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POTENTIAL FOR DESCENDING METEORIC WATER RECHARGE IN HYDROTHERMAL SYSTEMS AS A PATHWAY FOR CARBON DIOXIDE SEQUESTRATION

* John Victor Smith[1,2]

[1]Faculty, of Engineering, RMIT University, Australia; [2]CMW Geosciences, Australia

*Corresponding Author, Received: 4 June 2022, Revised: 4 Dec. 2022, Accepted: 24 Dec. 2022

ABSTRACT: The storage of carbon dioxide in secure underground sites has been identified as an important contributor to preventing climate change impacts of excessive CO_2 in the atmosphere. Significant quantities of carbon dioxide must be collected and stored for this technology to make the required contribution to climate change abatement. The carbon dioxide can be stored as a gas, supercritical fluid or a solid by mineralization. Most attention has been on reservoirs which are depleted in oil or gas or in saline aquifers more generally. Trapping gas or supercritical fluid requires a setting similar to the natural trapping of gases in sub-surface sedimentary rock reservoirs. The most advanced high-volume projects focus on the trapping of supercritical fluids. Pumping carbon dioxide into basalt host rocks to promote carbonate mineralization is also being undertaken. In this paper another environment in which CO_2 is naturally introduced into geological materials and stored below ground is reviewed. The environment is natural hydrothermal systems which are commonly found around magmatically active locations. In these areas it is common for the cool near-surface (meteoric) water to descend as a replacement (recharge) of the warm or hot magmatic water in the hydrothermal-magmatic system. The descending CO_2 can react with existing rocks and minerals to form carbonates of calcium and other cations. It is proposed that introduction of CO_2 into naturally descending meteoric water recharge in hydrothermal systems may be a mechanism for CO_2 sequestration by mineral trapping. Further investigation of this process and its potential is proposed.

Keywords: CO_2 Sequestration, Geological Storage, Saline Aquifers, Carbonate Mineralization, Geothermal Systems

1 INTRODUCTION

International studies of pathways to moderating climate change recognize the need for reduction in atmospheric carbon dioxide (CO_2) by use of geological storage [1]. The most advanced methods of geological storage of CO_2 are those that build on knowledge and technology of the oil and gas industries. In particular, the injection of supercritical CO_2 into sedimentary rock reservoirs has been demonstrated as a feasible technology [2]. The injection of dissolved gas into reactive rocks such as basalt has also been demonstrated as a feasible technology, albeit at a smaller scale [3].

Water plays an important role in the transfer of heat and matter in geothermal systems [4]. CO_2 can become fixed within carbonate minerals, for example calcite, dolomite and magnesite through hydrothermal alteration, which is a common occurrence in many parts of the Earth's crust [5]. Water also plays a significant role in the transfer of pressure and the initiation of movement on brittle structures [6]. In this way water can contribute to a fault-valve cycle and once fluid pressure exceeds the lithostatic load, accumulating shear stresses trigger failure on faults. This is supported by observed relationships of faults and other evidence of fluid-pressure cycling within gold veins and other metallic deposits [5-6].

The process of hydrothermal alteration which transforms large volumes of rock into different mineral assemblages has occurred through geological history including the formation of greenstone belts. Such belts, surrounded by extensive granite, may be 300 m to 12 km wide and 100 to 700 km long and the presence of carbonate minerals in greenstone belts is well known [7].

In this paper, challenges and opportunities for geological storage of CO_2 are reviewed. In particular, the need for greater application of knowledge of the permeability and reactivity of a range of geological settings is addressed. It is proposed that introduction of CO_2 into naturally descending meteoric water, which commonly recharges hydrothermal systems, may be a mechanism for CO_2 sequestration by mineral trapping. Further investigation of this process and its potential is proposed.

The transition to supercritical CO_2 with pressure means that efficient storage can be achieved at depths where pressure is sufficient to maintain the supercritical condition [8].

As supercritical CO_2 is injected into an aquifer flow initial follows Darcy's Law. However, Darcy flow soon gives way to Invasion Percolation flow behavior. Invasion Percolation flow is dominated by capillary and buoyancy effects. The unit-less Capillary Number can be used to define the transition of the flow mechanisms [8].

2 MINERALIZATION

Many of the existing projects intend to store CO_2 by trapping the gas within pore space. Some mineralization may incidentally occur. Other projects are focusing on the potential to form minerals with CO_2 as a form of storage (Fig. 1).

2.1 Shallow Underground

The CarbFix project in Iceland is a well known site where carbonate mineralization is the main focus as a carbon storage mechanism [9]. The CO_2 is typically not in a supercritical state as the depth of injection is down to about 750 m. The gas is mixed with and/or dissolved in water which is understood to enhance the mineralization process. The abundance of calcium in basalt promotes the formation of calcium carbonate (calcite).

Studies on the natural carbonate mineralization systems in the basalt have also contributed to understanding of the precipitation mechanism [10].

2.2 Deep Underground

Other projects have targeted gaseous storage and mineralization with supercritical injection of CO_2 into basalt [11]. In the Columbia River basalts brecciated zones between lava flows at depths between 800-900m were injected with supercritical CO_2 [12].

Approximately 1000 tonnes of CO_2 were injected over a 25-day period in July to August of 2013. Testing two years later identified free phase supercritical CO_2 fluid in the vicinity of the injection site. Carbonate mineral (ankerite, a mixed Ca-Mg-Fe carbonate) nodules were also recovered from the core sampling and could be directly linked to the injected CO_2 through analysis of isotopes [12].

2.3 Reactive Rocks

As pointed out by the proponents of the Carbfix project, Iceland is a location where oceanic crust is present above sea-level [9]. They emphasize that the entire oceanic crust is basaltic rock with CO_2 mineralization potential. There are other geological environments with potentially reactive rocks with permeability (Fig. 2).

3 FORMATION OF CARBONATE MINERALS

Geothermal systems such as Wairakei in New Zealand have been observed to form deposits of calcite mineral scale in wells [13]. Some other wells in geothermal systems produce no calcite scale and are inferred to be a result of waters entering the wells being undersaturated in CO_2 or suitable cations. It is inferred that, during recharge of shallow water deeper into the geothermal reservoir, an insufficient quantity of CO_2 was transported into the system to saturate the water with calcite. [4].

Carbonate minerals are commonly found within the rocks affected by hydrothermal systems. These minerals are found over a wide range of pH and temperature conditions. Carbonates are also found in association with a wide range of other minerals (Table 1). The types of carbonate minerals, as determined by their cations, can also vary with increasing fluid pH. This effect has been observed as zoning of carbonate minerals as has been observed in some hydrothermal systems [14]. Based on these observations, it has been inferred that Fe, Mn and Mg become less mobile as pH increases toward neutral.

The solubility of carbonate minerals in an aqueous solution at pH 4-8 can be represented by Eq. (1):

$$MCO_3+CO_2+H_2O=M^{2+}+2HCO_3^- \quad (1)$$

Where M represents the cations: Ca, Mn, Mg, Fe.

Table 1 Associations of carbonate and clay minerals [14]

Carbonate cations	Mineral names	Assoc. minerals	pH
Fe, Mn	Siderite, rhodocrosite	Kaolin, illite	Low
Ca, Mn, Mg, Fe	Rhodocrosite, ankerite, kutnahorite, dolomite	Illitic and chloritic clays	Mod.
Ca, Mg	Dolomite, calcite	Chlorite, calc-silicates	Neutral

At pH less than 4 carbonates are dissolved. At temperatures less than 100ºC and neutral conditions (pH 6-7) bicarbonate dominates over CO_2. The controls on carbonate solubility under high

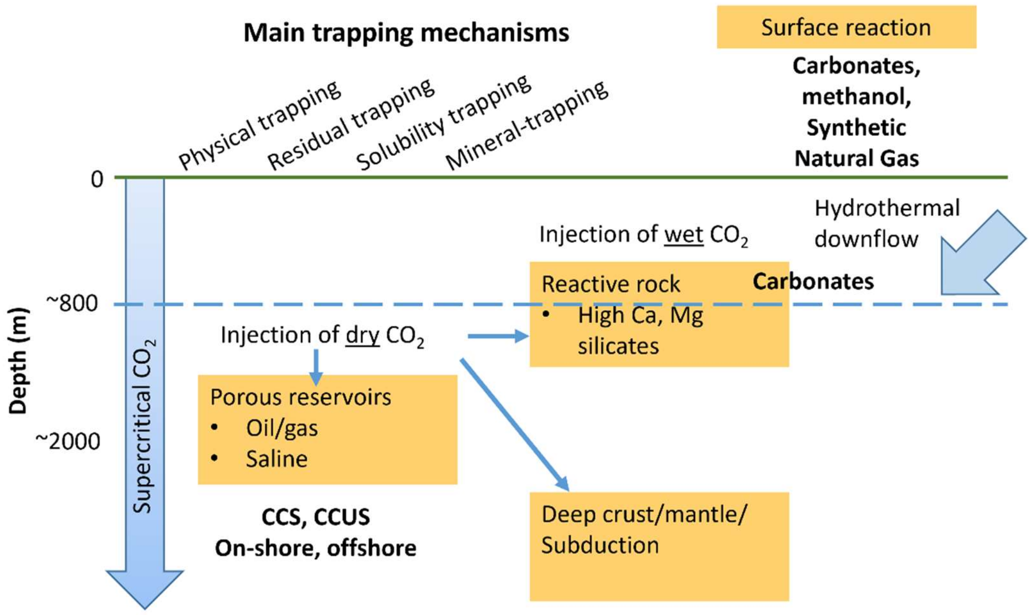

Fig.1 Conceptual summary of some methods of carbon dioxide trapping relative to depth

temperature saline conditions is the subject of current research. In general, it is known that calcium carbonate solubility decreases with increasing temperature [15].

Carbonate minerals are present in all levels in hydrothermal systems from low temperature surficial conditions to deep magmatic conditions. The dominant control on carbonate deposition is increasing temperature. In comparison, dilution and pressure drop are secondary factors in affecting carbonate deposition. Therefore, the heating of descending CO_2-rich waters leads to carbonate mineral deposition [14].

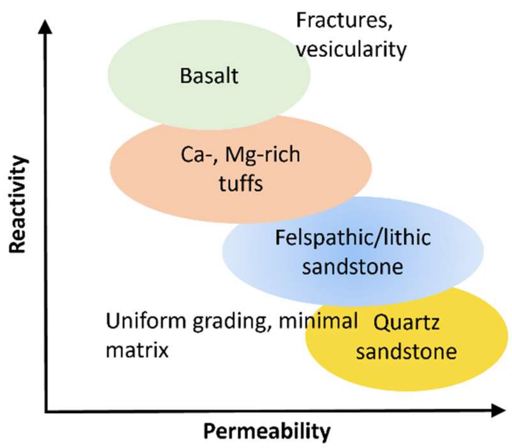

Fig.2 Conceptual summary of some examples of geological conditions for sequestration of CO_2

If a fluid with significant concentrations of dissolved CO_2 undergoes boiling, carbonate deposition can also occur. The most common cause of boiling in nature is the reduction in pressure rather than an increase in temperature. Such a reduction in pressure can occur gradually by ascending fluids having decreased overburden or rapidly by faulting or large-scale landslides [14].

A comprehensive review of hydrothermal systems in the southwest Pacific rim [14] showed that many of these systems incorporate meteoric water recharge.

4 METEORIC WATER RECHARGE IN HYDROTHERMAL SYSTEMS

Hydrothermal/geothermal systems can be characterized according to the chemistry of the water emanating from a magma intrusion. Geothermal systems can also be characterized by the depth of the magma intrusion and therefore the distance from the heat source to the geothermal activity observed near the surface. The class of geothermal systems known as 'epithermal' refer to the significant distance that can exist between the heat source (magma intrusion) and the ground surface. The epithermal system refers to the areas in which near surface waters circulate through the ground such that the chemistry of the waters is a mixture of juvenile magmatic water and groundwater known as 'meteoric' water. In these epithermal systems it is common for CO_2-rich water to descend from above an intrusion and mix with magmatic fluids in the conduits [14]. The conduits of fluid flow may be permeable rocks or structures such as faults. The magmatic fluids typically contain dissolved reactive gases. The chemistry of the fluids can be affected by interaction with rock and dilution by the circulating meteoric waters. The meteoric waters typically capture a significant amount of the magmatic gases, including CO_2, as the mixing occurs [14].

4.1 Mineral Deposition

The cool, descending, low pH, CO_2-rich meteoric water mixes with hot, silica-saturated, deep magmatic fluids resulting in deposition of carbonates in response to increasing temperatures [14]. Hydrothermal systems are commonly located in tectonically active areas such that major fracture zones and faults can be periodically reopened by fluid pressure permitting both the ascent of magmatic fluids and the descent of meteoric water to progressively deeper levels [6].

Where mineral precipitation occurs in permeable rocks, the rock is described as 'altered'. Where mineral precipitation occurs in fractures the rocks are described as 'veined'. The overall effect of these processes is to seal the most permeable zones by precipitation of minerals including carbonates.

A review of geothermal systems in the Philippines showed that cool, low pH, CO_2-rich water has been detected as deep as 2000 m [14]. Evidence that these CO_2-rich waters have mixed with warmer magmatic waters include the precipitation of carbonates with a range of cations including dolomite (Ca, Mg-carbonate), siderite (Fe-carbonate) and ankerite (Mg, Ca, Fe-carbonate).

Observations of the occurrence of various carbonate minerals at depths of 600-1200 m in the Broadlands hydrothermal system in New Zealand are also considered to be indicative of the drawing down of CO_2-rich waters into a saline aquifer dominated by magmatic water [14]. At Rotakawa in New Zealand, the occurrence of Mn-carbonates has been inferred to be the result of deep circulation of CO_2-rich surficial waters [14].

In summary, calcium carbonate (calcite) can be deposited from either magmatic or meteoric waters. However, magmatic fluids are typically depleted in magnesium and therefore the presence of Mg-carbonates (dolomite, ankerite) at deep levels in active geothermal systems is inferred to be related to the incursion of CO_2-rich meteoric waters into deeper parts of magmatic hydrothermal systems [14].

4.2 Evolution of Hydrothermal Systems

Hydrothermal systems pass through a series of stages, principally driven by the cooling of the intrusive pluton. According to [14] the stages are evidenced by the following features: 1) contact metamorphism, 2) release of a magmatic volatile plume, 3) convective hydrothermal alteration and 4) draw down of surficial waters.

Meteoric waters can be involved in each stage but become more dominant as the system evolves. In the earlier stages the descent of surface water occurs farther away from the thermal center and in the final stage the descent of water can occur directly on top of the waning thermal system [14].

This evolutionary model of hydrothermal systems provides a framework for studying and understanding individual examples of hydrothermal systems. The evolutionary model applies to active hydrothermal and geothermal systems and ancient systems preserved (fossilized) in rock. Both active and ancient systems are of great interest as they are a common host to the formation of metallic mineral deposits. Many of the ancient systems can be hosted within active hydrothermal areas. For example, a magmatic intrusion which has become fully cooled will be surrounded by its fossil hydrothermal system. If further magmatic intrusions occur, complex overprinting of the rock alteration and mineralogy will occur.

A summary of active and ancient hydrothermal systems in which evidence of descending meteoric waters has been observed is provided in Table 2.

4.2.1 Composite Volcanic Terrains

An example of descent of water toward a magmatic volatile plume is the Alto Peak system in northern Leyte, Philippines. In that system the magma is over 2 km below surface (Fig. 3). There is deep meteoric water recharge from multiple kilometers around the magma intrusion to around 2 km depth in the system. Concurrently, in shallower parts of the system meteoric water descends hundreds of meters to form a wide-spread zone of CO_2-rich water.

An example of descent of water toward a zone of convective hydrothermal alteration is the Tongonan system also located in northern Leyte, Philippines. In that system the magma is within 1 km below surface (Fig. 4). There is deep meteoric water recharge from multiple kilometers around the magma intrusion to around 2 km depth in the system. Concurrently, in shallower parts of the system, meteoric water descends hundreds of meters along major faults.

Examples of near-surface water being drawn down to deeper levels are the Bacon-Manito and Palinpinon geothermal fields in the Philippines. This final stage of geothermal waning occurs as the pluton cools. In this stage cool dilute meteoric waters and shallow, low pH and CO_2-rich waters descend to considerable depth within the reservoir of saline groundwater. The Palinpinon geothermal field is driven by the intrusion and cooling of a large monzonite intrusion [14].

One of the characteristics of a waning geothermal field is that CO_2-rich water can be descending directly at the center of the field as is occurring in the fault zones that mark the focus of the Palinpinon geothermal field. A pluton emplaced during the Miocene initiated the system and progressively cooled. Smaller and younger intrusions have occurred on tectonic structures. These younger intrusions and deeper extensions of them, provide the current heat source for the geothermal field.

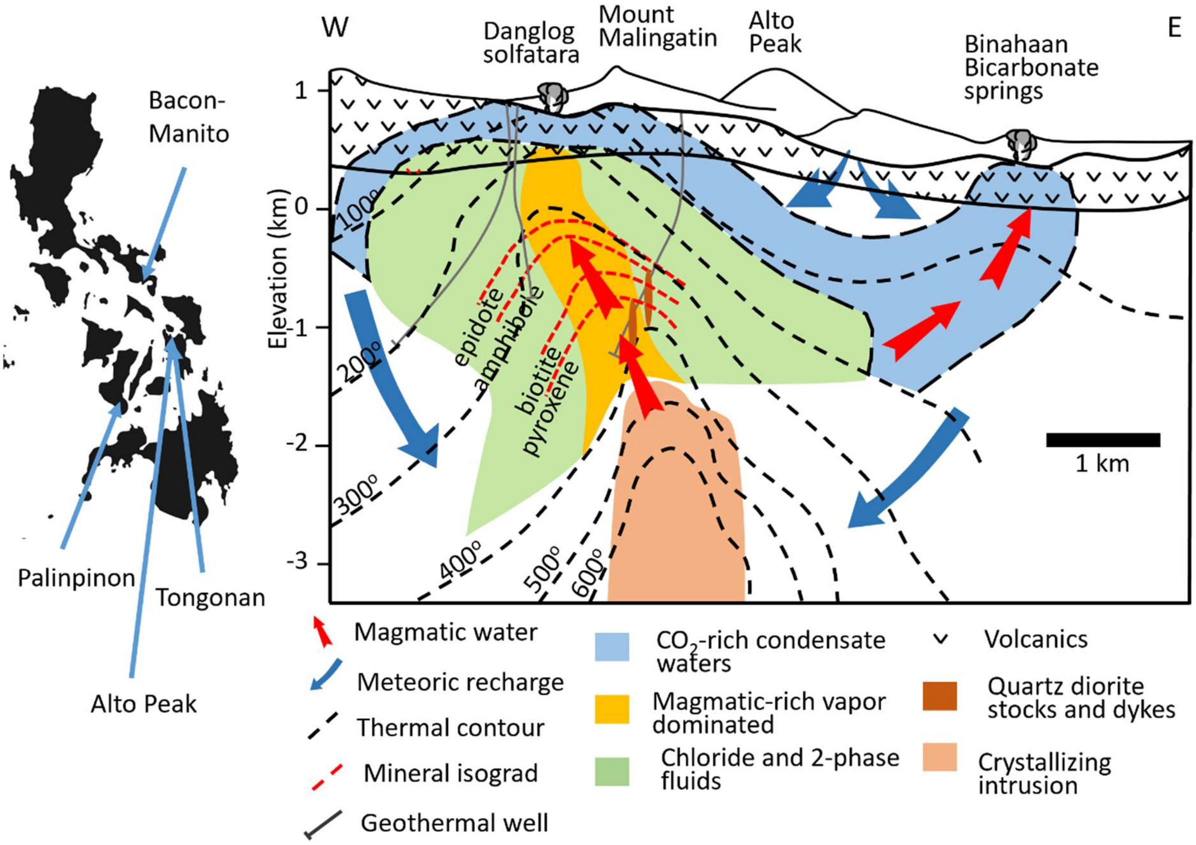

Fig. 3 Left) Map of the Philippines showing locations of examples. Right) Alto Peak hydrological model [14]. Descending meteoric waters shown in blue arrows. CO_2-rich waters shown in blue

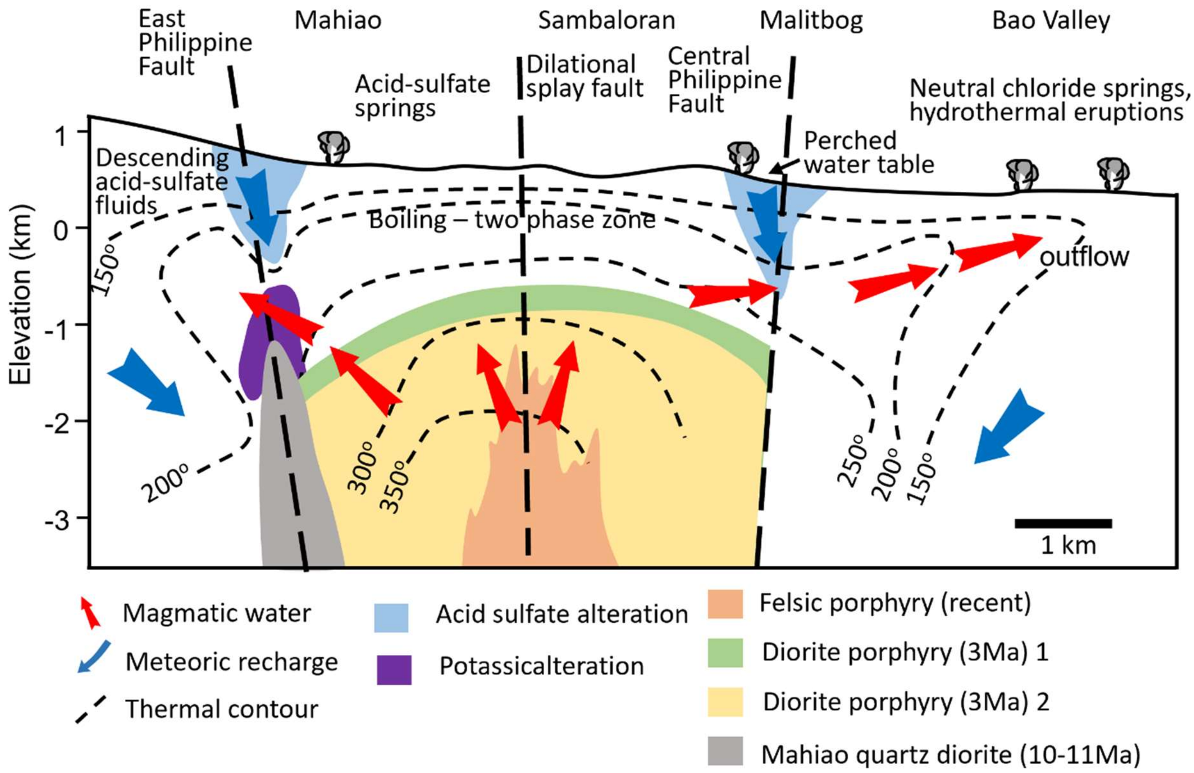

Fig. 4 Tongonan (Philippines) hydrological model [14]. Descending meteoric waters shown in blue arrows

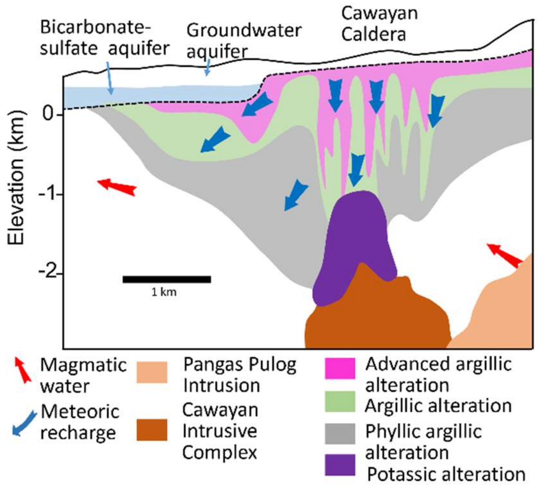

Fig. 5 Bacon-Minato (Philippines) hydrological model [14]. Descending meteoric waters shown in blue arrows

In the Palinpinon geothermal field, meteoric waters also move laterally through permeable strata toward the center of the geothermal field. According to [14] the Palinpinon geothermal field is in an early stage of waning or thermal collapse such that cool CO_2-rich meteoric waters are descending in response to pressure draw down. The dilute nature of the fluids in the central part of the system show that the meteoric waters are dominant over the residual presence of magmatic water.

The Bacon-Manito geothermal field in southern Luzon, Philippines, has a complex volcanic and intrusive geological history which has been interpreted as a waning system (Fig. 5). It has been observed that CO_2-rich waters move down structures and caldera margins to depths exceeding 1,500 m. This descent of cool waters into hotter environments has resulted in widespread deposition of carbonates and other minerals. As a consequence of mineral precipitation, the upper 1000 m is relatively low in permeability [14].

4.2.2 Cordillera-style Hydrothermal Systems

Volcanic mountain chains, commonly called cordillera are also associated with hydrothermal activity. In the Philippines, the northern Luzon and eastern Mindanao regions can be considered to be cordillera-style hydrothermal systems. In these areas upflow of magmatic water can be restricted to vertical channels. Mixing of magmatic fluid and CO_2-rich surficial water occurs near the surface.

An example of a Cordillera-style geothermal system is the Amacan geothermal system eastern Mindanao, Philippines. Abundant bicarbonate springs with as much as 4 wt% CO_2 occur in the area. This is inferred to be the result of meteoric waters intercepting CO_2 from the magmatic source and redistributing it within the geothermal system [14].

Table 2 Examples active and fossil hydrothermal systems with descending CO_2-rich/meteoric water [14]

Country	System	A/F
Philippines, northern Leyte	Alto Peak, Tongonan	A
Philippines, southern Negros	Palinpinon	A
Philippines, southern Luzon	Bacon-Manito	A
Philippines, eastern Mindanao	Amacan	A
Sumatra, Indonesia	Miwah	A
PNG, Lihir Island	Ladolam	A
Australia, Queensland	Kidston	F
Indonesia, Kalimantan	Kelian	F
PNG	Porgera	F
PNG	Morobe	F
PNG, Woodlark Island	Busai	F
PNG	Maniape	F
PNG	Mt Kare	F
Solomon Islands	Gold Ridge	F
New Zealand, Coromandel	Golden Cross	F
Japan, Kyushu	Hishikari	F

A=active geothermal system, F= fossil hydrothermal system

5 FOSSIL HYDROTHERMAL SYSTEMS

One of the most significant sources of information on hydrothermal systems is from those systems where metals have been deposited. Many of these systems are extinct or nearly extinct so they can be considered to be fossil hydrothermal systems.

Meteoric water played a significant role in many such hydrothermal systems. Of the mineral deposit-forming processes the epithermal or more specifically, the low sulfidation porphyry system is one environment in which meteoric water interaction is most crucial.

An example of such a system is the Ladolam gold mine on Lihir Island, Papua New Guinea. The gold mineralization is understood to have formed in an earlier phase of the evolving geothermal field. The current geothermal system is inferred to be in a waning stage. Currently, low pH, CO_2-rich waters are being drawn down into the geothermal system. These waters are understood to be groundwaters that have absorbed gases from the earlier magmatic systems. The temperature and pH of the water near the surface allowed dissolution of carbonate and later re-deposition of the carbonate deeper in the system as the descending water was heated [14].

The Kidston gold mine in Queensland Australia is hosted by a Permo-Carboniferous age volcanic breccia pipe. The sequence and distribution of minerals shows that descending cool, low pH, CO_2-rich waters descended into the hydrothermal system and deposited carbonate minerals [14].

There is an entire class of mineral deposit known as carbonate-base metal-gold systems. These systems have characteristics similar to other hydrothermal systems including the importance of the mixing of rising magmatic water with meteoric CO_2-rich water.

The fluid flow model for these deposits involves hot mineralized fluids which evolve from cooling intrusions. The fluids rise along permeable zones which may be fracture systems. At depth, these fluids mix with circulating meteoric waters. Gases evolve from these upwelling fluids by boiling and vapor loss and are absorbed by groundwater near the surface to form CO_2-rich waters. Cooling of the intrusions leads to draw down of these fluids deep into the hydrothermal system. Carbonate minerals are precipitated as a result.

An example of these processes is the Kelian mine, Kalimantan, Indonesia. In this system carbonate minerals are zoned with depth. Fe and Mn carbonates are encountered at shallow depth whereas Mg and Ca carbonates are found deeper in the deposit [14].

The Porgera deposit in Papua New Guinea is also classified as a carbonate-base metal-gold deposit. Carbonate minerals are zoned in a similar manner to the Kelian deposit. Carbon and oxygen isotope data from carbonates at Porgera support the model of progressive mixing of magmatic and meteoric waters to produce the observed zonation of carbonate minerals [16].

6 POTENTIAL DISPOSAL OF CO_2

It is known that geothermal systems can undergo steam eruptions due to changes in pressure [17-18]. Therefore, any ground engineering activities in a geothermal system must be conducted with careful risk assessment being applied.

Disposal of CO_2 usually involves pumping water through wells. In disposal into oil fields and saline reservoirs the depths of CO_2 introduction are very deep and the gas is in a supercritical state and dry of water. Shallower injection of CO_2 gas can be used where mineral carbonation is the aim. These gases are typically not in a critical state and water can be present with the gas.

It is proposed in this paper that the existing downward flow of meteoric water in natural geothermal systems introduces the possibility of using a syphon system to introduce CO_2 to the ground. This means that highly pressurized pumping may not be needed for the gas and/or gasified water to be introduced to the ground. It is anticipated that a well would be required to ensure that the gas becomes entrained in a downward flow. The depth of the well would depend on the nature of the natural flows.

It is anticipated that meteoric waters that are not currently saturated in CO_2 would be the most favorable for disposal of CO_2. Identification of suitable sites for CO_2 disposal would require a detailed understanding of the geothermal system. This assessment would include assessment of the presence of rock with sufficient cation potential for the formation of carbonate minerals in combination with the introduced CO_2.

Exploration of geothermal systems is a well developed field of research including the use of geochemistry [19] and geophysics [20]. The concept of using existing downward flowing water to sequester CO_2 through mineralization has not been previously considered and may have significant barriers and limitations. It is hoped that this review may prompt further investigation into this possibility.

7 CONCLUSIONS

The critical challenge for geological storage of CO_2 is to store large volumes of carbon so that a significant impact on atmospheric carbon can be achieved. This challenge can best be achieved by continuing with development of storage in large, deep offshore and onshore saline reservoirs. Continued experimentation with deep and shallow reactive rocks is also needed. High calcium and magnesium silicate rocks are abundant in the Earth's crust and where suitable permeability conditions exist – forming a considerable volume of mineralized carbon could be achieved.

Descending water with CO_2 occurs naturally and is common in areas of geothermal activity. It is known that carbonate minerals form abundantly in many of these systems.

It is proposed that active geothermal systems in which descending waters are not saturated in CO_2 could accommodate the introduction of CO_2 for the purpose of reducing atmospheric carbon. Because these are naturally down-flowing waters there is potential to use the syphon effect to draw water enriched in CO_2 into the descending flow without the requirement for highly pressurized pumping.

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