

Applications of Sulfur Isotopes in Characterizing Reservoir Processes at the Kawerau Geothermal Field, New Zealand

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ABSTRACT

Since its early development in 1957, some areas of the Kawerau reservoir have changed in response to production, mainly experiencing cooling brought about by inflow of marginal recharge and injection returns. The chemical signature of these reservoir processes are normally reflected in the fluid chemistry of the producing wells. However, due to the different characteristics of the Kawerau injection fluid (i.e. acid-dosed brine, oxygenated steam condensates or mixed brine and condensate) identifying the source and main reservoir process responsible for the change in fluid chemistry over time is a complex undertaking.

The use of isotopes to enhance the assessment of the reservoir processes affecting the Kawerau field was considered in this paper. Fluid samples from selected production wells and shallow/intermediate aquifer monitoring wells were collected and analysed for their isotopic composition. These include sulfur isotope ($\delta^{34}\text{S}$ in sulfate and sulfide in brine, and $\delta^{34}\text{S}$ in sulfide in steam) and stable isotopes such as deuterium ($\delta^2\text{H}$ in water in brine and steam) and oxygen-18 ($\delta^{18}\text{O}$ in water in brine and steam, $\delta^{18}\text{O}$ in sulfate in brine).

The isotopic sulfur composition of the production wells at Kawerau displayed similar features to many geothermal reservoirs in the TVZ, such as a homogeneous composition of $\delta^{34}\text{S}$ in sulfide of around 5 to 7 ‰ and an apparent sulfur isotopic disequilibrium state between the sulfate and sulfide species. The results of a mixing model using reservoir sulfate and the $\delta^{34}\text{S}$ in sulfate have shown that injection return is the prevailing reservoir process on the eastern side of the reservoir (e.g. in PK06 and PK07) while mixing with marginal recharge predominates on the western side of the field (particularly in KA30).

Used in conjunction with the hydrogeological, geological and geochemical data, sulfur isotopes proved to be very useful in the case of the Kawerau field, particularly in identifying the reservoir processes affecting the field. Moreover, the additional information from this study helped in creating a robust reservoir management plan for the field.

1. INTRODUCTION

The Kawerau geothermal field lies at the northern end of the Taupo Volcanic Zone (TVZ) of New Zealand. It covers an area of approximately 20 km². Exploration drilling began as early as 1951 and more than 70 wells have been drilled to date.

The Kawerau reservoir host rocks are comprised of a succession of volcanic lavas, pyroclastics and lacustrine/fluviatile deposits overlying a thick greywacke basement. While the permeability in the shallow part of the

reservoir appears mostly homogeneous in nature, the permeability in the greywacke is primarily fracture-controlled, associated with the structural features of the basement. Similar to the other high-temperature systems found in the TVZ, the deep reservoir fluid is dilute ($\text{NaCl} < 1,500 \text{ mg/kg}$), slightly alkaline (reservoir pH~6.8) and gas-rich with up to 1% by weight gas in total discharge. The measured reservoir temperature reached more than 300°C but the range of production temperature is between 230°C to 290°C based on Na-K-Ca geothermometer.

2. PRODUCTION HISTORY AND RESERVOIR PROCESSES

The Kawerau geothermal field begun production since the mid-1950's, initially supplying steam for heat process and electricity generation to the Tasman Pulp and Paper Company (now Norske Skog Tasman). Incremental development in electricity generation followed over the years, with Ngāti Tūwharetoa Geothermal Assets (NTGA) owning most of the wells and steamfield assets after the transfer from the Crown.

From 2008 onwards, development in the Kawerau geothermal field includes the following (Milicich et al., 2015):

- 100MWe KGL flash plant commissioned and operated by Mercury NZ Ltd.
- 8.3MWe Ormat binary plant commissioned by Geothermal Development Ltd. (GDL)
- 23MWe TOPP1 Ormat binary plant commissioned, geothermal fluid supplied and operated by NTGA;
- Resource consent for a 15-25 MWe power plant granted to Te Ahi O Maui Limited Partnership (TAOM)

A total of ~140MWe net generation are currently produced out of the Kawerau reservoir in addition to direct steam use. This corresponds to a total of 104,900t/d fluid extracted on average, with 61,850t/d re-injected and 17,280t/d discharged to the Tarawera River, using a network of 15 production wells and 13 injection wells (Figure 1).

The early production from the shallow section of the reservoir and the subsequent pressure drawdown induced cooler groundwater inflows. By the 1990's, shallow injection was implemented in Kawerau to mitigate the effects of cooling and dilution in early production wells, while subsequent drilling of production and injection wells were targeting the deeper reservoir within the greywacke basement. In addition, full reinjection of spent fluids was encouraged to provide pressure support in the reservoir. As a result, the recent production chemistry shows a dynamic combination of dilution (mixing with cool groundwater inflows/marginal recharge) and mixing with injection returns.

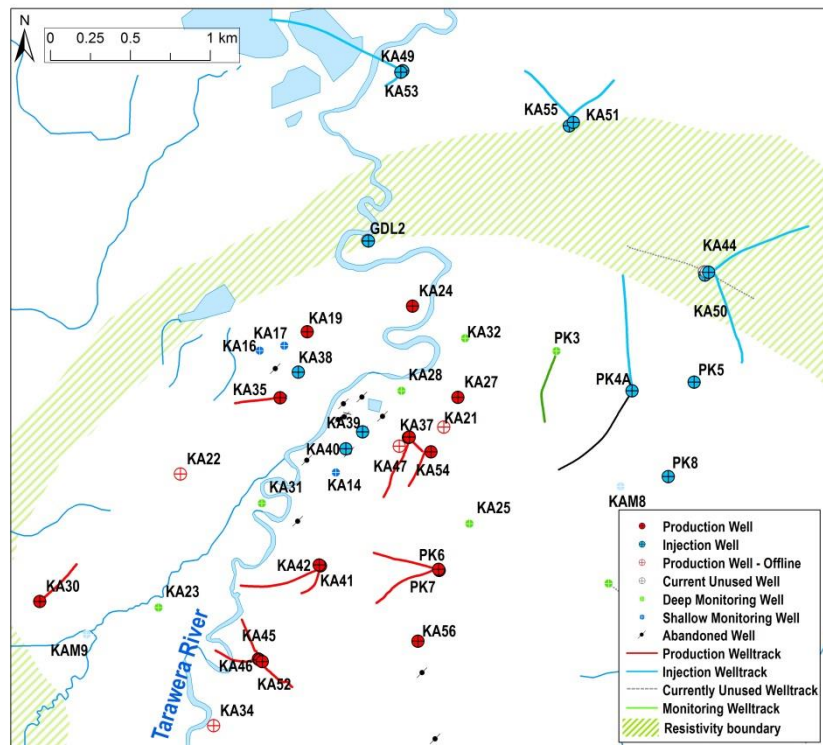


Figure 1: Layout of the Kawerau Geothermal Field and location of the main wells mentioned in the text (Resistivity boundary derived from Allis, 1997).

Characteristic of the injection fluids

For the KGL flash plant, two types of fluid -brine from the low-pressure (LP) separation and condensed steam from the cooling tower- are injected into wells (PK04A, PK5, PK8, KA44, KA50 and KA51). Since the LP flash temperature is low ($\leq 130^{\circ}\text{C}$), the brine is oversaturated with respect to silica and has high potential to deposit amorphous silica in the surface plant facilities, the injection wells and the reservoir formation. To manage this risk, the brine is dosed with sulfuric acid prior the LP separator to delay silica polymerisation by lowering the fluid pH. As a result, the reinjected LP brine contains a large amount of sulfate. On the other hand, the condensate is a very dilute fluid (e.g $\text{Cl} < 2 \text{ mg/kg}$) and can contain a substantial amount of sulfate as the cooling tower environment is prone to sulfur-oxidising bacterial growth. Similarly, the TOPP1 reinjection fluid (reinjected in KA53), which is a mix of brine and steam from the binary plant, is also dosed with sulfuric acid to manage the silica deposition (Figure 2).

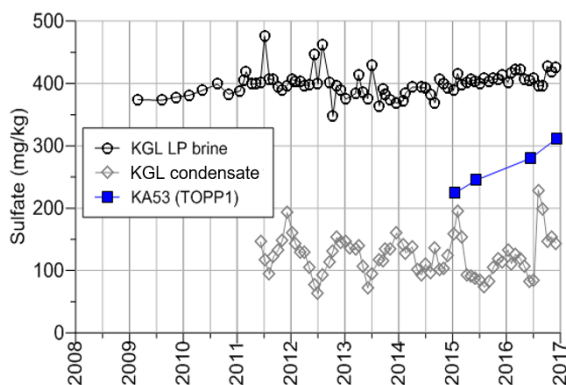


Figure 2: Sulfate concentration of various injectates at Kawerau.

Reservoir processes from the production chemistry

The level of development in Kawerau has changed the chemistry of fluids from the production wells, primarily because of mixing/diluting with marginal recharge and/or injection returns. Due to the large amount of injectates and potential diluting end-members, identifying the source of the mixing fluid in the production wells is not a straightforward undertaking.

Due to the nature of the injectates, an increasing sulfate concentration may indicate mixing with reinjection returns (e.g. Figure 3). However, increasing sulfate is also commonly associated with ingress of cooler, more diluted fluid, from shallow to intermediate depth, where the sulfate content is usually higher, due to increasing oxidation of geothermal H_2S (Nicholson, 1993). As exemplified by the sulfate concentration, the chemical composition of the production wells alone is insufficient to determine the source of the mixing fluid.

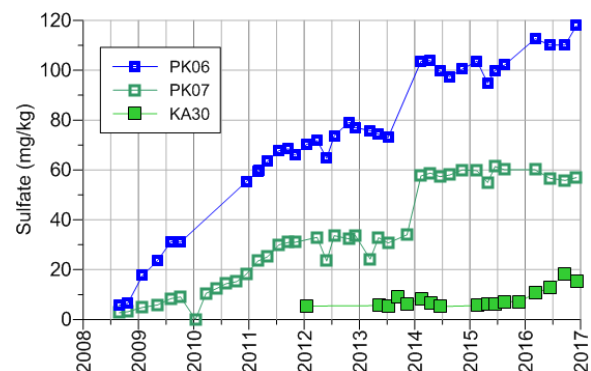


Figure 3: Sulfate concentration at reservoir conditions (TNaKCa) for selected production wells.

The water stable isotopes (^2H and ^{18}O , Figure 4, reported against the Vienna Standard Mean Ocean Water, referred to as SMOW herein) suggest also that mixing is one of the main reservoir process occurring in the Kawerau reservoir. Starting from an inferred isotopic composition of the parent fluid (i.e. data from KA07, the heaviest water discharged at Kawerau, Allis et al., 1995), the isotopic composition of the fluids from the Kawerau production wells are influenced by mixing with meteoric waters or injection returns (i.e. separated brine or condensate), the latter being by nature enriched in ^2H and ^{18}O . Among the production wells, KA30 and KA42 indicated mixing with marginal fluids while the rest of the production wells showed mixing with injectates.

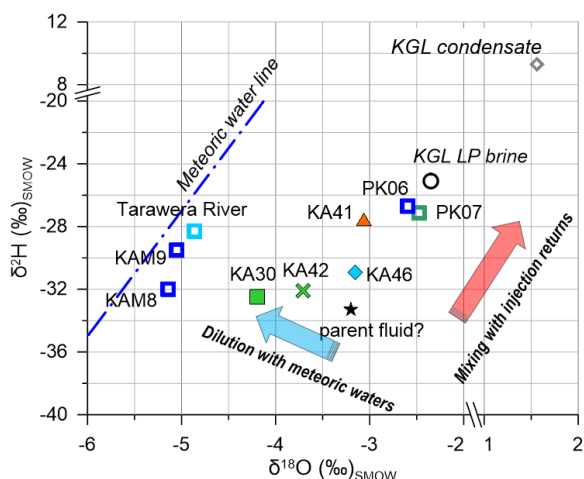


Figure 4: Stable isotopic composition of selected waters at Kawerau. The New Zealand meteoric water line is defined in Stewart and Taylor (1981).

Some degree of cooling was likewise observed in several production wells as shown in the enthalpy-chloride diagram, associated with mixing of injection returns (e.g. PK06 and PK07, Figure 5) or marginal recharge that is either steam-heated or conductively heated fluids (e.g. KA30). In addition to the diagram shown above, this mixing model supports the interpretation derived from the isotopic composition of the fluid from the production wells.

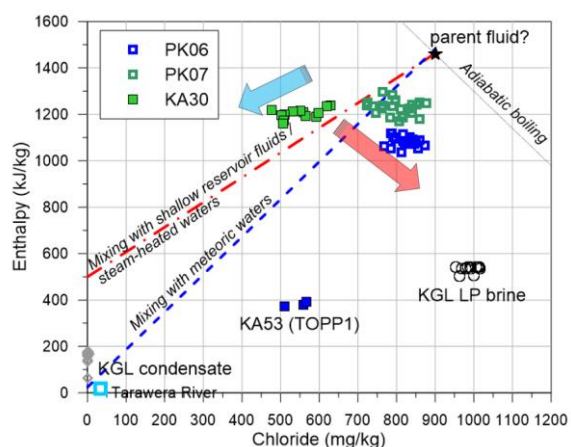


Figure 5: Enthalpy-chloride diagram for selected production wells (arrows indicate evolution over time)

In order to gain further understanding on the source of sulfate in the production wells and better constrain the current conceptual model and reservoir processes, sampling for sulfur isotope analysis in selected production wells and

relevant surface waters was conducted at Kawerau. The potential future use/application of the sulfur isotopes on reservoir performance monitoring was also investigated.

3. SULFUR ISOTOPES

Selected locations on the steamfield and at the power plant were identified and sampled for stable sulfur isotopic composition analysis (^{34}S).

3.1 Sampling

Sulfate (SO_4^{2-}) in brine was collected in a 2-litre bottle. Where meaningful, sulfide ($\text{H}_2\text{S}_{\text{aq}}$) in brine was also collected in a 2-litre bottle with an addition of 2 ml of cadmium-acetate. Sulfide ($\text{H}_2\text{S}_{\text{gas}}$) in steam was collected using a Rotoflo flask, prepared with sodium-hydroxide and cadmium-acetate to trap CO_2 and H_2S gases, respectively. The liquid samples were further preserved in ice prior to submission to the laboratory. Samples were analysed at the National Isotope Centre at GNS Science. The analytical precision for $\delta^{34}\text{S}$ is determined to be $\pm 0.3\text{‰}$. The $\delta^{34}\text{S}$ values are reported against the Vienna Canyon Diablo Troilite (VCDT) standard.

3.2 Results

The $\delta^{34}\text{S}$ results are presented on Figure 6. $\delta^{34}\text{S}$ values for pyrite and anhydrite minerals encountered in geothermal systems in the TVZ are also plotted for reference (based on values reported in Bayon, 1996; Marini et al., 2011, Chambefort, 2012). Table 1 in Appendix A provides the tabulated values.

Attempts were made to analyse $\delta^{34}\text{S}$ in sulfate in two shallow groundwater wells, KAM8 and KAM9 (<50 m deep), which were sampled using a N_2 lifting method. In both cases, the analyses yielded results nearly beyond the range of $\delta^{34}\text{S}$ composition commonly encountered in nature (53.5‰ and 31.5‰ respectively). It is inferred that post-sampling isotopic fractionation due to biological processes, such as sulfur oxidizing/reducing bacterial activities (usually favouring lighter sulfur isotopes), may have resulted in abnormally heavier residual samples. These were consequently discarded in the interpretation.

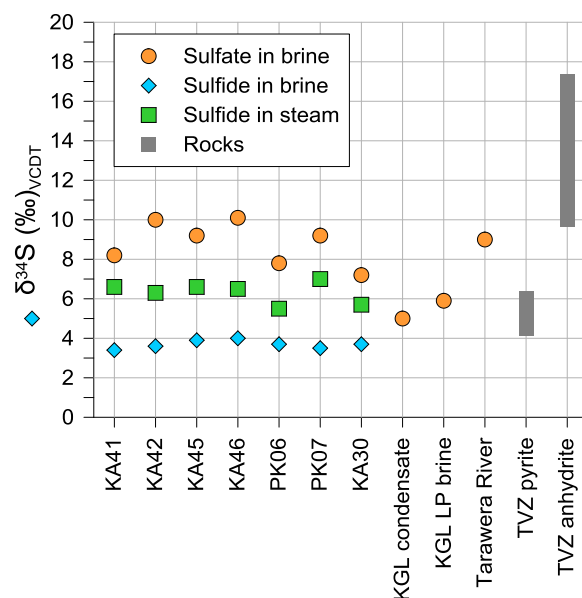


Figure 6: Distribution of $\delta^{34}\text{S}$ ratio in selected production wells, KGL injectates and Tarawera River.

³⁴S in sulfide

The range of the isotopic composition in sulfide in brine ($\text{H}_2\text{S}_{\text{aq}}$) is narrow (3.4‰ to 4.0‰) among the production wells. Values for the $\text{H}_2\text{S}_{\text{gas}}$ range from 5.5‰ to 7‰. Variations between H_2S in gas and liquid ($\Delta^{34}\text{S} \sim 1000 \ln \alpha_{\text{H}_2\text{S}_{\text{gas}}-\text{H}_2\text{S}_{\text{liq}}}$) are small (from 1.8 to 3.5‰) and may account for a small fractionation at atmospheric conditions (using the fractionation factor established by Szaran, 1996). The $\delta^{34}\text{S}$ in sulfide in the Kawerau geothermal fluid thus appears homogeneous and consistent with the H_2S isotopic composition commonly found in the geothermal reservoirs of the TVZ ($5 \pm 2\%$, Milicich et al., 2017), which also corresponds to the range measured in pyrite minerals within the TVZ. It is inferred that the isotopic composition of H_2S within the TVZ is mostly controlled by exchange of sulfur between the geothermal fluid and the greywacke basement via magmatic contamination (Milicich et al., 2017). In addition, the estimated half-time for this isotopic exchange reaction is more than 1000 years (Nicholson, 1993). As such, at the scale of the Kawerau geothermal reservoir, the isotopic composition of sulfide in the geothermal fluid does not directly provide meaningful insights on small-scale, short/medium term reservoir processes related to geothermal generation.

³⁴S in sulfate

The isotopic composition of sulfate in the sampled waters spans a larger range from 5.0‰ to 10.1‰. The KGL cooling tower condensate, with sulfate composition primarily controlled by sulfur-oxidizing bacteria activity, has the lowest $\delta^{34}\text{S}$ value (5.0‰). Similarly, the KGL LP brine also has a low $\delta^{34}\text{S}$ value of 5.9‰ with sulfate composition coming from the addition of sulfuric acid. The sulfuric acid used at KGL is sourced from a fertilizer supplier, where the sulfuric acid is produced from the combustion of elemental sulfur.

The production wells display higher value of $\delta^{34}\text{S}$. Using fractionation factors established by Robinson (1973) and Ohmoto and Rye (1979) at the reservoir temperature (240°C to 280°C using T_{NaKCa}), it appears that the sulfate composition is largely in isotopic disequilibrium with the sulfide composition (Figure 7). This disequilibrium may reflect mixing processes between an inferred heavy parent fluid in isotopic equilibrium with H_2S and lighter, shallower end-members. Understanding these mixing mechanisms can provide insights on the reservoir behaviour under development.

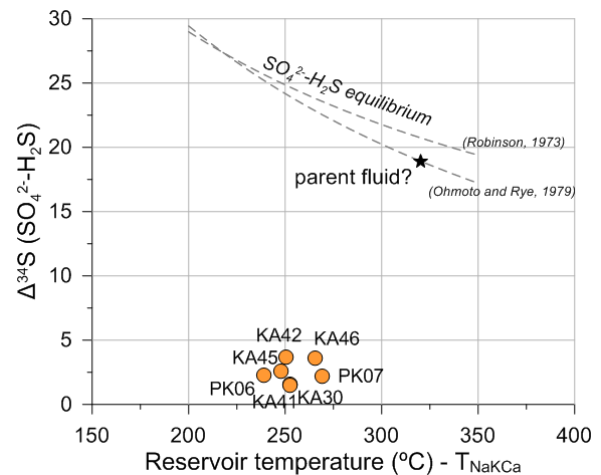


Figure 7: Isotopic fractionation factor between SO_4^{2-} and H_2S versus temperature.

Isotopic Geothermometry

The use of the common geothermometers to assess the reservoir temperature is somewhat difficult at Kawerau. The quartz geothermometer may be affected by injection returns: the reinjected LP brine provides an additional source of silica and it tends to overestimate the reservoir temperature calculated from the quartz geothermometer as a result. Similarly, injection returns can provide a source of additional calcium (as the cold injectates dissolve the calcite present in the reservoir rocks) and the empirical NaKCa geothermometer may be also underestimated. On the other hand, as mentioned by Christenson (1987), the albite ($\text{NaAlSi}_3\text{O}_8$)-feldspar/adularia (KAlSi_3O_8) equilibria used for the NaK geothermometer seems to be affected by the overall high gas content of the reservoir and the solubility of calcite, hence generally also give underestimated reservoir temperature.

In that context, the geothermometer based on the kinetic of oxygen exchange between sulfate and water (Figure 8) appears to yield relevant temperature for most of the production wells, ranging from 260°C to 314°C, down to 164°C for KA30. The latter temperature is much lower than estimated by the other aqueous geothermometers, and likely suggests the impact of mixing with a fluid containing much higher $\delta^{18}\text{O}$ (SO_4^{2-}) ratio, such as steam-heated/ H_2S oxidised fluids.

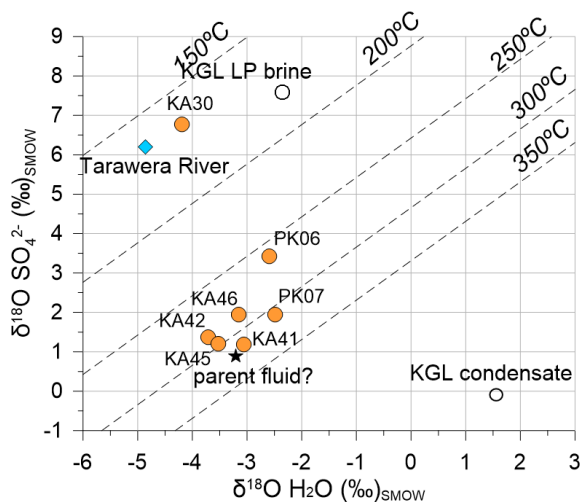


Figure 8: Equilibrium temperature of the oxygen fractionation in the SO_4^{2-} - H_2O system (after Mizutani and Rafter, 1969)

For most of the production wells however, this geothermometer appears consistent with the reservoir temperatures encountered at Kawerau (e.g. PK06, Figure 9, where the sulfate oxygen isotope geothermometer is in fact the closest to the observed temperature at the surface). The estimated half-time for this isotopic exchange reaction is about 1 year (Nicholson, 1993), slower than the common aqueous geothermometers previously mentioned (i.e. estimated equilibration time of a few hours to a few months). In that regard, this geothermometer provides a means of monitoring the reservoir temperature with a longer “memory” range and indicates the temperature at deeper depths in the reservoir.

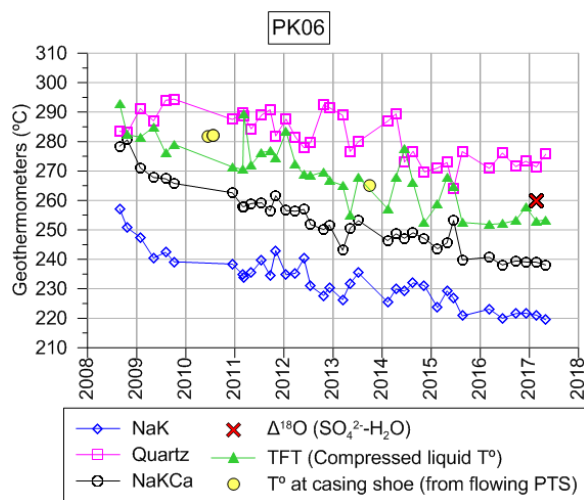


Figure 9: Comparison of physical and chemical measurements of temperature

The $\delta^{18}\text{O}/\delta^{34}\text{S}$ in sulfate cross-plot (Figure 10) is sometimes used to interpret the genesis of sulfate fluids (e.g. Bayon et al., 2005, Calibugan et al., 2015). In this interpretation diagram, both KGL injectates bear the signature of H_2S oxidation (while this is true for the KGL cooling tower, it is coincidental for the LP brine, as a result of the sulfuric acid addition); KA30 composition reflects some mixing with a low temperature, whose sulfate originates from H_2S oxidation. PK06 shows a similar trend

in this diagram, which also corresponds to a stronger influence of KGL LP brine injectate. The remainder of the production wells highlights a relatively homogeneous combination of the deep parent fluid with surficial $\delta^{34}\text{S}$ -depleted waters.

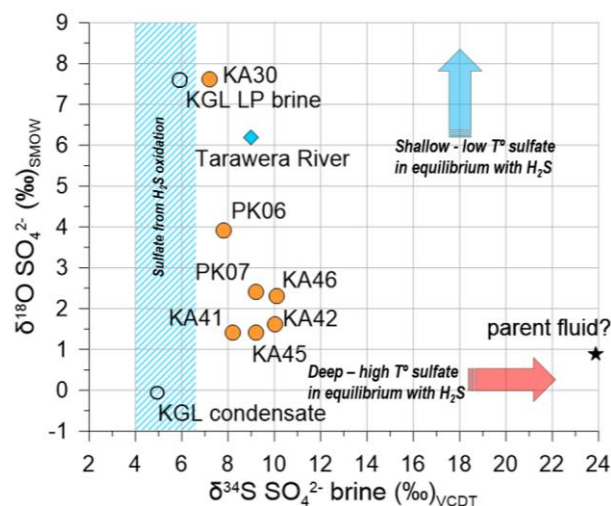


Figure 10: $\delta^{34}\text{S}$ versus $\delta^{18}\text{O}$ in sulfate

Isotopic mixing model

The isotopic composition of the production wells can be interpreted using a mixing model representing sulfate and its isotopic composition (Figure 11). On such a diagram, mixing lines are represented by hyperbolic curves because the mixture of two components that have different isotopic ratios and different concentrations does not vary linearly with the proportion of these two components (Kendall and McDonnell, 1998).

Three end-member compositions were used in the isotopic mixing models including: (i) a deep reservoir fluid for which sulfide and sulfate are in isotopic equilibrium (i.e. $\delta^{34}\text{S} \text{ SO}_4^{2-} \approx 24\%$ at 320°C using the fractionation factor from Ohmoto and Rye, 1979, Figure 7) and a sulfate concentration of about 15 mg/kg, (ii) the KGL injectates and (iii) a water with isotopic composition of H_2S at Kawerau ($\delta^{34}\text{S} \approx 4\%$) reflecting a steam-heated fluid or a H_2S -oxidised fluid, with a sulfate concentration of about 20 mg/kg. The latter end-member is likely to represent marginal recharge and/or a fluid from a shallow/intermediate depth where quantitative oxidation of H_2S has happened.

As shown in Figure 11, the isotopic composition in most of the production wells (KA41, KA42, KA45, PK06 and PK07) reflects the mixing between the deep reservoir and the KGL injectates. Results of the mixing model suggest 10 to 30% of reinjection returns in these wells, which is consistent with the estimated reinjection returns based on the results of the 2010 reservoir tracer tests (PK4a, PK08, KA44 and KA50) (Siega et al., 2012).

On the other hand, KA46 and KA30 isotopic composition can be explained by the mixing between the deep reservoir and the shallow/intermediate end-member (e.g. marginal recharge from a shallow/intermediate horizon). For KA30, the mixing ratio is about 70% to 80% of the dilute end-member, which is higher than the ratio suggested by a simple chloride mass balance ($\sim 50\%$, Figure 5), indicating

that this end-member could be chloride-rich. Similarly for KA46, the mixing model suggests up to ~60% dilute fluid which is not consistent with the ~20% dilute component based on the same simple chloride mass balance. Further analysis may be needed to better understand the discrepancy observed between the 2 mixing models. The Tarawera River sample also falls on this mixing line; the river is the major outlet of the Kawerau shallow groundwater system.

Generally, the isotopic signature of the production wells support the reservoir processes already suggested by the other techniques outlined in section 2. Accordingly, it provides a robust framework to constrain the origin of the sulfate observed in the production wells.

4. RESERVOIR IMPLICATIONS

Identifying reservoir processes is critical to effectively and sustainably manage the geothermal reservoir, particularly in improving the development strategy of the field.

This study suggests that the southwestern side of the Kawerau reservoir is affected to some degree by marginal recharge (i.e. ingress of a colder, more dilute fluid), probably from an intermediate and/or shallow depth. On the other hand, the eastern side of the field appears to be mostly affected by reinjection returns. The $\Delta^{18}\text{O}$ (SO_4^{2-} - H_2O) geothermometer also proves to be useful in providing a relatively accurate geothermometry temperature.

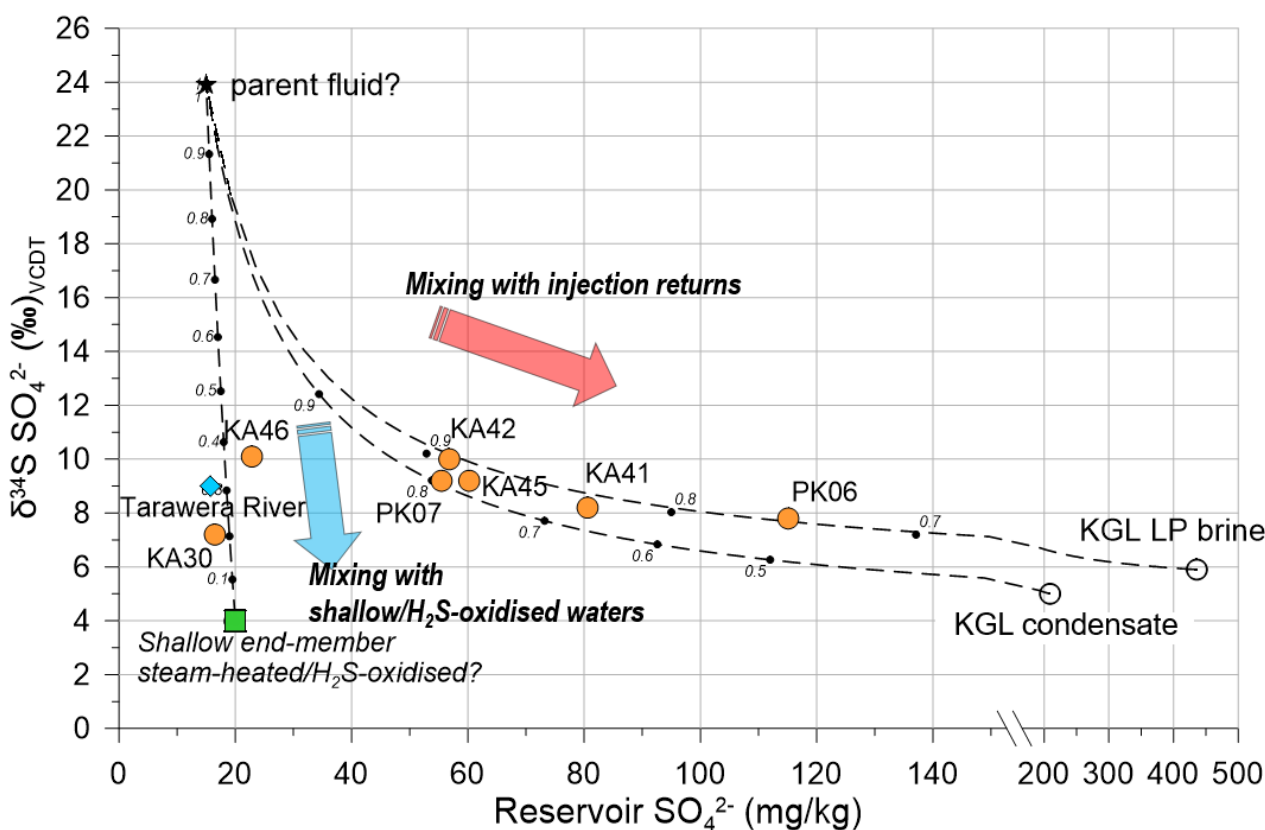


Figure 11: Isotopic mixing model between an inferred deep reservoir fluid, the KGL injectates and a dilute fluid end-members. Small italic numbers refers to the mixing ratio between end-members, relative to the parent fluid.

5. CONCLUSION

The use of sulfur isotopes at Kawerau provides valuable insights in the reservoir understanding as far as (i) estimating the fluid temperature of the production wells (using ^{18}O fractionation in the SO_4^{2-} - H_2O system) and (ii) determining the source of sulfate in the production fluid and inferring the reservoir processes leading to this composition.

Used in conjunction with the hydrogeological and chemical data, the sulfur isotope data provides additional information to confirm the reservoir processes identified through the others chemical data, in particular to differentiate marginal recharge from injection returns. In this context, this can be used as a natural tracer for the source of sulfate and types of fluid mixing with reservoir fluids.

On the other hand, isotopes analysis cannot be used independently to evaluate reservoir processes. The interpretative framework of the sulfur isotope measurements is only beneficial because additional subsurface measurements are available. In particular, the isotopic mixing model strongly depends on the choice of hypothetical end-members whose existence is supported by the production chemistry obtained so far, as well as the geological structure of the field.

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"He waka eke noa"

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APPENDIX A: TABLE OF VALUES

Table 1: Stable isotopes analyses for selected production wells, injectates and surface water at Kawerau

Well	Date	Brine $\delta^2\text{H}(\text{‰})$	Steam $\delta^2\text{H}(\text{‰})$	Brine $\delta^{18}\text{O}(\text{‰})$	Steam $\delta^{18}\text{O}(\text{‰})$	Brine $\delta^{18}\text{O}_{\text{SO}_4}(\text{‰})$	Brine $\delta^{34}\text{S}_{\text{SO}_4}(\text{‰})$	Brine $\delta^{34}\text{S}_{\text{H}_2\text{S}}(\text{‰})$	Steam $\delta^{34}\text{S}_{\text{H}_2\text{S}}(\text{‰})$
KA41	Feb-2017	-27.3	-28.9	-2.74	-4.86	1.4	8.2	3.4	6.6
KA42	Feb-2017	-32.3	-30.9	-3.39	-5.53	1.6	10.0	3.6	6.3
KA45	Feb-2017	-29.9	-30.7	-3.24	-5.34	1.4	9.2	3.9	6.6
KA46	Feb-2017	-30.9	-31.0	-2.81	-4.97	2.3	10.1	4.0	6.5
PK06	Feb-2017	-26.5	-28.1	-2.32	-4.57	3.9	7.8	3.7	5.5
PK07	Feb-2017	-27.1	-27.3	-2.11	-4.02	2.4	9.2	3.5	7.0
KA30	Feb-2017	-32.0	-36.5	-3.92	-6.44	7.6	7.2	3.7	5.7
KGL condensate	Feb-2017	9.3	-	1.56	-	-0.1	5.0	-	-
KGL LP brine	Feb-2017	-25.1	-	-2.35	-	7.6	5.9	-	-
Tarawera River ¹	Feb-2017	-28.3	-	-4.86	-	6.2	9.0	-	-

¹At the SH34 bridge in the Kawerau township