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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

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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.

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).
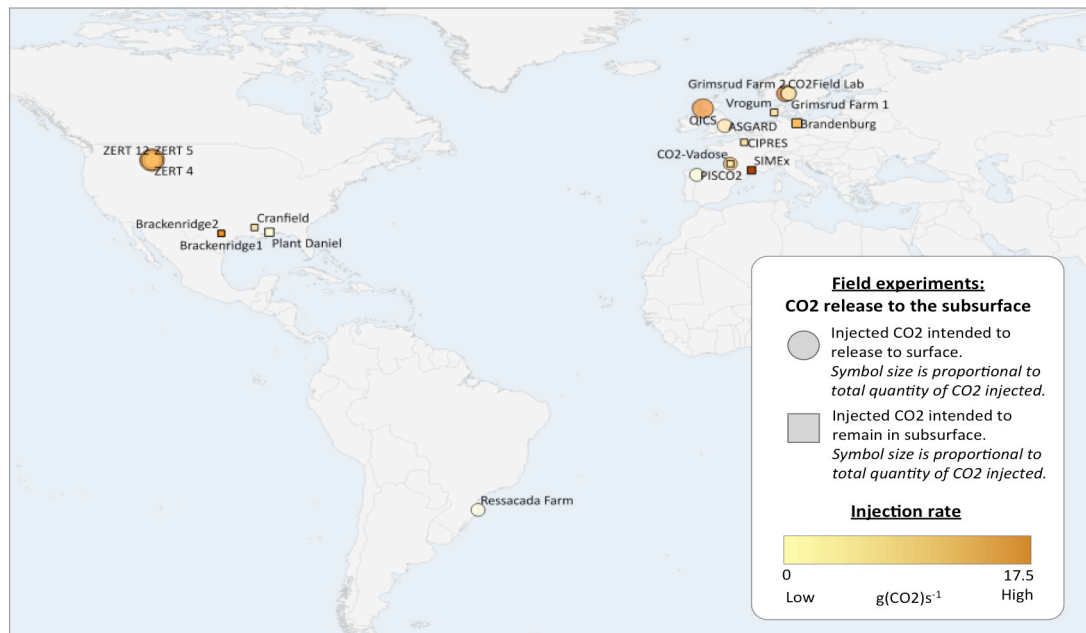

3.1. CO_2 release experiments: Where, when and why?

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).

Quantity of injected CO_2 and injection rate for experiments conducted each field site.
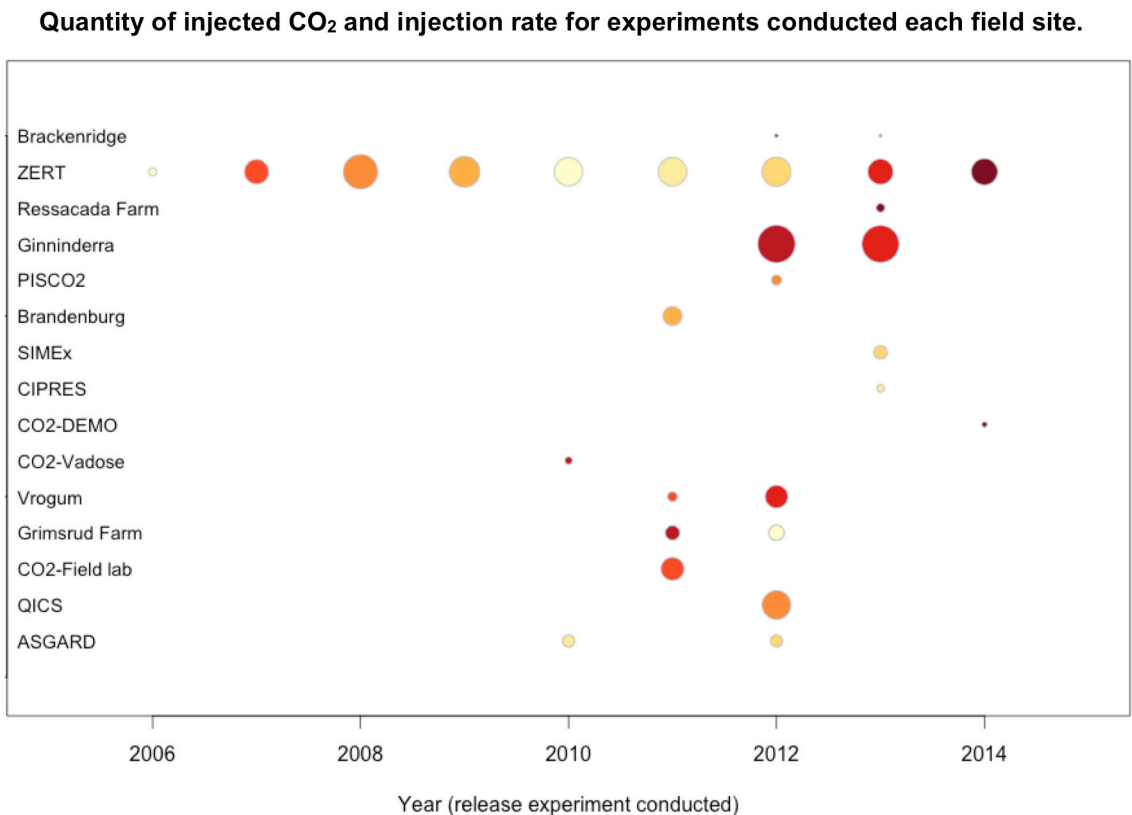
At the majority of the CO_2 release experiments, free-phase CO_2 gas was injected. These experiments commonly sought to mimic the effects of leakage of CO_2 gas, either from a point source or vertical feature (such as from poorly sealed well casing), or a linear feature (such as from a fault) – though 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.
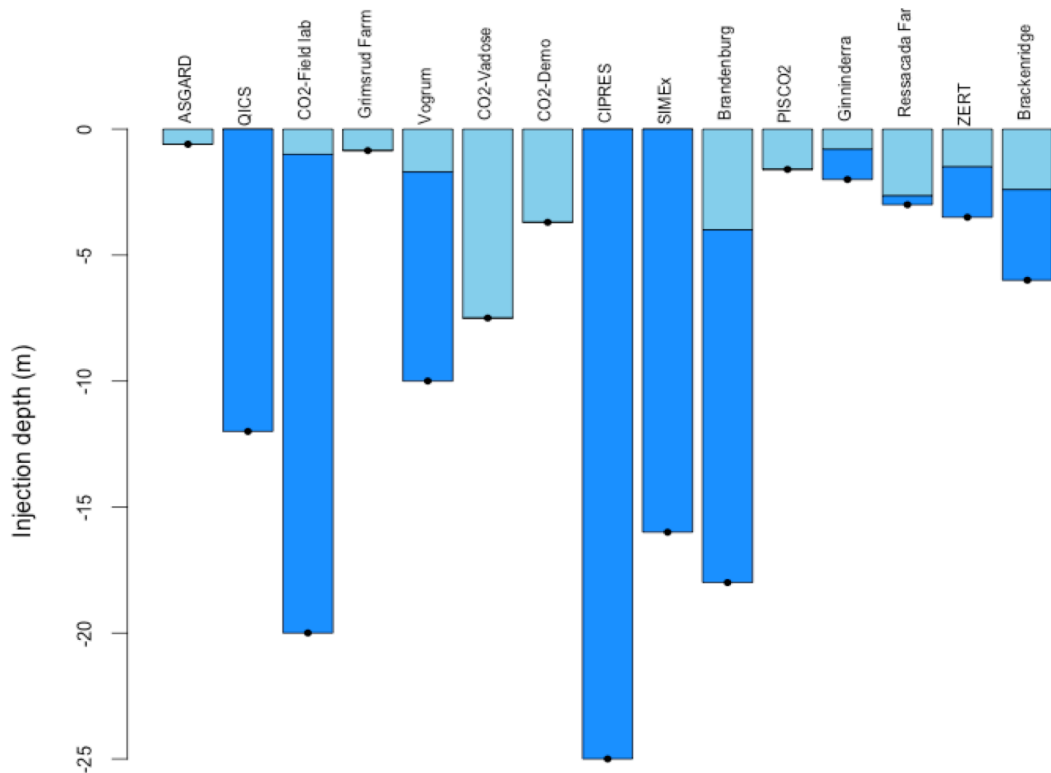
The depth of CO_2 injection ranges from 0.6 m (ASGARD) to 25 m (CIPRES). The deepest experiment to release CO_2 to surface is CO2 Field Lab at 20 m depth below surface. Fig. 3 shows the injector depth and the maximum depth of the water table by experimental site. Most experiments released CO_2 into sands 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.
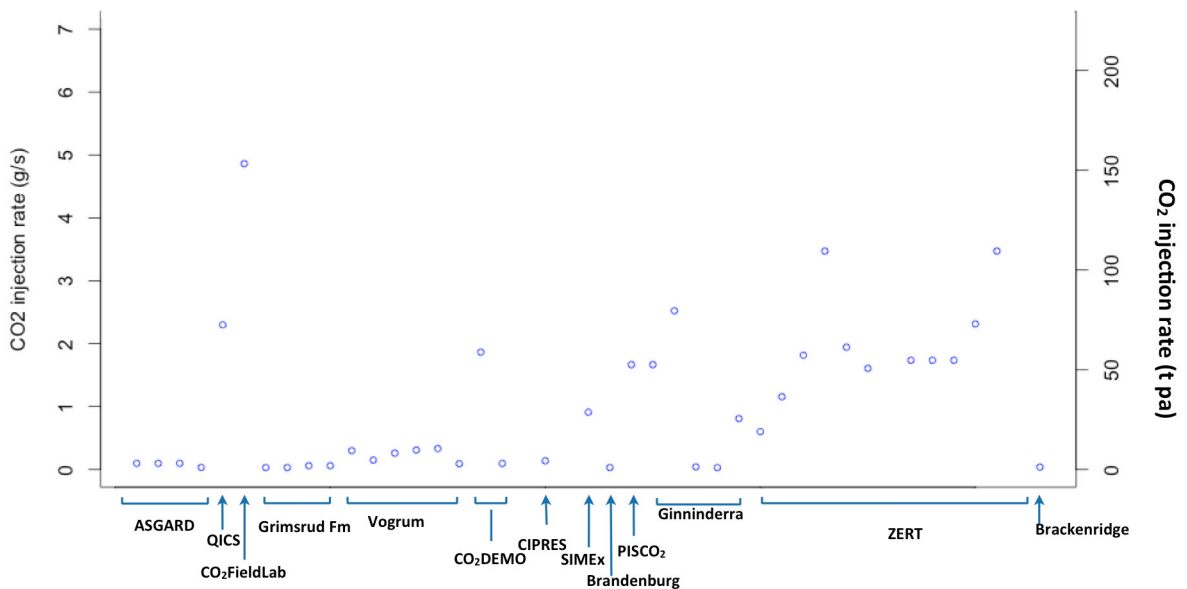
Free-phase CO_2 gas was released into the subsurface at all but two of the field experiments; CIPREs and Brackenridge injected CO_2-saturated water. As shown in Table 2 a steady or incrementally increasing injection strategy is favoured by most experiments at the site, though at SIMEx the rate varied 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.
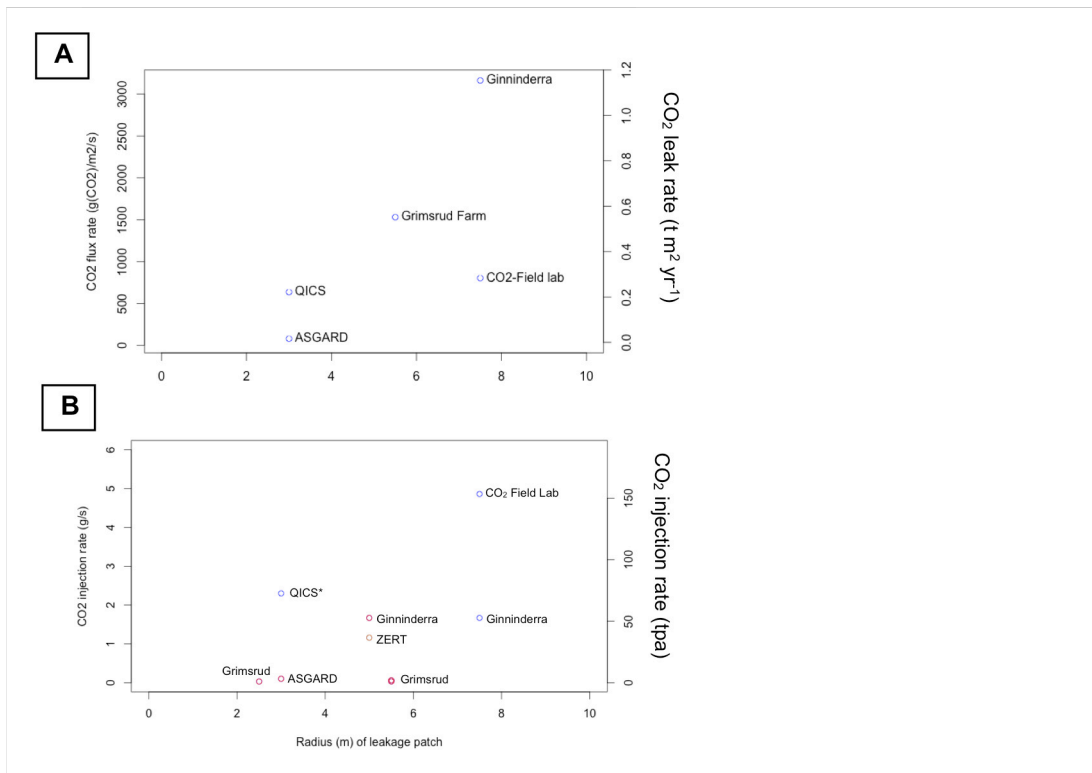
We note that there may be a correlation between the hotspot radius and the CO_2 injection rate, as shown in Fig. 5a, where the hotspot radius is larger at higher injection rates, however we find no relationship between injection rate and maximum CO_2 flux rate (graph not shown here). Both the wet-season 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].
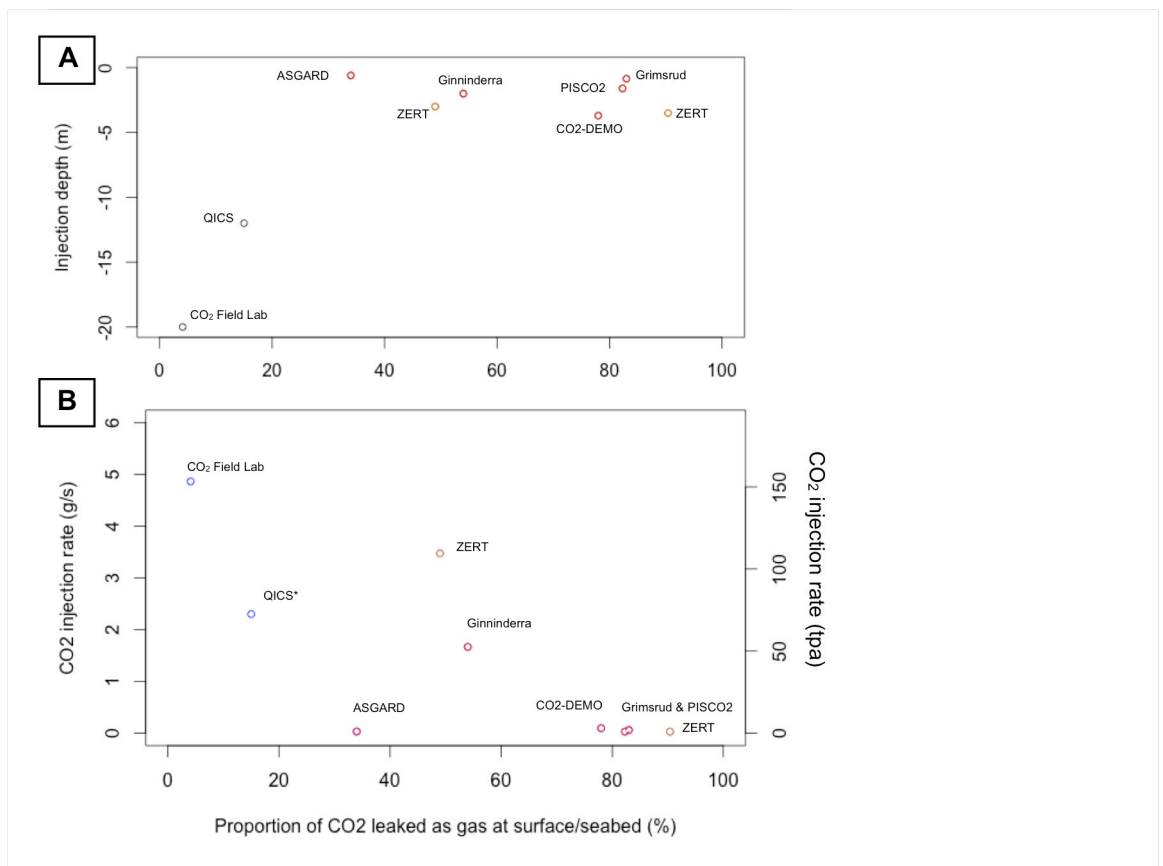
3.5. Quantifying CO₂ leakage

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 CO_2 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

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