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ScienceDirect

Energy Procedia 00 (2017) 000–000

Energy Procedia

www.elsevier.com/locate/procedia

13th International Conference on Greenhouse Gas Control Technologies, GHGT-13, 14-18 November 2016, Lausanne, Switzerland

# What have we learned about $CO_2$ leakage from field injection tests?

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**Abstract**

Legislation and guidelines developed for Carbon Capture and Storage (CCS) have set performance requirements to minimize leakage risk, and to quantify and remediate any leaks that arise. For compliance it is necessary to have a comprehensive understanding of the possible spread, fate and impacts of any leaked $CO_2$, and also the ability to detect and quantify any leakage. Over the past decade, a number of field scale $CO_2$ release experiments have been conducted around the world to address many of the uncertainties regarding the characteristics of near-surface expression of $CO_2$ in terms of the impact and quantitation of $CO_2$ leaks. In these experiments, either free phase or dissolved $CO_2$ is injected and released into the shallow subsurface so as to artificially simulate a $CO_2$ leak into the near-surface environment. The experiments differ in a number of ways, from the geological conditions, surface environments, injection rates and experimental set-up - including the injection and monitoring strategy. These experiments have provided abundant information to aid in the development of our scientific understanding of environmental impacts of $CO_2$ while assessing state of the art monitoring techniques.

We have collated a global dataset of field-scale shallow controlled release experiments that have released $CO_2$ at depths shallower than 25 m. The dataset includes 14 different field experiment locations, of which nine intended to release $CO_2$ to surface, and the remaining sites intended for $CO_2$ to remain in the shallow subsurface. Several release experiments have been conducted at half of these sites, and so in total, 42 different $CO_2$ release tests have taken place at the 14 sites we examine. These experiments and their results are scrutinised to establish: (i) the range of experimental approaches and environments explored to date (such as the environment, subsurface conditions, injection strategy and whether gaseous or dissolved $CO_2$ were injected and in what quantities); (ii) the range of $CO_2$ injection and surface release rates at these experiments; (iii) the collective learnings about the surface and subsurface manifestation of the $CO_2$ release, the spread and fate of the $CO_2$, rates of $CO_2$ flux to surface, and methods of measuring these; (iv) how successfully current approaches can detect and quantify $CO_2$. This allows us to highlight where uncertainties remain and identify knowledge gaps that future experiments should seek to address. We also draw

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Peer-review under responsibility of the organizing committee of GHGT-13.

on the collective experiences to identify common issues or complications, and so recommend 'best practice' guidelines for experiment design and reporting at future $CO_2$ release experiments.

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*Keywords:* $CO_2$ storage; monitoring; field experiments; leakage; environmental impact; $CO_2$ fate; experiment design.

## 1. Introduction

Carbon Capture and Storage (CCS) is a promising climate mitigation technology, whereby $CO_2$ emissions are captured at source, compressed and transported and then injected into deep geological formations where it is intended to remain for geological timescales. Small amounts of $CO_2$ leakage could be tolerated without negating the cost-effectiveness of CCS from both climate change mitigation and financial perspectives [1, 2], and the migration of $CO_2$ or brines from the $CO_2$ store may beneficially relieve reservoir fluid pressure [3]. However unintended leakage of $CO_2$ or formation fluids would impact on a number of stakeholders, incurring financial [4] and environmental costs [5] and also challenge the social and political acceptability of the technology [6]. As such any incidence of leakage from engineered stores could have ramifications for the CCS industry on a global scale, and so the viability of CCS depends on the reliable containment of injected $CO_2$ in the subsurface.

Legislation and guidelines developed for CCS set performance requirements that seek to minimize risk of leakage from the storage formation. The IPCC [7] recommend that $CO_2$ stores should operate with less than 1% $CO_2$ loss to the surface over 1,000 years. The US Department of Energy (US DOE) aims for 99% containment of $CO_2$ injected for the purpose of geological storage [8], whereas the EU CCS Directive [9] requires $CO_2$ to remain 'permanently' in the storage formation. Any $CO_2$ that leaks from the storage formation must therefore be quantified for reasons of performance assurance, as well as carbon accounting [10]. Furthermore, legislation permitting subterranean $CO_2$ storage in the US, EU and Japan or subseabed in the North Atlantic require appropriate assessment of risk of $CO_2$ leakage from the intended storage reservoir, the potential impacts of $CO_2$ leakage on the environment, and means of monitoring for leakage [5]. In this context, 'environment' includes the near-subsurface (such as underground sources of drinking water) or surface (terrestrial or marine) ecosystems, including human health. Environmental impacts might source from the $CO_2$ itself (free phase or dissolved) and any co-injected impurities, or brines displaced as a result of pressure perturbation from $CO_2$ injection and migration or degraded by geochemical interaction of $CO_2$ and the surrounding rock, or mobilization of other fluids (e.g. methane).

To comply with site performance and monitoring requirements it is necessary to have a comprehensive understanding of the possible spread and fate of $CO_2$ in the deep and shallow subsurface, and the potential impacts of such leakage. Since approximately 40% of global storage capacity is located offshore [11], it is important that the effect on both the marine and terrestrial environments are explored. It is also important to develop monitoring approaches that are capable of enabling any $CO_2$ leaks to be identified, attributed, and quantified (referred to as monitoring, measurement and verification, MMV, techniques). This presents challenges because $CO_2$ can be naturally present or generated in the subsurface, biosphere and atmosphere as part of baseline or background concentrations.

Over the past decade a number of field-scale controlled release experiments have been conducted around the world to further scientific understanding of environmental impacts and test MMV techniques. The experiments release free phase or dissolved $CO_2$ into the shallow subsurface to artificially simulate a $CO_2$ leak into the near-surface or surface. The experiments differ in regard to the geological and surface environments and experimental set-up, including the injection rate and monitoring strategy. Since $CO_2$ release is controlled, these experiments provide excellent opportunity to test methods of measuring and quantifying $CO_2$ fate, and compare changes to environmental conditions and ecosystem health. At the same time, methods can be calibrated and expertise and capability development occurs through learning-by-doing at the field site for future commercial-scale applications.

A number of recent reviews have excellently summarised the significant contribution that these experiments have made to current scientific understanding of environmental impacts and state of the art monitoring techniques [5, 12-

15]. However to date there has been no comprehensive examination of the collective learning at these sites with regards to the fate and spread of the $CO_2$ and the surface manifestation of the leakage. Similarly, there have been no syntheses of the injection rates and the leakage pathways that these sites mimic, nor a consolidation of lessons learned for the design of a successful experimental approach. To this end, we have collated a global dataset of field-scale controlled release experiments, detailing the experimental approach and findings about the surface and subsurface manifestation of the $CO_2$ release, the fate of the $CO_2$ and leakage quantification. The results are scrutinised to elucidate collective learnings, and are compared to observations from natural analogue and modelling studies. We highlight how these experiments have developed our understanding of $CO_2$ leakage processes and where uncertainties remain. Future release experimental design and reporting will benefit from this work as the scientific community continue to seek methods to best characterise and monitor storage sites most effectively.

## 2. Compiling the dataset of $CO_2$ release experiments

We compiled a dataset of field-scale shallow controlled $CO_2$ release experiments that have been conducted to date (prior to February 2016). We considered experiments that were conducted in the field, and injected/released $CO_2$ into the subsurface with the aim that it would reach the surface or shallow subsurface rather than remain trapped in the injection formation. Since we were interested in experiments releasing $CO_2$ into the near-surface, we only include projects where $CO_2$ was injected shallower than 25 m below surface.

Dataset variables, listed in Table 1, were populated through detailed review of the published literature complemented by personal communication/interview with some of the key research scientists.

In the published literature, $CO_2$ release rates and $CO_2$ fluxes can be reported in a range of different units. For example, in the experiments reviewed for this dataset, rate of $CO_2$ leakage was expressed in terms of mass (g, kg, tonnes) or volume (mL, L) or concentration (mol, mmol) per unit of time (which might be expressed as per second, per min, per hour, per day, per year). $CO_2$ flux, by definition, should be expressed as the rate of $CO_2$ leaked per unit area (usually $m^2$). If no area unit is provided, the reported value is the $CO_2$ leakage rate (rate of $CO_2$ leaked). Where possible, to facilitate comparison, we harmonised these values to report dataset parameters in standardised units (see Table 1). We express $CO_2$ leakage in terms of $CO_2$ flux as $g(CO_2)s^{-1}m^{-2}$ and total rate of $CO_2$ leakage as $g(CO_2)s^{-1}$. We also express $CO_2$ leakage rate as tonnes per annum, $t(CO_2)pa$, since this is the standard unit for carbon accounting. If specific information was not reported or available, values were inferred, calculated or estimated from the published information where possible; for example, seep width might be inferred from the spatial distribution of $CO_2$ flux, or vertical leak velocity calculated from the injection depth and surface arrival time. When converting, for example, from $CO_2$ volume to $CO_2$ mass, in the absence of specific temperature and pressure conditions at the site we assume $CO_2$ properties at STP.

For each experiment, key monitoring tools were also noted - particularly for leakage quantification, including the presence of any added chemical tracers since these are considered useful for $CO_2$ attribution and fate. For most $CO_2$ release experiments, the data sets may not be complete in the publications reviewed; either those data were not collected, or are not yet publically available.

Table 1. List of variables collected for each of the $CO_2$ release experiments. Data was collected from the published literature and from corresponding with site researchers. The dataset cannot be shown here in full for reasons of space, but some variables are shown in Table 2.

| Variable | Sub variable | Description | Units |
|---|---|---|---|
| **Basic descriptive information** | *Acronym* | Project name and acronym | |
| | *Location* | Long, lat, country. | degrees |
| | *Project aims* | Principal research aims. | |
| | | Release to surface intended or not. | |
| | *Funding body* | Source of funding. | |
| | | Funding total. | € |
| | *Project partners* | Incl. industry and academia. | |
| | *Key contact* | Name and contact email of Principle | |

| | | | |
|---|---|---|---|
| | | Investigator. | |
| | *Project status* | Project completed / more $CO_2$ releases intended. | |
| **Experiment set up and site information** | *Well information* | Depth below ground surface of $CO_2$ injector | meters |
| | | Borehole type (deviated, vertical or inclined) | |
| | *Geological information* | Properties of the intended injection formation | |
| | | Properties of any overlying rocks | |
| | | Properties of any soils | |
| | *Ecosystem type* | Terrestrial/marine | |
| | | The surface ecosystem at the site. | |
| | *Hydrological characteristics* | Water table depth and flow direction | |
| **$CO_2$ injection (for each experiment)** | *$CO_2$ properties* | $CO_2$ source | |
| | | d13C composition | ‰ |
| | | Injected phase ($CO_2$ gas or water with dissolved $CO_2$) | |
| | *Injection rate* | Steady, variable, or incremental | |
| | | Maximum (and minimum) injection rates | g(CO2)/s |
| | *Injection periods* | Date injection started & ceased | |
| | | Total injection period | days |
| | *Quantity of CO2 injected* | For each experiment | kg |
| | *Overall* | Number of experiments at the site | |
| | | Total quantity of $CO_2$ injected | kg |
| **Monitoring** | *Area of monitoring* | Area of surveillance | $m^2$ |
| | *Baseline* | Baseline monitoring period | days |
| | | What was monitored ($CO_2$ flux, soil gas, plant/ecosystem diversity) | |
| | *Surface* | Lag time since injection began | Hours |
| | | Flux rate* | $g(CO2)/s/m^2$ |
| | | Vertical leak velocity** | m/s |
| | | Proportion of injected $CO_2$ released to surface | % |
| | | Leakage style (patchy, uniform) | |
| | | Patch radius*** | Meters |
| | | Leak location (with respect to the injector) | above injector / deviated |
| | | Temporal changes | |
| | *Subsurface* | Maximum soil gas concentrations | % |
| | | Detection time | hours |
| | | Distribution style | |
| | *Recovery* | Post injection monitoring period | days |
| | | Time taken to return to baseline | days |

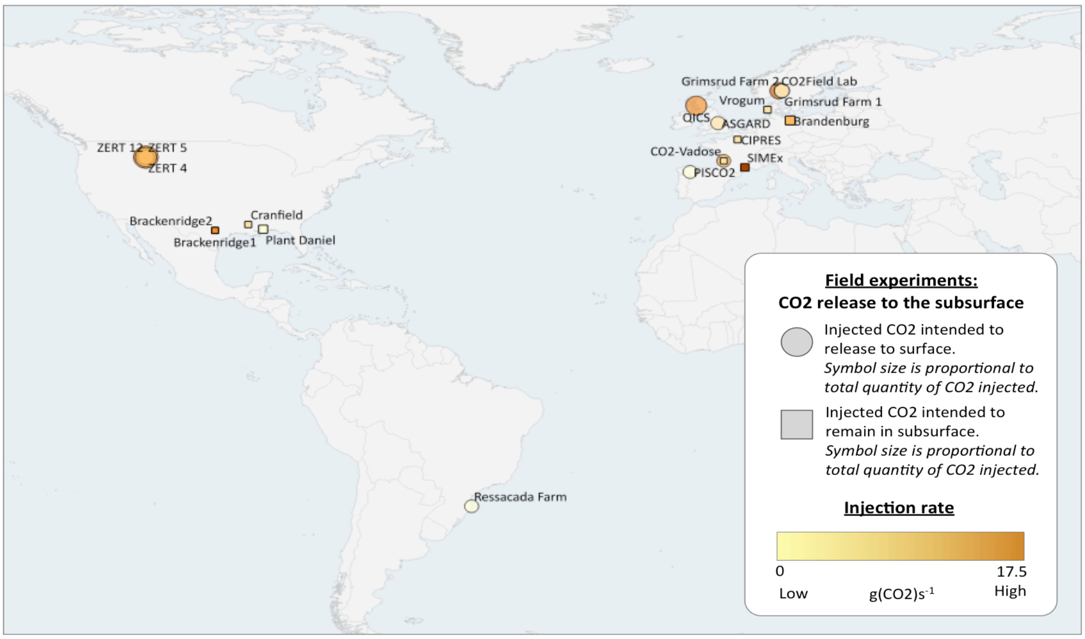
**Where $CO_2$ flux (rate of $CO_2$ leakage per unit area) was not reported, where possible, it was calculated from information about the leakage area and the reported total leakage rate; **Vertical leak velocity was calculated from the injection depth and the lag time between $CO_2$ injection and arrival at the surface (or near-surface); ***Where information about patch radius was not reported, if possible, it was estimated from flux measurements.*

## 3. Results and discussion

The detailed dataset includes 14 different field experiment locations around the world (we refer to each by their project acronym or their location). These are shown in Fig. 1, and a summary table of experimental parameters and results is provided in Table 2. Data have been collected using the framework in Table 1, used to generate our diagrams and for interpretive purposes but are not shown in tabulated form here.

A 25 m cut off was used to primarily to restrict our survey to a manageable number of case studies, and though two deeper have been reviewed as part of our activities (Plant Daniel [16] and Cranfield [17], both in the USA, and inject at 54 and 73 m, respectively) they are not included in the analysis we present here.

Fig. 1: Map of $CO_2$ release experiments around the world. Symbols are coloured according to the $CO_2$ injection rates at the site, and sized proportional to the total amount of $CO_2$ injected over the life to date of the sites (i.e. may be the sum of multiple injections).



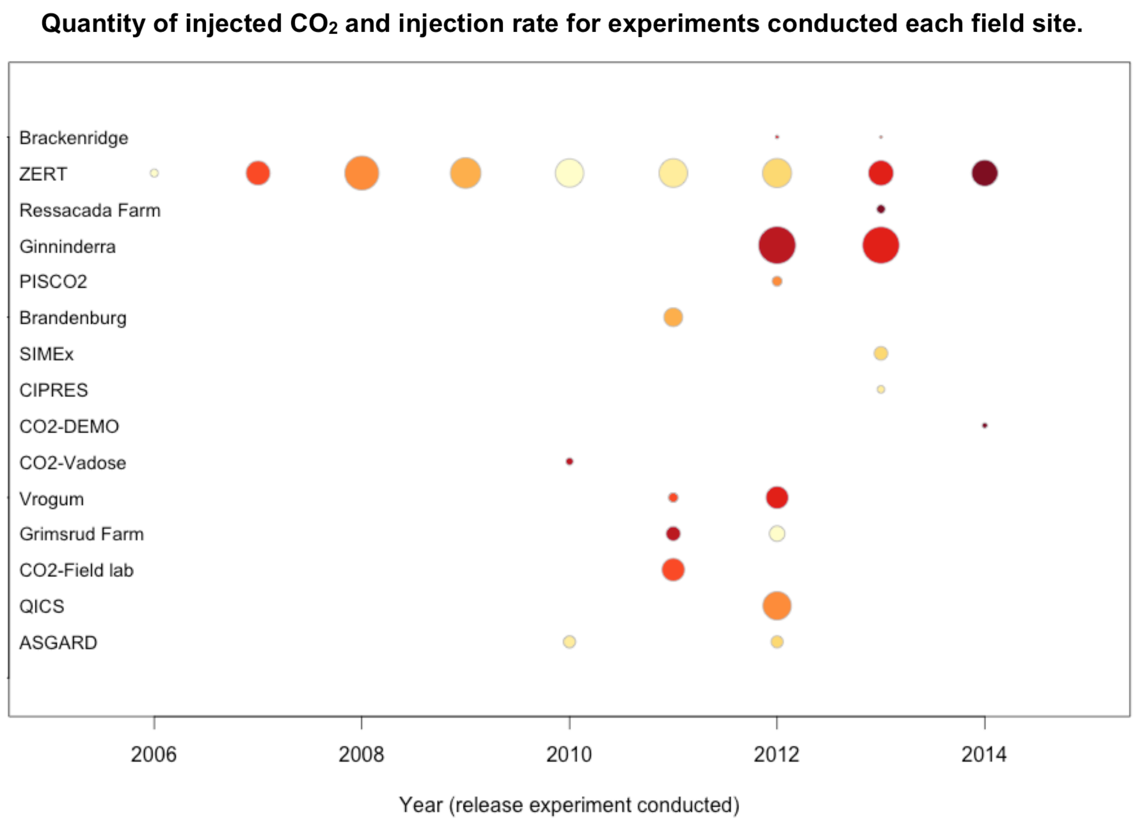

### 3.1. $CO_2$ release experiments: Where, when and why?

As can be seen in Fig. 1, the majority (10) of field experiments included in this dataset are located in Europe (ASGARD (1), QICS (2), $CO_2$ Field Lab (3), Grimsrud Farm (4), Vrøgum (5), $CO_2$-Vadose/DEMO (6), CIPRES (7), SIMEx (8), Brandenburg (9), PISCO2 (10)), with the remaining four located in Australia (Ginninderra, 11)), South America (Ressacada Farm, (12)) and the USA (ZERT (13) and Brackenridge (14)). Of all these field experiments to date, there has been only one subseabed $CO_2$ release; QICS (2). These global projects are made up of interdisciplinary teams, and budgets towards €1 M or greater, and typically endeavour to address one or more of the following broad aims:

- To investigate ecosystem responses to the injected $CO_2$ (half of the experiments conducted vegetation surveys on the indigenous grasses or on planted crops).
- To establish the fluxes, transformations and fate of $CO_2$ as it migrates from the injection point.
- To investigate geochemical interactions between $CO_2$ and groundwater.
- To test and calibrate models of $CO_2$ flow and fate.
- To test a broad or specific suite of monitoring techniques.

Nine of the $CO_2$ release facilities intended that the injected $CO_2$ be released to surface. For remaining six experiments the injected $CO_2$ was intended to remain in the shallow subsurface (see Table 2). Although $CO_2$ migration was intended at the $CO_2$-Vadose project, a clay layer in the subsurface prevented gas migration to surface [18, 19]. A subsequent project, the $CO_2$-DEMO project (6), successfully redesigned the experiment, injecting $CO_2$ above this clay layer [18]. Here, we mostly refer to the $CO_2$-DEMO project unless specifically stated.

Fig. 2: The quantity of $CO_2$ injected (circle size) and injection rate (colour) for each of the experimnets conducted at the shallow $CO_2$ release projects around the world, and the year that the experiments were conducted. The ZERT site has conducted the most experiments, and also the most varied injection rate (deeper colours indicate higher injection rate).




At the majority of the $CO_2$ release experiments, free-phase $CO_2$ gas was injected. These experiments commonly sought to mimic the effects of leakage of $CO_2$ gas, either from a point source or vertical feature (such as from poorly sealed well casing), or a linear feature (such as from a fault) – though some experiments did not focus on delivering $CO_2$ into the subsurface in a manner representative of a type of leak pathway. Two sites, CIPRES and Brackenridge, injected dissolved $CO_2$ by pumping water from the aquifer and saturated it with $CO_2$ before re-injecting into the same horizon [20, 21]. These were push-pull experiments that aimed explore the effect of $CO_2$ on groundwater quality.

All of the projects that we reviewed have been conducted in the past ten years, and as Fig. 2 shows, most of the $CO_2$ release experiments were conducted in the period 2011-13. At half of the sites, more than one release experiment was conducted, and the experiment phases often differed in length and rate of $CO_2$ release (see Table 2 also). For example, release experiments have been conducted typically each summer at ZERT since 2007, where the injection rate has ranged from 0.62 to 3.47 (g/s) (or 19 to 110 t($CO_2$)pa) [22]. In total, there have been 42 different

$CO_2$ release experiments completed at the 14 sites in the dataset, releasing a total of 82.8 t($CO_2$) into the subsurface over 994 days (i.e. 2.7 years). This is not a complete list, since some preliminary experiments may not have been reported, and some more recent experiments may not have been published yet. At least seven of the sites have been dismantled since the experiments were conducted with no intention to conduct future release experiments; a couple of sites are awaiting further funding for future experiments.

Table 2. Compilation of controlled $CO_2$ release experiments around the world conducted to date. Half of the sites have conducted more than one release experiment ('No. of exp'), some of them preliminary tests, and the experiment phases often differed in the length ('inj. length (days)') and rate of $CO_2$ injection ('Max inj. rate t($CO_2$)pa'). The style of the injection also varies between sites, injecting $CO_2$ as a gas (g) or dissolved in water (diss), and via a inclined, vertical (v) or horizontal (h) well, and at a steady, incremental (incr.) or variable (var.) injection rate.

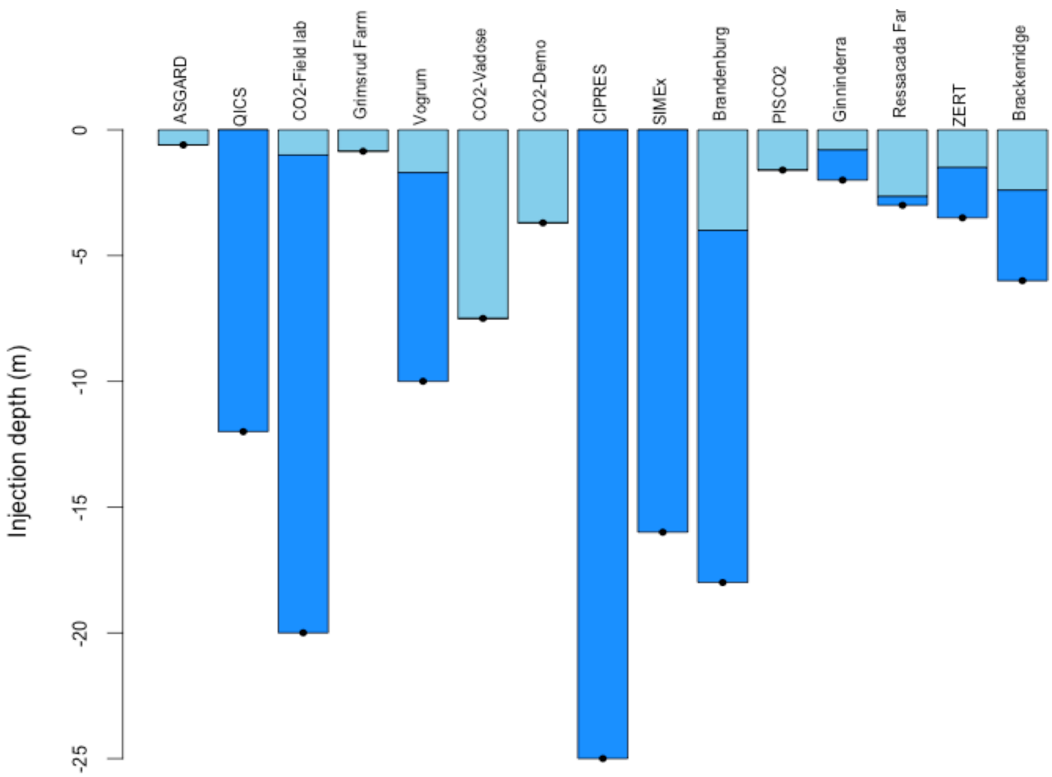
| N | Name or Acronym | Country | No. of exp | Year | Inj. depth (m) | $CO_2$ phase | Well orientation | Inj. style | Inj. length (days) | Max inj. rate t($CO_2$)pa | Surface leakage? | % of $CO_2$ leaked |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 1 | ASGARD | UK | 4 | 2006 - 2010 | 0.6 | g | 45° | Steady | 14-16 | 3.1 | Y | 34% |
| 2 | QICS | UK | 1 | 2012 | 12 | g | h | Incr. | 36 | 72.53 | Y | 15% as bubbles |
| 3 | CO2FieldLab | Norway | 1 | 2011 | 20 | g | 45° | Incr | 5 | 153.3 | Y | 5% |
| 4 | Grimsrud Farm | Norway | 4 | 2012 | 0.85 | g | h | Steady | 75 | 1.93 | Y | 82% |
| 5 | Vrøgum | Denmark | 6 | 2012 | 5-10 | g | 45° | Incr | 2-72 | 4.3-10.51 | N | 30-40% imaged |
| 6 | CO2DEMO | France | 2 | 2010-2014 | 3.7 | g | v | Steady | <1 | 3.06 | Y | 78% |
| 7 | CIPRES | France | 2 | 2013 | 25 | diss | v | Steady | 2 | 4.38 | N | |
| 8 | SIMEx | France | 1 | 2013 | 13-16 | g | v | Var. | 0.1 | 550.6 | N | |
| 9 | Brandenburg | Germany | 1 | 2011 | 18 | g | v | Steady | 0 | | N | |
| 10 | PISCO2 | Spain | 1 | 2012 | 1.6 | g | h | Steady | 46 | 0.96 | Y | 82.3% |
| 11 | Ginninderra | Australia | 5 | 2010 | 2 | g | h | Incr / steady | 56-80 | 21.8-79.6 | Y | |
| 12 | Ressacada Farm | Brazil | 1 | 2013 | 3 | g | v | Incr | 12 | 1.31 | Y | |
| 13 | ZERT* | USA | 5 | 2007-2014 | 1.1-2.5 | g | h | Steady / var. | 7-10 | 0.95-110.4 | Y | 90% |
| 14 | Brackenridge | USA | 2 | 2011-2012 | 6 | d | v | Steady | 2 | | N | |

*An experiment was conducted at ZERT in Autumn 2006 where $CO_2$ was released from a vertical pipe for 10 days to simulate leakage from well failure. Unless explicitly stated, when referring to the ZERT facility in the text we are referring to the subsequent horizontal injection experiments which is designed to simulate leakage via a line source such as a fault or fracture.

### *3.2. $CO_2$ release experiments: how?*

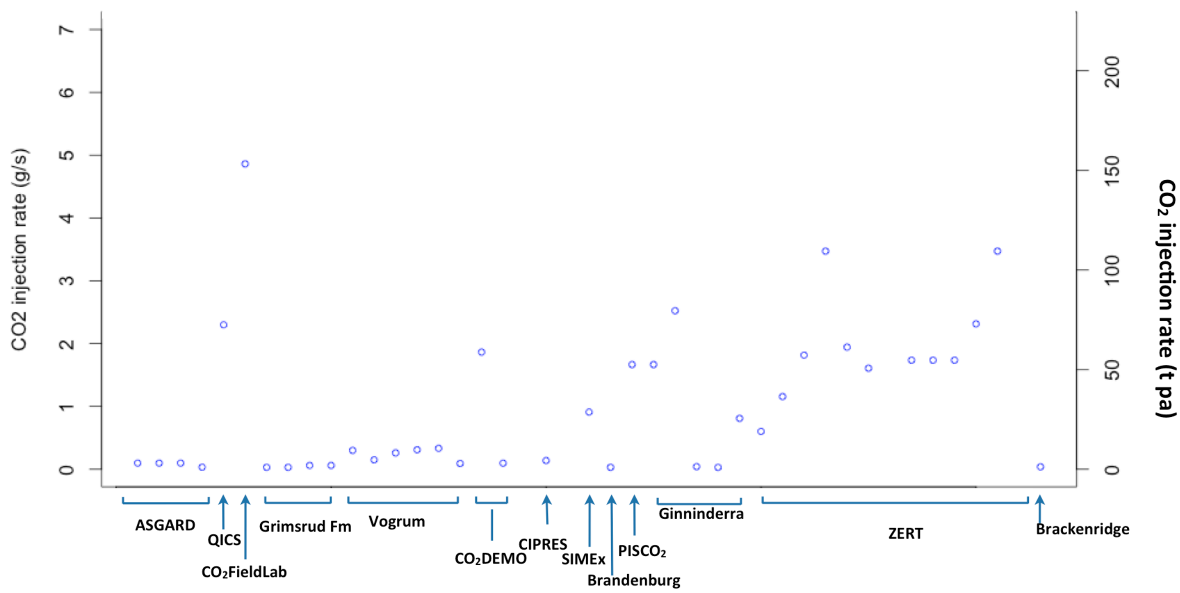
The site characteristics and experimental set-up of the $CO_2$ release experiments vary, though some projects mimic or build on the experimental design from other sites (e.g. the Ginninderra experimental set up is closely based on ZERT [23]). All of the projects injected high purity (99.9%) food grade $CO_2$ except $CO_2$-DEMO where a gas mixture of 90.57% $CO_2$, 5% Kr and 5% He was released [18]. $CO_2$ was often delivered by a borehole which was either horizontal (favoured at the shallower sites), at a 45° angle (ASGARD, CO2 Field Lab and Vrøgum) or vertical (favoured for deeper experiments; Table 2). Some set ups are more complex such as at PISCO2 where $CO_2$ is released from a horizontal grid arrangement of thin pipes. Several of the experiments have a number of physical blocks or plots for $CO_2$ release; for example the experimental area at ASGARD was divided into three blocks of eight replicate 2.5 × 2.5 m plots [24], at PISCO2 the $CO_2$ was injected through a grid with 16 pinholes [25], at ZERT the horizontal well, which is nearly 70 m long, is divided into six zones by inflatable packers [22] and Ginninderra is similar; the 100 m long pipe is partitioned into five 16-m long segments [23]. For experiments that inject $CO_2$ gas, the $CO_2$ is usually released via perforations along the pipeline rather than a single point source for injection. Indeed, perforations along the inclined well at Vrøgum aimed to simulate gas bubbling from a short fissure into flowing groundwater [26].

Fig. 3. For each project, the injector depth (black circle) and thickness of the vadose (pale blue) and the saturated zone (dark blue) is shown.




The depth of $CO_2$ injection ranges from 0.6 m (ASGARD) to 25 m (CIPRES). The deepest experiment to release $CO_2$ to surface is CO2 Field Lab at 20 m depth below surface. Fig. 3 shows the injector depth and the maximum depth of the water table by experimental site. Most experiments released $CO_2$ into sands or gravel. $CO_2$-DEMO is the only shallow release experiment to date to inject into a (lithified) carbonate formation, whereas $CO_2$ was released into soil at ASGARD ([27], and into an artificially constructed sand unit at $PISCO_2$ [25]. The overburden is often the same, or similar, to the injection formation, though inevitably there is more variation in the deeper injection experiments. At all the field locations the water table is relatively shallow, the vadose zone is less than < 4 m for all onshore experiments except the $CO_2$-DEMO site, where the water table is approximately 21 m depth [28], though the water table depth will vary seasonably. For example, the water table at ZERT is less than 1.5 m, and in springtime the water table can rise to surface level [22]. At four sites $CO_2$ is always injected above the water table (ASGARD, $CO_2$-DEMO, PISCO2, and Grimsrud Farm), whereas at ZERT, Ginninderra and Ressacada Farm the injection depth might be in the vadose or saturated zone, depending on the season. At all other sites the injection was below the water table. The majority of the experiments (particularly those that intended to release $CO_2$ to surface) were conducted in the dry season, with the exception of Ginninderra, which purposefully conducted experiments in both the dry and wet season to explore the effect of seasonality [23].

Fig. 4. The maximum injection rate at each experiment conducted at shallow $CO_2$ release projects worldwide. Projects show a range of $CO_2$ injection rates, and are typically selected to either represent a given leakage scenario from an engineered store, or to ensure the aims of the release experiment are achievable.




Free-phase $CO_2$ gas was released into the subsurface at all but two of the field experiments; CIPREs and Brackenridge injected $CO_2$-saturated water. As shown in Table 2 a steady or incrementally increasing injection strategy is favoured by most experiments at the site, though at SIMEx the rate varied unintentionally due to some challenges experienced during operation [29]. The maximum and minimum injection rate for each experiment at each $CO_2$ release site are shown in Fig. 4 (the maximum injection rate is illustrated visually in Fig. 2 and listed in Table 2). Fig. 4 shows that there is a wide range in the rate of $CO_2$ injection at these experiments; the highest rate, CO2 Field Lab is 4.9 $gs^{-1}$ (153.3 t($CO_2$)pa) and the smallest injection rates were at PISCO2 and Brackenridge, as 0.03, 0.04 $gs^{-1}$ respectively (0.95 and 1.3 t($CO_2$)pa). The majority of experiments however inject $CO_2$ between 0.05 and 2 $gs^{-1}$, which is equivalent to 1.6 - 63 t($CO_2$)pa. The values are largely selected based on possible permissible leak rates from engineered storage sites and modelled properties such as injectivity (e.g. Spangler et al., (2010)). This is less than 0.001% per year of a large scale CCS project injecting 1Mt($CO_2$)pa for 40 years, and are, these values are in the range of natural $CO_2$ emissions, for example, in Italy where there are hundreds of $CO_2$ seeps that most commonly emit between 10-100 t($CO_2$)pa [30]

The length of $CO_2$ injection at the experiments we reviewed varies from a couple of hours to several months. Most experiments inject $CO_2$ for periods shorter than 1 month, and indeed, the longest injection period at five of the sites (CO2 Field Lab, $CO_2$-Demo, Brackenridge, CIPRES, and SIMEx) lasted 5 days or less. This was not long enough for $CO_2$ leakage to reach steady state at CO2 Field Lab [31]. On the other hand, experiments at Grimsrud Farm and Ginninderra have lasted as long as ~3 months, and, though not included in this dataset, we note that the Plant Daniel field experiment is the longest conducted to date, lasting 5 months [16]. Generally, experiments investigating ecosystem responses to $CO_2$ injected for the longest periods. It is concluded by QICS researchers that the $CO_2$ release period (37 days) was not long enough, since the effect of $CO_2$ on pore water chemistry was only detected a couple of days before injection stopped [32, 33].

The field experiments find that change in stable carbon isotopic composition can be a sensitive indicator of the arrival of introduced/injected $CO_2$ [34-36]. $CO_2$ procured through chemical suppliers is often depleted in $^{13}C$ since the source is commonly from processes using hydrocarbons (e.g. natural gas to urea conversion; [37]). The $CO_2$ used at most experiments had $\delta^{13}C$ values towards -30 ‰, though the exact values are site and $CO_2$ source specific. This isotopic signature allowed the injected $CO_2$ to be distinguishable from biologically derived $CO_2$ in soil gas, and atmospheric $CO_2$, which are both less depleted (typical $\delta^{13}C(CO_2)$ values for biologically derived $CO_2$ are in the range of -20 ‰, and for atmospheric $CO_2$ are typically between -6 and -8‰), even though biological and atmospheric $CO_2$ varies spatially and temporally, for example with season, weather, and also any anthropogenic activity. The chemical signature of $CO_2$ used in the field experiments is in the range expected for captured $CO_2$ from most sources, and so the $CO_2$ that might be injected for storage [38]. As such, the simulated leaks can be considered isotopically representative of $CO_2$ that might leak from engineered stores. The only exception from this is $CO_2$ that sources from biomass combustion, which will be less negative than biologically derived $CO_2$, since $\delta^{13}C(CO_2)$ values from biomass are typically between <-6 - <-15 ‰ [38].

At a number of sites the monitoring period was much longer than the length of $CO_2$ injection, for example at Vrøgum the injection lasted 72 days but groundwater was monitored for 252 days [26]. The design of the monitoring array is site specific, but there are usually a number of monitoring boreholes of various depths to obtain regular soil gas and water samples, as well as devises for measuring $CO_2$ flux and possibly atmospheric monitoring methods such as Eddy Covariance towers, or geophysical tools such as Ground Penetrating Radar. At the QICS experiment a whole suite of monitoring approaches were deployed at and below seabed, sea surface and water column, and equipment installed by SCUBA divers [39]. The site can be designed to aid the management and recording of collected data, for example at ZERT the pipeline was laid at 45° North for ease of resolution of $CO_2$ transport, and a reference grid was laid over the ground surface [22], and subsequent projects followed suite (e.g. CO2 Field Lab, Jones et al., [31]).

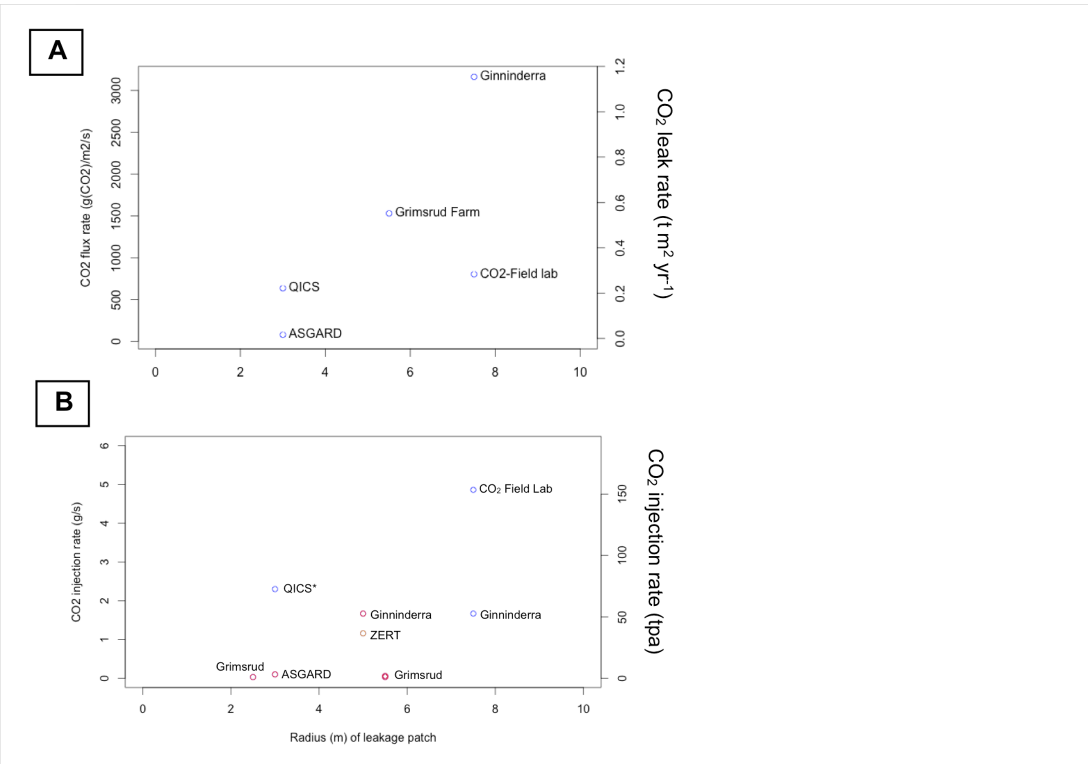
### *3.3. The surface characteristics of $CO_2$ leakage*

Injected $CO_2$ was rapidly detected at the surface at experiments where $CO_2$ release was intentional; often within 24 hours of the start of $CO_2$ injection (for example, at ZERT, $CO_2$ arrived within >5 hours of $CO_2$ release [40], and at the subseabed QICS experiment $CO_2$ bubble streams were observed within <3 hours of $CO_2$ release). The exact arrival time was not noted at a number of experiments due to the sampling frequency. The greatest lag time between injection and surface release was observed at Ressacada Farm and PISCO2 where surface flux of $CO_2$ was not detected for three or four days respectively [13, 25].

The surface leakage typically expressed as (several) patches or 'hot spots' showing $CO_2$ flux above background levels. The area of the surface hotpot is usually defined by $CO_2$ flux above baseline, where $CO_2$ flux and soil gas concentrations decrease radially from maximum levels at the centre of the hotspot. The hotspots that develop at the $CO_2$ release experiments are typically between 2.5 – 5.5 m radius, though leakage at Ginninderra was less patchy and occurred over a larger area than at other field sites [23]. These patches are often static once leakage has become established, for example, the leak patches at CO2 Field Lab coalesced as injection continued and then remained stable [31]. At QICS, which was located offshore, $CO_2$ bubble streams were mobile, but concentrated in two (static) patches [39] and the characteristics of the bubble stream (e.g. bubble density and bubble size) were affected by tidally induced changes to hydrostatic pressure [41].

At the patch center the soil gas $CO_2$ concentrations were observed to reach as high as 100% in some cases, though there is commonly more variability in soil gas concentrations towards the surface. Generally the $CO_2$ disperses quickly once it has degassed to surface but when there is little or no wind, $CO_2$ concentrations can become elevated above the soil. Plants and soil microbiology are affected by elevated $CO_2$ concentrations in the soil gas and land surface, showing effects within a couple of days, though some plant species are more resistant [5, 15]. Patchy $CO_2$ leakage matches observations at natural $CO_2$ seeps also [15, 42, 43], and observations at field and natural $CO_2$ release sites in a range of environments find that soil gas concentrations of 10% $CO_2$ and surface flux rates of 0.8 $kgm^2day^{-1}$ is the cut off above which the $CO_2$ begins to impact the ecosystem [15]. Hyperspectral imaging can detect the subsequent changes in e.g. chlorophyll levels, and so are a promising remote sensing monitoring tool [13, 44].

Fig. 5: Radius of the leakage patch and (a) maximum $CO_2$ flux rate and (b) maximum injection rate. The symbol colour indicates whether the $CO_2$ injection depth was into the saturated zone (blue) or the vadose zone (red) or if this was variable throughout the experiment (orange). QICS leakage radius might be considered to be either <3 m or >5 m, since bubble steams might be interpreted to occur in two patches on in one large cluster (Blackford et al., 2014). The patch width appears proportional to the flux of leaked $CO_2$.




We note that there may be a correlation between the hotspot radius and the $CO_2$ injection rate, as shown in Fig. 5a, where the hotspot radius is larger at higher injection rates, however we find no relationship between injection rate and maximum $CO_2$ flux rate (graph not shown here). Both the wet-season Ginninderra experiment and $CO_2$ Field Lab inject $CO_2$ into the saturated zone, and the hotspot radius is particularly large for these experiments. Interaction with the water table and soil conditions influences the way that $CO_2$ migrates through the subsurface, which we discuss further later in this paper. The flux rate and seep patch radius, shown in Fig. 5b, may also correlate, but there are too few data points with which to draw any reliable trend. The injection depth has no control on the $CO_2$ flux rate, nor does the injection rate control the vertical velocity of $CO_2$, though we do note that the vertical flow velocity is greatest for deepest experiments, where $CO_2$ will be less dense than the surrounding poor fluids.

At onshore experiments the location, width and intensity of hot spots were observed to be dependent on climatic conditions, including diurnal temperature (affecting airspeed), rainfall and pressure. At ASGARD, $CO_2$ flux was highest in drier periods, and stopped temporarily when the soil froze in winter [24]. $CO_2$ release performed at Ginninderra in the wet season and the dry season noted differences in the location, style and intensity of seepage. In the wet season leakage was restricted to one patch (16 m x 30 m) in a sandier region, and was characterized by more intensive gas flux, whereas in dry season leakage occurred in three smaller patches located in the more clay-rich regions. The dry season patches exhibited lower fluxes and were more steady state than the wet season patch [23]. Interestingly, observations of $CO_2$ driven mofettes in Italy also note seasonal changes in the location, size and style of $CO_2$ degassing [45].

These changes result from changes to the extent of the vadose zone and changes to soil properties from more rainfall. At Ginninderra, in the wet season the $CO_2$ was injected into the saturated zone whereas in the dry season it was injected into the vadose zone. It is hypothesised that in the dry season the greater extent of the vadose zone allows the $CO_2$, which is denser than air, to accumulate in the subsurface and so limiting $CO_2$ release to surface (see Schroder [46] of this conference). By contrast, experiments at other sites, such as ASGARD found $CO_2$ flux was impeded by rainfall events, although the difference in total $CO_2$ release to surface in different seasons has not been explore at other field sites. These variations are really important to consider because the period of surveying of a possible $CO_2$ leak above a CCS site (season, before or after rainfall, in the cool morning or hot afternoon) may then greatly influence the measured value and its eventual interpretation.

At a number of experiments the hotspots did not establish above the release point, and so there can be quite significant subsurface $CO_2$ migration - even when the injection depth is shallow. For example, at Ressacada Farm the hotspot was located ~30 m from the $CO_2$ release point (which was at 3 m depth) at a surface depression next to a road [13]. At the subseabed QICS site it seems that the patches of $CO_2$ bubble streams were located maximum of 10 m west of the $CO_2$ diffuser [39], along strike of the sediment structure [47]. At many of the field sites, the construction of the field facilities disturbed the subsurface structure such that the changes influenced the characteristics or location of the resulting $CO_2$ release – even though great effort and additional expense attempted to minimise subsurface disturbance. For example, at ZERT, despite the fact that horizontal directional drilling was used to install the $CO_2$ pipeline [22], the $CO_2$ leakage locations were controlled by small elevations in the horizontal well, and so it is thought that gas collected at high points within the pipe before leaking to the surface [48]. At Grimsrud Farm the $CO_2$ preferentially leaked along the border of the plots [35] and even the PISCO2 project, which used an almost entirely artificial set-up (a network of thin pipes release $CO_2$ into a sand unit which is boxed in by concrete that separates the sand from the underlying and adjacent soil though the top is open to atmosphere [25]), preferential $CO_2$ flow pathways quickly developed where injection and pumping tests performed prior to $CO_2$ injection are thought to have disturbed the sand structure [25]. However it is inappropriate to assume that these experiments are not valid or unrepresentative because of these issues. No soil structure will homogenous or undisturbed; soil surface is often altered or compromised through various practices such as farming, roads, laying of sewerage and other anthropogenic activities and also non-anthropogenic causes. Offshore the seabed sediment structure might be compositionally variable due to trawling, or due to storm disturbance or currents. The surface monitoring interval above a store will be traversed and surveyed during site characterisation and also monitoring design. The field experiments therefore usefully show that preferential flow pathways will usually channel $CO_2$ leakage, and these channels are sensitive to recent activity at the site and also might change with environmental factors

### *3.4. The sub-surface characteristics of $CO_2$ leakage*

At most onshore experiments the distribution of $CO_2$ gas in the subsurface was measured by shallow monitoring boreholes that allowed soil gas sampling, and complimented by other techniques such as ground penetrating radar or chemical tracers. The distribution of dissolved $CO_2$ can be detected by changes to groundwater chemistry, also collected via shallow boreholes or deeper monitoring wells.

At all sites that released $CO_2$ to surface, the extent of lateral spread of $CO_2$ in the subsurface was found to be greater than the area of surface degassing, $CO_2$ was detected first in soil gas prior to surface flux, and soil gas saturation was variable; affected by the soil/sediment structure and water saturation (rainfall/thickness of vadose zone). Observations at ASGARD found $CO_2$ moved preferentially through the more-permeable sandy and gravely deposits lying below the injection point [49]. Though there was no $CO_2$ flux to surface above the injection point at CO2 Field Lab, soil gas monitoring detected subsurface $CO_2$ in that region shortly after injected began, as $CO_2$ leaked up the well casing. Surface leakage emerged to the northeast (up dip) of the injector one day later once injection rate increased [31, 50]. Subsurface resistivity changes at Ressacada Farm (incurred as $CO_2$ gas moved through the aquifer, partially displacing the water in the pore space) were consistent with $CO_2$ leakage pathway to the hotspot location [51], which was 30 m from the injector.

At Ginninderra, soil gas measurements found that the season affected soil gas saturation, recording maximum of 80% saturation in the wet season, and 60% in the dry season when $CO_2$ flux was more distributed [23]. These surveys, aided by krypton tracer, found that in the wet season $CO_2$ spread 30 m from the horizontal well in the subsurface (nearly four times further than the surface flux) whereas in the dry season it only spread 5-10 m from the well (~ twice as far as the surface flux) (Feitz, pers. comm). Therefore, when $CO_2$ is injected into the saturated zone, $CO_2$ spreads further but gas intensity is more localised, whereas when $CO_2$ is released into the vadose zone $CO_2$ transport is more localised but gas intensity is more distributed.

Monitoring techniques deployed at Vrøgum found $CO_2$ favoured flow in more permeable rock formations; geochemical affects of $CO_2$ injection were faster and more uniform in the higher permeability sediments [26, 52, 53], the saturation of the gas was proportional to the grain size properties of the sediments [54], and the plume spread towards regions with higher permeability even overcoming groundwater flow to do so [52]. As such, the subsurface gas concentrations were heterogeneous, influenced by the permeability and structure of the subsurface [54]. Maximum gas saturation in the sediments at Vrøgum was estimated to be 7%, similarly, values at CIPRES were estimated to be in the range 2-7% [55]. Heterogeneous gas transfer is also supported by observations using noble gas tracers at the $CO_2$-DEMO project [18]. Interestingly, at QICS, repeat seismic reflection surveys found that, like at CO2 Field Lab, sub-surface $CO_2$ flow path was affected by sediment structure and also $CO_2$ flow rate [39, 47]. Sediment grain size controlled $CO_2$ flow initially until the gas pressure or gas volumes overrode the stratigraphic controls, spatially focussing the $CO_2$ flow via the formation of chimney structures [47].

All the experiments found that $CO_2$ induced changes in the groundwater chemistry. The spatial extent of geochemical impacts were also much wider than the extent of surface release, for example, while bubble vents at QICS were located within 10 meters of the subseabed injector, whereas the spatial extent of the geochemical impact of the injected $CO_2$ in the sediments and pore waters was contained to 25m of the injection point [33]. Monitoring techniques deployed at Vrøgum and Brandenburg observed a two-phase geochemical evolution of the $CO_2$ leak, where a pulse in ion concentrations is followed by persistent acidification [26, 52]. In the early stages of injection there is usually a delay before any chemical changes are detected, and it was hypothesized that this is because the gaseous $CO_2$ flows in discrete channels to start with, which limits the contact with the water-phase, and so restricting the amount of $CO_2$ that dissolves into the groundwater [54]. The Vrøgum researchers also noted that the unconfined aquifers were susceptible to recharge which cause rapid and inconsistent changes to the groundwater properties [26]. Where there is sufficient pressure from $CO_2$ release, $CO_2$ can flow against groundwater direction, as observed at Vrøgum where the $CO_2$ plume favoured spreading north-eastwards where there were higher permeabilities, however when injection ceased the plume followed groundwater gradient [52].

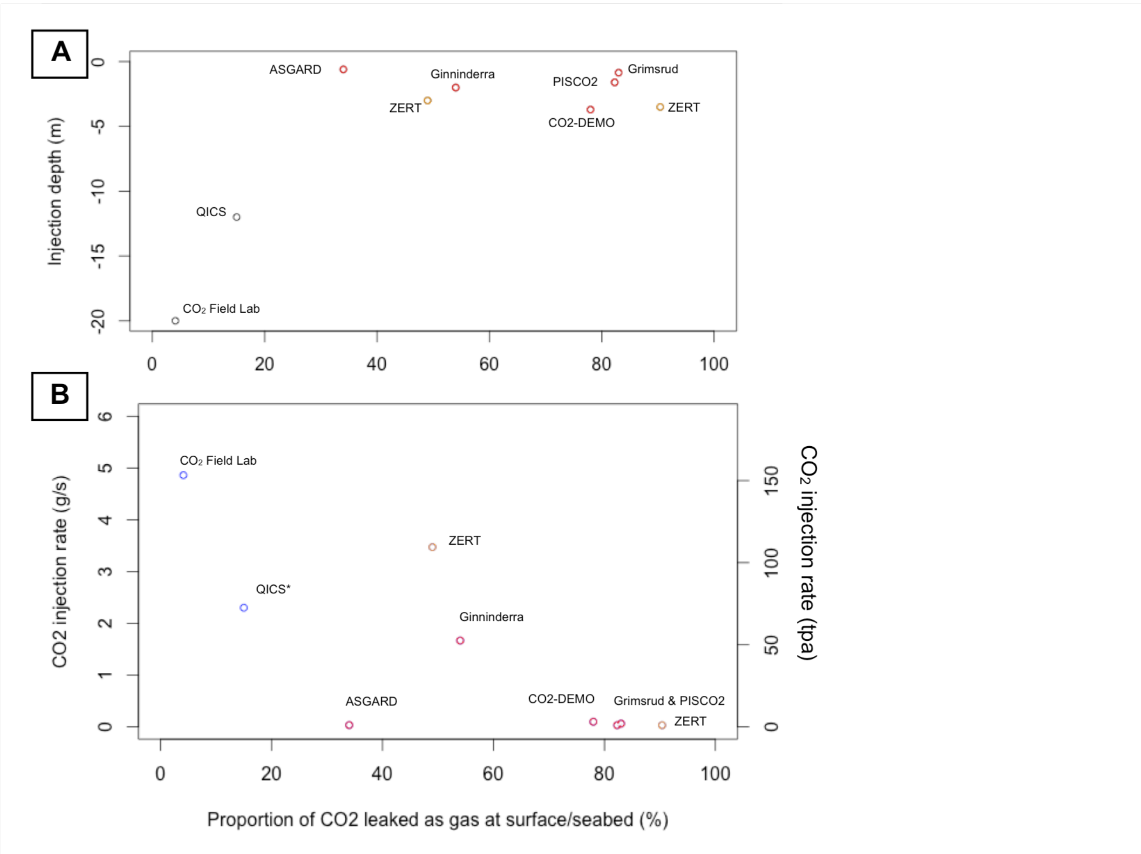
### *3.5. Quantifying $CO_2$ leakage*

Quantifying the proportion of injected $CO_2$ that is released to surface (atmosphere or seabed) has proven very challenging at release experiments [13]. Out of the 14 projects and 42 release experiments reviewed here, we find that only nine experiments (8 sites) report estimates of total $CO_2$ leakage to surface. These estimates were either extrapolated from flux measurements, or were modelled from multiple measurements over the duration of the release experiment. Estimates of total leakage range from 5% of the injected $CO_2$ (reported to be a likely underestimate [50]), up to 82-83% (at both Grimsrud Farm [56] and PISCO2 [25]), though not all reported estimates account for baseline $CO_2$ flux. In some cases, for example at ASGARD and ZERT, $CO_2$ migrated beyond the monitoring boundaries, making it difficult to estimate the relative proportions of $CO_2$ that leaked to surface or remained in soil gas or dissolved [40, 49].

The proportion of $CO_2$ that leaks to surface is observed to vary throughout the experiment duration, as various environmental factors affect $CO_2$ flux. For example, at Ginninderra, the proportion of $CO_2$ leaked to surface was higher in the wet season than the dry season (Feitz, pers. comm), whereas $CO_2$ flux at ASGARD was greatest in drier spells [57]. At QICS, 8-15% of injected $CO_2$ was released to seabed as a free phase during the QICS experiment, depending on the tide [39, 58]. Geochemical modelling based on pore-water observations at QICS find 14-63% remained dissolved within the sediment pore water [32]. The remaining proportion trapped in the sediment [47] could be imaged by repeat seismic surveys. Other remote methods were used to quantify subsurface gas saturation at several experiments. For example, at one of the Vrøgum experiments, cross-borehole ground penetrating radar imaged 30–40% of the injected $CO_2$ volume as free phase gas trapped in the sediment. The remaining $CO_2$ must have dissolved or migrated [54].

The experiments that found the smallest proportion of $CO_2$ leaked to surface/seabed as a gas also had the greatest injection depth ($CO_2$ Field Lab and QICS) find that the smallest proportion of $CO_2$ leaked to surface. It is therefore tempting to suggest a weak relationship between these factors, shown in Fig. 6a, however the leakage estimate at $CO_2$ Field Lab is likely to be an underestimate, and there is not other trend in the results. However, while there is no relationship between the injection rate and injection depths at the field experiments, the proportion of $CO_2$ that is released to surface is inversely related to the maximum injection rate (Fig. 6b). If this is the case, higher injection rates might encouraging lateral spread of $CO_2$ in the subsurface, perhaps because vertical spread is restricted by the soil properties.

Fig. 6: The estimated proportion of injected $CO_2$ that leaked to land surface or seabed as a gas at the field experiments plotted against (a) injection depth and (b) injection rate. The deepest experiments ($CO_2$ Field Lab and QICS) find that the smallest proportion of $CO_2$ leaked to surface, however the leakage estimate at $CO_2$ Field Lab is likely to be an underestimate. Instead, it is more convincing that a smaller proportion of gas leaks to surface when injection rates are higher.




Artificial tracers added to the injected $CO_2$ have been proposed as a potential method of quantifying $CO_2$ leakage rate [59, 60]. Several experiments have tested added tracers, including PFCs (ZERT), $SF_6$ (Brandenburg), noble gases (Krypton, Ginninderra; Helium and Argon, $CO_2$-DEMO), and future experiments at several sites also plan to use tracers. However no release experiment has yet attempted to quantify $CO_2$ leakage using chemical tracers. For tracers to quantify $CO_2$ leakage they must behave predictably and preferably conservatively (i.e. mimic the $_{CO2}$ behaviour). Krypton co-injected at the Ginninderra tests, and perfluorocarbon (PCF) tracers used at the 2007 ZERT experiments were found to correlate with soil–gas results and so track the injected $CO_2$ [48]. At ZERT, tracer concentrations were also affected by small changes in topography, and possibly also density or soil properties since away from the hotspot location, a 'reservoir' of tracer was detected at the soil-cobble interface at 1 m depth [22, 48]. Co-released noble gases at $CO_2$-DEMO included Helium and Argon, which both arrived ahead of the $CO_2$, and so behaved as precursor tracers for the leakage of $CO_2$ in the vadose zone [18]

### *3.6. Pre-and post-release monitoring*

Pre-release monitoring must be conducted to characterize the environmental baseline at the experimental field site to be able to establish which changes result from $CO_2$ release, and which are simply due to environmental variability unrelated to the release experiment. The environmental baseline must be assessed to comply with environmental regulation [5] and so the experience gained at $CO_2$ release experiments is extremely valuable.

Experience at field experiments have informed not only the importance of rigorous baseline monitoring, but also the type of baseline data collected and the appropriate time period over which baseline data is collected. Without adequate baseline, it becomes very difficult to, for example, report the flux of leaked $CO_2$. Further, there is no typical or standard baseline; background $CO_2$ flux and its variability are unique to each field experiment site. The variability of background is important to account for when interpreting $CO_2$ flux measurements. For example, at ASGARD, background $CO_2$ flux could vary by 3 to 4 fold, mostly in response to rainfall and air pressure [57].

At some experiments, baseline data was collected for only a couple of days, and concluded that this was not long enough [31, 40]. Subsequent projects have collected frequent or continuous baseline for longer, for example 2 weeks of continuous/daily monitoring was conducted at QICS and Ressacada Farm, but at QICS this was not deemed long enough. Some more recent projects such as $CO_2$-DEMO and Vrøgum collected data at intervals over a period of ~18 months prior to $CO_2$ release to allow a longitudinal baseline to be established. The spatial extent of baseline data should also be considered; for example, as Jones et al., [31] concludes, the location of the hotspots away from the point of $CO_2$ release shows how the areal coverage of baseline data must be spatially adequate, so that the hotspots do not establish where no baseline was collected.

Previous publications have little mentioned the value of continued monitoring of the field site once $CO_2$ injection has ceased. This is relevant also to environmental permitting at CCS projects, where post-release monitoring would be required for any leak arising from engineered $CO_2$ stores to establish when the leak has stopped (following any remediation efforts), and also to determine the longevity of any environmental impact, and remediate any long term ill-effects from any leakage. Further, post-release monitoring at these leak experiments to provide information about how $CO_2$ evolves in the absence of injection pressure is important for understanding $CO_2$ dispersion and fate. Our review also finds that, while most sites performed some post-injection monitoring, the length of monitoring period is widely variable, for example measuring for just one day only, or for a couple of days some time after injection ended. It was reported by some researchers that post-release monitoring was not sufficiently long enough for the decline in $CO_2$ concentrations to return to baseline conditions before sampling ceased. In fact, post-release monitoring at the ZERT 2008 experiments, QICS, and ASGARD are the only experiments to observe the return to baseline conditions. $CO_2$ flux at ASGARD was observed to return to baseline within 2-3 days [57], though at ZERT, it took 15 days to return to baseline at the hotspots (above the well) and only 5 days to recover further (5m) from the well [40] and at QICS, while $CO_2$ bubble streams stopped shortly after the $CO_2$ injection stopped, concentrations of all pore water constituents returned to background values within 18 days [33], and microbial species took 90 days to recover. Vegetation recovery may take longer, and be species dependent. Post injection monitoring at Vrøgum finds that 20 hours following the end of $CO_2$ injection much of the free phase gas had dissolved into the groundwater [54].

## **4. Common issues at $CO_2$ release experiments**

The information presented in the previous section illustrates the vast contribution that shallow $CO_2$ release experiments have made to current scientific understanding of near surface $CO_2$ flow pathways, $CO_2$ impacts, and methods of detecting $CO_2$ leakage. The observations at these field experiments (including patchy emissions, flux rates etc.), largely match those at natural $CO_2$ seeps also.

These experiments have allowed for testing of a range of monitoring techniques to identify and quantify $CO_2$ leakage. These experiments have highlighted the importance of establishing baseline, which can be highly variable. Current sampling approaches are high intensity, and the quantification of any leakage, as required by guidelines and legislation for CCS [10], has proven difficult. These experiments have therefore illustrated the need to develop more cost-effective detection and quantification techniques, and methods that are viable on the scale of the monitoring interval of $CO_2$ stores and over the time frame of the storage project. Release experiments present the opportunity to test remote detection methods, both airborne and ground-based, and these show promise as cost effective monitoring technologies [13, 44]. Similarly, the nature of the recovery period that follows $CO_2$ release has been little explored at $CO_2$ release experiments to date, and this is important for Environmental Impact Assessment and reporting procedure at $CO_2$ stores.

Knowledge exchange is extremely valuable for shaping future research, and the amount of knowledge exchange between shallow $CO_2$ release projects is laudable. As an example, a number of researchers from the ZERT project have been heavily involved with the design and set up of Ressacada Farm and Ginninderra. It is useful to summarise, for future work, some of the common issues and experiences that have occurred during the 14 projects that we have reviewed here:

- While every reasonable attempt might be made to minimise the disturbance to the subsurface structure during site construction and pre-release tests, it is difficult to avoid affecting the $CO_2$ flow pathways.
- The well bore is the primary source of unintentional leakage at $CO_2$ release experiments, much like the most likely leak pathways at CCS projects [7]. $CO_2$ leaked along the well bore / injection pipe at preliminary experiments at Ginninderra, $CO_2$ Field Lab, ZERT, SIMEx and Brandenburg, and possibly also at ASGARD, and some of these leaks required corrective engineering [15, 21, 50, 55].
- The location of degassing 'hotspots' can be hard to pre-empt prior to $CO_2$ injection, and this should be considered when designing the surface monitoring array and baseline survey. At several experimental sites where there are a number of experimental plots adjacent to one another, $CO_2$ has cross-contaminated neighbouring plots.
- Similarly, $CO_2$ breakout or injectivity may not occur as predicted by modelling and the pre-injection knowledge of the site [22, 35, 50]
- $CO_2$ flux rate at a single measurement point can vary due to a range of environmental factors. This is important to consider when interpreting results of $CO_2$ flux and using measured leakage rates to estimate total leakage quantities. It also highlights the importance of developing a robust understanding the baseline, and factors that influence the baseline. When reporting $CO_2$ flux, it should be clear whether the baseline has been subtracted from the measurements or not. Baseline should be subtracted before $CO_2$ flux values are used to estimate the proportion of leaked $CO_2$.
- Baseline surveys were not conducted for long enough at a number of $CO_2$ release experiments, and post-release monitoring was also not long enough to observe the return to baseline conditions. Environmental baseline must be assessed to comply with environmental regulation [5] – for leak detection and also to ensure that a site is returned to baseline after any leakage . Field experience of acquiring baseline and post-release information at the field experiments is extremely valuable to inform these monitoring protocols, as well as for identifying and quantifying leakage and $CO_2$ fate.
- If $\delta^{13}C(CO_2)$ analysis is a monitoring tool at the $CO_2$ release experiment, samples of $CO_2$ from every canister should be measured. The chemical signature might vary between canisters because suppliers can source $CO_2$ from different processes.
- Where that has been only one $CO_2$ release experiment conducted at a site, most researchers would choose to modify their experimental design to improve the experiment. For example, the period of injection at $CO_2$ Field Lab and QICS would ideally have been longer. This shows the value of conducting multiple releases at a site, such as at Ginninderra and ZERT.
- Quantifying the proportion of injected $CO_2$ that is released to surface (atmosphere or seabed) has proven very challenging at release experiments. This is also complicated by the need to integrate measured flux with the (variable) background biological $CO_2$ flux measurements.
- Many $CO_2$ release projects have not accurately established the $CO_2$ arrival time to surface or into soil gas. If the arrival time is an important parameter to establish for the field experiment then sampling frequency should be particularly intense during the first days of the experiment. Increase the sampling frequency.
- Ideally there would be a standard unit for reporting parameters such as $CO_2$ flux or arrival time and so on. Currently $CO_2$ might be reported in a range of units, including concentration. Similarly, there is no formal

definition of how the spatial extent of a $CO_2$ hotspot is determined; the outer limit of the hotspot is usually defined by $CO_2$ flux above baseline by an arbitrary value.

With these common issues in mind, future work could concentrate on, for example, exploring methods of rapid leak quantification, including trialing tracers for quantifying $CO_2$ leak rates, and also explore the role of topography, lithology and the water table on leakage distribution. Ginninderra is the only site to date that has explored the effect of the season with $CO_2$ leakage. The $CO_2$ Field Lab and $CO_2$-DEMO release experiments both used an old quarry site, whereas the remaining experiments were located on flat grassland - except QICS, which was subseabed. Therefore the effects of local topography have been little explored at these experiments. However, at ZERT, it was noted that the topography of the site, which rises slightly to the west and north, influenced the distribution of the PFC tracers (which are denser than $CO_2$) and at Ressacada Farm experiment leakage occurred at a surface depression [13]. Topography is observed to influence the characteristics of natural $CO_2$ seeps due to corresponding changes in depth to the water table [61], and also because topographic depressions are sheltered and so $CO_2$ dispersion can be limited, encouraging gravity-driven $CO_2$ ponding and so posing greater risk to human health [30, 43]. The field experiments find that the weather, namely windspeed, is the greatest factor affecting atmospheric $CO_2$ dispersion, which can also largely affect $CO_2$ measurements. Most currently operating onshore $CO_2$ storage projects (e.g. Quest, Boundary Damn, and In Salah are mostly located in relatively flat terrains) are in relatively uniform topographic settings, though future CCS projects might require monitoring of more topographic terrains. But the weather conditions at each site are distinctly different (temperate versus desert). For these projects, the effect of topography and annual weather conditions on the spread of $CO_2$ and tracers for $CO_2$ should be explored further. Similarly work should continue to characterise the effect of topography and infrastructure on wind speeds and $CO_2$ dispersion.

The hydraulic gradients at the sites are largely representative of groundwater flow systems in unconsolidated sandy aquifers with modest rainfalls [14]. Only one experiment, $CO_2$-DEMO, released $CO_2$ into lithified rock. Although we only analysed experiments injecting $CO_2$ at depths shallower than 25 m here, we found that only two field experiments were excluded from the analyses presented here, Plant Daniel and Cranfield, and these did not intend $CO_2$ to reach surface. It is tempting to recommend that future experiments release $CO_2$ at greater depths, and into a greater range consolidated rock formations that might comprise the shallow overburden above storage projects, with the aim to monitor $CO_2$ fate and spread and with the intention that $CO_2$ will leak to surface. However, experience at $CO_2$-Vadose and Vrøgum found that layers low permeability units such as fine sands or clays above the injector can prevent $CO_2$ from reaching surface, which demonstrated how subtle differences in lithology can significantly affect gas migration and dissolution [26]. Indeed, injection depth at Ressacada Farm was planned to be deeper, but a shallow depth was chosen after a preliminary survey found that clay lenses caused significant spreading of the $CO_2$ in the subsurface, and so a shallower release would result in less lateral spreading, shorter retention times, and earlier release to the atmosphere Moreira [62]. As such, conducting deeper $CO_2$ release experiments, with intent to release $CO_2$ to surface, would increase the cost and risk of the $CO_2$ release experiment, since deeper wells are more expensive to drill, and greater depths increase the risk that $CO_2$ will not reach to surface. $CO_2$ might have to be injected for a long time period and possibly in considerable quantities to allow the $CO_2$ plume to migrate distances that might allow the sealing horizon to be bypassed and so enable $CO_2$ leakage to surface – if at all. These scenarios might be useful to explore in future, longitudinal experiments.

## 5. Conclusions

Field-scale shallow $CO_2$ release experiments conducted around the globe in the past ten years have generated abundant data and contributed significantly to current scientific understanding of near surface $CO_2$ flow pathways, $CO_2$ impacts, and methods of detecting $CO_2$ leakage. We have collated and examined a global dataset experiments conducted to date, drawing on information in the published domain complimented with correspondence with researchers from specific sites. Preparing the data collected at each site into a uniform dataset was a non-trivial exercise, but allows us to illustrate and draw comparisons between the experimental procedure and results, and to identify future research needs. In this way, we examined 14 different $CO_2$ release projects, at which a total of 42 different $CO_2$ release experiments have been conducted. Other controlled release sites where injection depth was greater than 25 m were considered but not included in this paper. Collectively these experiments released 82.8

tonnes $CO_2$ over 994 days. Nine of the $CO_2$ release facilities intended that the injected $CO_2$ be released to surface, the remaining experiments intended for $CO_2$ to remain in the shallow subsurface (usually to investigate groundwater interactions). The experiments show a range of test approaches, including $CO_2$ release duration, modes of release (horizontal, angled, vertical pipes), and injection depths. Only one of the 14 sites has been located offshore (QICS), and since it is anticipated that 40% of commercial storage site capacity is located offshore [11] there is a need for more activity on this type of experimental investigation in the future.

A number of perils and pitfalls were identified from the collective experience at field experiments experiments. The main issues include leakage of $CO_2$ along the wellbore or pipeline, which in some instances require action to remediate, and disturbing the subsurface during construction of the experimental site in such a manner that these changes influence $CO_2$ spread and leakage. The importance of establishing baseline conditions for an appropriate time period cannot be underestimated, since this is important for estimating $CO_2$ impacts, fate, flux rates and total $CO_2$ leakage. Importantly, quantification of any leakage has proven difficult, despite intensive monitoring using multiple monitoring approaches at a number of the sites, and so more work is needed for any leaks to be quantified to an acceptable degree of confidence in the unlikely case of $CO_2$ leakage from an engineered store. Cost-effective approaches for doing so include remote sensing methods or mobile devices, or the use of chemical methods such as isotope tracers, and shallow $CO_2$ release experiments provide excellent opportunity to trial these methods.

## Acknowledgements

Special thanks to members of the research teams from the $CO_2$ release experiments discussed for their assistance in compiling and populating this dataset. In particular we would like to thank the following: Mike Steven, University of Nottingham (ASGARD); Maria Barrio, SINTEF (CO2 Field Lab); David Jones, British Geological Survey (ASGARD and CO2 Field Lab); Pete Taylor, Scottish Association for Marine Science, and Anna Lichtschlag, National Oceanography Centre (QICS); Jennifer Lewicki, James Thordsen, U.S. Geological Survey, and Laura Dobeck, Montana State University (ZERT); Andrew Feitz, Geoscience Australia (Ginninderra); Christophe Moni, Bioforsk (Grimsrud Farm); Julie Lions, BRGM (CIPRES); Phillippe Pezard, Montpellier University (SIMEx); Karsten H. Jensen, University of Copenhagen (Vrøgum).

We would like to acknowledge the financial support of the UK CCS Research Centre (www.ukccsrc.ac.uk) in carrying out this work by supporting Roberts' research placement at the National Geosequestration Laboratory (NGL). The UKCCSRC is funded by the EPSRC as part of the RCUK Energy Programme.

Stalker is funded by the Commonwealth Science and Industry Research Organisation (CSIRO). Roberts is funded by ClimateXChange, Scottish Government's Centre for Expertise on Climate Change, and the University of Strathclyde.

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