrease significantly upon further buffering of pH by basalt dissolution. However, in an oxic environment Cr(III) may oxidise to Cr(VI)O₄²⁻, which is toxic and much more mobile.

These are explained in more details in the following subsections.

1.3.4.1 Carbonate precipitation rates

Magnesite precipitation rates, measured at 100-200 °C increased with saturation state but decreased with increasing CO_3^{2-} activity and pH. Measured rates are consistent with a model that combines surface complexation and transition state theory and this can be readily implemented in speciation software for predictions. Determined reaction order agrees with magnesite precipitation controlled by a spiral growth mechanism (Figure 14). Comparison of measured magnesite precipitation rates with corresponding forsterite dissolution rates suggest that the relatively slow rates of magnesite precipitation could be the rate limiting step in mineral carbonation efforts in ultramafic rocks.

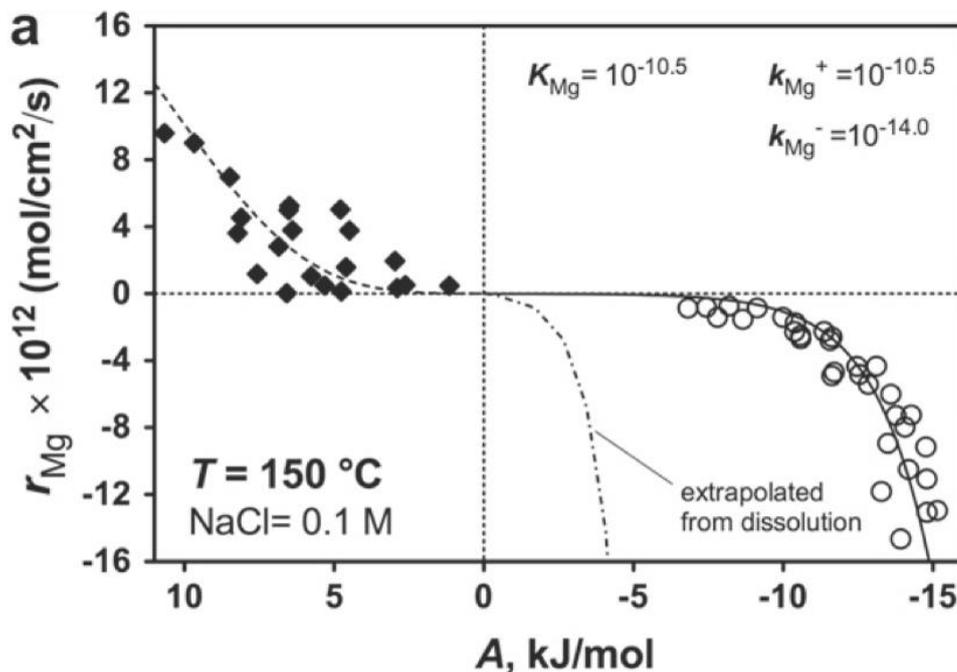


Figure 14: Precipitation rates as a function of chemical affinity. Rates are consistent with spiral growth but differ from those extrapolated from dissolution. This shows that large errors can arise in geochemical modeling from using the commonly adopted assumption that transition state theory derived dissolution rate equations can be used to predict mineral precipitation rates. Results are consistent with a pre-exponential factor equal to 5.9×10^5 mol/cm²/s and an activation energy, to 80.2 kJ/mol in the Arrhenius equation.

To study nucleation and growth on silicate minerals, calcite was precipitated in flow through experiments at 25 °C from supersaturated solutions (saturation index: 0.6, pH 9.1), in the presence of calcite or six common silicates. The onset of calcite nucleation and the amount of calcite precipitated depended on the identity of the substrate. In all cases, calcite precipitation occurs solely on the mineral substrate. We found no evidence of homogeneous nucleation. The results indicate that calcite nucleation and growth depend on the crystal structure of the silicate substrate. The implication for CO_2 mineralization in basaltic rocks is that trigonal carbonates nucleate more easily on crystalline rocks but they can form on glass surfaces as well.

At moderate supersaturation, calcite growth produces spirals where geometry reflects the relative growth rates of acute and obtuse steps. Because foreign ions, i.e. contaminants have different affinities for binding to the obtuse and acute growth sites, step growth velocity affects the extent of their incorporation. Experiments where supersaturation and $\text{Ca}^{2+}:\text{CO}_3^{2-}$ activity ratio were

systematically varied show that supersaturation significantly affects the growth velocity of the two step types, and causes marked differences in the geometry of the growth spirals.

Little is known about calcite recrystallisation at dynamic equilibrium. In sediments, this process leads to Ostwald ripening over longer time scales, which affects permeability as sediments are converted to rocks. Aging of synthetic, nanoscale calcite at up to 200 °C and 15 bar resulted in a 10 fold increase in grain size to lower free surface energy. During mineral carbonation, coarsening would move material to new sites and enlarge crystals, which might lead to fracturing, thus increasing surface area for further reaction.

Complex organic compounds are likely to be carried into the porous formation from the surface or be produced from microbial activity during lower temperature CO₂ injection. These can have a remarkable influence on CaCO₃ growth. In experiments with simple OH containing model compounds, X-ray photoelectron spectroscopy (XPS), molecular dynamics (MD) and density functional theory (DFT) show a correlation between adsorption energy, the length of the bonds between surface and adsorbate and the temperature at which the compounds desorb in vacuum.

Fe(II)-carbonates, such as siderite (FeCO₃), are likely to form during CO₂ sequestration but little is known about formation pathways. Mössbauer spectroscopy and pair distribution function (PDF) analysis of quickly precipitated Fe(II) carbonates showed that a previously undescribed, very poorly crystalline (near amorphous) phase formed. Replica exchange MD simulations of Fe(II) carbonate polynuclear complexes yield PDF peak positions that agree with those from experiments, indicating that the material could be a condensate of such complexes, assembled in a relatively unorganised fashion. The initial material transformed into siderite or chukanovite (Fe₂CO₃(OH)₂) with rates depending on pH (Figure 15).

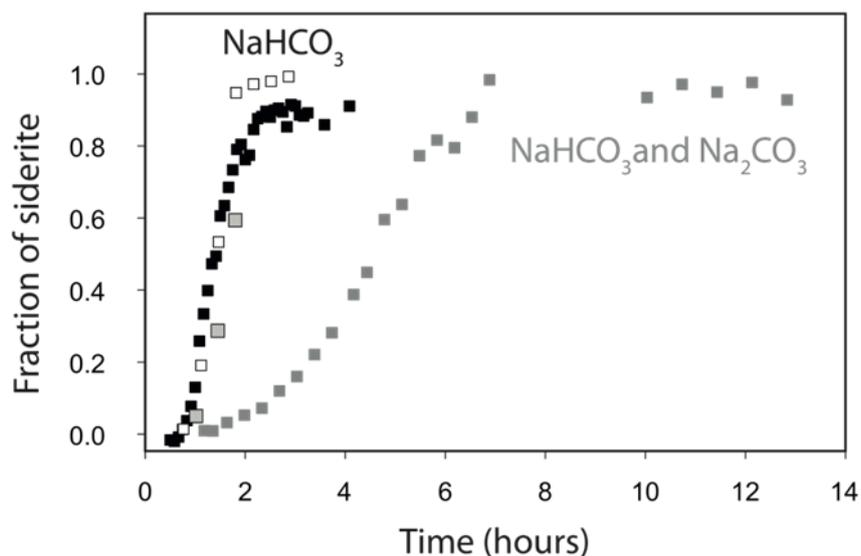


Figure 15: Fraction of siderite as a function of time from PDF.

1.3.4.2 Host rock and cap rock reactivity

The reactivity of host rocks has important implications for subsurface CO₂ storage. If CO₂ is to be trapped via mineral carbonation, divalent cations have to be released from primary silicate minerals

prior to their precipitation as carbonate minerals. In contrast, if CO₂ is to be injected into limestone, such as at Hontomin, CaCO₃ is expected to dissolve.

Steady-state element release rates from crystalline basalt dissolution at far-from-equilibrium were measured over a range of pH, 2-11, and temperature, 5-75 °C in mixed flow reactors. Si and Ca release rates increase at both acidic and basic conditions. In contrast, Mg and Fe release rates decrease continuously with increasing pH at all temperatures. Thus, crystalline basalt preferentially releases Mg and Fe relative to Ca at acidic conditions. The injection of the acidic CO₂ charged fluids into basalt could, therefore, favour the formation of Mg and Fe carbonates rather than calcite. In parallel column experiments to determine long term (1.5 year) chemical changes to the basalt during interaction with 5% CO₂, surface sensitive X-ray photoelectron spectroscopy showed decreased surface concentrations of Na, Fe, Ca, and Mg. This is consistent with a model involving initial, nonstoichiometric dissolution with preferential release of cations.

To probe the effect of carbonate precipitation on host rock reactivity, diopside dissolution rates were measured in neutral and basic solutions at 25 and 70 °C for up to 164 days. In solutions at 25 °C, supersaturated with respect to calcite, this mineral formed extensively on the diopside surfaces (Figure 16). In solutions strongly undersaturated with respect to calcite, no precipitates formed. Dissolution rates of diopside overgrown with calcite at 25 °C were 1) time independent and 2) the same as those in calcite free experiments. This indicates that diopside dissolution rates are unaffected by the simultaneous precipitation of calcite.

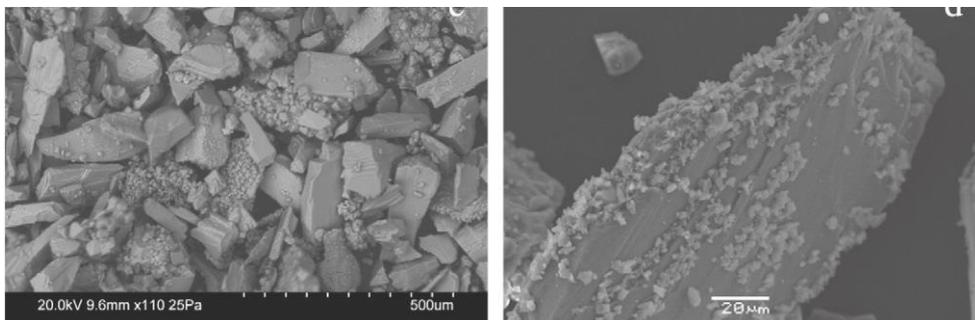


Figure 16: Scanning electron microscopy images of crushed diopside with secondary calcite precipitation.

Forsterite is abundant in mafic and ultramafic rocks. Its dissolution rate is relatively high and it is expected to transform to carbonate minerals quickly during mineral carbonation with rates limited by magnesite precipitation. During exposure to pure deionised water and supercritical CO₂ (120 °C, 80 bars) over 6-8 days, synthetic, Fe free forsterite transformed extensively to magnesite as expected (Figure 17). In identical experiments with natural, Fe bearing forsterite, magnesite formation was not detected, unless O₂ was excluded. XPS of natural forsterite, reacted in the presence of some O₂, showed that a thin surface layer formed that consisted of an outer, Si rich layer and an inner, Fe rich layer. Structurally, goethite matches forsterite, suggesting that a very thin layer of this mineral formed epitaxially over the forsterite and inhibited dissolution.

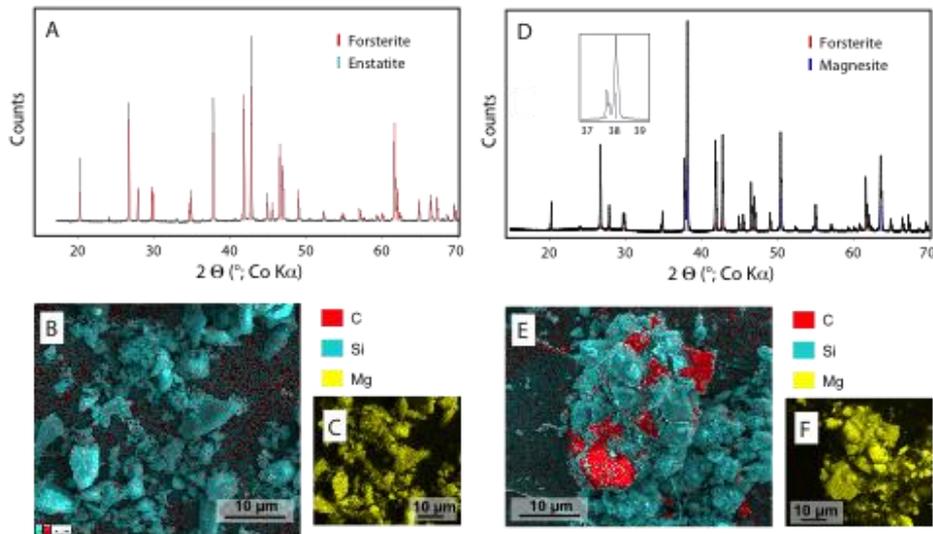


Figure 17: Structure, morphology and composition of two types of forsterite after reaction with supercritical CO₂. Natural forsterite: A) XRD pattern, B and C) EDXS maps. Synthetic forsterite: D) XRD pattern, E) and F) EDXS maps.

In a pilot project, CO₂ is to be injected into limestone situated at ~1500 m depth at Hontomin, Spain. To study the processes that are expected in the laboratory, limestone from the target formation was exposed to the conditions at the site (150 bar CO₂, 60 °C) for 1-7 days, using a SO₄ rich, synthetic brine or pure water. XRD of reacted material showed no significant structural evolution but SEM/EDXS showed a few ~ 50 nm, SO₄ rich particles, consistent with thermodynamic calculations. In the experiments, the Ca concentration had reached the values predicted for equilibrium, within 5 minutes when agitated and within a couple of hours when undisturbed. X-ray tomography of a sample of limestone before and after reaction indicates that material is dominantly dissolved from the fragment surfaces (Figure 18). This indicates that limestone dissolution is rapid, it depends on transport to and from surfaces and that dissolution from flow in fractures would dominate.

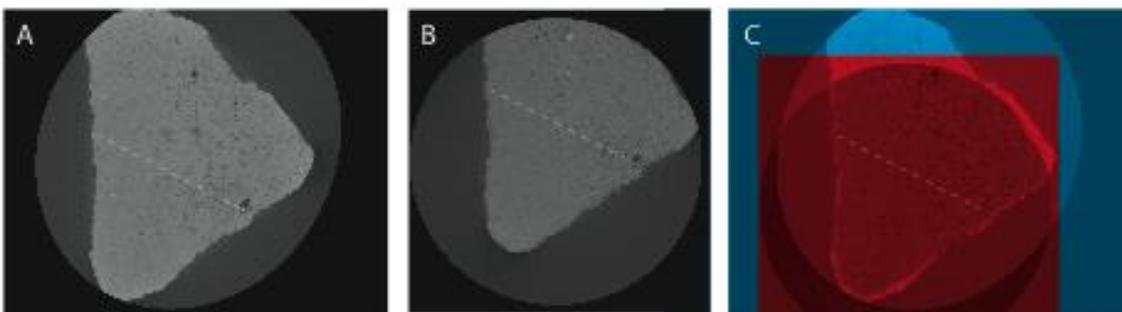


Figure 18: Cross sections from X-ray tomograms of a ~1 mm by 7 mm Hontomin limestone rod. A) Unreacted sample. B) sample after 7 days reaction. C) Overlay of the two images with the difference showing as bright red. The dotted line connects two well defined features in all images.

1.3.4.3 Release of toxic elements during interaction of rock with dissolved CO₂

Mineral dissolution rates are often pH dependent and the injection of low pH, CO₂ rich waters into the subsurface should result in extensive rock dissolution. Depending on rock composition, trace elements can be released and some of these are toxic.

To study the mobility of metals at the early stage of CO₂ injection into basalt, a large scale laboratory high pressure column flow reactor (HPCFR) was designed, built and tested (Figure 19). The HPCFR allows sampling of a pressurized, gas charged fluid along the flow path within a 2.3 m long titanium column filled with solid. In the first test, solutions with 1.2 M CO₂ (aq) were reacted with basaltic glass grains. The composition of solution samples, taken at discrete spatial intervals, under pressure, and in-situ measurement of dissolved inorganic carbon and pH, were successfully modelled with the PHREEQC geochemical speciation model.

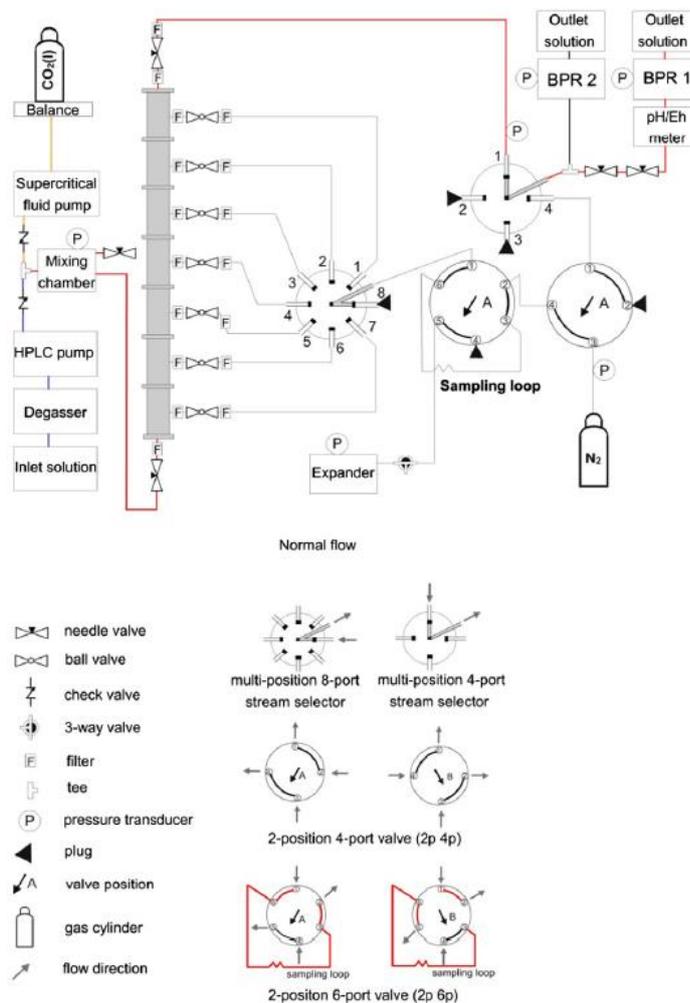


Figure 19: Schematic representation of the flow-through column. Set-up for normal flow. The CO₂ is mixed with the H₂O and pimped through the column. The reacted solution passes the multi-position 4-port stream selector through port 1, which is connected to the outlet port and flows through high temperature/high pressure electrodes followed by a back-pressure regulator (BPR 1). The multi-position 8-port stream selector is aligned with port 8. Both, the 2-position 6-port and 2-position 4-port valves are set on position A. All the compartment outlet ball valves are closed.

In a more detailed study, short lived pulses (50-100 hours) of CO₂ charged water were injected into the HPCFR, that had been filled with basaltic glass grains. Temperature was 22°C; total pressure was 8 MPa. The residence time of the water within the column ranged from 8 to 10 hours. The pH of the inlet CO₂ charged water was ~3.2 and the minimum pH measured in the column was 4.5, after less than 10 hours of water/rock interaction. The dissolved metal concentrations and their relative mobility increased dramatically during the CO₂ pulses. Increase was more than 100 times for Sr, Fe, Al, Ca, Ba, Mn, and Mg. Of these elements, all but Al can bind with CO₂ to form carbonate minerals. Only the dissolved Al, Fe, Mn and Cr concentrations exceeded allowable drinking water limits. After the CO₂ pulses, all of the elemental concentrations decreased close to, or even less than, what was measured during the conditioning of the column.

Using flow through column experiments, we determined the long term (1 year) release of toxic and trace elements from basaltic glass during interaction with pH ~4.5, CO₂ charged water. The dissolution rates of the glass were up to 4 orders of magnitude lower than rates derived from batch reaction experiments, such as are reported in the literature, reflecting lack of mixing and higher Si concentration in the column experiments because more available surface area. This suggests that the dissolution in the column experiments is transport limited and depends on the saturation state of the solution with respect to basaltic glass weathering products. The release of drinking water contaminating elements was generally low but concentration levels for Al, Cd, Mn, and especially As, exceeded the WHO recommended values for drinking water (WHO, 2008). At the high pH in the earlier stages of the experiment, solution composition was consistent with the formation of a range of secondary phases known for uptake of trace elements. As the pH decreased later in the experiment, some of these secondary phases dissolved giving rise to the observed, delayed release of toxic elements.

In experiments to probe the release of toxic elements from Hontomin limestone at the conditions expected at the storage site (CO₂ at 60 °C and 150 bar), concentrations of Mn, Zn and Ni correlated with Ca, indicating that these elements were released from dissolving calcite. Fe and Al were released also but decrease in concentration with time suggests their reprecipitation as poorly soluble Fe- and Al-(oxyhydr)oxides or silicate minerals. After reaction with the limestone, solutions were higher in Fe, Ni and Mn by a factor of up to ~2500, ~500 and ~125, than the EU drinking water directive. However, degassing and oxidation would increase pH and favour precipitation of CaCO₃ and Fe-Mn oxides, phases that are known to take up trace metals such as Ni.

The eruption of the Eyjafjallajökull volcano provided a unique opportunity to study the mobility of heavy metals during elevated aqueous CO₂ concentrations in basalt and the effect of subsequent carbonate precipitation. Near the volcano, the river water in Hvanná was rich in cations and dissolved CO₂. CO₂ degassing increased the pH of the water from 6.6 to 8.5 and calcite precipitated for hundreds of meters downstream. A clear drop in dissolved metal concentration along the flow path correlated directly with the precipitated calcite. Estimated partition coefficients (p_{me}) for Ba, Cd, Co, Cu, Mg, Mn, Na, Ni, Sr and Zn agreed surprisingly well with reported values from laboratory experiments. Other elements were also scavenged from the river water, including Al, Fe, K, P, S, Si, Ti,

V and the rare earth elements (REE). This demonstrates that toxic metal release from CO₂ storage in basalt should be considered but it is very likely that secondary carbonates would sequester significant amounts of the metals.

1.3.5 Work Package 5: Improvement of thermodynamic and kinetic databases for CO₂-rich fluids

Work package 5 (WP5) consisted of the creation and distribution of improved computer accessible databases describing the thermodynamics and kinetics of fluid-rock interaction during carbon-capture and storage efforts.

The following deliverables explained in detail the work carried out within WP5 over the course of the project.

- D5.1) Rate and thermodynamic data gathered and synthesised: [month 18].
- D5.2) Creation of improved thermodynamics and kinetic rate database: Incorporation of these databases into a computer assessable format that can be used by existing geochemical modelling codes [month 24].
- D5.3) Release of thermodynamic database: [month 30].

The main results obtained within this WP include:

- Creation of a thermodynamic database that describes CO₂-water-basalt interaction associated with CO₂ mineral sequestration.
- Creation of database of the dissolution rates of 110 common rock forming minerals as a function of temperature, and fluid composition.

These are explained in more detail in the following subsections.

1.3.5.1 Thermodynamic databases

A thermodynamic dataset describing 36 mineral reactions of interest for CO₂-water-basalt interaction associated with CO₂ mineral sequestration in was assessed and generated. A list of the minerals contained in this database is shown in Table 2. Mineral selection for the dataset was based on extensive review of natural analogs of water-basalt interaction at low and elevated CO₂ conditions. Widely used thermodynamic databases did not contain the mineral assemblage needed for successfully simulating the alteration processes observed in nature as important primary and secondary minerals were found to be missing. The EQ3/6 V7.2b database¹ was the primary source for aqueous equilibrium constants in the developed dataset but reactions for four missing Al-hydroxy complexes were added. Recently published thermodynamic data were compiled for most of the minerals considered in this study. Mineral solubility constants obtained directly from measurements were compiled to the dataset without modification but SUPCRT was used for computing solubility constants when such data was not available. To verify that the presented dataset can capture alterations observed in nature, simulations of CO₂-water-basalt interaction were carried out at low

¹ Wolery, T., 1992. EQ3/6: Software package for geochemical modeling of aqueous systems: package overview and installation guide (version 7.0). Report UCRL-MA- 210662. Lawrence Livermore National Laboratory, Livermore, Calif.

and elevated CO₂ conditions and compared to observed basalt alteration in Iceland and Greenland. Overall simulated and observed alteration are in good agreement, both at low and elevated CO₂ conditions, suggesting the dataset to be well suited for simulations of e.g. CO₂–water–basalt interaction associated with CO₂ mineral sequestration in basalts. A detailed description of this thermodynamic database has been made available to the public in an article published in Chemical Geology (Aradóttir et al. (2012) Chem Geol 304-305, 26-38).

Table 2: Chemical composition of minerals contained in the thermodynamic database developed for CarbFix.

Group	Mineral	Chemical composition	
Aqueous species ^a	Al(OH) ₃	Al(OH) ₃	
	Al(OH) ₂ ⁺	Al(OH) ₂ ⁺	
	Al(OH) ₂ ²⁺	Al(OH) ₂ ²⁺	
	Al ³⁺	Al ³⁺	
Primary minerals and glasses	Basaltic glass	SiAl _{0.36} Fe(II) _{0.19} Mg _{0.28} Ca _{0.26} Na _{0.08} O _{3.31}	
	Plagioclase	Ca _{0.70} Na _{0.30} Al _{1.70} SiO ₈	
	Pyroxene	Ca _{0.35} Mg _{0.42} Fe _{0.23} SiO ₃	
Silicates	Olivine	(Mg _{0.80} Fe _{0.20}) ₂ SiO ₄	
	Allophane	Al ₂ O ₃ (SiO ₂) _{1.22} (H ₂ O) ₅	
	Amorphous silica	SiO ₂	
	Antigorite	Mg ₃ Si ₂ O ₅ (OH) ₄	
	Ca-montmorillonite	Ca _{0.167} Al _{1.67} Mg _{0.33} Si ₄ O ₁₀ (OH) ₂	
	Celadonite	KMgAlSi ₄ O ₁₀ (OH) ₂	
	Fe-celadonite	KFeAlSi ₄ O ₁₀ (OH) ₂	
	Fe-chlorite	Fe ₅ Al ₂ Si ₃ O ₁₀ (OH) ₈	
	Imogolite	Al ₂ SiO ₃ (OH) ₄	
	Kaolinite	Al ₂ Si ₂ O ₅ (OH) ₄	
	K-montmorillonite	K _{0.33} Al _{1.67} Mg _{0.33} Si ₄ O ₁₀ (OH) ₂	
	Mg-Chlorite	Mg ₅ Al ₂ Si ₃ O ₁₀ (OH) ₈	
	Mg-montmorillonite	Al _{1.67} Mg _{0.5} Si ₄ O ₁₀ O ₁₀ (OH) ₂	
	Moganite	SiO ₂	
	Na-montmorillonite	Na _{0.33} Al _{1.67} Mg _{0.33} Si ₄ O ₁₀ (OH) ₂	
	Quartz	SiO ₂	
	Hydroxides	Amorphous Al hydroxide	Al(OH) ₃
		Fe(II) hydroxide	Fe(OH) ₂
		Fe(III) hydroxide	Fe(OH) ₃
Carbonates	Calcite	CaCO ₃	
	Dolomite	CaMg(CO ₃) ₂	
	Magnesite	MgCO ₃	
	Siderite	FeCO ₃	
Zeolites	Analcime	Na _{0.96} Al _{0.96} Si _{2.04} O ₆ ·H ₂ O	
	Ca-chabazite	CaAl ₂ Si ₄ O ₁₂ ·6H ₂ O	
	Ca-heulandite	CaAl ₂ Si ₇ O ₁₈ ·6H ₂ O	
	Laumontite	CaAl ₂ Si ₄ O ₁₂ ·4.5H ₂ O	
	Mesolite	Ca _{0.666} Na _{0.666} Al ₂ Si ₃ O ₁₀ ·2.667H ₂ O	
	Na-chabazite	Na ₂ Al ₂ Si ₄ O ₁₂ ·6H ₂ O	
	Na-heulandite	Na ₂ Al ₂ Si ₇ O ₁₈ ·5H ₂ O	
	Natrolite	Na ₂ Al ₂ Si ₃ O ₁₀ ·2H ₂ O	
	Stellerite	Ca ₂ Al ₄ Si ₁₄ O ₃₆ ·14H ₂ O	
	Stilbite	Ca ₂ NaAl ₅ Si ₁₃ O ₃₆ ·16H ₂ O	

^a Four Al-hydroxy complexes were added to the EQ3/6 database, which is the primary source of equilibrium constants for aqueous species in this study.

1.3.5.2 Database describing the dissolution and precipitation rates of natural minerals.

The overall goal of this task was to create a general database of the current state of knowledge of the rates of mineral dissolution and precipitation reactions. Through our efforts over the past three years the database currently contains more than 110 rock forming minerals, and has been put into general form such that it can be readily applied by the scientific community to assess the potential of fluid-rock interactions during carbon capture and storage efforts.

This database was created by first collecting and correlating the existing kinetic rate data available in the scientific literature. In cases where adequate fits of these data were already available in the literature, such fits were adopted in this study. In other cases we have sought to provide a quantitative description of these data consistent with what is known about the dissolution and/or precipitation mechanism of the minerals. The resulting and adopted equations have been incorporated into a PHREEQC enabled computer script to all it to be applied directly in conjunction with this geochemical modelling code. Note that PHREEQC is widely available geochemical computer modelling code, originally created at the US Geological Survey and available free to any potential user via the web,

The kinetic database was established from a thorough literature review of the existing data on precipitation and dissolution rates. Most rates were fit to the following equation:

$$r = A_A(-E_A/RT) \sum a_i \left(1 - \exp\left(-\frac{A}{RT}\right)\right)$$

where r signifies the steady state dissolution rate, A_A refers to a pre-exponential factor², E_A designates an activation energy, R represents the gas constant, and T denotes absolute temperature, a stands for the activity of the subscripted aqueous species, and A refers to the chemical affinity of the dissolving phase. It is this latter term that assured that calculated dissolution rates approach zero as equilibrium is approached. Note that within this formalism precipitation rates are, consistent with transition state theory assumed to be the inverse of dissolution and can be calculated directly from the dissolution rate constants. Experimental dissolution rates obtained from the literature datasets were assessed and the values for the reaction rate constant (A), the activation energy (Ea), the reaction order with respect to the exponent on the activity of aqueous species that are involved in the dissolution or precipitation rate mechanisms (e.g. H^+ , Mg^{2+} , SiO_2 , Na^+ , Al^{3+} and/or K^+). This file is organized by mineral, glass phases; a list of solids currently included the database is listed in Table 3.

Table 3: List of minerals and glasses currently incorporated into the CarbFix kinetic database.

Silicates	Carbonates
Albite	Aragonite
Almandine	Calcite
Andesine / Labradorite	Dawsonite
Andradite	Dolomite
Anglesite	Magnesite
Anhydrite	Rhodochrosite
Anorthite	Siderite
Anthophyllite	Witherite
Augite	
Biotite	Phosphates
Bronzite	Apatite (fluorapatite)
Bytownite	Apatite (hydroxylapatite)
Celestite	Struvite
Chlorite	Variscite
Christobalite	
Chrysotile	Oxides/hydroxides
Cordierite	Bayerite
Diopside	Boehmite
Enstatite	Brucite
Epidote	Corundum
Fayalite and other non-forsteric olivines	Diaspore
Forsterite	Gibbsite
Glaucosite	Goethite
Glaucophane	Hematite
Grossular	Ilmenite
Hornblende	Magnetite
Illite	Uraninite
Muscovite	
Quartz	Glass
Jadeite	Basaltic
Kaolinite	Rhyolitic
Oligoclase	
Prehnite	Other
Pyrophyllite	As ₂ S ₃ – Orpiment
Serpentine minerals	Barite
Smectite - Montmorillonite	Fluorite and other divalent metal fluorides.
Spodumene	Gypsum
Staurolite	Halite
Talc	Sphalerite
Tourmaline	
Tremolite	
Wollastonite	
Zoisite	

1.3.6 Work package 6: Geochemical and hydrological modelling

WP6 deals with the model prediction and calibration of the injection of CO₂ at Hellisheidi. The final objective is the development of robust simulations able to reproduce the observations made during the life of the CarbFix project and to predict the behaviour and fate of the injected CO₂ after the end of the operational stage. These calculations allow the quantification of the amount of CO₂ permanently and safely stored as mineral carbonates.

The following deliverables explain in detail the work carried out within WP6 over the course of the project:

- D6.1) Re-evaluation of conceptual model with respect to Hellisheidi field observations: [month 12]
- D6.2) Re-calibration of hydrological parameters using extensive field data from Hellisheidi: Parameters re-calibrated. [month 18]
- D6.3) Updated version of the CarbFix reactive transport model: [month 24]
- D6.4) Effect of re-injection of H₂S on subsurface transport and mineralogy: [month 30]
- D6.5) Feasibility study of full scale CCS at Hellisheidi Power Plant: [month 36]

The main results obtained within this WP include:

1. Simulations predict calcite to be the most abundant among carbonates to form at Hellisheidi at intermediate depths but magnesite dominates in deep system. This is in agreement with natural basalt alteration.
2. Simulations of 1200 ton pilot injection at Hellisheidi predict 85% mineral CO₂ capture within 5 years and 100% within 10 years. These results are in excellent agreement with monitoring results which indicate about 80% mineralization 1 year from injection.
3. Simulations of full-scale (400,000 tons) injection for 10 years predict 80% mineral capture after 100 years, indicating industrial scale CO₂ injection into basalts to be a viable option in reducing anthropogenic CO₂ emissions.
4. Sequestration of CO₂ and H₂S in the deep reservoir predicted to be rapid in all simulated scenarios. In pilot injection scenarios, 85% H₂S sequestered within 1 year and 100% within 5 years, and all CO₂ mineralized within 1 year. In up-scaled injection of 75% CO₂ and 25% H₂S, 97% H₂S and 100% CO₂ mineralized after 5 years, and all H₂S mineralized after 20 years. Similar results for up scaled injection of 25% CO₂ and 75% H₂S.
5. More rapid sequestration at greater depths and higher temperatures.
6. Numerical modelling indicates in situ CO₂ mineral sequestration in basalts to be a viable option in reducing anthropogenic CO₂ emissions as they predict fast mineralization of injected resulting in permanent mineral storage within a few years compared to 100-10,000 years in supercritical CO₂ storage.
7. Concerning modelling of injection in sedimentary aquifers, 3D full-scale injection model has been completed considering non-miscibility between CO₂ and native brine.
8. Adaptive meshing is proved to be very efficient when calculating very big domains with thousands of elements.
9. 2D miscible simulations predict faster CO₂ dissolution than commonly expected. Fingering effect sinks CO₂ to the bottom of the aquifer.
10. Significant differences are predicted depending on the location of the injection well.

These are explained in detail in the following subsections:

1.3.6.1 Numerical models developed of Hellisheidi injection sites

Pilot scale injections were carried out at two different injection sites in Hellisheidi in CarbFix (see Figure 20). Well HN-02 penetrates a storage formation that lies between 400-800 m depth and temperatures of 30-80°C. Well HN-08 on the other hand, penetrates the high temperature geothermal system below 800 m depth, consisting of temperatures above 270°C. Groundwater flow within the shallower aquifer has been identified as being predominantly matrix dominated, whereas flow within the deep reservoir is fracture dominated. These findings are based on numerous tracer tests that have been carried out at the injection sites. Two different models have been developed of the pilot injection sites at Hellisheidi.

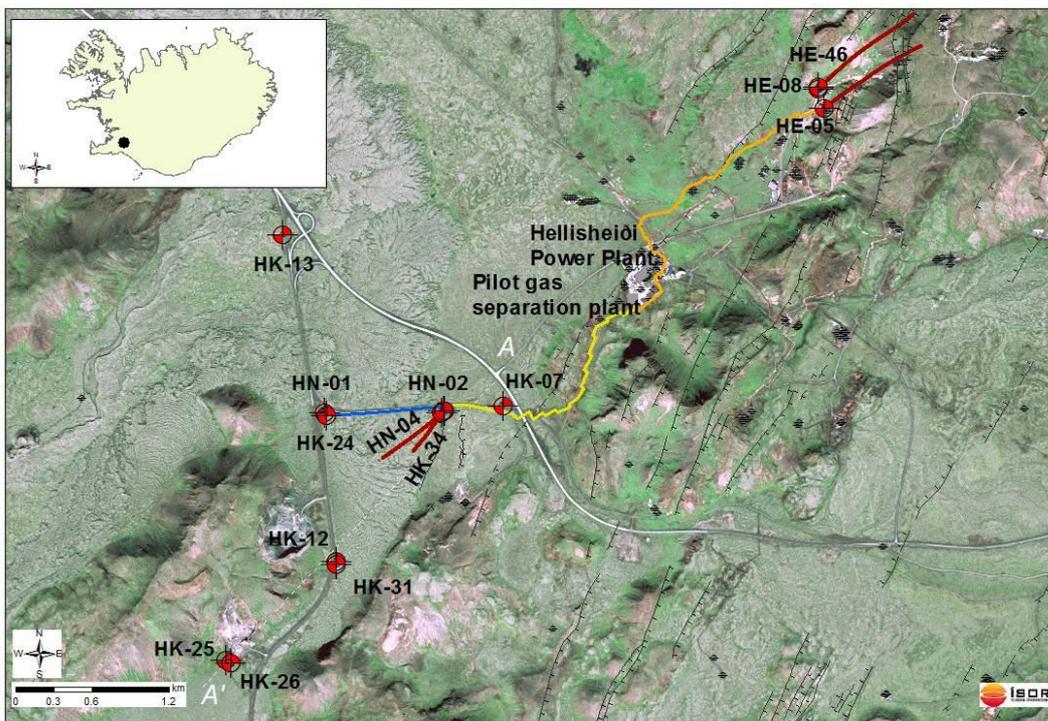


Figure 20: Aerial map of CO₂ injection sites in Hellisheidi, SW-Iceland. A CO₂-H₂S gas mixture containing trace amounts of H₂ will be transported in pipelines (yellow and orange) towards two different injection sites. HN-02 and HE-08 are injection wells in basaltic storage formations located between 400-800 m depth and below 1000 m depth, respectively. Fresh groundwater, used for dissolving gases during injection into HN-02 (storage between 400-800 m depth) will be pumped from well HN-01 and transported towards HN-02 (blue line). Separated geothermal water and/or condensate water will be used for dissolving gases prior to injection into well HE-08 (storage below 1000 m depth). Wells HK-13, HK-07, HK-24, HK-12 and HK-25 are shallow monitoring wells, whereas wells HN-04, HK-34, HK-31, HK-26 and HE-08 are deep monitoring wells that penetrate the two different storage formations. Well deviation is depicted by red lines. Combed lines represent faults.

Shallower reservoir

Tracer tests at the shallower (400-800 m depth) injection site indicated the storage formation to consist of a relatively homogeneous porous media. In view of this, it was decided to develop a single porosity numerical model representing the dominant matrix flow in the storage formation. The model is three dimensional, consisting of eight layers, from 100 to 900 m depth. The model is aligned

in direction of regional groundwater flow, which is from NE-SW. Hydrological properties of the storage formation were calibrated against a large set of field data using iTOUGH2. Resulting principal hydrological properties are lateral and vertical intrinsic permeabilities of $300 \times 10^{-12} \text{ m}^2$ and $1700 \times 10^{-12} \text{ m}^2$, respectively, effective matrix porosity of 8.5% and a 25 m/year estimate for regional groundwater flow. Figure 21 shows an aerial view of the developed model.

An extensive literature review of water-basalt interaction at low and elevated CO_2 conditions was carried out with the objective of predicting which minerals are likely to dissolve and precipitate in *in situ* CO_2 mineral sequestration. Widely used thermodynamic databases did not contain the mineral assemblage needed for successfully simulating the alteration processes observed in nature as important primary and secondary minerals were missing. We thus developed and evaluated a thermodynamic dataset describing 36 mineral reactions of interest for CO_2 -water-basalt interaction. All minerals in the database (Aradóttir et al. 2012, Chem Geol. 304-305, 26-38) were compiled to the CarbFix numerical models along with kinetic variables describing their dissolution and precipitation. Dissolution and precipitation of most minerals is kinetically controlled in simulations, with kinetic rates calculated as a product of the rate constant and reactive surface area. Aradóttir et al (2012) provides details on the parameters used for calculating kinetic rates of mineral dissolution and precipitation (Aradóttir et al., 2012, Int J Greenh Gas Cont 9, 24-40).

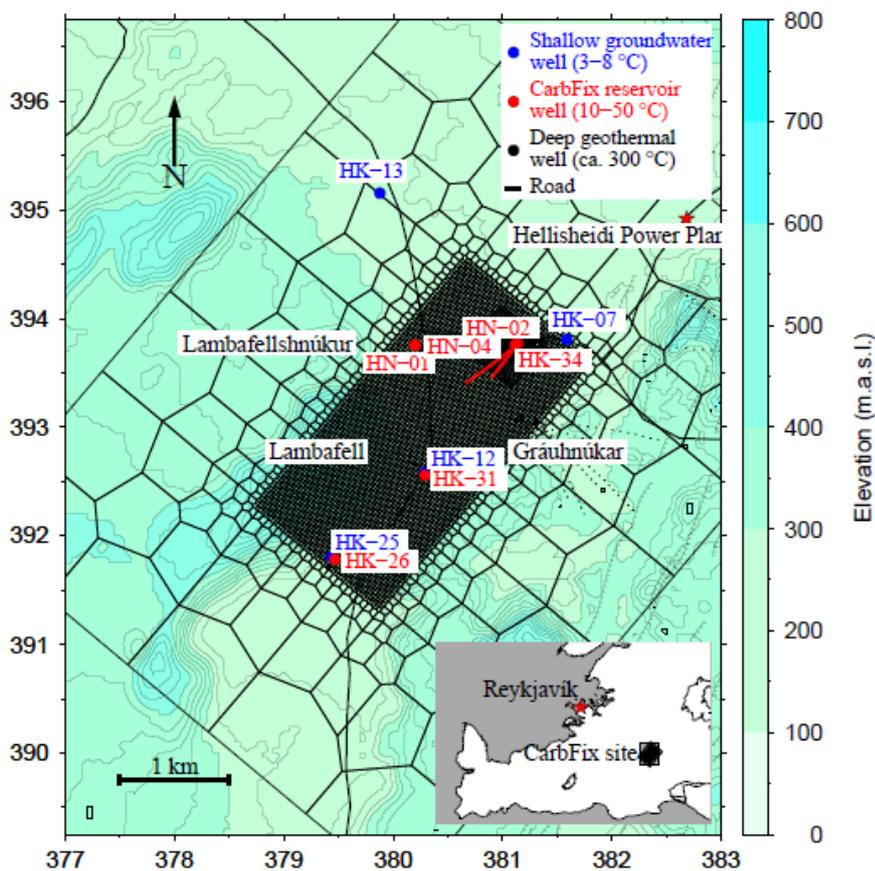


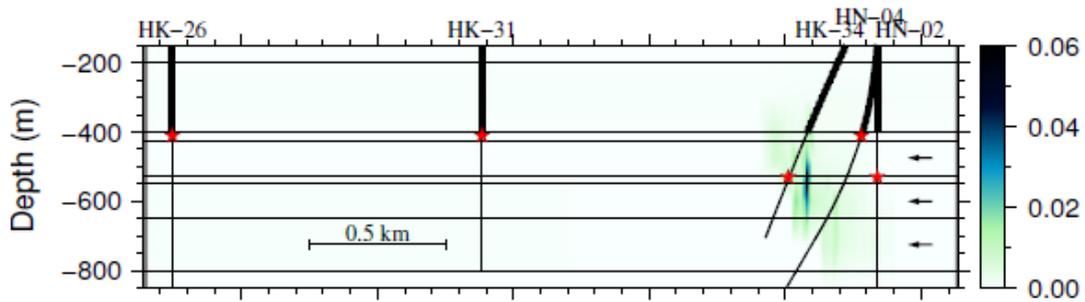
Figure 21: Geographic location of the CarbFix field model and distribution of elements in the model's mesh. The model is aligned with the regional groundwater flow direction (NE-SW) and covers all CarbFix wells.

Reactive chemistry was coupled to calibrated hydrological models and predictive mass transport and reactive transport simulations carried out for both a 1200 tons pilot CO₂ injection and a full-scale 400,000 tons CO₂ injection scenario at Hellisheidi power plant. Pure CO₂ was assumed to be injected in the simulations and the effects of H₂S and H₂ in the gas mixture that is actually being injected were thus neglected.

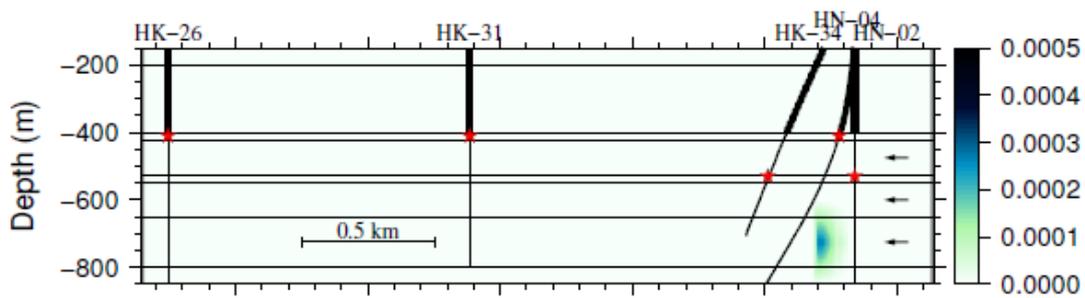
Simple hydroxides, silica phases, zeolites and carbonates are all predicted to form in pilot and upscaled injection scenarios at the shallower injection site. Carbonate formation increases gradually and calcite is most predominant among carbonates, which is in agreement with natural basalt alteration in Iceland. Magnesite-siderite solid solution also precipitates but in significantly lower quantities. Antigorite precipitation is generally associated with calcite precipitation in the simulations. Increased silica precipitation is observed behind the moving CO₂ plume.

Figure 22 shows a cross sectional reservoir view of selected precipitated minerals after ten years simulation time in the pilot injection scenario. SiO₂ (am), quartz and antigorite precipitation is associated with calcite precipitation in the simulations which is in agreement with natural alteration in the vicinity of mineral springs in Iceland. Ca leached from primary minerals is modelled to preferentially form calcite rather than zeolites while clay minerals, simple oxides and oxyhydroxides are predicted to compete with magnesite-siderite solid solution for leached Mg and Fe.

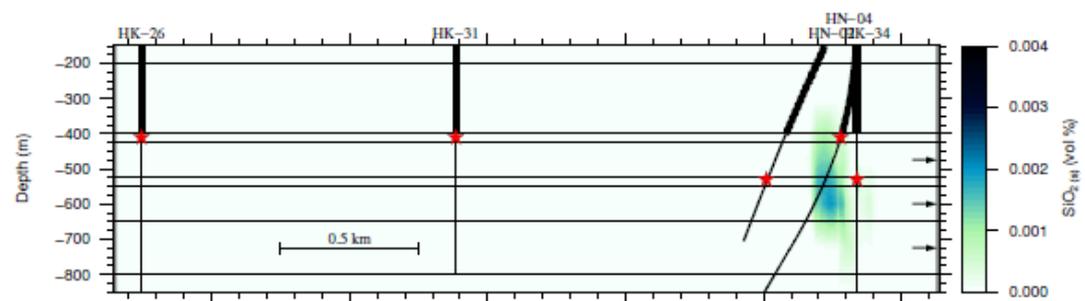
Reactive transport simulations of the 1200 ton pilot injection predict 85% CO₂ mineral capture within 5 years and 100% capture within 10 years. Simulations of the full-scale injection scenario predict 80% CO₂ mineral capture after 100 years. CO₂ sequestration rate is predicted to range between 1200 and 22,000 tons/year in both scenarios, a value in good agreement with the natural sequestration rate of the Hellisheidi geothermal system, which has been estimated to be 410023,500 tons/year. Relatively fresh basaltic formations are thus predicted to be promising geological CO₂ storage formations.



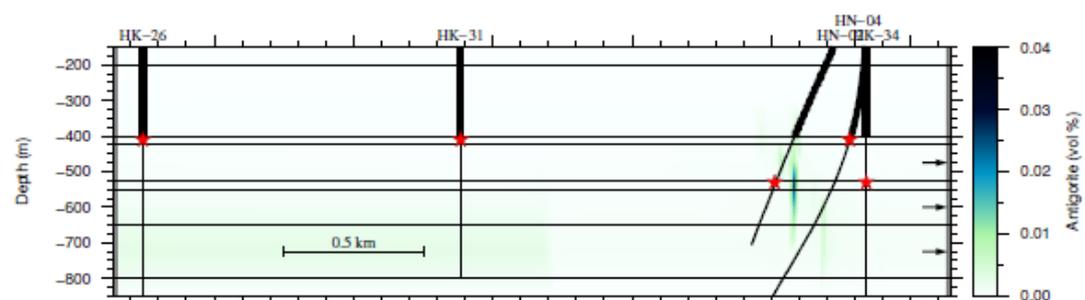
(a) Calcite



(b) Magnesite-siderite



(c) SiO₂ (s)



(d) Antigorite

Figure 22: Cross sectional reservoir view of selected precipitated minerals in the CO₂ pilot injection 10 years from injection, as predicted by three-dimensional reactive transport simulations. The cross section is drawn along A-A' on Error! Reference source not found.. The figure shows different minerals forming at different temperature and pressure conditions.

Deep reservoir

A tracer test carried out in 2010 indicated fracture flow to be dominant in the deep storage formation. The fractures are, however, connected to a porous matrix that contains the majority of the reactive surface area in the storage formation. In view of this, it was decided to use a one-dimensional dual porosity model as a first approach to simulating injection into the deep reservoir. Figure 23 shows a simple schematic of the conceptual model along with an illustrative representation of elements and connections. Injection well HE-08 is located at $x=202.5$ and monitoring well HE-46 is located at $x=652.5$. Both wells are positioned within the fracture continuum of the model. The model's fracture permeability was calculated to be $733 \times 10^{-12} \text{ m}^2$, by assuming laminar flow and fracture diameter of 2 mm. Matrix porosity was assumed to be 2.5% and the assumed matrix permeability of $0.02 \times 10^{-12} \text{ m}^2$ is that of the Hengill reservoir model at a corresponding depth (Gunnarsson et al (2011) *Transp Porous Med* 90:3-22).

The thermodynamic dataset described above was used as a basis thermodynamic database for simulations of injection into the deep reservoir. Several minerals needed to be added to the dataset as the alteration mineralogy associated with the injection is different to that in the shallower reservoir due to additional H_2S -water-basalt interaction and a considerably higher reservoir temperature. Minerals added to the database were goethite, grossular, magnetite, prehnite, pyrite, pyrrhotite, sulfur and wollastonite. Dissolution and precipitation of most minerals is kinetically controlled in simulations, with kinetic rates calculated as a product of the rate constant and reactive surface area. Mineral precipitation was represented using the same kinetic expression as that for dissolution. Kinetic variables describing dissolution and/or precipitation of sulfur, pyrite and pyrrhotite at reducing conditions were not found in the literature. These minerals had thus to be assumed to precipitate and dissolve under equilibrium.

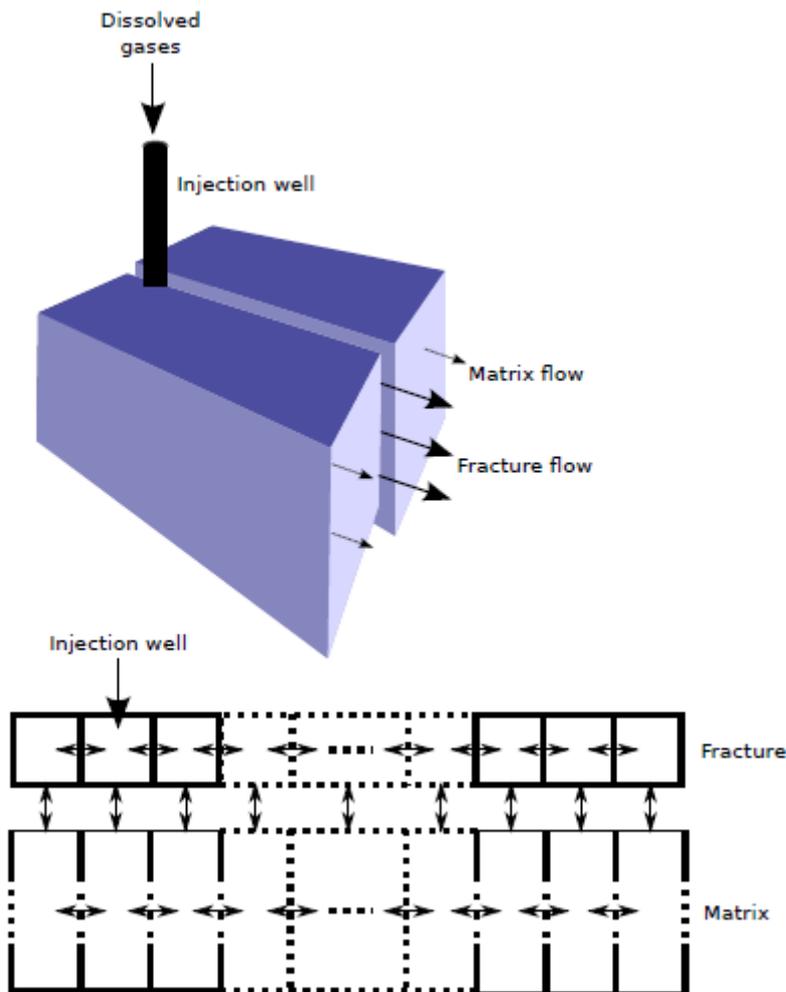


Figure 23: Schematic of the conceptual one-dimensional dual porosity model used for simulations of the deep Hellisheidi reservoir (top) and illustration of elements and connections in the developed model (bottom).

Predictive simulations were carried out for both a 6 month continuous pilot injection and up scaled 5 years continuous injection into well HE-08. In the pilot scale scenario, 20 g/s of a gas mixture containing 75% CO₂ and 25% H₂S were dissolved in 10 kg/s separated geothermal water. In the up scaled simulation scenarios, 400 g/s of the same gas mixture were dissolved in 50 kg of either separated geothermal water or condensate. Identical simulations were carried out for a gas mixture consisting of 25% CO₂ and 75% H₂S. Simulation of water-rock interaction associated with the injection scenarios were carried out for 50 years.

Alteration mineralogy is predicted to be similar in all injection scenarios into the deeper reservoir. Al-hydroxide, antigorite, pyrite and pyrrhotite are predicted to precipitate within the fracture domain of the model, while Al-hydroxide, analcime, antigorite, celadonite, magnesite, magnetite, pyrrhotite, quartz and wollastonite are predicted to form within the matrix domain. Precipitation mainly occurs within the matrix. Magnesite and pyrrhotite mainly form in the vicinity of injection well HE-08 but

other minerals are predicted to form throughout the SulFix reservoir. Figure 24 and Figure 25 show predicted mineral precipitation in pilot and up scaled injection scenarios, respectively.

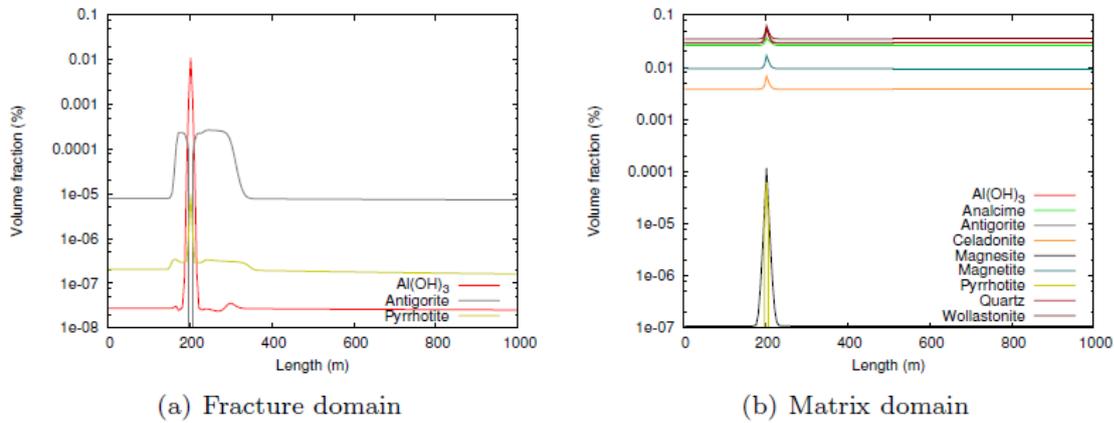


Figure 24: Predicted mineral precipitation in simulation of a 6 month continuous SulFix pilot injection, in which 20 g/s of 75% CO₂ and 25% H₂S gas mixture was dissolved in 10 kg/s separated geothermal water. Simulation was carried out for 10 years. The figure shows precipitation after 1 year simulation time. SulFix injection well HE-08 is located at x=202.5 within the fracture continuum of the model.

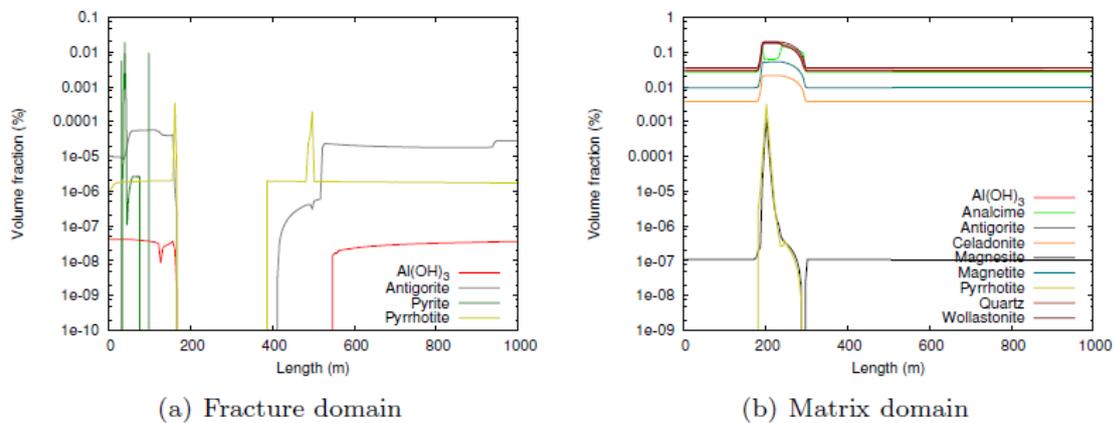


Figure 25: Predicted mineral precipitation in simulation of a 5 years continuous up scaled SulFix injection scenario, in which 400 g/s of 25% CO₂ and 75% H₂S gas mixture was dissolved in 50 kg/s condensate. Simulation was carried out for 50 years. The figure shows precipitation after 5 years simulation time. SulFix injection well HE-08 is located at x=202.5 within the fracture continuum of the model.

Sequestration of injected CO₂ and H₂S is predicted to be rapid in all simulated scenarios, indicating basaltic formations to be promising for sequestering sour gas emissions. In the 6 month pilot injection scenario, where 20 g/s of 75% CO₂ and 25% H₂S gas mixture was dissolved in 10 kg/s separated geothermal water, 85% of injected H₂S is predicted to be sequestered within 1 years' time and 100% within 5 years' time. All injected CO₂ is predicted to be sequestered within 1 year time. In simulations of up scaled injection where 400 g of 75% CO₂ and 25% H₂S gas mixture was injected for 5 years, 97% of injected H₂S and 100% of injected CO₂ is predicted to be sequestered after 5 years simulation time and all H₂S within 20 years. Similar results were obtained in simulations of up scaled injection where 400 g of 25% CO₂ and 75% H₂S gas mixture was injected for 5 years. Simulation

results were not affected by the fact whether separated geothermal water or condensate was used for dissolving the injected gas mixture.

Injected H₂S is mainly predicted to be sequestered into pyrrhotite while only a negligible amount goes into pyrite. Magnesite acts as the only CO₂ sink in the deeper reservoir. Magnetite is predicted to compete with pyrrhotite and pyrite for Fe leached from the primary rock matrix, while antigorite and celadonite compete with magnesite for Mg leached from the matrix. Ca is predicted to predominantly precipitate into wollastonite.

1.3.6.2 Numerical models developed on future injection in Los Páramos site

A number of areas have been selected either for scientific investigation or commercial purposes in the Duero basin, in North Spain (Figure 26). The CarbFix consortium has selected for model prediction of CO₂ injection in deep saline aquifer a large structure capable of storing ~200Mt of CO₂, called Los Páramos, located in the San Pedro tectonic band. The San Pedro area has been studied in the past for hydrocarbon exploration and a large body of information is available from seismic profiles (over 170 km) and 3 deep wells (Figure 27).

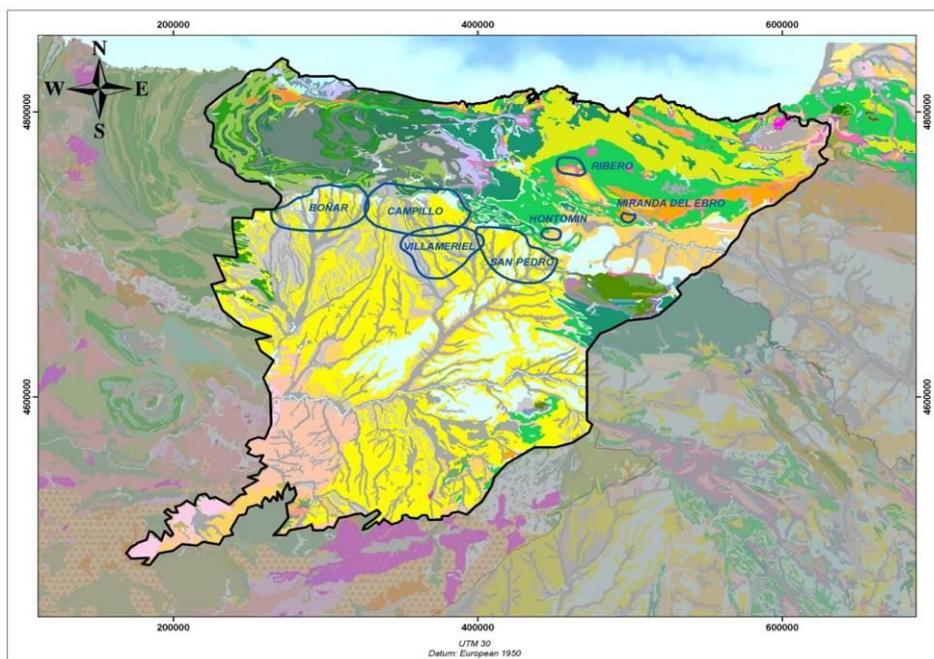


Figure 26: Location of the favourable structures for CO₂ storage in deep saline aquifers in the Duero basin (from IGME, 2010). The San Pedro structure hosts the Los Páramos site which has been selected in the CarbFix project for comparison with injection in basaltic aquifers in Hellisheidi.

The Los Páramos site is emplaced in the San Pedro Folded Band (SPFB) that consists mainly of thick-skinned thrusts of Mesozoic rocks (Triassic and Upper Cretaceous) sealed by a thick (1200-1500 m), undeformed cover of Tertiary claystones. Dome-like structures are related to thrusts leading to favourable reservoirs (Figure 27). The potential target horizon for CO₂ storage is the Utrillas Fm sandstone with high porosity (13-20%) and thickness (225-250 m). This aquifer is saline, up to 50 g·L⁻¹

¹, according to the data from drill wells. The presence of saline groundwater is explained by water interaction with Triassic evaporite layers just underlying the Utrillas Fm sandstones.

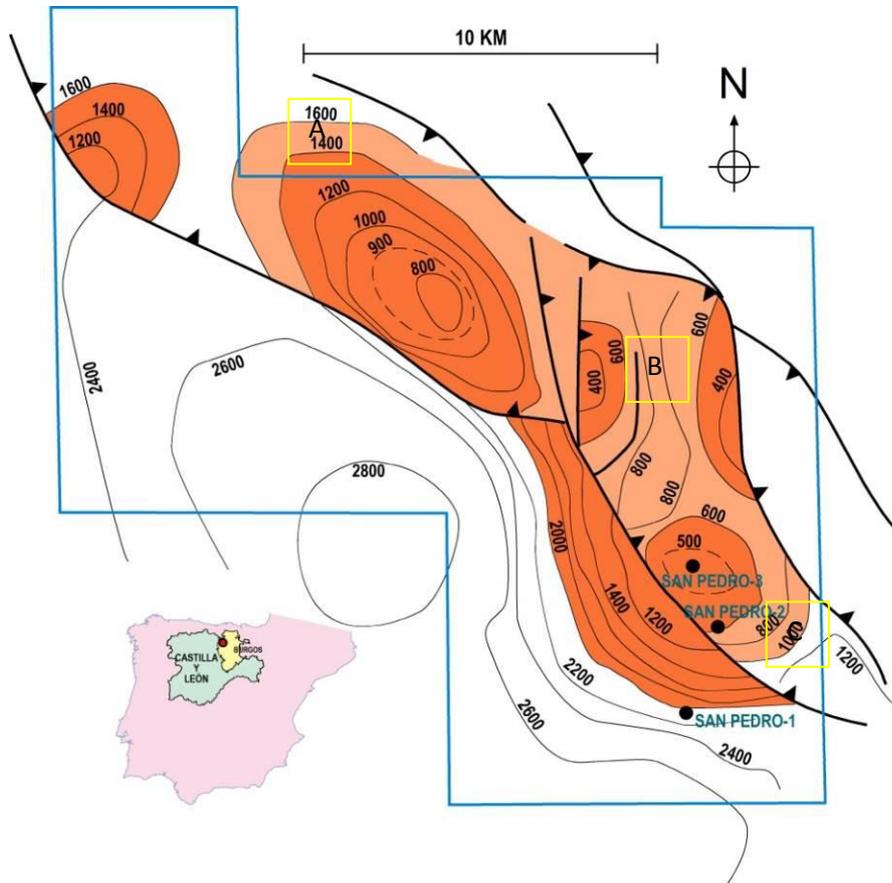


Figure 27: Geological map of the Los Páramos permit in the Duero basin (NE Spain). Red-coloured surfaces show the top of the Utrillas Fm, which is the major target for injection in the site. The three reservoirs for favourable CO₂ storage are indicated as A, B and C. The location of the existing wells is also shown (San Pedro wells).

An essential parameter for safety and pre-injection assessment of a geological storage is the estimation of the dominant mechanism in the fixation of the CO₂. The relevance of these mechanisms will vary during time due to CO₂ migration, dissolution and reaction. The aim of this model is to quantify the CO₂ trapped in free phase, by capillarity and dissolved, assessing the influence of the injection point location on the evolution of trapping mechanisms.

A dome shape 2D miscible model (Figure 28) in which CO₂ is allowed to dissolve into the native brine has been built. The model was developed applying finite element approximation and fully implicit method with Comsol Multiphysics (COMSOL, 2013. Comsol Multiphysics. *Version 4.3a.*). Comsol can handle all kind of PDE-ADE physics systems in a coupled manner. Although it provides some already built-in interfaces it is also possible to introduce your own set of differential equations as it was done in this project. Despite the heat equation was not specifically solved, the geothermal gradient was considered and the thermodynamics properties for both gas and liquid phase were adapted (Pruess (2005) Lawrence Berkeley National Laboratory Report LBNL-57592, Berkeley, CA).

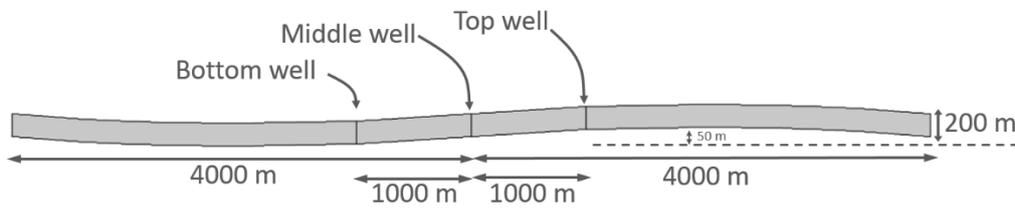


Figure 28: Model sketch used for fluid injection with miscibility between CO₂ and native brine.

The models simulate the fate of supercritical CO₂ during 100 years in a synthetic 2D dome at a depth of 2500 m. The CO₂ was injected with a rate of 0.3 kg s⁻¹ during half year, which leads to a total amount of CO₂ of 4.4 of tons. Clearly the total amount of CO₂ injected is far below those expected in a real commercial storage but this simulation is primarily intended to assess the competence between trapping mechanisms. The rate of injection, however, is equivalent to an industrial project, considering that the model is a 2D section.

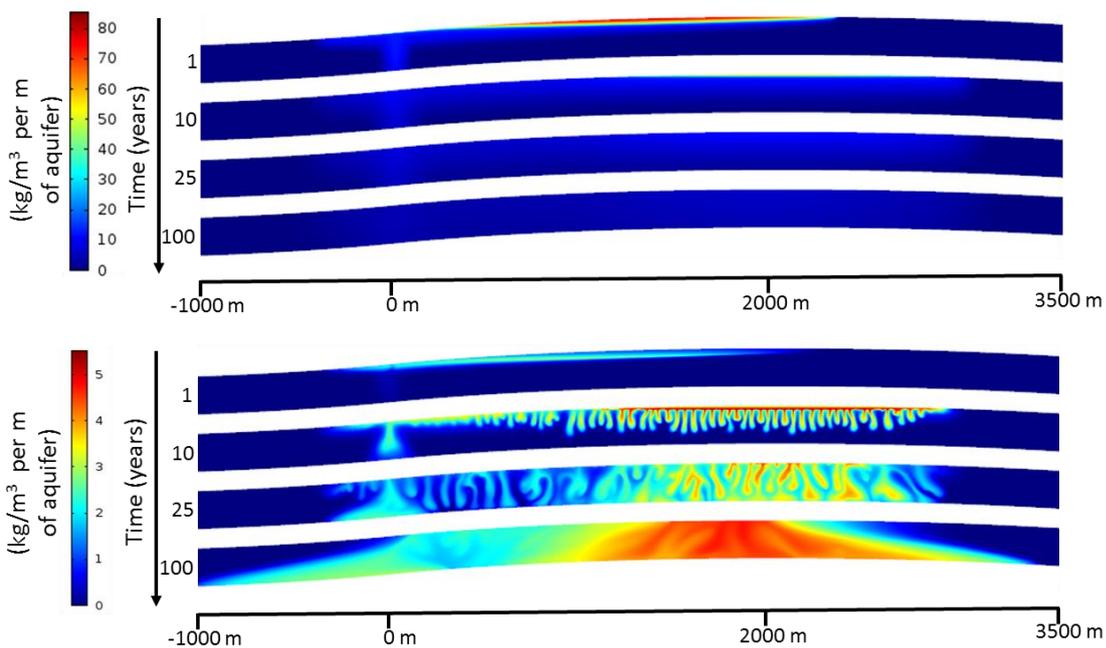


Figure 29: Evolution of the amount of CO₂ trapped in gas phase (top) and dissolved CO₂ (bottom) for the middle well injection case at 1yr, 10yr, 25 yr and 100 yr. Injection takes place at coordinate 0.

The model of the CO₂ injection into a section of the Dome B of Los Páramos site reproduce the fate of the supercritical plume and the formation and evolution of denser CO₂-rich brine fingers. As can be seen in Figure 29 the CO₂ bubble (supercritical) migrates quickly upward and after two years the bubble stops, occupying the top of the dome.

In the interphase between the supercritical CO₂ and the brine, some of the CO₂ is dissolved in to the brine, increasing the its density. This leads towards a situation where the denser CO₂-rich brine is on top of lighter fluid, causing gravitational instability. “Fingers” of denser fluid migrate downwards generating a convective mixing between undersaturated brine and CO₂-rich brine which enhances the dissolution of the CO₂. This phenomenon has been demonstrated experimentally, analytically and with numerical simulations. However, and despite its relevance in the evolution of the CO₂, numerical models showing convective mixing has been typically conducted on small-scale, being rare the models able to reproduce this process in a large scale reservoir model. After 30 years, much of the CO₂ is already dissolved and free gas can only be found trapped by capillarity.

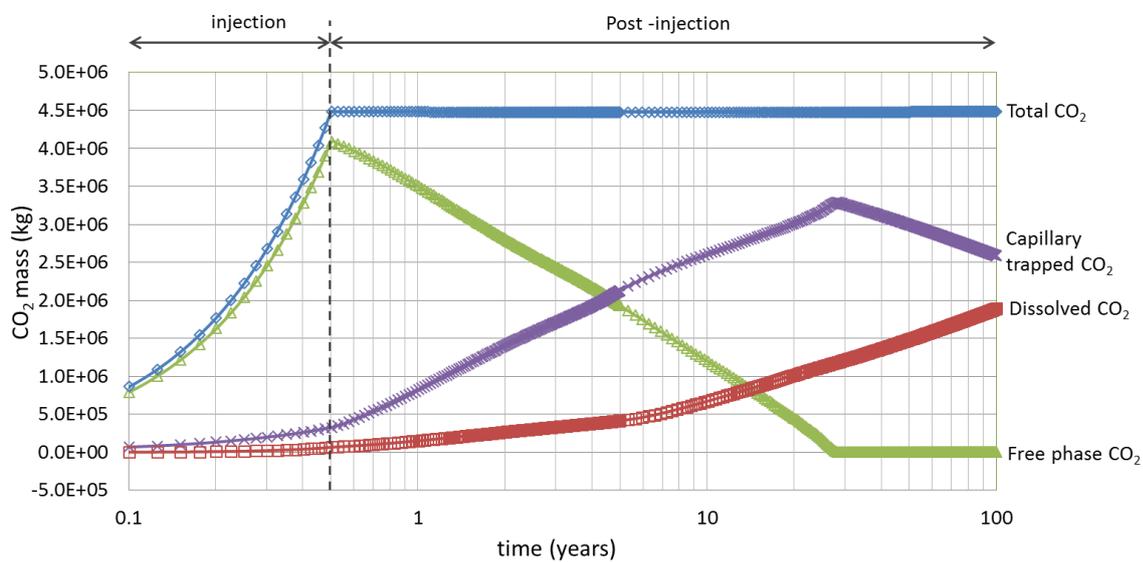


Figure 30: Predicted evolution of CO₂ trapping mechanisms (as mass of CO₂ trapped) through time in the whole 2D section for the middle well injection case. Note that the mass of CO₂ is preserved along the simulation after injection stops.

Figure 30 shows the contribution of the trapping mechanisms computed in the whole 2D section (excluding mineralisation, since no reactive mineralogy was included in the model). As expected, structural trapping dominates at early times, with a progressive dissolution onwards. The amount of CO₂ trapped by dissolution and capillarity increase until the free CO₂ is depleted (around 30 years after injection). Then, due to the dissolution of the CO₂ trapped by capillarity, the amount of dissolved CO₂ continues growing, becoming responsible for around 40% of the total CO₂ stored. These model results had been tested for different meshes to ensure its numerical stability and preserve through the simulation the total amount of CO₂ injected.

These results have significant implications in terms of safety assessment since the majority of the CO₂ is dissolved in a relative short period of time (less than 30 years), significantly decreasing the risk of leakage in case of fracture of the caprock. In addition, interesting conclusions from the monitoring point of view can be drawn, since the extent of potential leakage at surface can be envisaged.

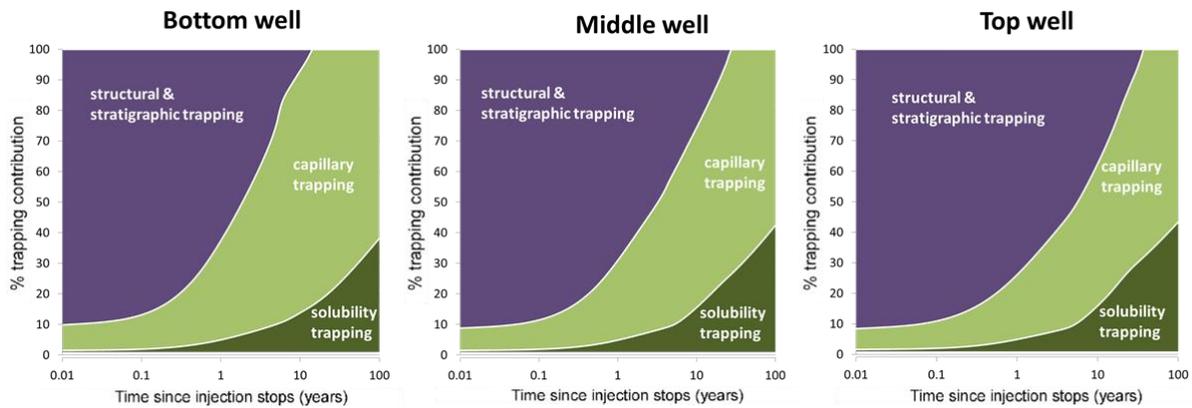


Figure 31: Evolution of the competition between trapping mechanisms for three different injection locations.

As seen in Figure 34, even small changes in the injection well location lead to different competition between trapping mechanisms. Injecting the CO₂ far from the top of the storage formation, the migration of the supercritical plume is higher and the “fast” capillary trapping is enhanced. This leads to an increase of the storage safety. Solubility trapping, however, is mostly function of time, being almost independent of the injection location.

1.4 The potential impact and the main dissemination activities and exploitation of results (not exceeding 10 pages)

1.4.1 Potential impact

While CCS is acknowledged by experts as having the potential to substantially contribute to the mitigation of greenhouse gas impact, it is, as of yet, scarcely demonstrated and has yet to be accepted by the wider public and by the prospective regulative authorities. The EU commitment to reduce greenhouse gas emissions by at least 80% by 2050 requires the refinement and commercialization of proven carbon capture and storage approaches. Storage methods must be safe and environmentally sound. Over the course of the project, CarbFix has significantly contributed to this task by developing and demonstrating a new CCS technology involving the whole CCS chain at Hellisheidi power plant in SW Iceland.

Most notably, it has been demonstrated that the CarbFix carbon storage method leads to 1) the immediate solubility storage of CO₂ and 2) the transformation of the injected CO₂ into stable carbonate phases on a time scale of months to years. This method is therefore significantly safer than traditional geological storage methods, where a single gas phase is injected into the subsurface.

First, in traditional geologic storage, the injected gas phase is distinct and is likely to remain buoyant for hundreds of years or more and thus escape back to the surface is a risk. This contrasts with the CarbFix method, where the gas is dissolved during injection. Gas dissolved in water is denser than the formation water, so the gas is carried to the bottom of the porous unit, avoiding the possibility of escape. Second, basaltic rocks rapidly neutralize the acidity of the injected fluid, leading to precipitation, limiting the extent of toxic metal transport associated with acidic pore fluids. Third, the CarbFix method has been demonstrated to rapidly transform the injected carbon into carbonate minerals that are stable for millions of years or more. Once the carbon is transformed into carbonate minerals, further site monitoring is unnecessary. By providing this safer alternative to traditional geological storage, which minimises the risk of leaking for generations, monitoring requirements disappear, and we anticipate that public acceptance for carbon storage can be improved throughout Europe.

Thus, a major impact of CarbFix is decreased costs by cutting the long-term monitoring costs of the geological carbon storage site. The estimated cost of transport and storage of CO₂ at the CarbFix site, performing at maximum reservoir exploitation, is 17 €/ton (Ragnheidardottir et al. (2011) Int J Greenhouse Gas Control, 5, 1065). This is twice the common estimates for traditional subsurface transport and storage but even this is a factor of about 5 times lower than the current total estimated cost for CO₂ capture and separation. The lack of long term site monitoring makes the CarbFix approach a promising one as a stepping-stone for wider deployment.

A further impact of CarbFix is that it has demonstrated the success of storing CO₂ in basaltic rocks. Carbon storage in basalt has generally been ignored because these rocks are not as common as sedimentary rocks on the continents. Over the past years, however, public opinion in Europe has moved against geologic storage on land. Almost all proposed on-land carbon storage sites have been shut down as a result. It is likely that future European carbon storage will move off shore. In

contrast to the continents, the global seafloor is primarily comprised of basaltic rocks – only a small fraction of submarine rocks are consolidated sedimentary rocks. Porous basaltic rocks, covered by impermeable, altered basalt cap rocks are common adjacent to the continents and these have huge storage capacity. CarbFix has demonstrated a potential new option for submarine geological carbon storage, off shore, which is what is more accepted to the European public. We have done the testing and development on shore, in the Iceland basalts, because it offers easier working conditions but the method can be implemented off shore, in the same type of rock.

It is notable that the CarbFix approach is portable and not limited to the power industry. The CarbFix method involves the injection of gas dissolved in water into reactive rocks and the formation of stable carbonate minerals. The CarbFix gas capture/separation technology is independent of the gas source; the CarbFix capture/separation pilot plant runs independently of the Hellisheidi power plant. Therefore, we envision that it can be adopted directly by non-power generating industries throughout the world.

In addition to direct development and demonstration of CCS technology and infrastructure, the CarbFix team has furthermore focused on other inter-related activities such as development of laboratory, field and simulation equipment and tools that are expected to have a notable impact along with the aforementioned industrial scale CCS methods and equipment. These include:

- Design of a geochemical and tracer monitoring program to determine efficiency of CO₂ trapping mechanisms and the possibility of leakage.
- Design of a tracer injection system to label injected CO₂, providing methods for geochemical monitoring of injection site.
- Design of a high pressure down-well bailer sampling device to use for monitoring injection sites.
- Improved thermodynamic and kinetic databases describing phases of interest for carbon mineral and solubility trapping and are used in (hydro)geochemical modelling.
- Improved conceptual and mathematical modelling tools for predictive simulation and reservoir management of injection sites and carbon trapping processes.
- Design of a high pressure column flow reactor for laboratory studies on subsurface processes in carbon geological storage.

These developments will be beneficial for many industrial and scientific applications beyond the CO₂ underground sequestration.

1.4.2 Main dissemination activities and exploitation of results

The results of CarbFix provide novel and effective methods to capture and store carbon dioxide directly from its industrial source. To insure the maximum impact of the CarbFix results, we created a detailed dissemination plan at the beginning of the project, tailored to the many actors involved and interested in CCS. This dissemination plan is outlined in the following and is detailed in the following sections.

- Launch and maintenance of an external CarbFix website as well as an internal worksite.

- Development and publication of educational material for children regarding a) climate change and the challenges it poses and, b) CarbFix as a contribution to the solution.
- Maintenance of Press office/writing of press releases to support media coverage.
- Three international public outreach forums.
- Presentations and publications at conferences and meetings.
- On-site demonstration and education.
- Market research and business plan.
- Exploitation plans and reports.

The following deliverables describe in detail the work carried out within work package 7, which focused on dissemination and exploitation.

- D7.1) Website online [month 1]
- D7.2) Annual dissemination report 1 [month 12]
- D.7.3) Updated exploitation plan [month 12]
- D.7.4) Market research [month 12]
- D.7.5) Mid-term public outreach meeting [month 18]
- D7.6) Updated exploitation plans [month 18]
- D7.7) Annual dissemination report 2 [month 25]
- D.7.8) Business plan for industrial exploitation [month 30]
- D.7.9) Final public outreach meeting [month 36]
- D.7.10) Final dissemination and exploitation reports [month 36]

1.4.2.1 CarbFix website

The external CarbFix website (www.carbfix.com) was launched and corresponding deliverable 7.1 was handed in at end of month 1. The website was maintained and updated on a regular basis. An internal worksite (<http://vd-ext.or.is/verkefni/carbfixec>) for CarbFix partners was set up and all participants as well EC representatives had access to the site.

1.4.2.2 Development of educational material for children

The purpose of the educational material for children was to educate them on climate change and the challenges it poses as well as CarbFix as a contribution to the solution. After discussion and research of the teaching material, it was decided that the focus group for the material would be 11-12 years old children.

A group of three students with background in education/pedagogy, design/computer sciences and geology/natural science developed the material in 2013. The educational material was officially finished in the end of August that year, and was launched on the CarbFix website (www.carbfix.com). The National Centre for Educational Materials in Iceland was kept informed on the progress of the work.

1.4.2.3 Press release and coverage in the media

Dr. Sigurður Reynir Gíslason, head of the CarbFix scientific steering committee and leader of WP3, and Dr. Eric Oelkers, member of the CarbFix scientific steering committee and leader of WP2 and

WP7 published a paper on CarbFix in *Science* in April 2014. Considerable media coverage followed the publication, e.g. in Icelandic news³.

Several other media have shown interest in the project. CarbFix was published as a success story on the Horizon 2020 website in September 2014⁴ and climate reporting project 'Bracing for Impact' published a story on CarbFix on Nov 19th 2014⁵.

The CarbFix consortium has furthermore been visited by several film crews as well as editors/journalists that wish to visit the CarbFix injection site and write a story on the project. These include:

- A film crew on behalf of the Top-Level Research Initiative, a Nordic venture for climate, energy and the environment
- Lead geology reporter for the New York Times
- Senior editor for *Science* at Columbia University

Christiana Figueres, the Executive Secretary of the United Nations Framework Convention on Climate Change (UNFCCC), visited Hellisheidi Power Plant at the end of the CarbFix project in order to learn the project results. Ms. Figueres was very interested in the CarbFix injection method as it has been proven to result in immediate solubility storage of injected CO₂ followed by between 80-90% mineral storage within only 1 year from injection, hence greatly increasing the safety of geological carbon storage compared to conventional supercritical storage of CO₂. In the case of supercritical CO₂ storage, solubility storage is generally believed to take ca. 100 years whereas mineral storage takes 10,000-100,000 years, if it occurs at all.

1.4.2.4 International public outreach forums

Initial plans called for running two international public outreach forums, mid-term and at the end of this project. The CarbFix consortium has, however, been more active in public outreach than originally planned as four international public outreach forums were held over the course of the project. Following sections describe the different public outreach forums in more detail.

Combined CarbFix/Delta-Min conference and workshop (Feb-March 2012)

The first international public outreach forum consisted of a combined conference/workshop between the CarbFix and the Delta-Min network which was held in Oviedo, Spain February 29th – March 2nd 2012. About 70 scientists and students participated in the workshop, which included dozens of interesting oral and poster presentations. Three talks were given on the CarbFix project at the conference. In addition, two CarbFix consortium members gave talks on research focusing on mineral reactions occurring in CarbFix experiments and field injections.

³ See e.g. Energy News: Storing Greenhouse Gases by Petrifying Them. 24. april 2014, <http://www.technologyreview.com/news/526896/storing-greenhouse-gases-by-petrifying-them/>, <http://www.ruv.is/frett/timamotanidurstodur>,

http://www.mbl.is/frettir/taekni/2014/04/25/sigurdur_reynir_i_science/

⁴<http://ec.europa.eu/programmes/horizon2020/en/news/quick-fix-underground-carbon-storage>

⁵<https://www.beaconreader.com/flux/a-planetary-fix>

The 2013 Icelandic Geothermal Conference (March 2013)

CarbFix and related R&D projects were presented at the 2013 Icelandic Geothermal Conference, which took place at the Harpa Conference Centre in Reykjavík, Iceland, between 5th and 8th March 2013. Roughly 600 participants, delegates, and exhibitors from over 40 countries attended the conference to discuss changes and forward thinking within the energy industry, with 55 presentations given by global figureheads within the industry. Keynote speakers included Sri Mulyani Indrawati, Managing Director of the World Bank; Bjarni Bjarnason, CEO of Reykjavík Energy; Gunther Oettinger, the European Commissioner for Energy and Professor Jefferson W. Tester, Cornell University. Össur Skarphéðinsson, Iceland's Minister for Foreign Affairs, furthermore gave an opening speech and the closing presentation was made by President Ólafur Ragnar Grímsson, who briefed the challenges posed by global warming. CarbFix was presented in two presentations at the conference. An informative brochure on CarbFix and related R&D projects ongoing within Reykjavík Energy was also prepared for the conference and made available to conference attendees at Reykjavík Energy's exhibition booth.

Brainstorming day in association with the TCCS-7 conference in Trondheim (June 2013)

The third planned international public outreach forum consisted of a brain-storming day on the long term fate of geologically stored CO₂. The brainstorming day was planned in cooperation with four other EU-funded CCS projects; MUSTANG, ULTIMATECO₂, CO₂CARE and PANACEA. Over 200 specialists on Carbon Capture and Storage met on June 3rd 2013 in Trondheim to debate key issues, innovations and best practices related to CCS.

The workshop focused on a number of key areas including:

- Processes affecting the stabilization of the stored CO₂
- Mechanical impacts (seal integrity, fault reactivation, plug alteration) and induced seismicity
- Monitoring technologies during site operation and decommissioning
- Modelling and validation
- Leakage detection and mitigation
- Risk assessment and management

Proceedings of the Brainstorming Day 2013 are available at:

<http://www.bsdt2013.org/proceedings.cshtml>

CCS conference and workshop in Iceland (August 2014)

A large public outreach meeting was held in Iceland in August 2014. The venue was a joint EU and Nordic network conference between CarbFix, CO₂-REACT, MINSC, MetTrans and NORDICCS. The event was named The International Carbon Conference (ICC 2014) and was held in Iceland from August 25-29⁶. The event involved 1) Network management meetings, 2) Field sampling course for students attending the event, 3) ICC 2014 conference with 15 invited presentations and about 70 posters and 4) Field trip to S-Iceland.

⁶ See conference website at: <http://www.or.is/en/projects/international-carbon-conference-2014>

About 160 participants from all over the world attended the event, including officials from ministries and environmental agencies in Iceland. 13 invited speakers gave talks on CCS and related topics and about 70 posters were presented. The President of Iceland, Mr. Ólafur Ragnar Grímsson, one of the founders of CarbFix, opened the conference with an influential speech about the importance of taking immediate action against climate change and that excuses will no longer suffice.

1.4.2.5 Exploitation plans and reports

Carbon Capture and storage (CCS) as a potential industrial method is currently on the decline. Two of the major reasons why CCS is declining is that 1) it costs far more than the value of the carbon dioxide that is removed from the environment through CCS, and 2) there are very strong concerns that the carbon injected into the ground will remain for sufficient time to stem the potential detrimental effects of the increasing CO₂ concentration in our atmosphere. The CarbFix project has addressed directly these two challenges in a number of fundamental ways:

- **Cost benefits of subsurface carbon mineralization:** A major result of CarbFix is to accelerate the transformation of carbon dioxide injected into the subsurface to stable carbonate minerals. Once fixed as a stable carbonate mineral, it is not possible for the injected gas to re-migrate to the surface; one can walk away from the storage site with no further risk. This eliminates all monitoring costs once carbon mineralisation is confirmed.
- **Increasing storage security:** There has been increasing concern over the detrimental effects of the leaking of injected carbon back to the surface. Published estimates indicate that if less than 1% of injected carbon is lost annually the whole of the CCS effort will be wasted. More recent reports have prompted a debate if CCS is worth the effort at all, as induced seismicity may likely occur at all carbon injection sites, providing pathways for buoyant supercritical CO₂ to re-escape to the atmosphere. CarbFix has addressed these concerns in several distinct ways. First, the CarbFix method injects the gas dissolved in the water phase, rather than a separate single phase (e.g supercritical CO₂). Water, saturated with CO₂ is denser than formation waters and will tend to sink rather than rise. Second, the CarbFix method promotes the rapid mineralisation of the injected gas. Once CO₂ is mineralized it is immobile and is no longer at any risk to escape.

The CarbFix method is easily applicable wherever industrial waste-water is being injected into the subsurface or where point sources of CO₂-rich emissions are located close to a water source (including seawater). Moreover it may be most economically favourable for industry having the additional challenge of disposing other polluting water-soluble gases such as H₂S, including the geothermal industry and gas sweetening processes. It is thus anticipated that the CarbFix method can be commercialized throughout the geothermal industry and beyond. The following measures will allow for commercializing the CarbFix injection method:

- OR is collaborating with other geothermal energy companies in Iceland on geothermal gas capture, treatment and injection. It is anticipated that the CarbFix method will be implemented by these other companies following its successful demonstration at Hellisheidi together with its economic optimisation. Note that the removal of sulphur from power-plant flue gases are likely be required by other power plants in Iceland in the following years due

to a new regulation on atmospheric H₂S concentration, providing a powerful incentive to implement this method throughout the country.

- Commercialization will take advantage of the contact and collaborations among CarbFix partners and paternal external industry end users. OR and the UI are members of NORDICCS, a nordic CCS competence centre, where the CarbFix method is discussed in detail. NORDICCS is a virtual carbon capture and storage (CCS) networking platform aiming for increased CCS deployment in the five Nordic countries. The success of NORDICCS is dependent on active involvement from industrial partners, e.g. Statoil Petroleum AS, Gassco, CO₂ Technology Centre Mongstad and Norcem AS in Norway as well as Vattenfall in Sweden. OR also has a dissemination cooperation with 15 Scandinavian energy and district heating companies. OR also cooperates with the International Energy Association (IEA) and with Nordvarme, Euroheat & Power, Nordenergy and Eurelectric as a member of Samorka, a federation of the Icelandic electricity industry, district heating, waterworks and sewage utilities in Iceland. The UCPH group also has close partnerships with Maersk Oil and Gas A/S and BP p.l.c., both major oil and gas producers. A21 similarly has a close cooperation with ENDESA, CIUDEN and Foster Wheeler, all of which are dedicated to the development and promotion CCS technology. The above connections will be utilized to successfully present results of CarbFix to different end users, and facilitating the commercialization of CarbFix technology and techniques.
- OR along with Icelandic authorities is working on increasing understanding and acceptance of the CarbFix method within the EU as a better solution for injecting and storing CO₂ than conventional supercritical CO₂ storage. As mentioned above, the benefits of dissolving CO₂ during injection are considerable as risk of leaks due to buoyancy are eradicated, potentially leading to greater public acceptance.
- In addition, the CarbFix method will continue to be extensively promoted through presentations at local and international meetings, outreach forums and media, scientific peer-reviewed papers and online educational material.

1.5 The address of the project public website, if applicable as well as contact details

The external CarbFix website (www.carbfix.com) has been launched and corresponding deliverable 7.1 was handed in at end of month 1. The website is maintained and updated on a regular basis. An internal worksite (<http://vd-ext.or.is/verkefni/carbfixec>) for CarbFix partners has also been set up and all participants as well EC representatives have access to the site.