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Extended abstract

Storing CO_2 in buried volcanoes

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Abstract. Australia contains rich natural gas resources, but many of Australia's currently producing and undeveloped gas fields contain relatively high CO_2 contents; if not captured and stored, the venting of co-produced CO_2 could hinder efforts to meet Australia's emission reduction targets. The most mature technology for isolating produced CO_2 from the atmosphere is by containing it in deep sedimentary formations (e.g. saline aquifers or depleted oil and gas reservoirs). The effectiveness of this approach is dependent on factors such as reservoir capacity, the presence of low-permeability seals that physically impede vertical migration of injected CO_2, the chemical reactivity of both reservoir and seal minerals, the risk for leakage, and a gas-entrapping structure. An alternative and attractive mechanism for permanent storage of CO_2 is geochemical or mineral trapping, which involves long-term reactions of CO_2 with host rocks and the formation of stable carbonate minerals that fill the porosity of the host rock reservoir. Natural mineral carbonation is most efficient in mafic and ultramafic igneous rocks, due to their high reactivity with CO_2. Here we review the outcomes from a series of recent pilot projects in Iceland and the United States that have demonstrated high potential for rapid, permanent storage of CO_2 in basalt reservoirs, and explore the practicalities of geochemical trapping of CO_2 in deeply buried basaltic volcanoes and lava fields, which are found in many basins along the southern (e.g. Gippsland Basin) and northwestern (e.g. Browse Basin) Australian margins, often in close proximity to natural gas fields with high CO_2 content.

Keywords: CCS, CO_2, reservoirs, basalts, buried volcanoes.

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Introduction

Australia contains abundant natural gas resources and is a leading global exporter of liquified natural gas (LNG) (Geoscience Australia 2019). Many of Australia's producing and undeveloped natural gas fields contain relatively high CO_2 contents, with particularly high values (>20%) reported from wells and fields in the Otway, Cooper-Eromanga, Gippsland, Bonaparte, and Carnarvon basins (Boreham *et al.* 2001). For some producing fields, most notably Gorgon (~7–27% CO_2: Maftei *et al.* 2013), CO_2 is separated and injected into deep saline aquifers (Michael *et al.* 2010). In many fields, however, reservoir CO_2 is not captured but directly released to the atmosphere, contributing significantly to national greenhouse gas emissions. The Ichthys Field in the Browse Basin contains high CO_2 contents in the Brewster (8%) and Plover (17%) reservoirs, and it is anticipated that ~96 Mt of reservoir CO_2 will be emitted over the 40-year lifetime of the Ichthys LNG project (INPEX 2010).

The most mature technology for the long-term carbon capture and storage (CCS) is underground sequestration of supercritical CO_2 in sedimentary rock formations such as deep saline aquifers or depleted hydrocarbon reservoirs deep enough (typically 1 km or deeper) for the CO_2 to remain in a supercritical state (Bunch *et al.* 2014; National Academies of Sciences Engineering and Medicine 2019). The critical requirements for CCS in sedimentary formations are (i) a thick reservoir (usually sandstone or carbonate) with sufficient porosity and permeability to contain large volumes of CO_2 at commercially meaningful injection rates, (ii) an overlying thick seal (usually shale) with sufficiently high capillary entry pressure and low permeability to retain the injected CO_2 over geological timescales, and (iii) a gas-trapping structure that, due to gas buoyancy, concentrates large volumes of CO_2 and prevent its migration and leakage (Dempsey *et al.* 2015; National Academies of Sciences Engineering and Medicine 2019). Additional important geological considerations include the absence of permeable faults and fractures through the seal and favourable stresses and pore pressures to avoid reservoir or seal fracturing during injection

(Nicol *et al.* 2017; National Academies of Sciences Engineering and Medicine 2019). The combined underground storage capacity in saline aquifers and depleted hydrocarbon reservoirs is estimated between 5000 and 25 000 Gt CO_2 (Kelemen *et al.* 2019).

Carbon mineralisation in igneous rocks

An alternative option for the secure, long-term underground storage of CO_2 is carbon mineralisation in mafic and ultramafic igneous rocks, which could potentially host up to 60 000 000 Gt CO_2 (Kelemen *et al.* 2019). Carbon mineralisation involves the formation of stable carbonate minerals (e.g. calcite, magnesite, dolomite) through the reaction of CO_2 (gas, liquid, dissolved in water or supercritical) with rocks that are rich in calcium or magnesium (Wolff-Boenisch and Galeczka 2018). Mg-rich, Ca-bearing rocks include ultramafic peridotites and mafic basalts, which contain highly reactive minerals including olivine and pyroxene (National Academies of Sciences Engineering and Medicine 2019). Whilst *in situ* carbon mineralisation in sandstone reservoirs can occur over timescales of thousands of years, peridotites and basalts may mineralise and sequester 90% of the injected CO_2 in a few months to decades (Fig. 1*a*; National Academies of Sciences Engineering and Medicine 2019).

The rate of carbon mineralisation is influenced by factors including the available CO_2 dissolved in solution, temperature and variations in pH, with low pH promoting mineral dissolution and high pH accelerating carbonate precipitation (Kelemen *et al.* 2019). Olivine (Mg_2SiO_4), which is the major mineral in peridotite, has amongst the highest reaction rates with CO_2-bearing aqueous fluids, whilst plagioclase feldspar (the major constituent of basalts) has somewhat lower reaction rates, meaning that CO_2 mineralisation may be expected to occur more rapidly in peridotites than basalts (Fig. 1*b*; Kelemen *et al.* 2019; Seyyedi *et al.* 2020). However, an exception may occur where basalts contain amorphous glass horizons, which are thought to provide exceptionally good reactants for carbon mineralisation (National Academies of Sciences Engineering and Medicine 2019). Glass forms when basalt cools very quickly and is often abundant in submarine lavas and hyaloclastite breccias (National Academies of Sciences Engineering and Medicine 2019).

Basaltic rocks are extremely abundant; most of the ocean floor (comprising ~70% of Earth's surface) and >5% of the continents is basaltic, with extensive flood basalt fields present in central India, Siberia, the United States, and Canada (Snæbjörnsdóttir *et al.* 2020). Counter to many petroleum geologists and engineers' assumptions, volcanic rocks often have high porosity and

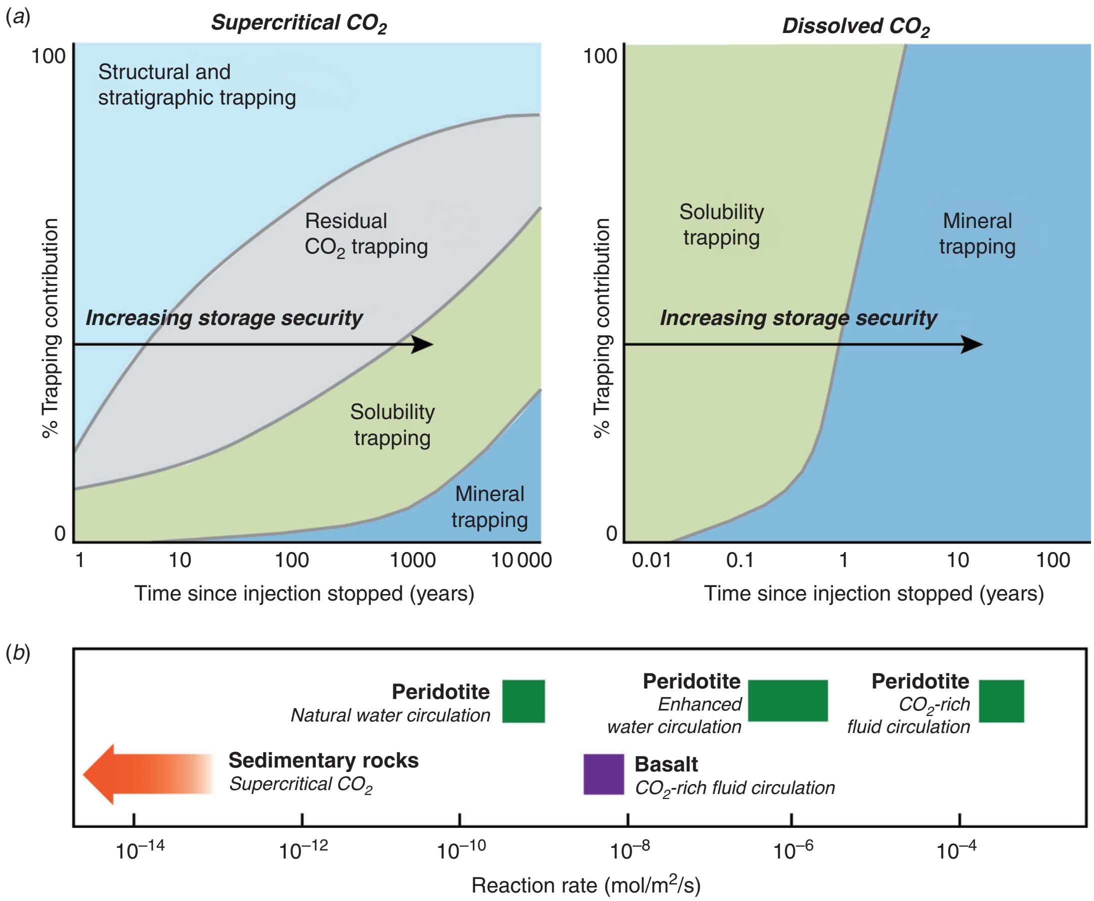

Fig. 1. (*a*) Comparison of CO_2-trapping mechanisms over time when injecting pure supercritical CO_2 into sedimentary basins (left) and water-dissolved CO_2 for mineralisation (right). Modified after Snæbjörnsdóttir *et al.* (2020). (*b*) Reactions rates of *in situ* mineralisation of CO_2 in peridotites, basalts and sedimentary rocks. Modified after Kelemen *et al.* (2019).

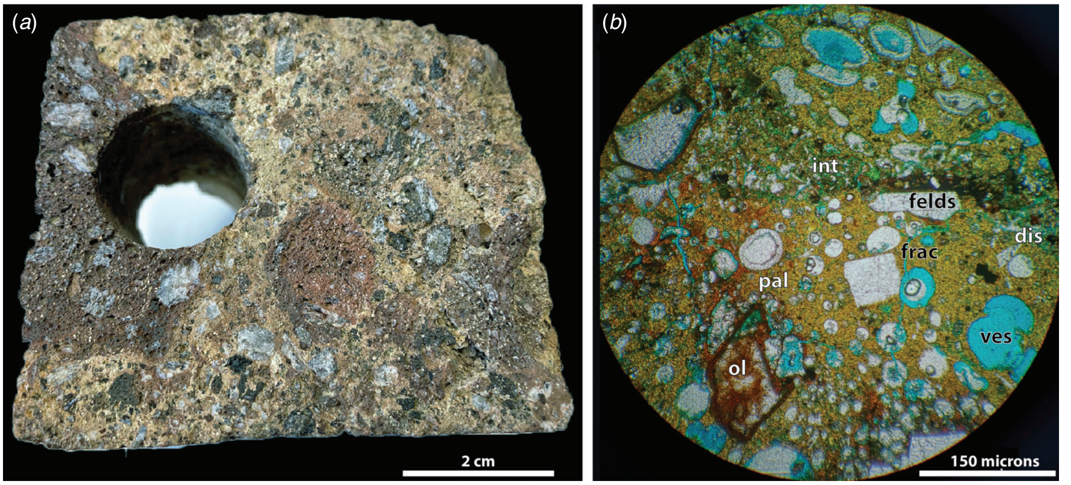

Fig. 2. (*a*) Hand sample and (*b*) plain-polarised thin section of a trachybasalt crystal-rich lapilli tuff from Banks Peninsula volcanic complex, New Zealand. The rock mineral assemblage comprises olivine (ol) and feldspar (felds) immersed in a glassy matrix altered to palagonite (pal), which is a typical product of devitrification of basaltic magmas. The primary porosity (47%) and permeability (800 mD) are controlled by an intense degree of material fragmentation that forms intergranular pores (int) associated with high vesicles content (ves) and quenching fractures (frac). Secondary processes of mineral alteration creates dissolution pathways (dis), interconnecting primary pores.

permeability (Fig. 2), particularly when fractured (Massiot *et al.* 2017; Kelemen *et al.* 2019; Bischoff *et al.* 2020; Millett *et al.* 2020). Two recent pilot projects have demonstrated the potential for CO_2 storage through carbonate mineralisation in basalt reservoirs. In the Wallula project in Washington State, 977 tons of pure CO_2 was injected into high-permeability basalt flow tops at depths between 882 and 887 m over 3 weeks in August 2013 (McGrail *et al.* 2017). Whilst it is not yet known what proportion of injected CO_2 has formed carbonate minerals and how much remains within pore fluids, drill core from the injection zone indicates high rates of basalt dissolution and CO_2 mineralisation, consistent with laboratory experiments (National Academies of Sciences Engineering and Medicine 2019).

In addition, during phase I of the CarbFix project in Iceland, ~200 tCO_2 (co-produced with SO_2 in geothermal fluids) was injected into highly permeable, fractured basalts with ~10% porosity at a depth of 500 m (ambient temperature ~20–50°C). Because CO_2 is not supercritical at this depth, CO_2 and H_2O were separately injected with proportions adjusted to ensure complete solubility of CO_2 into aqueous fluid at the target depth (National Academies of Sciences Engineering and Medicine 2019). Quantification of mineral carbonisation using reactive and non-reactive tracers and isotopes has demonstrated rapid mineralisation of the injected CO_2, with >95% of the injected gas mineralised within 2 years (Snæbjörnsdóttir *et al.* 2020). Following the success of CarbFix phase I, the project has considerably upscaled, injecting 10–20 $ktCO_2$/year into basaltic reservoirs at a depth of ~1500 m (ambient temperature ~250°C), with tracer results continuing to indicate nearly complete loss of carbon along a ~2000 m flow path (National Academies of Sciences Engineering and Medicine 2019).

Despite the success of these projects, a number of uncertainties remain, including determining the precise mineral assemblage (including alteration phases) that are reacting in the reservoirs, the passivation of reactive mineral surfaces over time, the potential clogging of pore space, and the spatial distribution of carbonate mineral precipitation versus dissolution (National Academies of Sciences Engineering and Medicine 2019). Today, the cost of storing CO_2 in basalts is estimated to be US\$20–30/tCO_2 as compared to <US\$20 for storage in sedimentary reservoirs (Kelemen *et al.* 2019). However, if the technology can be further proven, storage in basaltic reservoirs may become a preferred choice in volcanic provinces (Kelemen *et al.* 2019).

Buried basaltic sequences in Australian basins

Buried basalt lava flows and volcanoes are commonly found at depths amenable to dissolved and/or supercritical CO_2 injection (i.e. >1 km) in many Australian sedimentary basins. These include the Browse and Carnarvon basins along the North West Shelf (Holford *et al.* 2013), and the Bight, Otway, Bass, and Gippsland basins along the southern Australian margin (Holford *et al.* 2012; Meeuws *et al.* 2016; Reynolds *et al.* 2017, 2018; Watson *et al.* 2019) and in the Eromanga Basin in central Australia (Hardman *et al.* 2019). In some of these basins, buried basaltic sequences are located in close proximity to gas-bearing reservoirs with high CO_2 content (Holford *et al.* 2012).

Well-preserved basaltic submarine volcanoes are common within the Miocene Torquay Group of the Bass Basin (Holford *et al.* 2017; Reynolds *et al.* 2018), with a large (~5 km diameter) volcano overlying the Yolla Gas Field, where reservoir fluids contain ~17–20% CO_2 (Holford *et al.* 2012; Watson *et al.* 2019). Core from the volcanic section penetrated by the nearby Bass-1 well, which penetrated the flank of a Miocene volcano, contains clast-supported conglomerate of dark grey microvesicular basalt interpreted to represent reworked hyaloclastite and volcaniclastic material, whilst log data and thin sections from the Tasmania Devil-1 well, which penetrated the crest of a volcano, indicate the presence of fine grained crystalline basaltic material interpreted as pillow lavas (Watson *et al.* 2019).

Volcanic rocks have been intersected by many wells in the hangingwall of the basin-bounding Rosedale Fault System in the

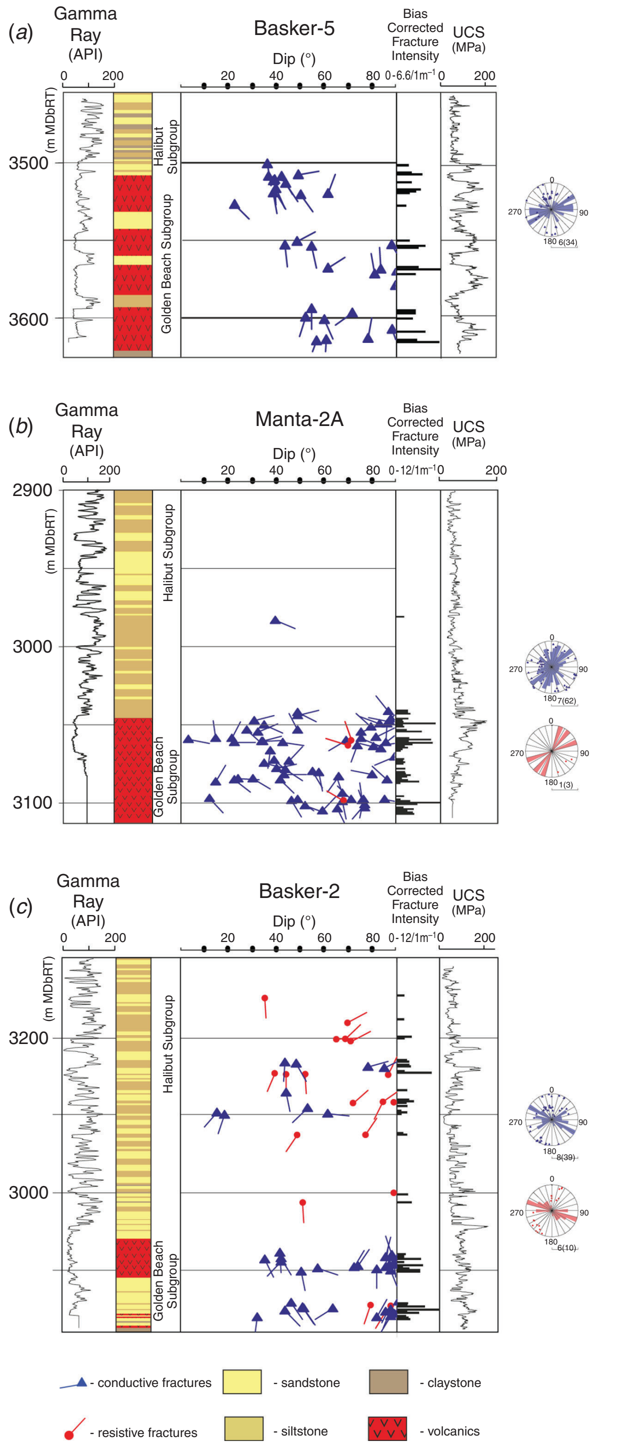

Fig. 3. Image log-based observations of natural fractures in the (*a*) Basker-5, (*b*) Manta-2A, and (*c*) Basker-2 wells presented along with detailed lithologies determined from drill cuttings and petrophysical logs, indicating substantial fracturing within volcanic sequences. Natural fractures are plotted as tadpoles (conductive fracture – blue triangles and electrically resistive fractures – red circles) where marker location present the amount of dip on fracture plane, with the tick corresponding to the dip direction of the plane, followed by bias corrected fracture intensity, and dynamic UCS log. Rose diagrams are plotted natural fractures orientation on lower hemisphere, equal-area projection of poles to natural fractures (both conductive – blue colour and resistive – red).

northern Gippsland Basin, where numerous gas accumulations with high-CO_2 content (some exceeding 30%) occur (O'Brien *et al.* 2008; Holford *et al.* 2012). The majority of these volcanic rocks are Campanian-age basalts that have been subject to various degrees of alteration (Holford *et al.* 2012). In some cases (and in spite of the alteration), these basalts provide the top seals to underlying gas accumulations, most notably at the Kipper field where a 328 m gross gas column (~10% CO_2) within Golden Beach Group sandstones is sealed by ~98 m of volcanic rocks, interpreted to be basaltic lava flows (Sloan *et al.* 1992; O'Halloran and Johnstone 2001). Volcanic rocks also act as seals at Remora-1 (oil) and Tuna-4 (oil and gas) (Holford *et al.* 2012). Petrological and petrophysical studies of these rocks have been limited, though cuttings and sample descriptions indicate that they comprise fine-grained basaltic lavas and tuffs (McPhail 2000). However, our analysis of borehole image logs acquired through volcanic sequences in the Manta-2A, Basker-2, and Basker-5 wells demonstrates an abundance of conductive fractures (Fig. 3), raising the possibility of significant fracture-related permeability.

Hardman *et al.* (2019) have recently described a province of Jurassic extrusive and intrusive rocks within the Eromanga Basin, overlying the Nappamerri Trough of the Cooper Basin, where the highest CO_2 contents (up to 40%) in gas samples are found (Boreham *et al.* 2001). 3D seismic interpretation coupled with log analyses indicates the presence of multiple mafic monogenetic volcanoes that extend into tabular basalt lava flows (Hardman *et al.* 2019). The Lambda-1 well intersected 283 m of basalt directly underlying the Lower Jurassic Birkhead Formation, with the upper 33 m comprising heavily weathered, fractured, and vesicular facies, and the remaining 250 m comprising fresh and crystalline rock (Hardman *et al.* 2019). In other parts of the province, basalts have been extensively altered; Kappa-1 intersected a 120 m succession of mostly fine-grained basalt with evidence for extensive chlorite replacement of ferromagnesian minerals (Hardman *et al.* 2019).

Conclusions

Given the promising results from both experimental studies and pilot projects, and the close proximity of basaltic sequences to many high-CO_2 content gas fields in onshore and offshore Australia, we propose that the possibility of storing CO_2 through *in situ* carbon mineralisation in buried volcanic rocks in sedimentary basins merits further consideration. In addition to the aforementioned uncertainties related to CO_2–rock–fluid interactions, further challenges specific to sedimentary basins include defining the first-order stratigraphic and permeability architecture of buried volcanic sequences in order to identify potential reservoir facies (e.g. Bischoff *et al.* 2021), quantifying the hydraulic and petrophysical properties of buried basaltic volcanoes and lava flows, and defining their petrological characteristics and diagenetic histories.

Conflicts of interest

There are no conflicts of interest associated with this research.

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