THE POTENTIAL OF GEOTHERMAL EMISSIONS STORAGE IN THE TAUPŌ VOLCANIC ZONE, NEW ZEALAND

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ABSTRACT

New Zealand's low carbon aspirations are in line with the goals of the Paris Agreement to constrain anthropogenic warming to 1.5–2°C. The enhanced utilisation of renewable geothermal resources is crucial to reduce the greenhouse gases emissions sourced from the energy sector that is the third largest industrial emitter in the country after national transport and manufacturing industries. Although more than a magnitude lower in CO_2 release comparing to coal-burning power plants, New Zealand geothermal power stations emit on average 76 gCO_2eq/kWh, which equals to about 530 $ktCO_2/yr$. As a result, necessary efforts must be undertaken to transform the geothermal industry into a neutral or negative carbon emissions energy source. Two solutions are currently developed worldwide to mitigate anthropogenic geothermal CO_2. The first one is the reinjection of the emissions back to the reservoir with the reinjection waters. The second one that is yet to be tested in New Zealand is the CarbFix mineral storage method. The latest is currently developed in Iceland where about 12 kt of CO_2 and 8 kt of H_2S are captured and injected annually into the subsurface. The CO_2 and H_2S are mineralised into carbonate and sulphide minerals as a result of interaction between the reinjected gases and the basaltic host rock that is enriched in divalent cations (Ca, Mg, Fe).

This study aims to investigate three geothermal areas for their CO_2 and H_2S storage potential: Wairakei, Kawerau, and Rotokawa geothermal fields, all located in the Taupō Volcanic Zone, New Zealand. In contrast to the basaltic reservoir at the CarbFix injection site, here, the subsurface consists of silicic tuff, andesitic greywacke, andesite, and rhyolite that are depleted in divalent cations, possibly limiting the CO_2 mineral storage. To address these limitations, geochemical reaction path models were created using the compositions of the geothermal reservoir fluids and rocks. The outcome of the simulations was used to assess the maximum mineral storage capacity of these geothermal sites.

1. INTRODUCTION

Geothermal power production provides renewable, sustainable and green energy. Since 2015, the installed worldwide capacity has grown by 28% to reach 15,950 MW_e in 2020 (Huttrer, 2020). The inevitable part of geothermal power production is the emission of carbon dioxide (CO_2) and other gases such as hydrogen sulphide (H_2S) and methane (CH_4) that occurs naturally in any volcanic derived geothermal system. Although lower comparing to a typical fossil fuel burning plant, which emits ~1100 gCO_2/kWh to the atmosphere, a typical geothermal power plant emits ~122 gCO_2/kWh (Fridriksson et al., 2017). The current world total emissions from the geothermal activity are estimated to be about 10 $MtCO_2/yr$ (Marieni et al., 2018). For comparison New Zealand utilised geothermal systems emit on average 76 gCO_2eq/kWh, that equals to about 530 $ktCO_2/yr$ (McLean et al., 2020). Among the solutions for the anthropogenic CO_2 levels decrease in the atmosphere is to maximise the geothermal utilisation together with carbon capture and storage technologies.

One carbon capture and storage approach is through CO_2 mineralisation in the subsurface (e.g., Oelkers et al., 2008; Snæbjörnsdóttir et al., 2020). For example, the CarbFix method that is based on the separation of the condensable and non-condensable gases from the emission stream and their subsequent dissolution in condensed steam has been developed in Iceland to reduce the emissions during geothermal power generation (e.g., Gislason et al., 2010; Gunnarsson et al., 2018; Matter et al., 2016). The CO_2 and H_2S are dissolved in condensed steam from the power plant and injected back into the geothermal system together with the reinjection water. Subsequently, the CO_2-H_2S mixture dissolves the reservoir host rocks liberating cations including Ca^{2+}, Mg^{2+} and Fe^{2+}. These cations react with dissolved carbonate and sulphide to form calcite ($CaCO_3$) and pyrite (FeS_2). In New Zealand, the reinjection of geothermal gases is currently investigated as a result of the net-zero carbon emissions by the 2050 regulatory framework (Kissick et al., 2021).

The aim of this study is to investigate three geothermal areas within the Taupō Volcanic Zone (TVZ): Rotokawa, Wairakei, and Kawerau geothermal fields for their potential for carbon and sulphur storage. These three locations represent variable geological settings of the reservoir and distinct hydrothermal fluid compositions (e.g. Bixley et al., 2009; McNamara et al., 2016; Milicich et al., 2016, 2020). Chemical reaction path models were used to calculate the sequestration efficiency. This efficiency was compared with the efficiency observed at the CarbFix injection site. In addition, potential consequences of the CO_2 injection on the reservoir composition within TVZ are discussed.

2. STUDY AREA

2.1 Rotokawa

The Rotokawa geothermal field is one of seven geothermal fields utilised for power generation. The steam field is owned by Rotokawa Joint Venture (a 50:50 joint venture between Tauhara North No.2 Trust and Mercury Energy), whereas the Rotokawa Generation (100% Mercury Energy) owns the generation plant. Mercury Energy operates both the station and the steam field. The installed capacity of the Rotokawa power plant is 35 MW_e with an annual generation of about 270 GWh. The Mercury Energy/Tauhara North No 2 partnership also developed the 140 MW_e Nga Awa Purua power station commissioned in 2010 (Sewell et al., 2015). Currently,

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14 wells drilled down to depths of 2000-3000 mVD serve as production wells, and 6 are used as reinjection wells for the two power stations (Pogacnik et al., 2019).

Rotokawa is one of the hottest geothermal systems in the TVZ, with a maximum measured temperature of 337 °C (Hedenquist et al., 1988; Sewell et al., 2015). There are three aquifers identified in the system: (a) shallow meteoric groundwater; (b) intermediate aquifer containing steam heated groundwater, acid-sulphate-chloride fluids, and boiled reservoir fluids; and (c) a geothermal reservoir with temperatures >300 °C below 1000 m depth (Addison et al., 2015; Giggenbach, 1995; Winick et al., 2011). The Rotokawa andesite, with a thickness of up to 2200 m, hosts the geothermal reservoir that consists of a succession of interbedded volcaniclastic, sedimentary and pyroclastic units of the Tahorakuri and Waikora Formation (Milicich et al., 2020; Figure 1).

The main alteration assemblage in the geothermal reservoir is propylitic, generated in the host rocks by high-temperature and near-neutral pH chloride fluids. The alteration mineralogy includes quartz, chlorite, illite, calcite, pyrite, adularia, albite, epidote, clinozoisite, wairakite, and accessory hematite (Krupp et al., 1986; Krupp and Seward, 1987; Price et al., 2011). This mineral association overprints a higher temperature assemblage which includes actinolite and biotite (Chambefort et al., 2011). Most of the sulphate minerals are also identified to depths of ~1600 m (Hedenquist et al., 1988), although deep anhydrite has been recognised below 2000 mVD in the Rotokawa Andesite (Chambefort, 2021). The estimated surface diffuse degassing of CO_2, and H_2S are 441 t/ d and 31 t/d, respectively (Bloomberg et al., 2014). The Rotokawa and Nga Awa Purua geothermal power plant emissions are equal to 65 and 181 tCO_2/day, respectively (McLean et al., 2020).

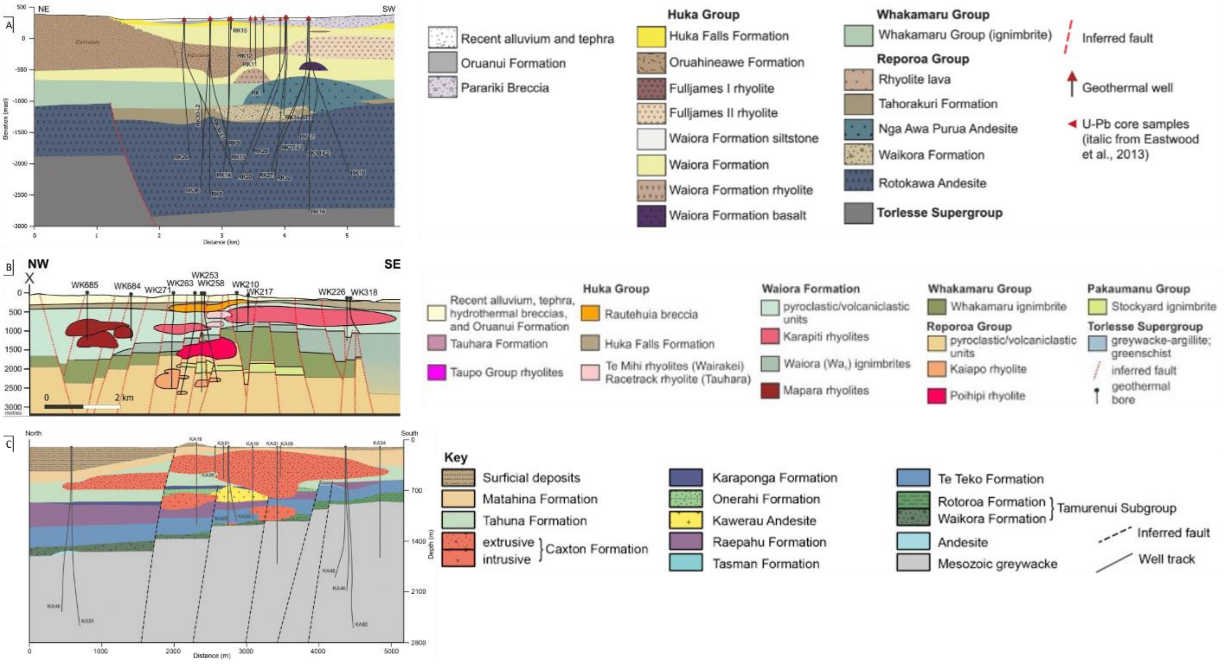

Figure 1: Geological cross-sections of the Rotokawa (A), Wairakei (B), and Kawerau (C) geothermal fields, together with the production and reinjection wells. Modified from (Milicich et al., 2016; Milicich et al., 2020; Rosenberg et al., 2020).

2.2 Wairakei

Wairakei geothermal field is New Zealand's oldest utilised field with an installed capacity of 362 MW_e. The Wairakei, Te Mihi and Poihipi power stations are owned and operated by Contact Energy Ltd. Currently, there are 73 production and 19 reinjection wells (NZGA, 2021). Most of the early wells were fed by the shallow aquifers <1000 m, whereas the newest wells reach depth to about 3000 m (Glover and Mroczek; 2009 Thain et al., 2009). The deepest feed zone is at 2880 m. The highest measured temperature was 272 °C at 1100 m (Ardid et al., 2021 and references therein). The total pressure in the main aquifer after 60 years of production decreased by 26 bar_g, whereas temperature declined on average by 20 °C (Glover and Mroczek, 2009). The shallow aquifer temperatures have decreased by 100 °C. In addition, the pressure drawdown in the deep reservoir triggered compaction within the clay-altered cap rock and ground subsidence.

The rhyolite lavas, rhyolitic tuffs and breccias of the Waiora Formation are the major production aquifers at the Wairakei–Tauhara and Ohaaki fields (Rosenberg et al., 2009). Propylitic alteration is predominant in the Waiora Formation strata and rhyolite lavas with chlorite, quartz, epidote, albite, adularia, wairakite, titanite, clinozoisite and/or zoisite. The highest rank assemblage includes wairakite, epidote and prehnite that indicates water-rock interaction with near-neutral (to slightly acidic) pH fluids at temperatures above 240–280 °C. An overprint of the propylitic-type alteration was observed through illite and/or calcite altering hydrothermal albite and adularia and illite replacing chlorite (Rosenberg et al., 2009).

Table 1: Key characteristics of the geothermal systems that are investigated in this study. Data were taken from McLean et al. (2020) and Millicich et al. (2016).

Parameter	unit	Rotokawa	Wairakei	Kawerau
Reservoir temperature	°C	<320	240-260	225-308
Field area	km^2	18	25	22
Generation capacity	MW$_e$	175	362	160
CO_2 emissions	kt/d	246	244	394

Table 2: The whole-rock compositions in oxide wt% used for reaction path modelling.

	Rotokawa	Wairakei	Kawerau
		wt%	
SiO_2	54.8	75.4	61.4
Al_2O_3	16.5	12.5	15.6
FeO*	6.09	2.53	6.61
MnO	0.15	0.07	0.11
MgO	9.03	0.26	2.3
CaO	3.30	1.96	4.67
Na_2O	2.36	3.06	4.23
K_2O	0.99	3.74	1.88
TiO_2	0.69	0.26	0.79
P_2O_5	0.13	0.04	0.17

2.3 Kawerau

There are four major consent holders that utilise Kawerau geothermal resources: Mercury NZ Limited, Ngāti Tūwharetoa Geothermal Assets Limited, Te Ahi O Māui Partnership, and Geothermal Developments Ltd. They operate Kawerau (KGL), TOPP1, TAOM, and GDL geothermal power plants, respectively (McLean et al., 2020). The total installed capacity of these plants is 151 MW$_e$. To date, more than 70 wells have been drilled, although no more than 6–7 have been in production at any one time, with many of the older wells now used for field monitoring or plugged and abandoned (Milicich et al., 2016). Production reservoirs in the older boreholes are at around 1 km depth, while the recent wells produce from the greywacke basement.

The greywacke at Kawerau with low matrix porosity hosts both geothermal production and reinjection, with fluid flow localised in fault and fracture networks (Milicich et al., 2016). It consists of Mesozoic medium-grained sandstone dominated by subangular volcanic clasts (mainly andesite lava, rare rhyolite lava and ignimbrite), with minor argillite (Wood et al., 2001). The mineral alteration is characterised by a ‘typical’ neutral-pH hydrothermal assemblage consisting of epidote, quartz, calcite and illite, which indicate temperatures exceeding 240 °C (Bignall, 2010). Commonly, the occurrence of these high-temperature minerals is consistent with measured well temperatures. Similar to those from Rotokawa, the reservoir fluids are gas-rich (1.2-4.5 wt% gas in the steam) with the chloride concentration up to about 880 ppm and calculated deep temperature from 225 to 308 °C (Christenson, 1997). The composition of the production fluids has changed in response to utilisation. This includes dilution, declining temperatures, particularly in the shallow reservoir, and precipitation of calcite in wells and associated feed zones due to reservoir degassing. Increasing chloride, silica, calcium, and decrease in the gas content at a relatively consistent enthalpy are observed in some production wells as the response from the deep reinjection (Milicich et al., 2016). The estimated power plant emissions are 394 tCO_2/day, with the KGL contributing about 318 tCO_2/day at the generation capacity of 107 MW$_e$ (McLean et al., 2020).

3. METHODOLOGY

Reaction path calculations were carried out to assess: 1) changes in fluid chemistry and mineralogy due to the reinjection of a CO_2 and H_2S rich fluid between the reinjection and production wells, and 2) the efficiency of CO_2 and H_2S sequestration as a function of reaction progress. In these calculations, the host rock that represents the geothermal reservoirs in the studied areas was dissolved stoichiometrically into the reinjection fluid in fixed steps, and secondary minerals were allowed to form at local equilibrium. The mass of host rock dissolved into 1 kg of H_2O was used to define the reaction progress. The chemical formula of the rock is consistent with 1) $Si_{1.0}Ti_{0.009}Al_{0.354}Fe_{0.093}Mg_{0.246}Ca_{0.065}Na_{0.083}K_{0.023}Mn_{0.002}O_{3.008}$ for altered andesite in Rotokawa with the molar mass of 103 g/mol, 2) $Si_{1.0}Ti_{0.003}Al_{0.195}Fe_{0.028}Mg_{0.005}Ca_{0.028}Na_{0.079}K_{0.063}Mn_{0.001}O_{2.431}$ for altered rhyolite tuff in Wairakei with the molar mass of 79 g/mol, and 3) $Si_{1.0}Ti_{0.010}Al_{0.299}Fe_{0.090}Mg_{0.056}Ca_{0.081}Na_{0.134}K_{0.039}Mn_{0.002}O_{2.784}$ for greywacke in Kawerau with a molar mass of 96 g/mol. The kinetics of mineral dissolution and precipitation was not considered in the calculations due to uncertainties related to the kinetic rate equations and the surface areas of the primary and secondary minerals. The secondary minerals allowed to form in the reaction path simulations were those observed in the reservoirs. These minerals included carbonates (calcite, ankerite, siderite), sulphides (pyrite and pyrrhotite), chlorites, actinolite (tremolite and ferroactinolite), albite, K-feldspar, epidote, clinozoisite, prehnite, hematite, titanite, and quartz. The chemical composition of reactive fluids was represented by the geothermal separated waters that are currently reinjected in Rotokawa, Wairakei, and Kawerau. The plans call for injecting the CO_2 and H_2S gases together with the reinjection fluid. As no data exists for New Zealand, the concentration of CO_2 and H_2S in the reinjected fluid was equal to these concentrations used at the CarbFix injection site. There, the CO_2-H_2S charged fluid reaching the target aquifer has a pH of 4.7 at about 100 °C with the concentration of dissolved CO_2 and H_2S of 50.6 mM and 31.3 mM, respectively. The reservoir temperature in the subsurface varies across the geothermal reservoirs. Therefore, to compare the carbon and sulphur mineral storage capacity between the reservoirs, the reaction temperature was chosen to be 260 °C which is the reinjection aquifer temperature at the CarbFix site (Marieni et al., 2021).

4. RESULTS

The CO_2 and H_2S charged reinjection fluids have pH of 5.83, 4.85, and 5.50 at 260 °C for Rotokawa, Wairakei, and Kawerau, respectively. These fluids are calculated to be supersaturated with respect to sulphides and undersaturated with respect to calcite, tremolite, and chlorites. The saturation state with respect to other minerals of interest depends on the reinjection site. At Rotokawa, where the reinjection will target Rotokawa Andesite, the reinjection fluid is supersaturated with respect to albite, epidote, ferroactinolite, hematite, K-feldspar, clinozoisite, and wairakite. It is close to equilibrium with respect to ankerite, prehnite, and siderite. At Wairakei, where the reinjection reservoir is hosted by a rhyolitic tuff, the fluid is undersaturated with respect to the minerals of interest apart from albite, K-feldspar, and quartz that are calculated to be close to equilibrium. At Kawerau, where the reinjection aquifer is the Waipapa terrane greywacke, the fluids are supersaturated with respect to albite, K-feldspar, and wairakite, and undersaturated with respect to other minerals. This suggests that the CO_2 and H_2S reinjection fluids have the potential to dissolve Ca, Fe and Mg bearing silicate minerals including epidote, prehnite, chlorites, and clinozoisite at the Wairakei and Kawerau upon the reinjection.

The CO_2-H_2S charged reinjection fluid was reacted with the host rock at a temperature of 260 °C. The reservoir rock was titrated into the reinjected fluid, and the secondary minerals were allowed to precipitate at local equilibrium if they attained saturation with respect to the fluid phase at each step of the reaction path model. No mixing of the reinjection water with the reservoir fluids in the subsurface is assumed in the model. A close match between some of the constituents in the modelled fluid composition after the dissolution of 0.2 moles of host rock and the reservoir fluid (Mroczek et al., 2015) was observed. For instance, the modelled aqueous Na concentration equals 1100 ppm at Wairakei, and 605 ppm at Kawerau comparing to 905 and 518 in their production aquifers, respectively. In contrast, the modelled fluid concentrations are somewhat higher for Mg and lower for SiO_2 than those observed in the production aquifer. The poor agreement with some of the reactive fluid constituents and the reservoir fluids likely stems from the effects of reinjection fluid mixing with the formation fluids. The resulting mineral assemblages in the reaction path modelling agree to those seen in the geothermal reservoirs of the TVZ, such as quartz, albite, sulphides, and chlorites. Quartz constitutes the greatest molar abundance among these secondary phases.

The CO_2 and H_2S mineralisation efficiency is defined here as the ratio between molar abundances of carbonate and sulphide minerals after dissolution of 0.2 mol_r/kg_w of host rock to the initial CO_2 and H_2S concentrations in the CO_2-H_2S-charged reinjection fluid. At 260 °C, and with chosen secondary minerals the calculated **CO_2 mineralisation efficiency** is **0.7 %** for Rotokawa, **0%** for Wairakei, and **12%** for Kawerau. **The H_2S mineralisation efficiency** is almost **100%** for Rotokawa and Kawerau, and **34%** for Wairakei.

5. DISCUSSION

The maximum CO_2 mineralisation of the injected fluid is calculated for Kawerau (12 %), whereas for Wairakei no CO_2 is predicted to mineralise. For comparison at the CarbFix injection site, the CO_2 mineralisation is about 60%, as observed after 3.5 year of continued injection (Clark et al., 2020). The low mineralisation potential of the TVZ host rocks, comparing to CarbFix injection site, stems from the differences between the chemical compositions of the dissolving host rock. The CarbFix site is characterised by a basalt-hosted reservoir with the general chemical formula of $SiTi_{0.024}Al_{0.358}Fe_{0.188}Mg_{0.281}Ca_{0.264}Na_{0.079}K_{0.008}O_{3.370}$ when normalised to one Si (Marieni et al., 2021). It has a higher molar mass and contains greater amounts of the divalent cations, Ca, Mg, and Fe, than any of TVZ host rocks. In addition, basalts are basic rocks and therefore, the reactive fluid pH increases substantially when dissolving, providing sufficient pH conditions for carbonate precipitation. The pH after the dissolution of 0.2 mol_r/kg_w of basalt increases to about 7.5 (Marieni et al., 2021), whereas the pH after the dissolution of the same amount of host rock is 4.3 using Rotokawa Andesite, 5.6 using Wairakei Rhyolitic tuff, and 6.4 using Kawerau greywacke. At these lower pH, the dominating CO_2 species is the $CO_{2(aq)}$. Note that similar secondary minerals were chosen to precipitate during the simulation of the CarbFix injection (Marieni et al., 2021). Here significant amounts of calcite were calculated to form at pH >7.3, consistent with the observations reported by Clark et al. (2020) where calcite precipitated when sufficient amount of basalt dissolved to neutralise the acidic reinjection fluids. Due to the formation of Ca-bearing silicate minerals, carbonate minerals precipitation is limited at temperatures of 260 °C.

The results of the simulations show that the reinjection into the production aquifers within TVZ at 260 °C will result in insignificant CO_2 mineralisation. Studies on the natural occurrence of carbonate in basalt-hosted geothermal fields show that occurrence of carbonate minerals decreases at greater depths and higher temperatures (e.g. Wiese et al., 2008). Therefore, the reinjection into lower temperature parts of the geothermal areas within a host rock containing the highest amount of divalent cations might be an optimum solution for CO_2 sequestration in the TVZ. This might not correspond to the current locations of reinjection wells. Lower temperatures enhance mineralisation of carbon and sulphur as fewer Ca-bearing non-carbonate minerals and fewer Fe-bearing non-sulphide minerals form (Marieni et al., 2021).

Further efforts are also needed to assess the degree to which the reinjection of CO_2 and H_2S into TVZ geothermal reservoirs will affect the carbon and sulphur budget of this system. One might expect an increase in CO_2 fluxes in the fluid phase by the reinjected CO_2 amounts in active volcanic systems. These increased fluxes might result in the increased CO_2 soil degassing and increased CO_2 content in the production fluids. As such, efforts to enhance CO_2 mineralisation may be necessary to maximise the long-term viability of carbon storage in this volcanic system. In contrast, as the H_2S mineralisation efficiency is close to 100% in Rotokawa and Kawerau and it is likely that the reinjection of this gas will have a little long-term effect on the current H_2S gas emissions from the geothermal system.

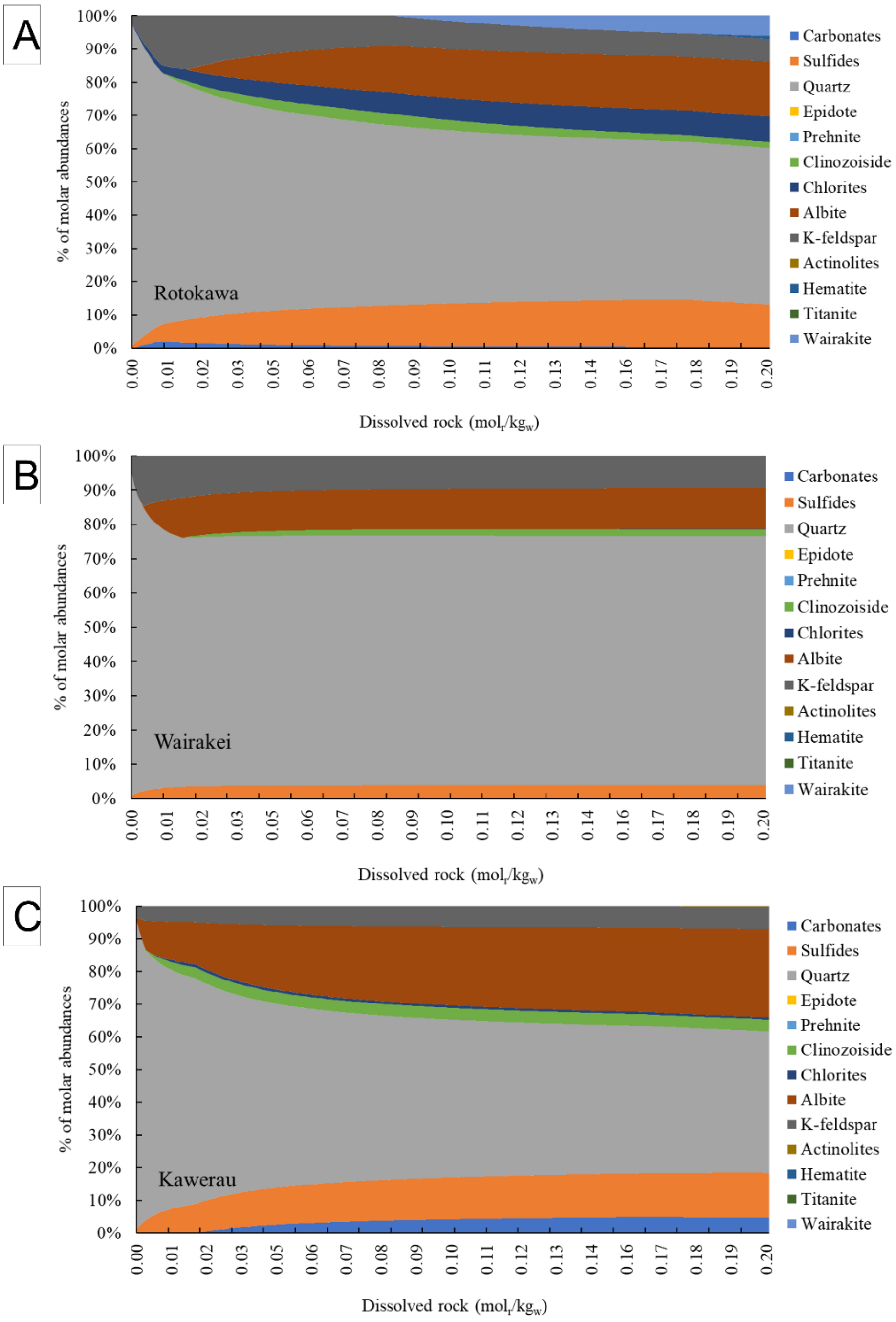

Figure 2: Results of the reaction path model calculations simulating the CO_2-H_2S charged reinjection water interacting with host rock along the flow path from the reinjection well towards the production reservoir. Note that these models do not take into account the subsurface fluid mixing of the reinjected fluid with the reservoir fluid.

6. SUMMARY

The results of this study provide following conclusions:

- The mineralisation efficiency calculated for Rotokawa, Wairakei, and Kawerau at the chosen reservoir conditions is insufficient to consider mineralisation as the efficient CO_2 storage method. The greatest CO_2 mineralisation efficiency was calculated to be about 12% for Kawerau reinjection site.
- Low mineralisation efficiency might result in the increased CO_2 content in the reservoir. This can result in higher than currently CO_2 surface emissions over time.
- Therefore, efforts need to be made to maximise reinjected gas mineralisation before the gas-charged reinjection fluids reach production wells.
- The optimisation of the CO_2 and H_2S reinjection conditions involves choosing the reinjection wells that tap lower temperature hydrothermal reservoir with the host rock that contains greater abundances of divalent cations. This might be not possible with currently used reinjection wells.

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