Supplementary Information for "Estimating geological CO² storage security to deliver on climate mitigation", by Alcalde, Flude et al. (2018)

Contents

Supplementary Note 1: Overview of the Storage Security Calculator (SSC)

This Supplementary Information document describes how we derived the model inputs (Supplementary notes 2 to 9) along with the rationale for the values selected, and the structure of the computational program. A summary of the inputs is provided in the methods section of the main manuscript. Details of the sub-models and of the integrated model (Supplementary Notes 10-12) are also described to aid the reader in understanding the program.

This document is designed to be used as a reference text rather than read cover-to-cover, and thus involves a certain amount of repetition of information, in order to be user-friendly. Figure 6 of the main text shows how the different sections of the program interact, along with the sections of this document that discuss the relevant input parameters.

The SSC combines an immobilisation model (1), comprised of residual trapping (1a) and chemical trapping (1b) sub-models; and a leakage model (2), comprised of active well (2a), abandoned well (2b) and natural pathways (2c) leakage sub-models.

Three storage environment scenarios are explored by the SSC:

1) Scenario A: "Offshore Well-Regulated" - Storage in an offshore environment that takes advantage of a mature, well-regulated hydrocarbon industry in the region.

2) Scenario B: "Onshore Well-Regulated" - Storage in an onshore environment that takes advantage of a mature, well-regulated hydrocarbon industry in the region.

3) Scenario C: "Onshore Poorly-Regulated" - Storage in an onshore environment that has experienced an extensive, but historically poorly-regulated hydrocarbon industry in the region.

The required information to characterise these scenarios was obtained from certain geographical regions that could be described as examples of these scenarios. These include the North Sea (Scenario A), Texas, USA (Scenario B), and Pennsylvania, USA (Scenario C). Scenarios A and B are the most likely targets for CO_2 storage sites, providing a balance of the benefits of existing knowledge with the increased potential risk of leakage pathways due to legacy wellbores; but we include Scenario C as a worst-case scenario to investigate $CO₂$ storage security in the event of sub-optimal regulation.

In Section 2 we discuss how values were derived for each model input parameter. Each parameter has a maximum and minimum value and a base case value, which we consider to be a reasonable and likely, albeit often conservative estimate. These parameters are used to calculate the base case scenarios. For a sensitivity analysis we employ a Monte Carlo method to select random values for each parameter, from within a defined range of values or distribution, and run the model 10,000 times. These ranges and distributions are based on real data, where possible, and are described on a case-by-case basis in Section 2.

For parameters that describe average values (e.g. the amount of $CO₂$ leaked per well), the Monte Carlo analysis picks a random number from a distribution defined by the mean and the standard error (=σ/sqrt(*n*), where "*n*" is defined either as the number of observations in the data set that the distribution is based on, or as the minimum number of virtual samples that will be returned by the model). This ensures that the random values are statistically representative and do not return an extreme value with very low probability. For example, continuous leakage in active wells is expected to result in loss of between 102 and 215 t of $CO₂$ per year from each leaking well. Continuous leakage is calculated from the frequency (i.e. % of wells that leak) and the average amount of $CO₂$ leaked per leaking well. Random sampling of a normal distribution that fits the minimum and maximum values for amount leaked per leaking well, defined by a mean and standard deviation, would produce leakage values equivalent to a single well, rather than an average of multiple wells, and would result in over-representation of extreme values. Random

selection of the high value (215 t $CO₂$ year⁻¹) would imply that every single leaking well leaks the maximum amount of $CO₂$, which is very unlikely. To avoid this, where parameters are average values, we define the distribution based on the mean and the standard error, instead of the mean and standard deviation.

Supplementary Note 2: General Parameter Definitions

General parameters comprise our basic assumptions, including injection targets, injection rates, and the areal extent of the subsurface $CO₂$ plume. These are parameters that influence all three models.

2.1 Injection Targets

In 2013, the IEA published estimates of the amount of $CO₂$ required to be stored by 2050, to meet the IEA's 2 °C (2DS) scenario, in which there is an 80% chance of limiting average global temperature increase to 2 $^{\circ}$ C ¹. Regional 2050 storage targets range from 3.5 Gt for non-OECD Latin America, up to 42.2 Gt for China, with a total of over 120 Gt CO stored globally by 2050.

Here, we apply the SSC to regional scenarios and we select a 2050 storage target of 12 Gt CO₂, in keeping with the storage target of OECD Europe. However, we note that the nature of the calculations employed by the SSC scales the leakage impact according to the amount of $CO₂$ injected, and so changing the storage target makes no difference to the proportion of $CO₂$ leaked. To achieve this target, we make a simplistic assumption that full CCS chain will be in place and operational by 2020 and that injection will take place over 30 years, from 2020 to 2050. Injection is assumed to take place gradually; programmatically this involves addition of $1/30$ of the $CO₂$ target per year (i.e., 400 Mt year⁻¹) to the reservoir.

2.2 Injection rate and number of injection wells

Assuming that adequate storage capacity is available and that the injection targets will be met, the number of injection wells needed will depend on the injectivity of each well.

There are three main sources of data regarding $CO₂$ injectivity: enhanced oil recovery with $CO₂$ $(CO₂-EOR)$, pilot $CO₂$ storage projects and commercial $CO₂$ storage projects. Pilot storage projects often inject small volumes, verifying injectivity before scaling up to larger projects. As such, their associated injection rates are lower than those likely to be applied at commercial storage sites; therefore, we do not consider pilot $CO₂$ storage site injection rates to be representative of a CCS industry implemented at a full, global-scale. At commercial CO2-EOR injection sites, large tonnages of $CO₂$ may be injected, but these are often spread over a large number of wells for operational reasons and so do not necessarily represent optimised single-well injection rates. For instance, initial injection rates at Weyburn were up to 5000 t of $CO₂$ per day, equivalent to 1.8 Mtpa², but this was spread over nine injection wells² to optimise $CO₂$ contact with the residual oil. Hence, these existing injections are likely to be lower than those used in commercial storage sites.

Our injection rate parameter is based on measured $CO₂$ injection rates from commercial-scale storage projects. $CO₂$ injection rates are available for six commercial scale $CO₂$ storage sites (Supplementary Table 2) and range from 0.2 to 1.1 Mtpa. We assume that $CO₂$ storage sites will be designed to optimise injectivity, and we adopt a range of injection rates from 0.5 to 1 Mtpa, with a base case value of 0.75 Mtpa.

The number of injection wells needed to achieve the 12 Gt injection target is calculated based on the storage target, the injection period, and the injectivity. Our injection target is a constant 12 Gt $CO₂$ over 30 years. This equates to 400 Mt $CO₂$ per year, which gives a base case scenario requirement of 533.3 injection wells.

For the sensitivity analysis, we assume that well injectivity can be described by a normal distribution with a mean of 0.75 Mtpa (the base case value) and where the maximum (1 Mtpa) and minimum (0.5 Mtpa) values represent 3 standard deviations from the mean. This gives a standard deviation of 0.083.

The injection rate parameter is used to calculate an average injectivity value for hundreds of wells, and so we use the mean and standard error to create a distribution for the Monte Carlo analysis. To ensure we are not under-estimating the errors, we assume the smallest value of *n* that will be sampled by our program. In this case, *n* represents by the number of injection wells, which is defined by the injectivity. The highest injectivity (1 Mtpa) will result in the smallest number of wells, giving $n=400$. The standard error is thus 5.21×10^{-7} .

2.3 Injection Plume Area

Within the SSC program, the areal extent of the injection plume has a significant impact on leakage from abandoned wells, and via natural pathways, because the plume area will determine how many potentially leaking structures (e.g. abandoned wells or open faults) will be contacted by the injected CO2. Plume area will depend on the geometry of structural traps and the mass of $CO₂$ injected. There are not yet enough large-scale $CO₂$ storage projects to assess the likely area impacted by a CO_2 plume during CO_2 storage, and so we use natural gas fields as an analogy. We use the data listed in Appendix 1 of Gluyas and Hitchens³ and concentrate on fields for which area, recoverable gas volume, and gas expansion factor data are available. We use the recoverable gas and the gas expansion factor data to calculate the volume of gas in each reservoir. We then assume an in-reservoir CO_2 density of 700 kg m⁻³ to calculate the equivalent mass of CO_2 for each field. We then divide the field area by the mass of $CO₂$ to obtain an area-to-mass ratio. Data for individual fields are displayed in Supplementary Table 3. A histogram of the area-to-mass ratio values is displayed in Supplementary Figure 1. The data do exhibit a normal distribution, but can be interpreted as an incomplete lognormal distribution where the natural logs of the data have a mean of -0.7595 ± 0.8815 (one standard deviation), based on 25 data points. This gives a standard error on the mean of the logged data of 0.1763. We thus use this mean (i.e. $e^{-0.7595}$ km² / Mt) as the base case, and use the standard error (i.e. $e^{0.1763}$) for sensitivity analysis.

Measured and modelled $CO₂$ injection rates per well.

Supplementary Figure 1: Area-to-mass ratios of natural gas fields

Histograms of area-to-mass ratios of natural gas fields (grey boxes) and fitted lognormal distributions (blue lines). Black vertical lines represent the minimum, mean, and maximum values.

Supplementary Note 3: Leakage through Active (Injection) Wells

3.1 Background on well leakage

Wells, both active and abandoned, present pathways for the leakage of $CO₂$ from geological storage. Wells developed for production have a steel casing inserted which is sealed in place with cement ¹³ (Supplementary Figure 2), while wells abandoned at the exploration stage consist of a simple well bore through rock and may or may not be sealed by a series of cement plugs. Well blowouts are rare but significant events that transfer volumes of fluids from geological depth to the surface, and can occur through both active (injection and / or production) and abandoned wells. Less dramatic, low-seepage rate leaks associated with wells may also occur. Numerous potential leakage pathways exist for wells^{14,15} (Supplementary Figure 2), although modern well design incorporates numerous blowout prevention mechanisms to mitigate and control any unplanned fluid flow¹⁶. In the case of $CO₂$ storage, considerable attention has been paid to longterm well integrity as $CO₂$ -associated corrosion of steel and cement casing has been cited as a cause of blowouts and well failure^{17,18}.

However, laboratory studies of well casing materials and samples taken from decades-old $CO₂$ injection wells suggest that corrosion may precipitate as well as erode material, and does not always lead to enhanced permeability¹⁹⁻³⁰. Steel corrosion can be minimised in newly commissioned wells by the use of corrosion resistant carbon steel and by good cementing practice²³, but corrosion may present a significant leakage risk that increases over time in preexisting wells. When present, cement corrosion most often exploits pre-existing cracks and defects, thus modifying pre-existing pathways such as a poor seal between the wellbore wall-rock and the casing cement²⁰ (potential leakage pathway $#3$ in Supplementary Figure 2). The risk of poor cement seals can be minimised by good, modern industrial practice.

We assume that any wells drilled for the application of CCS will employ best-practice and operate in a well-regulated industry and hence above average failure rates are not expected. We also assume that best practice will involve installation of blowout prevention and control equipment and training for well operation personnel¹⁸. Pre-existing legacy (presumed abandoned, for simplicity) wells within a CCS site may have been drilled, completed, and abandoned without adequate regulation and using materials less suitable for $CO₂$ -bearing fluids. Hence, these wells are expected to have a greater than average risk of leakage, that will vary by geographical region (the different levels of leakage risk associated with regions with differing industrial histories are assessed by comparing our Scenarios 2 and 3). However, we also assume that $CO₂$ storage site operators will have a duty to monitor and remediate any leakage on pre-existing wells and that any significant leakage will be mitigated on timescales typical of dealing with an active well blowout.

We have reviewed the scientific and agency literature and databases for frequency of leakage and for leakage rates or total amount of material leaked to estimate expected leakage from active and abandoned wells. Leakage frequency is reported in the literature as a range of units and scales, including incident rates within an entire industry, incidents or leakage from a single site or field, as incidents per well or as wells per site. Industry-wide and site-scale data are reviewed for context, but such data are only included in our calculations where it is possible to convert the data to incidents per well per year for discrete events, or proportion of leaking wells for continuous leakage. When estimating the amount of $CO₂$ expected to leak, data for gas leakages (instead of leakage of other fluids) are considered the most appropriate. The amount of material leaked is collected as a volume value where available, or converted to a volume assuming standard

temperature and pressure ("STP" – 0° C, 1 atm), if appropriate. We report volumes in the units quoted in the source publication and convert these to cubic meters $(m³)$. These volumes are then converted to an equivalent mass of $CO₂$ (tonnes, t, or megatonnes, Mt, as appropriate) assuming a pure, ideal gas at standard temperature and pressure. This approach (calculating equivalent mass of $CO₂$) differs from other similar studies such as that of Loizzo et al³¹ who convert reported leakage volumes to masses of the gas leaked in the case study (e.g. CH4).

The data used to derive leakage estimates for these wells was extracted from a range of sources. The data from the underground gas storage (UGS) industry are the most pertinent as they describe scenarios of injecting gas into geological reservoirs for storage. Unconventional hydrocarbon production (hydraulic fracturing/EOR) often involves injection of fluids (including $CO₂$) into the reservoir and is also relevant, but the available well-failure data of hydrocarbon production often does not distinguish between conventional and unconventional operation wells. Furthermore, data relating to blowout or leakage frequency do not always distinguish between gas and oil production.

In terms of leakage from wells, two modes of leakage can be distinguished: 1) continuous, low level leakage of gas that is not considered to pose an acute hazard to operational staff or the environment; such leaks are monitored but, in traditional hydrocarbon industry practice, may not be remediated until the well is abandoned; 2) acute events involving unplanned and uncontrolled release of fluid (gas / oil / water) from the well (blowouts) that require rapid assessment and remediation as soon as possible. These two leakage modes will be reviewed separately.

Supplementary Figure 2: Leakage pathways along wellbores

Leakage pathways along plugged and abandoned wellbores. 1: Corrosion or fracture of the inwell cement plug. 2: Poor contact between the stainless steel well casing and the cement plug. 3: Poor contact between the wall rock of the well bore and the casing cement. 4: Poor contact between the casing cement and the stainless steel casing. 4: Fracture or corrosion of casing cement. 6: Corrosion of the stainless steel well casing. Leakage pathways for active wells are similar and include pathways 3-6 with additional possibility of unintended gas flow back up the wellbore (blowout) during operation. Modified from ³².

3.2 Continuous Leakage from active (injection) wells

Minor continuous leakage events are considered to occur at wells that develop sustained casing pressure (SCP), sustained casing vent flow (SCVF) or small leaks, where the flow rate is low enough to not require remediation. Bachu and Watson³³ suggest that gas leaking during SCP is derived from shallower horizons than the target injection reservoirs, and so such leakage rates may not be relevant for our calculations. However, deep sources of gas have been identified as the source of SCP in some cases³⁴. A reasonable amount of information regarding slow leakage of hydrocarbon production wells is available, but less so for injection wells. Bachu and Watson³³ noted the occurrence of SCVF on CO₂ injectors in Alberta, but these were documented as discrete events, rather than continuous leakage, and were quickly remediated. Rather than assume that an absence of evidence is evidence of absence, we review the frequency and level of continuous leakage in hydrocarbon production wells and incorporate this data into our models. This allows us to assess the impact of slow leaks from injection wells on overall $CO₂$ storage security. Supplementary Table 4 lists data on hydrocarbon wells that have reported leaks, casing failures, or SCP, in nineteen different studies, for onshore and offshore fields, along with the minimum, maximum and average proportion of wells that are leaking. The data suggest that offshore wells have a higher leakage risk (see histogram in Supplementary Figure 13). This is unsurprising given the added difficulties of cementing wells in offshore environments compared to onshore. We thus select different leakage frequencies for onshore and offshore environments.

Offshore well leakage frequencies range from 0.02 (2%) to 0.6 (60%), with a mean of 0.145 (14.5 %). However, these data do not form a normal distribution (Supplementary Figure 3), and are better described by a lognormal distribution with a mean of the natural logs of -2.17 ± 0.6 (one standard deviation). This distribution produces a reasonable fit for both the logged and original values (Supplementary Figure 3). We use this lognormal mean (i.e. $e^{-2.17}$) as the base case value.

Onshore well leakage frequencies range from 0.013 (1.3%) to 0.22 (22%), with a mean of 0.075 (7.5%). As for the offshore wells, we describe the data as a lognormal distribution with a mean of the natural logs of -2.89 ± 0.7 (one standard deviation). This distribution produces a reasonable fit for both the logged and original values (Supplementary Figure 4), and we use the mean (i.e. e-2.89) as the base case value.

A comprehensive survey on gas leakage through well cement documented a range of leak rates from $<$ 5000 cf (142 m³) to > 250,000 cf (7080 m³) per day³⁵, while other documented leak rates associated with non-hazardous SCP are on the order of 142 m^3 (5000 cf)¹⁹ to 200 m^{3 36} per day (see Supplementary Table 5). The lower estimate seems to be \sim 5000 cf per day, which equates to 102 t $CO₂$ year⁻¹. The upper limit for allowable leakage without the need for well reparation in Alberta is 300 m³ per day, which equates to 215 t $CO₂$ year⁻¹. The mid-point of these two values is 158.5 t $CO₂$ year⁻¹, which we take as the base case.

For the sensitivity analysis, we assume a normal distribution with a mean of 158.5 t CO₂ year⁻¹ (the base case value) and where the maximum (102 t CO_2 year⁻¹) and minimum (215 t CO_2 year⁻¹) ¹) values represent three standard deviations from the mean. This gives a standard deviation of 18.83. To calculate the standard error, we calculate the lowest number of wells likely to be leaking by multiplying the lowest leak frequency (1.3% - onshore leakage) by the smallest number of injection wells (400 wells at the maximum 1 Mt year⁻¹ injection rate) to give a standard error of 5.2.

Supplementary Figure 3: Leakage frequency for offshore injection wells.

Offshore injection well leakage frequency. Histograms (grey boxes) and fitted lognormal distributions (blue lines) for the percentage of injection wells that exhibit continuous leakage. Data from Supplementary Table 4).

Supplementary Figure 4: Leakage frequency for onshore injection wells

Onshore injection well leakage frequency. Histograms (grey boxes) and fitted lognormal distributions (blue lines) for the percentage of injection wells that exhibit continuous leakage. Data from Supplementary Table 4.

3.3 Leakage via discrete events from active wells (blowouts)

3.3.1 Sources of data

Uncontrolled release of fluids (blowouts) from a well are rare but ubiquitous events in the hydrocarbon industry. Many blowout incidents are associated with exploratory drilling and well completion¹⁶; the frequency of blowouts associated with exploratory and developmental drilling is up to two orders of magnitude greater than for production wells³⁷ due to the risk of drilling into an unexpectedly over-pressured reservoir. In the case of $CO₂$ storage, drilling related blowouts will occur prior to the $CO₂$ injection and thus will not result in leakage of the stored $CO₂$ back to the surface, hence we focus on production and injection blowout rates, where available.

Several studies have collated data on well failure and pollution incidents associated with hydrocarbon wells, but few distinguish between well operation phase and not all sources distinguish between pollution incidents associated with well failure and those associated with surface processes such as improper waste disposal. Hence, many quoted data (Supplementary Table 6) are likely to be over-estimates of the expected incident frequency. Closer inspection of the information sources cited by many well failure review papers^{16,38} shows that, in some cases,

overly-simplified data has been used to calculate failure rates. For example, frequency of documented pollution incidents is often used as a proxy for well failure, but leakage of fluids from depth only accounts for a minority of incidents in such cases. A further complication is the imprecision of terminology used to describe subsurface fluid releases. Terms such as "blowout", "well failure", "well / fluid release", "loss of well control" and "leak" may all be used to describe an uncontrolled release of fluids from the subsurface, but many of these terms can also describe "near misses"39,40. For example, some studies consider a "blowout" and a "well release" to be different types of events, the latter being an unintended flow of fluid from the well that was stopped by the well barrier system while "blowout" is reserved for complete failure of the well barrier system^{39,41}. The term "leak" may refer to a discrete incident, or to continuous leakage, as described in the previous section. Another problem is the imprecise definition of incident frequency in the literature. For instance, when quoting frequencies as percentages, it is rarely clear whether percentage of wells experiencing failure is being discussed, or it refers to the percentage of well-years that a failure occurs in. Collating and comparing discrete leakage event (here referred to as "blowouts", even if minor, and including both surface and subsurface blowouts) frequency data is thus not straightforward. Published and calculated blowout frequencies are listed in Supplementary Table 6 and relevant examples are discussed below. Supplementary Table 7 lists examples of blowouts along with amount of material leaked and blowout duration, where available. Case studies where high well failure rates are due to production from poorly consolidated reservoirs (e.g. Malacca Straight, Indonesia⁴²) are not included in our analysis.

Where available, plotting blowout frequencies against the number of wells in each case study (Supplementary Figure 5) shows that blowout frequency tends to decrease with increasing study size, suggesting that smaller studies are unlikely to be representative of the whole industry.

The highest documented blowout frequency (0.0693 releases per gas well year⁻¹) comes from analysis of the UK Health and Safety Executive (HSE) North Sea population⁴³ and hydrocarbon release databases⁴⁴. Wellhead Process release incidents were selected to exclude non-blowout incidents (such as surface spills or storage container leaks), but it is possible that this high blowout rate is an over-estimation due to not fully excluding other release sources and mechanisms. The durations of the selected incidents ranged from one minute to 70 days, with an average of \sim 3.5 hours. Total volumes of gas released range from 9.80 x 10^{-5} m³ (1.9 x 10^{-7} t CO₂ equivalent) to 1.98 x 10⁶ m³ (3889 t CO₂ equivalent), with an average of 1.93 x 10³ m³ (3.8 t CO₂ equivalent). Only two of the 1,597 gas release incidents were of a volume greater than 2.55 x 10^5 m³ (500 t $CO₂$ equivalent). The vast majority of these incidents are thus volumetrically small and it is possible that the difference in blowout frequency between this database and other North Sea databases³⁹ reflects a difference in reporting standards and/or how a blowout or well release is defined. However, other offshore production blowout frequencies suggest that there is a greater risk of blowout for offshore wells than for onshore wells (Supplementary Figure 5).

Few data exist for CO₂ injection well failure frequencies, and those available are on small data sets. One study, summarising events in acid (H_2S) and CO_2 injection wells in Alberta, Canada³³, discusses the difference in blowout rates between injection and production wells. Here, well failure events were found to be due to general well operation issues and were not caused by injection³³. They found a greater risk of well failure in wells converted from production to injection wells, rather than wells initially completed for injection, and in wells drilled before improved regulations came into force in 1994. On the other hand, well integrity failures were found to be twice as common in injection wells as in production wells on the Norwegian Continental Shelf (17% of 526 production wells vs 29% of 185 injection wells experienced well integrity issues in 2007 ⁴⁵, although these did not necessarily result in leakage.

The next most relevant case studies are those of UGS experiences, where the gas is stored in saline aquifers or depleted hydrocarbon fields. Blowout frequencies are quoted from a number of studies addressing the worldwide UGS industry. Well numbers are generally not available, but these values are expected to be representative of the industry. These values fall well within the range of blowout frequencies for hydrocarbon production (Supplementary Figure 5).

The volume of material lost during a blowout is reported for a number of case studies, but no studies currently exist that consider the relative frequencies of different blowout magnitudes. One study³¹ estimates frequency of blowouts of varying severity but bases severity on the environmental and health and safety impact of the incident, which is not necessarily proportional to mass of material (e.g. $CO₂$) lost. Supplementary Table 7 summarises leakage details from a selection of blowouts. Mass lost during a blowout has a positively skewed distribution (Supplementary Figures 7 and 8) with most of the blowouts releasing less than 10 t $CO₂$ equivalent. Supplementary Figure 6 combines the blowouts from Supplementary Table 7 with the HSE blowout data⁴⁴ and shows that the vast majority of blowouts release less than 500 t $CO₂$ equivalent.

Supplementary Figure 5: Well blowout frequencies

Figure 5: Blowout frequencies plotted against the number of wells involved in each case study. Where absolute well numbers are not available, they are estimated by dividing well years by the number of years of study. Some case studies for offshore and UGS blowouts do not provide number of wells or number of years – the blowout frequencies for these are represented as horizontal bands on the chart. Data from Supplementary Table 6.

Supplementary Figure 6: Blowout magnitude (mass lost)

The magnitude of blowout release (as volume equivalent mass of $CO₂$ leaked in a single year – for blowouts lasting less than a year this is the total leaked) plotted against blowout duration.

3.3.2 Active well blowouts – Parameter definitions

Given the above discussion, we consider blowout frequencies in terms of volumetrically minor and major incidents. Minor blowouts are based on the UK North Sea HSE database with a blowout frequency of 0.0693 events per well per year and are considered to leak between 1 and 500 t $CO₂$ equivalent per event. Blowout data are mostly from conventional hydrocarbon wells. Remediation of a CO² blowout may take longer and/or emit proportionally more gas due to complications resulting from CO_2 flow (rapid flow rate due to gas expansion coupled with dry ice formation¹⁸). To ensure that we are not under-estimating likely leakage due to the extra difficulties in remediating blowouts on $CO₂$ wells, the values of mass lost during a blowout have been increased by 50% to account for the expected more rapid flow and longer remediation timescales. We acknowledge that this is an arbitrary increase but, given the lack of data comparing $CO₂$ to conventional blowouts, we believe this provides a conservative likely estimate. This increases our boundary between minor and major blowouts from 500 t CO_2 equivalent to 750 t CO_2 equivalent.

For sensitivity analysis, the data are described in terms of a lognormal distribution where the natural log of the values is taken twice (i.e. $Ln(Ln(x))$, where x represents the data in Supplementary Table 7 and from the HSE database⁴⁴ that release between 1 and 500 t $CO₂$ equivalent, multiplied by 1.5 to account for the 50% increase). For this lognormal distribution, the mean of the logs is 1.27 ± 1.21 (one standard deviation), which provides a representative fit for the original data, but over-estimates the logged data (Supplementary Figure 7). We use this mean (i.e. $e^{e^{1.27}}$) for the base case value.

Major blowout rates are treated separately for onshore and offshore cases due to the higher frequency of offshore blowouts. Representative ranges are taken as 1.63 x 10⁻⁶ to 1.33 x 10⁻⁴ blowouts per well per year for onshore and 4.74×10^{-5} to 2.48×10^{-4} blowouts per well per year for offshore environments (Supplementary Figure 5). These are based on the studies with the largest data sets; for production wells, studies involving fewer than 10,000 wells have been excluded. In order to model a realistic worst case scenario we then multiply the frequencies by two, to allow for the potential doubling of risk associated with injection rather than production⁴⁵. For the sensitivity analysis, we assume a normal distribution where the maximum and minimum values represent three standard deviations from the mean. This results in well blowout frequencies of 1.35 x $10^{-4} \pm 4.4$ x 10^{-5} blowouts per onshore well per year, and 1.48 x $10^{-4} \pm 3.3$ x 10^{-5} blowouts per offshore well per year. We use these mean values as the base case values.

The blowout data in Supplementary Table 7 inform our estimate of the maximum and minimum amount of leakage during a major blowout. Documented blowout (or other significant leakage) durations range from 1 second to decades, with total fluid volume leaked ranging from 9.8×10^{-5} to 350 million $m³$. Supplementary Figure 6 plots the magnitude of blowout releases against the blowout duration. Most blowout events last less than a year, many lasting less than day. This is corroborated by a study into Gulf of Mexico offshore blowouts between 1971-1978, which found 16 blowouts, unrelated to drilling, of durations between 1 hour and 55 days, with an average of 8 hours and a median of 2.5 days⁴⁶. Three of the fluid release events compiled in Supplementary Table 7 lasted much longer than a year: the Bečej field $CO₂$ leak (39 years), the Kalle UGS facility (5 years), and the 22/4b North Sea blowout (21 years). In the case of the Bečej field, the blowout itself lasted 209 days, but leakage continued until the well was remediated 39 years later; as far as we can tell, no attempt was made to remediate the well before this time. The Kalle UGS facility in Germany was shown to have leaked for at least 5 years, but again, it seems that no attempt was made to remediate this. In these two cases, the lack of well remediation was an operational issue that would not occur in the case of a $CO₂$ storage site. The 22/4b North Sea blowout continued for 21 years before successfully being remediated. This was a drilling-related gas blowout that was particularly difficult to plug because of the shallow depth of the reservoir. This scenario is therefore not analogous to a $CO₂$ storage scenario, where $CO₂$ storage and associated blowouts would occur from greater depths. We thus consider the yearly leak volumes shown in Supplementary Figure 6 to be representative and assume that blowouts taking place on injection wells will not continue for more than one year.

For a minimum / major blowout mass-loss, we adopt our minor / major blowout boundary of 500 t per event for the hydrocarbon industry data. The greatest volume of material released during a blowout of less than one-year duration that we have identified is an estimated 5.98×10^8 m³ of oil and gas (-1.17 Mt CO_2) equivalent) released over 84 days during the Macondo / Deepwater Horizon blowout; we adopt this as a representative maximum mass-loss from documented blowouts. As discussed above, we then increase these values by 50% to account for greater losses from $CO₂$ wells, resulting in minimum and maximum values of 750 t and 1,749,687 t, respectively. We note that recent work⁴⁷ has concluded that $CO₂$ blowouts are likely to result in smaller gas losses than natural gas blowouts, due to the greater density of $CO₂$ in the reservoir and resulting lower discharge rate. However, we have chosen not to factor this possible reduction into our data compilation until this phenomenon has been independently verified and can be quantified for a wider range of conditions. For sensitivity analysis, the data are described in terms of a lognormal distribution where the natural log of the values is taken twice (i.e. $Ln(Ln(x))$, where x represents the data in Supplementary Table 7 and from the HSE database ⁴⁴ that release between more than $500 \text{ t } CO_2$ equivalent, multiplied by 1.5 to account for the 50% increase). For this lognormal distribution, the mean of the logs is 2.57 ± 0.045 (one standard deviation), which provides a fit that envelopes, but over-estimates the original data (Supplementary Figure 8). To calculate the standard error, we take the number of data points (17) as n, giving a standard error of 0.011. We use this mean (i.e. $e^{e^{2.57}}$) as the base case value.

Supplementary Figure 7: Minor blowout losses

Active well minor blowout leakage. Histograms (grey boxes) and fitted lognormal distributions (blue lines) for the mass leaked during a minor blowout. Data from Supplementary Table 7. Black lines show the minimum, base case, and maximum values.

Active well major blowout leakage. Histograms (grey boxes) and fitted lognormal distributions (blue lines) for the mass leaked during a major blowout. Data from Supplementary Table 7. Black lines show the minimum, base case, and maximum values.

Supplementary Note 4: Estimating Leakage in Abandoned / Legacy Wells

4.1 Background and context

Abandoned wells pose different risks to those associated with active wells. While abandoned wells do not experience the operational stresses of active wells, it may not be possible to identify and monitor all abandoned wells within a storage site, and so leakage may go unnoticed, preventing its remediation. As frequency of pre-existing wells decreases with depth, the risk of leakage along an abandoned well can be reduced by selecting injection formations deeper than historically producing formations⁴⁸.

The identification and monitoring of all abandoned wells in a storage site is particularly relevant for regions with a long history of hydrocarbon exploitation. In such regions, many wells were drilled before comprehensive record keeping and regulation standards began, meaning that no records exist of the wells and their abandonment status (plugged / unplugged / plug integrity) is unknown. A recent survey of abandoned wells in Pennsylvania estimated that the majority of abandoned wells are not documented 49 . The two main field-techniques used for identifying undocumented abandoned wells are magnetic surveys and measurement of methane concentrations $50,51$. However, magnetic surveys will not identify wells where the steel casing has been removed or was never fitted. Methane will only leak from wells that penetrate formations containing buoyant fluids. Most wells are abandoned because they no longer produce or have never produced hydrocarbons and so methane surveys are unlikely to identify all abandoned wells. Furthermore, measurement of fluid flow up an abandoned well is the main way of testing abandoned well integrity, without requiring costly re-entry of the well. However, this method will only work if the well is penetrating pressurised reservoirs. If the reservoir is depleted, fluid flow up the well will be reduced⁵², leakage potential will be under-estimated, and well integrity will be over-estimated. For many wells, it may be impossible to assess the integrity of well plugs and cement until the well bottom is pressurised by injection or migration of fluids into the reservoir(s) penetrated by the well. While this is unlikely to be an issue for regions with a highly regulated CCS industry, where we anticipate that regulators will require poorly documented wells to be reentered, tested, and repaired, it may be an issue if CCS is developed in a region with a poorly enforced regulatory system.

4.2 Sources of abandoned well data

The amount of $CO₂$ that may leak from an abandoned well depends on a number of factors, including: the areal density and depth of pre-existing wells, proximity to the injection well, injection pressure, the $CO₂$ plume geometry, the permeability of the wells and the reservoir, the ability of the $CO₂$ to flow up the well (hydraulic head), and whether the well is open to overlying aquifers that may act as a sink for the leaking $CO₂$. Precise modelling of potential leakage from abandoned wells at a given storage site requires detailed constraints on all of these parameters, and also on the permeabilities of the geological formations, temperature and pressure (thus phase density and viscosity), injection volume and pressure, and appropriate model-grid spacing and equations of state^{13,53,54}. For a more generalised model, we estimate the amount of $CO₂$ that may leak from abandoned wells via two sources: 1) Data of natural gas leakage or blowouts from abandoned well bores. 2) Mathematical models of $CO₂$ leakage along abandoned wells.

Gas leakage data from abandoned wells are available for some hydrocarbon fields, but these are unlikely to be representative of leakage from $CO₂$ storage reservoirs because wells tend to only be abandoned once they are no longer producing (i.e. reservoir pressure is low). As a result, such data would vastly under-estimate the amount of $CO₂$ likely to leak from an abandoned well. For example, a study on abandoned wells in Pennsylvania, USA, found that leakage rates were higher for abandoned gas wells than for abandoned oil wells⁵², indicating that leakage strongly depends on the presence of buoyant material in the reservoir. However, a more recent study on abandoned wells, also in Pennsylvania⁴⁹, found no correlation for high-emitters between abandoned well leak rate and proximity to underground gas storage, suggesting that the highest emitting wells may actually be representative of wells leaking from a gas-rich reservoir. For comparison, the highest emitter was an unplugged gas well from a non-coal area with a methane flow rate of 3.5×10^5 mg h^{-1} ⁴⁹; assuming standard temperature and pressure conditions, this is the equivalent of 8.45 t CO₂ year⁻¹. The average methane emissions from plugged gas wells in non-coal areas was 5.4×10^2 mg / h/ well⁴⁹, equivalent to 0.01 t CO₂ well⁻¹ year⁻¹. A mean leakage rate of 0.27 kg of methane well⁻¹ year⁻¹ (volumetric equivalent of 271 kg $CO₂$ well⁻¹ year⁻¹) was measured for abandoned wells during another study in Pennsylvania⁵⁵.

Data for blowouts occurring on abandoned wells in fields undergoing injection $(CO₂)$, steam or water) may be more representative, but data are sparse. A study investigating the timing and mechanism of abandoned well leakage due to nearby steam injection in California found that most well failures leaked shortly after being abandoned and / or impacted by steam. This indicates that leakage is most commonly due to initial well defects⁵⁶; while the timing of some blowouts indicates that they were caused by well degradation over time, these were a minority⁵⁶. This study estimated that inactive wells contacted by steam floods would blowout at a rate of one per several thousand wells for initial defects during or shortly after injection⁵⁶. For aging-related defects, the risk of blowing out over the longer term decreased over time, due to improvements in cementing practices, from $1/10,000$ well years in the 1990s to $1/100,000$ well years in the year 2000^{56} .

Various studies have modelled the amount of $CO₂$ expected to leak along abandoned wellbores and consider a range of storage and migration conditions. Many of these studies, which are summarised in Supplementary Table 7, model leakage of $CO₂$ out of the storage formation, and not necessarily leakage to the surface.

To assess how representative the models are of field conditions, we compare the model parameters to values experimentally derived for effective permeabilities for leaking wellbores (Supplementary Figure 9). One study used gas flow rates to determine effective well permeability for plugged (0.4 mD) and unplugged (17 mD) wells⁵², but also noted significant difference in flow rate between gas and oil wells, and calculated higher effective permeabilities for gas wells than for oil wells. In their study, Kang et al noted that the effective permeability model assumes an infinite supply of gas and that, for wells with low gas contents the effective permeabilities will be an under-estimate⁵². For this reason, we take the upper ranges of their calculated effective permeabilities, rather than the quoted means, and assume that 10^0 and 10^2 mD are more appropriate values for plugged and unplugged wells, respectively.

Supplementary Figure 9: The relationship between permeability and leakage

Modelled CO² leakage along abandoned wells, plotted against well effective permeability used in the models, compared to measured well permeabilities. Red symbols represent models of a pressurised reservoir. Blue symbols represent models where buoyancy is the sole driver of leakage (no overpressure). Black symbols are effective well permeabilities measured along entire wellbores. Grey symbols are local permeabilities measured in samples of well cement, either sampled from CO₂ wells or corroded in the laboratory. Letters refer to letters in Supplementary Tables 8 and 9. Measured leaks (circles) refer to the values described in the above text.

4.3. General abandoned well parameters - well density and condition

For simplicity, we make the unrealistic but conservative assumption that all abandoned wells penetrate the storage formation. The areal well density and proportion of degraded and leaking wells is likely to vary depending on past and contemporary drilling and abandonment history and regulations. Assuming that regions with an established hydrocarbon industry are targeted, we expect the areal density of abandoned wells to be different between offshore and onshore environments. The IPCC report on CCS (Figure 5.27)⁵⁷ estimated a hydrocarbon well density for the North Sea of up to $4,400$ wells per 10,000 sq km $(0.44 \text{ wells km}^{-2})$. This figure is assumed to be reliable as hydrocarbon exploration in the North Sea has been regulated (and thus documented) since the industry began in the 1960's. We take this value as an estimate of abandoned well density for offshore CO_2 storage regions. A 2009 study ⁵⁸ estimated that Texas contains more than 125,000 wells over 50,000 km², equating to a well density of 2.5 wells / km². We use this value as an estimate of abandoned well density in onshore $CO₂$ storage regions.

We note that, in regions with an exceptionally long-lived hydrocarbon industry, there may be instances of wells not being recorded and being improperly abandoned. A study of Pennsylvanian hydrocarbon industry wells recently revised estimates of well numbers from 350,000 wells to up to $750,000$ wells, based on studying historical records and historical aerial photographs⁴⁹. In this case, an under-estimation factor of ~ 2.1 describes the difference between recorded and existing wells. We have built an under-estimation factor into our model, that allows us to quantify the impact of unidentified abandoned wells. In reality, the under-estimation factors will be lower than that found for Pennsylvania, because $CO₂$ storage is unlikely to be promoted in regions with such high uncertainty and potential risk of leakage. For our Offshore, and Onshore Well-Regulated scenarios, the under-estimation factor is 1. For our Onshore Poorly-Regulated scenario, we adopt a base case under-estimation factor of 1.55, with a minimum and maximum of 1.1 and 2.0, respectively, and assume a uniform distribution between these values.

To estimate well integrity, we consider the wells in terms of unplugged, degraded, and intact wells. We assume that all known wells are plugged (or at least are remediated with high quality plugging procedures), and so unplugged wells are only an issue where the well under-estimation factor is greater than 1. For our Onshore Poorly-Regulated scenario, we estimate the proportion of unknown abandoned wells that are unplugged to be 30%, based on estimates of abandoned well status in Pennsylvania⁵². We use the frequency of continuously-leaking active wells as a proxy for degraded wells, giving base case scenarios of 14.5% and 7.5% for offshore and onshore wells, respectively. The remaining wells are assumed to be intact. For sensitivity analysis, we assume the same lognormal distribution as applied to continuously leaking active wells, discussed in Section 2.2.2.

Because leakage from abandoned wells depends on the number of abandoned wells and well integrity issues, both of which vary over time, we have calculated two different abandoned well leakage rates to be applied to the injection (AB1) and post-injection (AB2) phases of the program. We assume that: (1) the storage site is monitored during the injection period; (2) any blowouts are remediated with high quality plugging; and (3) all known abandoned wells are monitored, allowing rapid identification and remediation of degraded wells. Once injection ceases, the injection wells are converted to abandoned wells. During AB2, all known wells are assumed to be plugged and intact, with any degraded wells having been repaired during AB1.

4.4 Abandoned well continuous leakage parameters

We expect smaller leaks to occur, similar to continuous leakage in active wells, with the amount of $CO₂$ leaked dependant on well integrity. Estimating the amount of $CO₂$ that may be leaked from wells with different levels of integrity is difficult as this also depends on local reservoir conditions. Maximum modelled leak rates that would not be classed as blowouts (as defined above) are \sim 300 t CO₂ year⁻¹. This is comparable to the higher documented gas flow rates from abandoned wells⁵² and from active wells (Supplementary Table 5). We use this value (300 t $CO₂$) year-1) as a proxy for leakage from degraded wells and assume that this leak rate is low enough that it might not be detected without frequent monitoring and is thus constant during AB1. To estimate leakage from relatively intact abandoned wells, we refer back to the continuous leakage rates for active wells, presented by Marlow³⁵, who found that up to 5.4% of wells leaked up to 230 t $CO₂$ equivalent per year (Supplementary Table 5). To avoid under-estimating leakage from abandoned wells, we thus assume that 5.4% of intact wells will leak 230 t $CO₂$ year⁻¹ and the remainder will leak 0.004 t $CO₂$ well⁻¹ year⁻¹ (the minimum modelled leak rate for wells experiencing overpressure in Supplementary Figure 9 and Supplementary Table 8).

4.5 Abandoned well discrete event parameters

For short term blowouts (one per several thousand wells⁵⁶ over the 30 year injection period), we consider minimum, maximum, and mid-point probabilities of one per 9,000 wells $(1/9000/30 =$ 3.7×10^{-6}), one per 2,000 wells $(1/2000/30 = 1.7 \times 10^{-5})$, and one per 5,500 wells $(1/5500/30 = 1.7 \times 10^{-5})$ 6.1×10^{-6}), respectively. For the sensitivity analysis, we assume a lognormal distribution based on these minimum, maximum, and base case values that is described by the mean of the natural $\log s = -8.6125 \pm 0.23$ (one standard deviation). We use this mean (i.e. $e^{-8.6125}$) as the base case. The model has been constructed to include the possibility of unplugged wells being present (for scenarios with unidentified wells – i.e. well under-estimation factor >1), and we assume that any unplugged wells will also blowout during the injection period.

For the long term blowout rate (AB2), we consider minimum, maximum, and mid-point probabilities of one per $100,000^{56}$ well years $(1/100,000 = 1 \times 10^{-5})$, one per 10,000 well years⁵⁶ $(1/10,000 = 1 \times 10^{-4})$, and one per 50,000 well years $(1/50,000 = 2 \times 10^{-5})$, respectively. For the sensitivity analysis, we assume a uniform distribution between these minimum and maximum values.

We also assume that abandoned well blowout events are similar to those of active wells, but that there may be delays in identifying the location and thus the remediation of smaller blowouts (c.f. up to 750 t vented in total for active wells). Discounting the three longest lasting blowouts described in Supplementary Table 7, the median blowout duration is 22 days and we use this to calculate a likely CO_2 venting rate of 34 t CO_2 day⁻¹ during a small blowout, and scale this to a scenario where identifying and remediating the blowout takes 1.5 years. This gives a conservative minimum $CO₂$ loss of 18,615 t $CO₂$ per abandoned well blowout. For a mid-point, we use the venting rate of the Bečej Field $CO₂$ leak (678,500 t $CO₂$ year⁻¹) and assume that such a large blowout would be identified and remediated within a year. For a maximum blowout loss, we adopt the maximum value for active well blowouts $(1,749,687 \text{ t } CO₂)$ in the Macondo / Deepwater Horizon event).

For sensitivity analysis assessing the impact of the mass of $CO₂$ lost during an abandoned well blowout, we describe the minimum, maximum, and mid-point values in terms of a lognormal distribution where the mean of the logged values is 13.4 ± 0.35 (one standard deviation). In this case, we work with the standard deviation, rather than the standard error, because the number of blowouts per year is expected to be so small that the standard deviation and standard error will be comparable.

Supplementary Table 2: General abandoned well input parameters

General parameters used to estimate abandoned well leakage in all scenarios.

Supplementary Note 5: Natural Leakage

 $CO₂$ may potentially leak out of a geological storage site via natural pathways (i.e. diffusion through the cap rock, or advection along faults and fractures). However, in a review of global natural and man-made CO_2 geological subsurface accumulations, Miocic et al⁵⁹ noted that only 10 out of 76 known $CO₂$ fields show conclusive or inconclusive evidence of leakage.

Work by numerous authors^{60–63} has concluded that diffusive losses through the caprock matrix will be negligible under CO_2 storage timescales. Busch et al⁶¹ and Deming⁶⁴ have shown that capillary pressure $CO₂$ breakthrough of the caprock occurs after hundreds to thousands of years for medium to low permeability caprocks with a realistic thickness of 100m. Hence, diffusion of CO² through the cap rock is not expected to result in significant loss from the storage reservoir.

Fractures are the most likely route for $CO₂$ leakage from the reservoir through the caprock. Preexisting fractures may be present, or overpressure as a result of $CO₂$ injection may cause hydraulic fracturing or re-activation of fractures within the seal rock^{65,66}. During monitoring of the world's first commercial scale on-shore CO_2 storage project, In Salah, evidence for CO_2 migration into the lower, shaly seal rock layer was observed. This was interpreted to have occurred as a result of tensile opening of a fracture zone in response to pressurisation during $CO₂$ injection⁶⁷. As with leakage along abandoned wells, at the individual site-scale, leakage through fractures will be influenced by a host of parameters, including the permeabilities of the conduits, the reservoir rocks, and the pressure in the reservoir and in all of the formations contacted by the conduit; flow through fractures is influenced by thermo-hydraulic-mechanical and chemical (THMC) processes. These processes are intrinsically linked such that one process affects the initiation and progress of others⁶⁸.

Predicting leakage along natural flow pathways is difficult due to uncertainties in whether a fault or fracture will act as a migration conduit or as a seal, and this may also vary over time^{69–71}. Using this information to quantify a natural leakage parameter is challenging even when $CO₂$ fluxes from point sources are quantified - e.g. gas seeps, along fractures, of methane \pm CO₂ \pm heavier hydrocarbon gases from hydrocarbon fields, volcanoes, mud volcanoes, and along faults^{72–81}. This is due to difficulties in extrapolating from point source data to predict areal fluxes, due to the uncertainties in fracture density, and fracture permeability. Even in a highly faulted area, only a very small proportion of the surface will be acting as a gas flow conduit at the square-kilometer scale^{74,82}. Estimates of gas fluxes from entire fields or regions are thus more appropriate to inform our natural leakage parameter, but are less common. Compiled data on gas flux are shown Supplementary Figure 10 and their applicability to a $CO₂$ storage model is discussed below.

Italy is a tectonically active country noted for a high rate of geological $CO₂$ emissions. It is estimated to have a flux rate of geological $CO₂$ of 20-60 Mt $CO₂$ year^{-1 83}. Assuming an area of 294,140 km^{2 84}, this gives geological CO_2 fluxes of between 68 and 204 t km⁻² year⁻¹. The tectonically active Iceland also has a high degassing rate, and is estimated to emit 0.16 to 2 Mt $CO₂$ per year ⁸⁵. Assuming an area of 100,250 km^{2 84}, an average geological $CO₂$ flux of between 1.6 and 20 t km⁻² year⁻¹ is estimated for Iceland. Both Iceland and Italy's high degassing rates are a symptom of tectonics creating flow pathways (fractures and faults) in the crust. Such regions are useful for determination of possible $CO₂$ fluxes in tectonically active areas, but as these will likely not be the target of $CO₂$ storage sites due to the prevalence of active faults and fractures, their degassing rates are not relevant here.

At a global scale, degassing of $CO₂$ from the deep Earth via volcanism is a large-scale proxy of degassing via faults in a sedimentary basin. Estimates of global degassing of $CO₂$ via volcanoes range from 65 Mt per year, based on SO₂ fluxes⁸⁶, to 6×10^{12} moles of C (264 Mt CO₂), based on $CO₂$ and He degassing from ridges, island arcs and plumes. Assuming a global area of 148.3 million km^{2 87}, this gives an average global CO_2 flux of between 0.4 and 1.8 t CO_2 km⁻² year⁻¹.

Similarly, total global seepage of methane from petroleum systems has been estimated at 14-28 Mt methane per year emitting from an area of 8×10^6 km^{2 88}. Converting this mass of methane to a volume at standard temperature and pressure and calculating the equivalent mass of $CO₂$, gives a total of 38.5 to 77 Mt, equivalent to between 5 and 10 t CO_2 km⁻² year⁻¹. As most petroleum systems are associated with sedimentary basins, we consider this generalised gas flux to represent the high end of the likely range of $CO₂$ leakage rates along natural pathways.

As noted by Miocic et al⁵⁹, the majority of naturally occurring $CO₂$ accumulations do not show evidence of CO_2 leakage to the surface. For example, at the Farnham CO_2 field, measured CO_2 fluxes were $0.5 - 3.7$ g m⁻² d^{-1 89}, equivalent to 183-1,351 t CO₂ km⁻² year⁻¹; these fluxes were concluded to be shallow and biogenic in origin, based on their similarity in flux rate to biogenic sources in arid regions 89 . Background fluxes of surface $CO₂$, due to biological action and respiration, are on the order of 60-1260 g of carbon per $m²$ per year (equivalent to 220 to 277,200) t CO₂ km⁻² year⁻¹)⁹⁰. Measurements of CO₂ fluxes must, therefore, rule out and / or correct for biological $CO₂$ when quoting flux rates for deep-sourced gases. If this is undertaken, the resulting flux rates are often very small or imperceptible. Thus, we do not consider the $CO₂$ fluxes measured at Farnham to represent natural leakage of deep $CO₂$.

Natural gas seeps in the Upper Ojai Valley in Southern California leak an estimated 55 m^3 of gas per day 75 . The gas is a mixture of $CO₂$, methane, and heavier hydrocarbons and emanates from seeps and vents within the valley associated with numerous faults. We estimate an area of 30 km² for the Upper Ojai Valley, measured via Google Earth, and using the topographic trace of the San Cayetano Fault as the northern boundary, and the crest of the Sulphur Mountain ridge as the southern boundary. This gives an emission rate of 2 m^3 of gas per km² per day, equivalent to 1.31 t CO₂ per km² year, assuming standard temperature and pressure.

Twenty three Mt $CO₂$ have been injected for enhanced oil recovery at the Rangely Field, Colorado, and surface CO₂ fluxes were measured to quantify micro-seepage of the injected $CO_2^{\,91}$. Fluxes of 8,600 t CO_2 per year were measured over the 73 km² of the field, but carbon isotope data revealed that the vast majority of this gas was shallow and biogenic in origin, with geological seepage contributing an estimated 170 t CO₂ year⁻¹ for the site ⁹¹. This equates to 2.2 t CO₂ km⁻² year-1 .

Our approach to incorporate leakage rates into the SSC draws on gas flux data from multiple scales - from the regional (Rangely Field and Ojai Valley) and global (methane seepage from total petroleum systems, and global emissions of deep-sourced $CO₂$) – and adapts this for a $CO₂$ storage setting. We adopt a most likely natural leakage rate of 2 t km⁻² year⁻¹, based on the areal fluxes from the Ojai Valley natural seeps and the Rangely EOR field. As a lower limit, we take the lowest estimate of global fluxes of geological $CO₂$ (0.44 t km⁻² year ⁻¹), and as an upper limit, we take the highest estimate of average areal flux from total petroleum systems (10 t $km⁻²$ year- $\left(\frac{1}{2} \right)$

For the sensitivity analysis, we describe the minimum, maximum, and most likely values in terms of a lognormal distribution where the mean of the logs = 0.693 ± 0.37 (one standard deviation), and take this mean (i.e. $e^{0.693}$) as the base vase value.

Supplementary Figure 10: Natural leakage fluxes

Ranges of documented natural leak rates for CO₂, or for hydrocarbon gasses converted to equivalent mass of CO_{2.}

Supplementary Note 6: Reduction of leakage rate with time

The leakage model is based on measured and modelled surface fluxes and is not directly linked to subsurface conditions. However, leakage is not expected to be constant over time but expected to decrease as the buoyancy of the $CO₂$ decreases by reduction of the mobile $CO₂$ remaining in the plume (through leakage, immobilisation and pressure dissipation). To assess the rate of leakage decay, we compared gas flux rates over time from $CO₂$ leakage models, natural gas production, and from a large natural gas blowout.

Zahasky and Benson⁹² carried out TOUGH2 simulations to model the evolution of $CO₂$ leakage out of a storage reservoir with various different mitigation techniques. Their model outputs (Figures 11 and 12 of their paper) plot the leakage rate in $Kg s^{-1}$ over 500 years. The "passive mitigation" result (i.e. injection stops once leakage is identified, but no other mitigation measures are carried out) shows a roughly exponential decrease in leakage rate over time. Results are available for two scenarios: (1) where a leak is discovered and the injection stopped after 5 years of injection; and (2) after 10 years of injection. This decrease in modelled leak rate is plotted on Supplementary Figure 11 as a proportion of maximum leakage rate.

Jordan et $al⁹³$ used a Monte-Carlo approach with a multiphase reservoir simulator to investigate CO² flux to the surface along leaking wellbores over 200 years. Their Figure 16a plots the leakage rate in tonnes per year over time and all simulations show a decrease in leakage rate. Almost half of the simulations approximate an exponential decrease in leakage rate, while others show plateaus and an overall step wide decrease in leakage rate over time. The decrease in leakage rate for Jordan et al's 50% of realisations is plotted as a proportion of maximum leakage over time on Supplementary Figure 11.

Réveillère et al⁹⁴ investigated using pressure control to mitigate $CO₂$ leakage into an overlying aquifer, using TOUGH2/ECO2N multiphase flow transport simulations. While this simulation considers leakage to another aquifer, rather than to the surface, we consider the passive leakage decay rate to be a valid approximation for leakage to the surface. We have plotted the decrease in leakage rate over time, based on Réveillère's Figure 11, in Supplementary Figure 11.

The Aliso Canyon natural gas blowout, California, began in October 2015 and continued for over three months. Methane emissions from the blowout were measured along plume transects by research aircraft on thirteen flights over the course of the blowout⁹⁵. These emissions data suggest an initial period of maximum leakage rate, followed by an exponential decrease in leakage rate, beginning after almost 1.5 months. The leakage rate appeared to plateau at less than 50% of the initial leakage rate, \sim 1 month before the well was finally brought under control and the blowout stopped⁹⁵.

Eight years' worth of gas production data from Alberta, Canada were compiled by Samson⁹⁶, to investigate whether production trends were sufficient to meet future demand for natural gas. These data are from multiple gas fields and multiple wells, and incorporate commissioning and closure of wells each year. It is possible that the decline in gas production reflects outside market forces, such as the price of gas, or regulation issues that decrease the number of active wells, but given the context of an increasing demand for natural gas, this seems unlikely and we assume that the decrease in productivity is mostly controlled by depletion of the gas reservoirs. The rate of decrease in production (based on Samson's Figure 12 – calculated daily production rate) is plotted on Supplementary Figure 11. The decline in production rate is similar to the decline in leakage rate modelled by Réveillère et al, and by the decline in leakage rate during the Aliso Canyon blowout.

Plotting these leakage decay rates against time (Supplementary Figure 11) shows that all modelled and measured leakage rates show an approximately exponential decrease over time, albeit with different exponential rates.

To incorporate leakage decay into our model, we created two exponential decay curves that envelope the data. The longest data set is the Zahasky and Benson⁹² model that runs for 500 years, while other models and measurements are for much shorter timescales. To avoid inaccuracies in extrapolating the leakage reduction curves forward in time beyond the range of data, our curves assume that leakage decreases to a point, and then remains constant over time. These curves are of the form:

% "" = + (100 −) ∗ Eq. [1]

Parameters A and B were iteratively determined to produce curves that envelope the data and parameter A represents the minimum long-term leakage rate as a percentage of the maximum. For the high leakage reduction rate envelope (i.e. leakage reduces quickly), $A = 3$ and $B = 0.5$. For the low reduction rate envelope (i.e. leakage reduces more slowly and plateaus at a higher level), the curve was fitted to the Jordan et al data, and resulted in $A = 53$ and $B = 0.03$. We also fitted a curve to the Zahasky and Benson⁹² data, which gave $A = 3$ and $B = 0.0143$.

For sensitivity testing of these leakage reduction curves in our model, we take maximum and minimum values of A and B based on the three curves $(A = 3-53; B = 0.0143-0.5)$. For parameter A, we notice that the majority of the data suggest that leakage will plateau at less than 20% of the original leak rate, and we consider that a skewed distribution is the most appropriate to apply to this data range. We thus apply a triangular distribution to Parameter A with minimum, maximum, and most likely values of 3, 53, and 12, respectively. The most likely value (12) is taken as the base case value. For Parameter B we apply a uniform distribution from 0.0143 to 0.5, and take the midpoint (0.257) as the base case.

When carrying out a Monte Carlo analysis, we use 10,000 realisations. To show the range of leakage reduction curves that will be produced during the Monte Carlo analysis, a sample of 10,000 curves produced by selecting random numbers (within the defined ranges) for Parameters A and B are shown in Supplementary Figure 11 as red lines.

A) Modelled and measured leakage reduction rates (symbols), enveloping and base case scenario curves (black lines) and 10,000 realisations of leakage reduction curves based on a triangle distribution for Parameter A and a continuous distribution for Parameter B (red lines). Black dotted line shows the minimum reduction curve based on the Maximum Parameter A (envelope) and minimum Parameter B (base case). B) Histogram of 10,000 randomly selected values of Parameter A assuming a triangular distribution with minimum and maximum values of 3 and 53, respectively, and a most-likely value of 12.

Supplementary Note 7: Residual Saturation Trapping

To model residual trapping we use the data compiled by Burnside and Naylor 97 for published residual saturation values (Supplementary Table 10). These data form a normal distribution (Supplementary Figure 12), described by a mean of 0.5800 (58.00%), which we take as the base case value, and a standard deviation of 0.1897 (18.97%). The data compilation comprises 44 data points, giving a standard error of 0.0286. Please see the main text for a discussion on the relevance of laboratory and simulated residual trapping values.

Histogram of the residual trapping data, approximating a normal distribution.

Supplementary Note 8: Chemical Trapping

Chemical trapping refers to both solubility and mineral trapping, where free-phase (gaseous or supercritical) $CO₂$ dissolves into the groundwater and / or precipitates as carbonate minerals.

Estimates on the proportion of gaseous $CO₂$ that can dissolve into reservoir brines vary from 6.5% 98 to 90%⁹⁹ on geological timescales. Estimates for mineral trapping range from 2%¹⁰⁰ to 43%⁹⁸. These two processes are likely to interact and occur in equilibrium with each other. While various studies have investigated rates of solubility or mineral trapping individually, studies investigating the rates of both processes occurring together, and over geological timescales, are rare. The most comprehensive model we have found is that from Xu et al 101 , which models the variation in proportions of gaseous, dissolved, and mineralised $CO₂$ over 10,000 years. This model uses the mean rock composition of the Frio formation (Gulf Coast, USA) as target unit. The hydrogeological parameters (e.g., pressure, temperature, brine composition, porosity, permeability, etc.) were selected to be representative of the storage conditions at 1km depth.

The evolution of the total CO_2 injected into the reservoir, and how the CO_2 is sequestered into different phases, according to the Xu et al model (Figure 5 their paper¹⁰¹) shows the evolution of the total $CO₂$ injected into the reservoir and how the $CO₂$ is sequestered into different phases (gaseous, dissolved, mineralized). Mineral trapping begins at a significant rate at 500 years and increases with time. After 10,000 years, the proportion of $CO₂$ sequestered by mineral trapping is comparable to $CO₂$ dissolution in pore waters. The Xu et al¹⁰¹ model results are expressed in absolute $CO₂$ values. We converted these into relative values by dividing them by the total injected $CO₂$, and determined equations that describe the $CO₂$ partitioning as a function of time.

 Xu 's modelling results predict that dissolution of the $CO₂$ will increase during the first decades of the model and stabilise its effect at \sim 100 years, expressed with the following approximation:

% *Solubility Trapping* (*t*) =
$$
0.204 \cdot t^{0.0342}
$$
 Eq. [2]

where t is the time in years. The mineral trapping increases steadily with time until reaching a trapping level similar to the solubility trapping $(2-5 \text{ kg of CO}_2 \text{ per m}^3 \text{ of reservoir})$, following the expression:

% Mineral Trapping $(t) = (1.67 \cdot 10^{-13} \cdot t^3) + (2.90 \cdot 10^{-9} \cdot t^2) + (1.40 \cdot 10^{-5} \cdot t)$ Eq. [3]

As the Xu et al^{101} simulation is the most comprehensive chemical model available, we use it to simulate chemical trapping in the SSC. Details on how these rates are built into our chemical trapping model are provided in Section 4.

Supplementary Note 9. The Leakage Model (2)

The leakage model - or Leakage model (2) - calculates the amount of $CO₂$ expected to leak from storage, based on measured and expected surface fluxes. It combines leakage calculated for active wells, abandoned wells, and natural pathways, and reduces over time once injection ceases (Section 2.5). This section describes how the active well, abandoned well, and natural pathway leakage rates are calculated.

9.1. Active Well Leakage (2a)

Below is a step-by-step description of and list of the parameters used to calculate active well leakage. Labels in [*square brackets and italics*] refer to the label used in the R code, which are also listed in the methods section of the main text.

Scenario specific inputs:

- Slow leak frequency (% wells leaking) [*ActiveWellFreq*]
- Major blowout frequency (events well⁻¹ year⁻) [*MajorBlowFreq*]

General inputs:

- Injection target
- Injection rate per well
- Injection period Slow leak rate (t CO₂ well⁻¹ year⁻¹) [*SlowLeakInjector*]
- Minor blowout frequency (events well⁻¹ year⁻¹) [*MinorBlowFreq*]
- Minor blowout mass (t CO₂ event⁻¹) [*MinorBlowout*]
- Major blowout mass (t CO₂ event⁻¹) [MajorBlowMass]

Calculate the number of injection wells:

• Injection target / injection period / injection rate $=$ Number of injection wells

Calculate loss via slow leakage:

• Slow leak frequency $*$ slow leak rate = average t $CO₂$ well⁻¹ year⁻¹

Calculate loss via blowouts:

- Minor blowout frequency $*$ minor blowout mass = average t $CO₂$ well⁻¹ year⁻¹
- Major blowout frequency * major blowout mass = average t $CO₂$ well⁻¹ year⁻¹

Calculate Active Well Leakage

• Slow leak + minor blowout + major blowout

9.2 Abandoned Well Leakage (2b)

Below is a step-by-step description of and list of the parameters used to calculate abandoned well leakage. Labels in [*square brackets and italics*] refer to the label used in the R code, which are also listed in the methods section of the main text.

Scenario specific inputs for initial model set-up:

- Number of injection wells (see 3.1)
- The areal extent of the CO₂ plume [*MeanPlumeArea*]
- Measured abandoned well density (wells km-2) [*KnownWellDensity*]
- Well under-estimation factor [*wellUnderEst*]
- Proportion of wells that are plugged / unplugged [*UnPlugWells%*]
- Proportion of plugged wells that are intact / degraded [*DegradWells*[%]]
- Proportion of intact wells with a high continuous leak rate

Calculations for initial model set-up:

- Calculate the density of known and unknown plugged and unplugged wells.
- Calculate the density of known and unknown degraded and intact wells.
- Convert known plugged wells to known intact wells.

At end of initial model set up we should have:

- Unknown, unplugged well density
- Unknown, degraded well density
- Unknown, intact well density
- Total unknown well density
- Known intact well density
- Total known well density

AB1 &AB2 General Inputs:

- Short term blowout rate for plugged wells (events well⁻¹ year⁻¹) [*PlugBlowoutYear*]
- Long term blowout rate for plugged wells (events well⁻¹ year⁻¹) [*BlowoutWellYear*]
- CO2 loss per blowout (t CO₂ event⁻¹) [*CO2largeBlowout*]
- Proportion of intact wells with a high leak rate [*IntactHighRate%*]

AB1 &AB2 Scenario Specific Inputs:

- Injection period (years)
- Degraded well leak rate (t CO₂ well⁻¹ year⁻¹) [*CO2degraded*]
- Intact well, high leak rate [*CO2intactHigh*]
- Intact well, low leak rate [*CO2intactLow*]

AB1 blowouts:

- Calculate number of blowouts
	- o 100% unplugged wells blow out during injection period
		- Unplugged wells / Injection years = unplugged blowouts (events $km⁻²$ year⁻¹)
	- \circ Blowout rate $*$ unknown plugged wells = blowouts from unknown plugged wells (events km-2 year-1)
	- o Blowout rate * known wells = blowouts from known wells (events km^{-2} year⁻¹) o Sum the above
- Calculate the mass of CO₂ lost during blowouts: (Loss event⁻¹) * (events km⁻² year⁻¹)

AB1 continuous leakage:

- From degraded wells:
	- \circ Calculate the total degraded well density (known + unknown)
	- o Multiply by degraded well leak rate = loss from degraded wells (t $CO₂$ km⁻² year-1)
- From intact wells:
	- \circ Calculate the total intact well density (known + unknown)
	- o Calculate the proportion of high and low leak wells (multiply by proportion of high leak rate intact wells) = high and low leak rate intact wells km^{-2}
	- o Calculate the loss from high leak rate intact wells (*intact well high leak rate)
- o Calculate the loss from low leak rate intact wells (*intact well high leak rate)
- Sum the continuous leak rates

AB1 Leakage:

- Sum the continuous and blowout leakage to calculate the total leakage.
- This represents the maximum leakage rate, which assumes that the injected $CO₂$ plume has reached its maximum extent and highest leakage risk. This condition is not appropriate for the early stages of injection; therefore, we calculate AB1 leakage to increase linearly from 0 to the maximum over the injection period.

AB1-AB2 transition – well conversions

- All active injection wells are converted to known, intact, abandoned wells.
- All unplugged wells convert to known, intact wells.
- Unknown plugged wells that blew out convert to known intact wells. These are assumed to be from unknown degraded wells, the density of which is reduced accordingly.
- All known wells undergo monitoring and remediation $=$ intact wells.
- Unknown intact wells remain constant.

AB2 Blowouts

- All wells are now plugged, so blowouts / year $=$ total well density $*$ long term blowout rate.
- Multiply by the loss per blowout = $t CO₂ km⁻² year⁻¹$.

AB2 Continuous leakage

- From degraded wells:
	- o Multiply degraded well density by yearly loss.
- From intact wells:
	- o All known wells are monitored (and remediated if necessary) and have the low leak rate.
	- o Calculate the density of unknown intact wells with the low leak rate.
	- o Sum the above for the total well density for intact, low leak rate wells.
	- \circ Multiply by the low leak rate for loss in t CO₂ km⁻² year⁻¹.
	- o Calculate the density of unknown intact wells with the high leak rate and multiply this by the high leak rate, for loss in t $CO₂$ km⁻² year⁻¹.
- Sum the above for total continuous loss.

9.3 Natural Leakage (2c)

Natural leakage for each scenario is as determined in Section 2.4.

9.4 Combined Leakage Model (2)

The combined leakage model is split into injection and post-injection phases.

Injection Phase:

- The number of injection wells are calculated by dividing the injection target by the injectivity per well.
- The total yearly leakage from active wells is calculated by multiplying the active well leak rate by the number of injection wells.
- The area impacted by the $CO₂$ plume is calculated by multiplying the injection target by the area:mass ratio of the plume.
- Total leakage from abandoned wells and natural pathways are calculated by multiplying the leak rates (in t km^{-2} year⁻¹) by the area impacted by the plume.
- Total yearly leakage for the injection period is calculated by:
	- \circ Total active well leakage + Total abandoned well leakage [AB1] + Total natural pathways leakage.
- To account for lower leakage levels while the reservoirs are being filled, the leakage rate increases from zero to maximum levels over the 30-year injection period, via a linear progression.

Post injection phase:

- Total yearly leakage for the post-injection period is calculated by:
	- o Total abandoned well leakage [AB2] + Total natural pathways leakage.
- This yearly leakage rate constitutes the maximum leak rate.
- The true leak rate for a given year is calculated by multiplying the maximum leak rate by the proportional leak, as defined by Equation 1 in Section 2.5.
- The leakage amount of $CO₂$ leaked per year is calculate for years 1 to 10,000, along with the cumulative leakage for each year.

Supplementary Note 10: The immobilisation models (1)

10.1 Residual Trapping (1a)

We assume that residual trapping takes place as the plume of injected $CO₂$ migrates through the reservoir, and it reaches its maximum at the end of injection period (i.e., when the $CO₂$ plume has likely reached its maximum extent):

Residual Trapping (t) =
$$
\begin{cases} (I * RES\%) \cdot \left(\frac{t}{30}\right) & \text{if } t < 30\\ I \cdot RES\% & \text{if } t \geq 30 \end{cases}
$$
 Eq. [5]

where *t* is the time in years, *I* is the CO₂ injection targets (1.2 x 10¹⁰ t CO₂) and *RES%* is the percentage of residual trapping calculated from the data in Supplementary Table 10.

During the post injection stage, the proportion of $CO₂$ that is residually trapped will decrease over time, as chemical trapping consumes both mobile and residually trapped free-phase $CO₂$. To simulate this, residual trapping is applied to the injection target minus the calculated chemically trapped CO₂.

10.2 Chemical Trapping (1b)

The equations used to calculate the chemically trapped $CO₂$ are described in Section 2.7 and are applied to the amount of $CO₂$ remaining in the reservoir after leakage has been subtracted. The chemically trapped $CO₂$ consumes both residually trapped and mobile free-phase $CO₂$ (see Section 4.1).

Supplementary Note 11: The integrated model (1)

The Storage Security Calculator combines the Leakage and Immobilisation models by calculating the amount of $CO₂$ leaked and immobilised for each year and summing these values. This value is subtracted from the total amount injected (i.e. the injection target, once injection has ceased) to give the amount of mobile (i.e. leakable) CO₂ remaining in the reservoir. The model is projected forwards until 10,000 years, or until no mobile $CO₂$ remains, whichever occurs earlier.

Supplementary Note 12: The R-code for the Storage Security Calculator

Example code for the SSC is provided for the Offshore Scenario, and included in the Supplementary Information as a separate file (Offshore-SSC.R).

Area: Mass ratios $(CO₂$ equivalent) of gas fields in the UK North Sea. Data from Gluyas and Hitchens³.

Frequency of continuously leaking hydrocarbon wells in the literature (Table extends over 2 pages).

Supplementary Table 4 – continued from previous page

Mass flow rates of continuous leakage reported on hydrocarbon wells in the literature.

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Blowout frequency data reported in the literature (Table extends over 2 pages)

Supplementary Figure 13: Blowout frequencies

Histogram of blowout frequencies; data from Supplementary Table 6. $* = \text{large data sets}$ only; for studies where well numbers are specified, this is >10,000 wells per study for production wells. The Onshore large data set also includes the largest injection well study, which involved 978 injection wells.

Leakage details of various documented blowouts with volume-equivalent tonnage of $CO₂$ leaked. Where blowouts continue for more than a year, the yearly leak rate is also presented. (Table extends over 3 pages)

Supplementary Table 7 continued from previous pages

Modelled CO₂ leakage rates along abandoned wellbores of varying permeability. [Letters] indicate data labels in Supplementary Figure 9.

Experimentally derived well permeabilities. [Letters] indicate data labels in Supplementary Figure 9.

Published experimental values for residual saturation trapping $(R, in \%)$ of the $CO₂$ trapped). Modified from ⁹⁷. From these 43 residual trapping values we calculate a mean and standard deviation of 58.45 ± 18.96 %. (1 page)

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