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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
on the collective experiences to identify common issues or complications, and so recommend ‘best practice’ guidelines for experiment design and reporting at future CO₂ release experiments.

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Keywords: CO₂ storage; monitoring; field experiments; leakage; environmental impact; CO₂ fate; experiment design.

1. Introduction

Carbon Capture and Storage (CCS) is a promising climate mitigation technology, whereby CO₂ 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 CO₂ 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 CO₂ or brines from the CO₂ store may beneficially relieve reservoir fluid pressure [3]. However unintended leakage of CO₂ 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 CO₂ 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 CO₂ stores should operate with less than 1% CO₂ loss to the surface over 1,000 years. The US Department of Energy (US DOE) aims for 99% containment of CO₂ injected for the purpose of geological storage [8], whereas the EU CCS Directive [9] requires CO₂ to remain ‘permanently’ in the storage formation. Any CO₂ 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 CO₂ storage in the US, EU and Japan or subseabed in the North Atlantic require appropriate assessment of risk of CO₂ leakage from the intended storage reservoir, the potential impacts of CO₂ 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 CO₂ itself (free phase or dissolved) and any co-injected impurities, or brines displaced as a result of pressure perturbation from CO₂ injection and migration or degraded by geochemical interaction of CO₂ 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 CO₂ 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 CO₂ leaks to be identified, attributed, and quantified (referred to as monitoring, measurement and verification, MMV, techniques). This presents challenges because CO₂ 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 CO₂ into the shallow subsurface to artificially simulate a CO₂ 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 CO₂ release is controlled, these experiments provide excellent opportunity to test methods of measuring and quantifying CO₂ 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-
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.

<table>
<thead>
<tr>
<th>Variable</th>
<th>Sub variable</th>
<th>Description</th>
<th>Units</th>
</tr>
</thead>
<tbody>
<tr>
<td>Basic descriptive information</td>
<td>Acronym</td>
<td>Project name and acronym</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Location</td>
<td>Long, lat, country.</td>
<td>degrees</td>
</tr>
<tr>
<td></td>
<td>Project aims</td>
<td>Principal research aims.</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>Release to surface intended or not.</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Funding body</td>
<td>Source of funding.</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>Funding total.</td>
<td>€</td>
</tr>
<tr>
<td></td>
<td>Project partners</td>
<td>Incl. industry and academia.</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Key contact</td>
<td>Name and contact email of Principle</td>
<td></td>
</tr>
<tr>
<td>Investigator.</td>
<td>Investigator.</td>
<td></td>
<td></td>
</tr>
<tr>
<td>-------------</td>
<td>--------------</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Project status</td>
<td>Project completed / more CO₂ releases intended.</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

### Experiment setup and site information

<table>
<thead>
<tr>
<th>Well information</th>
<th>Depth below ground surface of CO₂ injector meters</th>
</tr>
</thead>
<tbody>
<tr>
<td>Borehole type (deviated, vertical or inclined)</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Geological information</th>
<th>Properties of the intended injection formation</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Properties of any overlying rocks</td>
</tr>
<tr>
<td></td>
<td>Properties of any soils</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Ecosystem type</th>
<th>Terrestrial/marine</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>The surface ecosystem at the site.</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Hydrological characteristics</th>
<th>Water table depth and flow direction</th>
</tr>
</thead>
</table>

### CO₂ injection (for each experiment)

<table>
<thead>
<tr>
<th>CO₂ properties</th>
<th>CO₂ source</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>d13C composition %</td>
</tr>
<tr>
<td></td>
<td>Injected phase (CO₂ gas or water with dissolved CO₂)</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Injection rate</th>
<th>Steady, variable, or incremental</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Maximum (and minimum) injection rates g(CO₂)/s</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Injection periods</th>
<th>Date injection started &amp; ceased</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Total injection period days</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Quantity of CO₂ injected</th>
<th>For each experiment kg</th>
</tr>
</thead>
<tbody>
<tr>
<td>Overall</td>
<td>Number of experiments at the site</td>
</tr>
<tr>
<td></td>
<td>Total quantity of CO₂ injected kg</td>
</tr>
</tbody>
</table>

### Monitoring

<table>
<thead>
<tr>
<th>Area of monitoring</th>
<th>Area of surveillance m²</th>
</tr>
</thead>
<tbody>
<tr>
<td>Baseline</td>
<td>Baseline monitoring period days</td>
</tr>
<tr>
<td></td>
<td>What was monitored (CO₂ flux, soil gas, plant/ecosystem diversity)</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Surface</th>
<th>Lag time since injection began Hours</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Flux rate* g(CO₂)/s/m²²</td>
</tr>
<tr>
<td></td>
<td>Vertical leak velocity** m/s</td>
</tr>
<tr>
<td></td>
<td>Proportion of injected CO₂ released to surface %</td>
</tr>
<tr>
<td></td>
<td>Leakage style (patchy, uniform)</td>
</tr>
<tr>
<td></td>
<td>Patch radius*** Meters</td>
</tr>
<tr>
<td></td>
<td>Leak location (with respect to the injector) above injector / deviated</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Subsurface</th>
<th>Maximum soil gas concentrations %</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Detection time hours</td>
</tr>
<tr>
<td></td>
<td>Distribution style</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Recovery</th>
<th>Post injection monitoring period days</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Time taken to return to baseline days</td>
</tr>
</tbody>
</table>

*Where CO₂ flux (rate of CO₂ 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 CO₂ 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 CO₂ release experiments around the world. Symbols are coloured according to the CO₂ injection rates at the site, and sized proportional to the total amount of CO₂ injected over the life to date of the sites (i.e. may be the sum of multiple injections).

3.1. CO₂ 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), CO₂ Field Lab (3), Grimsrud Farm (4), Vragum (5), CO₂-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 subsea bed CO₂ 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 CO₂ (half of the experiments conducted vegetation surveys on the indigenous grasses or on planted crops).
- To establish the fluxes, transformations and fate of CO₂ as it migrates from the injection point.
- To investigate geochemical interactions between CO₂ and groundwater.
- To test and calibrate models of CO₂ 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 experiments 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
CO₂ release experiments completed at the 14 sites in the dataset, releasing a total of 82.8 t(CO₂) into the subsurface over 994 days (i.e. 2.7 years). This is not a complete list, since some preliminary experiments may have not 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 CO₂ 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 CO₂ injection (‘Max inj. rate t(CO₂)pa’). The style of the injection also varies between sites, injecting CO₂ 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.

<table>
<thead>
<tr>
<th>N</th>
<th>Name or Acronym</th>
<th>Country</th>
<th>No. of exp</th>
<th>Year</th>
<th>Inj. depth (m)</th>
<th>CO₂ phase</th>
<th>Well orientation</th>
<th>Inj. style</th>
<th>Inj. length (days)</th>
<th>Max inj. rate t(CO₂)pa</th>
<th>Surface leakage?</th>
<th>% of CO₂ leaked</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>ASGARD</td>
<td>UK</td>
<td>4</td>
<td>2006-2010</td>
<td>0.6</td>
<td>g</td>
<td>45º</td>
<td>Steady</td>
<td>14-16</td>
<td>3.1</td>
<td>Y</td>
<td>34%</td>
</tr>
<tr>
<td>2</td>
<td>QICS</td>
<td>UK</td>
<td>1</td>
<td>2012</td>
<td>12</td>
<td>g</td>
<td>h</td>
<td>Incr.</td>
<td>36</td>
<td>72.53</td>
<td>Y</td>
<td>15% as bubbles</td>
</tr>
<tr>
<td>3</td>
<td>CO2FieldLab</td>
<td>Norway</td>
<td>1</td>
<td>2011</td>
<td>20</td>
<td>g</td>
<td>45º</td>
<td>Incr</td>
<td>5</td>
<td>153.3</td>
<td>Y</td>
<td>5%</td>
</tr>
<tr>
<td>4</td>
<td>Grimsrud Farm</td>
<td>Norway</td>
<td>4</td>
<td>2012</td>
<td>0.85</td>
<td>g</td>
<td>h</td>
<td>Steady</td>
<td>75</td>
<td>1.93</td>
<td>Y</td>
<td>82%</td>
</tr>
<tr>
<td>5</td>
<td>Vrøgum</td>
<td>Denmark</td>
<td>6</td>
<td>2012</td>
<td>5-10</td>
<td>g</td>
<td>45º</td>
<td>Incr</td>
<td>2-72</td>
<td>4-3-10.51</td>
<td>N</td>
<td>30-40% imaged</td>
</tr>
<tr>
<td>6</td>
<td>CO2DEMO</td>
<td>France</td>
<td>2</td>
<td>2010-2014</td>
<td>3.7</td>
<td>g</td>
<td>v</td>
<td>Steady</td>
<td>&lt;1</td>
<td>3.06</td>
<td>Y</td>
<td>78%</td>
</tr>
<tr>
<td>7</td>
<td>CIPRES</td>
<td>France</td>
<td>2</td>
<td>2013</td>
<td>25</td>
<td>diss</td>
<td>v</td>
<td>Steady</td>
<td>2</td>
<td>4.38</td>
<td>N</td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>SIMEx</td>
<td>France</td>
<td>1</td>
<td>2013</td>
<td>13-16</td>
<td>g</td>
<td>v</td>
<td>Var.</td>
<td>0.1</td>
<td>550.6</td>
<td>N</td>
<td></td>
</tr>
<tr>
<td>9</td>
<td>Brandenburg</td>
<td>Germany</td>
<td>1</td>
<td>2011</td>
<td>18</td>
<td>g</td>
<td>v</td>
<td>Steady</td>
<td>0</td>
<td>0</td>
<td>N</td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>PISCO2</td>
<td>Spain</td>
<td>1</td>
<td>2012</td>
<td>1.6</td>
<td>g</td>
<td>h</td>
<td>Steady</td>
<td>46</td>
<td>0.96</td>
<td>Y</td>
<td>82.3%</td>
</tr>
<tr>
<td>11</td>
<td>Ginninderra</td>
<td>Australia</td>
<td>5</td>
<td>2010</td>
<td>2</td>
<td>g</td>
<td>h</td>
<td>Incr/steady</td>
<td>56-80</td>
<td>21.8-79.6</td>
<td>Y</td>
<td></td>
</tr>
<tr>
<td>12</td>
<td>Ressacada Farm</td>
<td>Brazil</td>
<td>1</td>
<td>2013</td>
<td>3</td>
<td>g</td>
<td>v</td>
<td>Incr</td>
<td>12</td>
<td>1.31</td>
<td>Y</td>
<td></td>
</tr>
<tr>
<td>13</td>
<td>ZERT*</td>
<td>USA</td>
<td>5</td>
<td>2007-2014</td>
<td>1.1-2.5</td>
<td>g</td>
<td>h</td>
<td>Steady/var.</td>
<td>7-10</td>
<td>0.95-110.4</td>
<td>Y</td>
<td>90%</td>
</tr>
<tr>
<td>14</td>
<td>Brackenridge</td>
<td>USA</td>
<td>2</td>
<td>2011-2012</td>
<td>6</td>
<td>d</td>
<td>v</td>
<td>Steady</td>
<td>2</td>
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*An experiment was conducted at ZERT in Autumn 2006 where CO₂ 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 horizon.

3.2. CO₂ release experiments: how?

The site characteristics and experimental set-up of the CO₂ 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 CO₂ except CO₂-DEMO where a gas mixture of 90.57% CO₂, 5% Kr and 5% He was released [18]. CO₂ was often delivered by a borehole which was either horizontal (favoured at the shallower sites), at a 45º 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 CO₂ 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 Vrangum 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 seasonally. 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\textsubscript{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\textsubscript{2} release projects worldwide. Projects show a range of CO\textsubscript{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\textsubscript{2} gas was released into the subsurface at all but two of the field experiments; CIPREs and Brackenridge injected CO\textsubscript{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\textsubscript{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\textsubscript{2} injection at these experiments; the highest rate, CO\textsubscript{2} Field Lab is 4.9 g s\textsuperscript{-1} (153.3 t(CO\textsubscript{2}) pa) and the smallest injection rates were at PISCO2 and Brackenridge, as 0.03, 0.04 g s\textsuperscript{-1} respectively (0.95 and 1.3 t(CO\textsubscript{2})pa). The majority of experiments however inject CO\textsubscript{2} between 0.05 and 2 g s\textsuperscript{-1}, which is equivalent to 1.6 - 63 t(CO\textsubscript{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\textsubscript{2})pa for 40 years, and are, these values are in the range of natural CO\textsubscript{2} emissions, for example, in Italy where there are hundreds of CO\textsubscript{2} seeps that most commonly emit between 10-100 t(CO\textsubscript{2})pa [30]

The length of CO\textsubscript{2} injection at the experiments we reviewed varies from a couple of hours to several months. Most experiments inject CO\textsubscript{2} for periods shorter than 1 month, and indeed, the longest injection period at five of the sites (CO\textsubscript{2} Field Lab, CO\textsubscript{2}-Demo, Brackenridge, CIPRES, and SIMEx) lasted 5 days or less. This was not long enough for CO\textsubscript{2} leakage to reach steady state at CO\textsubscript{2} 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\textsubscript{2} injected for the longest periods. It is concluded by QICS researchers that the CO\textsubscript{2} release period (37 days) was not long enough, since the effect of CO\textsubscript{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 \(^{13}\)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 \(^{13}\)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 \(^{13}\)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 Vrågum 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\(^{-2}\)day\(^{-1}\) 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 streams might be interpreted to occur in two patches or 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 molettes 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 CO₂ 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 CO₂, which is denser than air, to accumulate in the subsurface and so limiting CO₂ release to surface (see Schroder [46] of this conference). By contrast, experiments at other sites, such as ASGARD found CO₂ flux was impeded by rainfall events, although the difference in total CO₂ 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 CO₂ 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 CO₂ migration - even when the injection depth is shallow. For example, at Ressacada Farm the hotspot was located ~30 m from the CO₂ 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 CO₂ bubble streams were located maximum of 10 m west of the CO₂ 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 CO₂ 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 CO₂ pipeline [22], the CO₂ 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 CO₂ 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 CO₂ 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 CO₂ flow pathways quickly developed where injection and pumping tests performed prior to CO₂ 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 treading, 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 CO₂ 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 CO₂ leakage

At most onshore experiments the distribution of CO₂ 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 CO₂ can be detected by changes to groundwater chemistry, also collected via shallow boreholes or deeper monitoring wells.

At all sites that released CO₂ to surface, the extent of lateral spread of CO₂ in the subsurface was found to be greater than the area of surface degassing, CO₂ 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 CO₂ moved preferentially through the more-permeable sandy and gravely deposits lying below the injection point [49]. Though there was no CO₂ flux to surface above the injection point at CO2 Field Lab, soil gas monitoring detected subsurface CO₂ in that region shortly after injected began, as CO₂ 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 CO₂ gas moved through the aquifer, partially displacing the water in the pore space) were consistent with CO₂ 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 CO₂ flux was more distributed [23]. These
surveys, aided by krypton tracer, found that in the wet season CO\textsubscript{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\textsubscript{2} is injected into the saturated zone, CO\textsubscript{2} spreads further but gas intensity is more localised, whereas when CO\textsubscript{2} is released into the vadose zone CO\textsubscript{2} transport is more localised but gas intensity is more distributed.

Monitoring techniques deployed at Vrøgum found CO\textsubscript{2} favoured flow in more permeable rock formations; geochemical affects of CO\textsubscript{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\textsubscript{2}-DEMO project [18]. Interestingly, at QICS, repeat seismic reflection surveys found that, like at CO2 Field Lab, sub-surface CO\textsubscript{2} flow path was affected by sediment structure and also CO\textsubscript{2} flow rate [39, 47]. Sediment grain size controlled CO\textsubscript{2} flow initially until the gas pressure or gas volumes overrode the stratigraphic controls, spatially focussing the CO\textsubscript{2} flow via the formation of chimney structures [47].

All the experiments found that CO\textsubscript{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 subs seabed injector, whereas the spatial extent of the geochemical impact of the injected CO\textsubscript{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\textsubscript{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\textsubscript{2} flows in discrete channels to start with, which limits the contact with the water-phase, and so restricting the amount of CO\textsubscript{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\textsubscript{2} release, CO\textsubscript{2} can flow against groundwater direction, as observed at Vrøgum where the CO\textsubscript{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\textsubscript{2} leakage

Quantifying the proportion of injected CO\textsubscript{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\textsubscript{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\textsubscript{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\textsubscript{2} flux. In some cases, for example at ASGARD and ZERT, CO\textsubscript{2} migrated beyond the monitoring boundaries, making it difficult to estimate the relative proportions of CO\textsubscript{2} that leaked to surface or remained in soil gas or dissolved [40, 49].

The proportion of CO\textsubscript{2} that leaks to surface is observed to vary throughout the experiment duration, as various environmental factors affect CO\textsubscript{2} flux. For example, at Ginninderra, the proportion of CO\textsubscript{2} leaked to surface was higher in the wet season than the dry season (Feitz, pers. comm), whereas CO\textsubscript{2} flux at ASGARD was greatest in drier spells [57]. At QICS, 8-15% of injected CO\textsubscript{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\textsubscript{2} volume as free phase gas trapped in the sediment. The remaining CO\textsubscript{2} must have dissolved or migrated [54].
The experiments that found the smallest proportion of CO₂ leaked to surface/seabed as a gas also had the greatest injection depth (CO₂ Field Lab and QICS) find that the smallest proportion of CO₂ leaked to surface. It is therefore tempting to suggest a weak relationship between these factors, shown in Fig. 6a, however the leakage estimate at CO₂ 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 CO₂ 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 CO₂ in the subsurface, perhaps because vertical spread is restricted by the soil properties.

Fig. 6: The estimated proportion of injected CO₂ 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 (CO₂ Field Lab and QICS) find that the smallest proportion of CO₂ leaked to surface, however the leakage estimate at CO₂ 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 CO₂ have been proposed as a potential method of quantifying CO₂ leakage rate [59, 60]. Several experiments have tested added tracers, including PFCs (ZERT), SF₆ (Brandenburg), noble gases (Krypton, Ginninderra; Helium and Argon, CO₂-DEMO), and future experiments at several sites also plan to use tracers. However no release experiment has yet attempted to quantify CO₂ leakage using chemical tracers. For tracers to quantify CO₂ 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 CO₂ [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 CO₂ hotspot is determined; the outer limit of the hotspot is usually defined by CO₂ 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 CO₂ 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 CO₂ leakage. The CO₂ Field Lab and CO₂-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 CO₂) and at Ressacada Farm experiment leakage occurred at a surface depression [13]. Topography is observed to influence the characteristics of natural CO₂ seeps due to corresponding changes in depth to the water table [61], and also because topographic depressions are sheltered and so CO₂ dispersion can be limited, encouraging gravity-driven CO₂ 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 CO₂ dispersion, which can also largely affect CO₂ measurements. Most currently operating onshore CO₂ storage projects (e.g. Quest, Boundary Dam, 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 CO₂ and tracers for CO₂ should be explored further. Similarly work should continue to characterise the effect of topography and infrastructure on wind speeds and CO₂ 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, CO₂-DEMO, released CO₂ into lithified rock. Although we only analysed experiments injecting CO₂ 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 CO₂ to reach surface. It is tempting to recommend that future experiments release CO₂ 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 CO₂ fate and spread and with the intention that CO₂ will leak to surface. However, experience at CO₂-Vadose and Vrøgum found that layers low permeability units such as fine sands or clays above the injector can prevent CO₂ 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 CO₂ 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 CO₂ release experiments, with intent to release CO₂ to surface, would increase the cost and risk of the CO₂ release experiment, since deeper wells are more expensive to drill, and greater depths increase the risk that CO₂ will not reach to surface. CO₂ might have to be injected for a long time period and possibly in considerable quantities to allow the CO₂ plume to migrate distances that might allow the sealing horizon to be bypassed and so enable CO₂ leakage to surface – if at all. These scenarios might be useful to explore in future, longitudinal experiments.

5. Conclusions

Field-scale shallow CO₂ 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 CO₂ flow pathways, CO₂ impacts, and methods of detecting CO₂ 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 CO₂ release projects, at which a total of 42 different CO₂ 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 CO₂ over 994 days. Nine of the CO₂ release facilities intended that the injected CO₂ be released to surface, the remaining experiments intended for CO₂ to remain in the shallow subsurface (usually to investigate groundwater interactions). The experiments show a range of test approaches, including CO₂ 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 CO₂ 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 CO₂ spread and leakage. The importance of establishing baseline conditions for an appropriate time period cannot be underestimated, since this is important for estimating CO₂ impacts, fate, flux rates and total CO₂ 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 CO₂ 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 CO₂ release experiments provide excellent opportunity to trial these methods.

Acknowledgements

Special thanks to members of the research teams from the CO₂ 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 Dobbeck, 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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