

Hydrochemical and isotopic characterisation of groundwater at an Australian underground mine

Connor Verrall ^{a,*}, Antoinette Stryk ^b, Devmi Kurukulasuriya ^c

^a Hi! Engineers, Australia

^b BHP, Australia

^c Valenza Engineering, Australia

Abstract

A mine scale water sampling campaign was undertaken at the iron oxide, copper, gold (collectively IOCG) sublevel cave (SLC) asset to ascertain the presence of a possible third, upwelling source of water within the mine and its contribution to inrush hazards. The campaign consisted of 98 water samples testing for $\delta^2\text{H}$, $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ -dissolved inorganic carbon (DIC) isotopes of which 76 were hydrochemically sampled; testing for potassium, sodium, calcium, magnesium, lithium, bicarbonate, chloride and pH.

The results of this campaign were cross-referenced with historic isotopic water samples ($\delta^2\text{H}$ and $\delta^{18}\text{O}$) from the baseline environmental characterisation of the mine site. Through Hydrochemical and isotopic water analysis, water expressions in the mine were classified as belonging to the 2 overlying sedimentary aquifers, 2 previously unknown sources, and 2 mixtures of the newly classified sources.

Using the data from the sampling campaign, historic datasets, and comparison of the fluid fingerprints against waterbodies sourced from the literature; two of the additional water sources were characterised as active hydrothermal systems, both created by radiogenic geothermal heating of evaporated sea water. The results of this paper can be used to categorise any influx of water observed reporting through the drawpoints or intercepted throughout the mine in relation to those that have been characterised in this paper.

Keywords: isotopic analysis, hydrochemical analysis, underground mine, inrush, sublevel cave

1 Introduction

A caving operation, be it sublevel or block cave (SLC or BC), is a complicated and technical endeavour. The creation of a large muck pile, from which ore is drawn, harbours a set of critical hazards seldom encountered on a day-to-day basis in traditional open pit or underground mines. Engulfment, otherwise known as inrush or mud-rush, is one such hazard. The engulfment hazard pertains to being overrun or enveloped by a flow of material, be it rocks, fines or water and is often fatal. In caving mines, it is experienced at the drawpoints of the cave where the production charging and bogging activities take place. It is generally constituent of a mixture of fines and water, forming mud, destabilising and mobilising down the drive.

In order to attain a better grasp upon the hydrogeology of the mine, a hydrochemical and isotopic mine water sampling campaign was undertaken to characterise the water, its evolution, and its relation to an inrush hazard at the mine.

Meteorological processes influence stable isotopes in water, such as ^{18}O and ^2H , imparting a distinctive fingerprint that is essential for tracing the origin of groundwater (Clark & Fritz 1997). Temperature dictates the distribution of isotopes in precipitation, with variations arising from evaporation, altitudinal and seasonal effects (Craig 1961). While the isotopic composition of groundwater assists in identifying its source by

* Corresponding author. Email: verrall@hiengineers.com.au

comparing it against local precipitation, this signal can undergo modification upon entry into the groundwater system due to evaporation or temperature changes, resulting in unique compositions for water following similar recharge pathways. Consequently, isotopic data can effectively discern between different water sources, revealing potential mixing scenarios (Kharaka & Thorsden 1992).

Furthermore, stable carbon isotopes ($\delta^{13}\text{C}$) are assessed in dissolved inorganic carbon (DIC) to elucidate water-rock interactions and subsurface geochemical processes. The $\delta^{13}\text{C}$ values also provide insights into carbon-related geochemical processes that may have contributed to the distinctive compositions observed in different water sources. This study has collectively used these isotopic tracers and general hydrochemical parameters to characterise the water observed within the mine.

Water flow mapping links the characterisation results to the operational observations and facilitates the interpretation of groundwater pathways, however this is outside the scope of this paper but is a logical next step for any mine site.

In this instance, the water characterisation reaffirmed the presence of the two main aquifers, revealed the presence of three additional unknown sources mine-wide and two water mixtures occurring in the active SLC. Of the three additional sources, only two are discussed in this paper. These are shown to be active radiogenic hydrothermal mixtures.

This paper details the sampling campaign, presents the results, and discusses their interpretation.

2 Methodology

2.1 Water sampling

Over eight months, two sampling campaigns were conducted, 98 water locations were sampled for isotopes and 76 of these were further sampled hydrochemically. Of the two batches, 27 samples were taken in Batch 1 and 71 in Batch 2. Hydrochemical analysis was primarily conducted on samples from batch 2 with five samples in batch 1.

Sampling was undertaken by collecting the water at the expression point. As such, the sampling categories were a direct reflection of where the water was found and sampled from and are defined as:

- Base of muck (water sampled from cave muck pile expressions)
- Drillhole (water sampled from an identified drillhole)
- Dripping (water sampled from an overhead dripping water source)
- Floor Weep (water sampled from a spring or water upwelling in the floor of underground workings)
- Geological formation (water sampled from an identifiable geological structure)
- Mine Water (water that was sampled from mining infrastructure [i.e. poly – dropper or tap])
- Wall Seep (water sampled from wall rock of underground workings)
- Surface Bore (water sampled from a drillhole or bore on the surface).

Samples were collected in 500 mL HDPE bottles from ALS Laboratories. Samples were collected using a funnel, rinsed with water from the source prior to collection. Samples were taken with care as to prevent contamination, and bottles were filled to the brim such that a meniscus was observable prior to capping. This was done to minimise the amount of isotopic contamination introduced by the air.

The samples were then sent to ALS laboratories for filtration and hydrochemical analysis. They were then on freighted to the School of Earth and Environmental Sciences at the University of Queensland's Stable Isotope Laboratory, where they underwent isotopic analysis.

Twenty seven historic isotopic samples from known aquifer sources were also included in this analysis, these were sampled using a Hydra-sleeve and extracted from bores prior to the commencement of mining at the asset.

3 Data

3.1 Hydrochemical data

The hydrochemical data returned from ALS consisted of a full suite of analysis for the following element concentrations and physical parameters: potassium, sodium, calcium, magnesium, sulphates, lithium, bicarbonate, chloride, cadmium, chromium, copper, fluoride, zinc, lead, total dissolved solids (TDS) and pH. In Figure 1 is a ternary piper plot depicting the major ion characterisation of the shallow aquifers from historical monitoring events, along with the water collected from the underground mine for the current isotopic study.

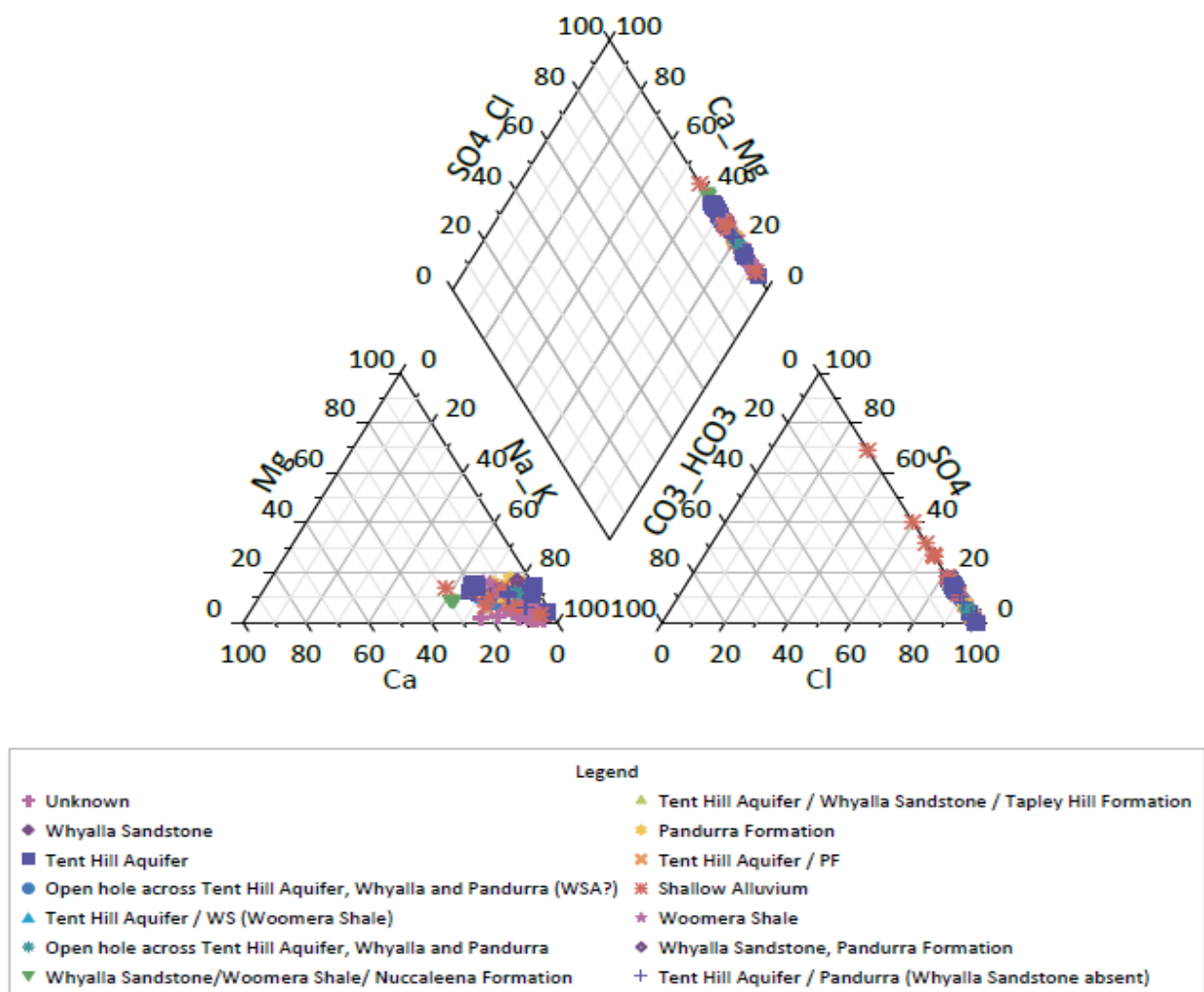


Figure 1 Ternary piper plot depicting major ion characterisation of the assets shallow aquifers

The pH of the water collected from the underground mine varied between 4.7 and 7.7 pH units with an average of 6.9. The TDS ranged between 18 to 340 g/L indicating a highly saline groundwater system. The overall major ion composition of groundwater suggests it is of the Na-Cl type, with minor variations in Ca, and Mg and sulphate concentrations. Figure 2 plots the Tent Hill Aquifer (THA) and Whyalla Sandstone Aquifer (WSA) against the sampled water from the mine.

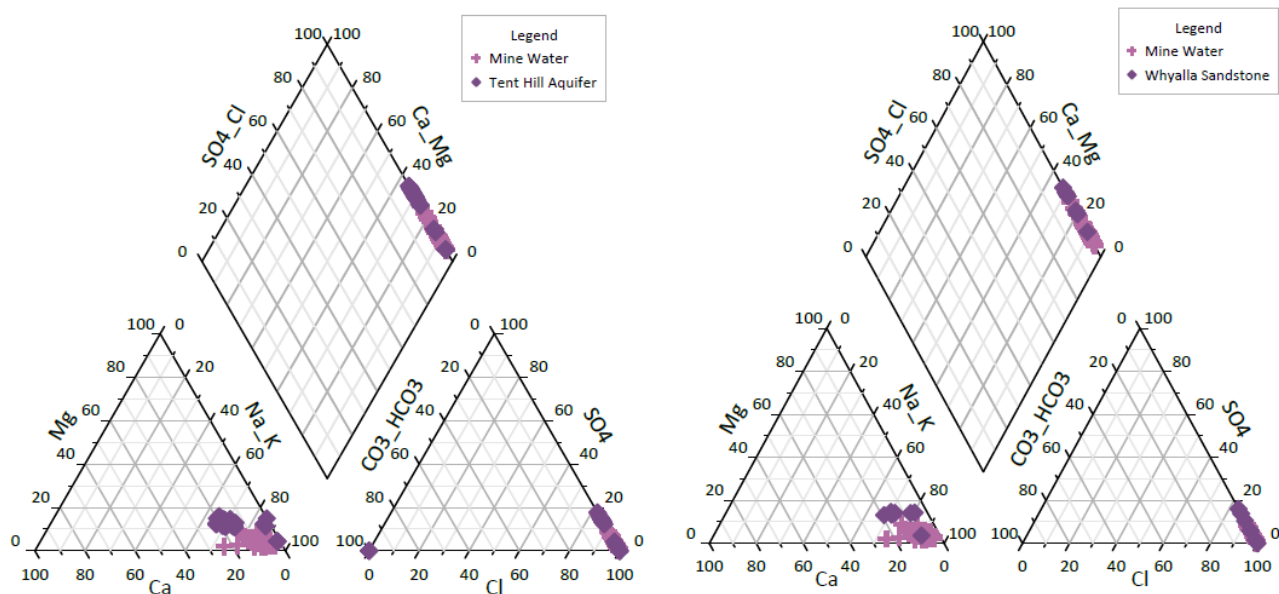


Figure 2 Ternary Piper Plots of the Tent Hill Aquifer (THA) and Whyalla Sandstone Aquifer (WSA) aquifers against the water sampled from the mine

Given the similar characteristics of THA and WSA with minor changes in Mg and Ca, it appears that WSA may have evolved from THA. However, WSA being a linear mixing scenario between the suspected deep unknown source of water (Na-Cl type) and THA seems unlikely.

The water collected from the mine on the other hand appears to exhibit a different composition compared to both THA and WSA water types. A potential source, likely dominated by sodium and chloride (suspected deep hydrothermal water), may be mixing with either the THA or WSA waters found in the mine.

The hydrochemical data was plotted against chloride concentration to better compare and contrast against fluids from the literature, this is presented in Section 4. Not all elemental compositions are discussed in this paper, only those that pertain to the commentary of the discussion.

3.2 Isotopic data

The isotopic composition data returned from the University of Queensland was for $\delta^2\text{H}$, $\delta^{18}\text{O}$ and $\delta^{13}\text{C-DIC}$. The $\delta^{13}\text{C}$ (DIC) were analysed using a Thermo Delta V with Gasbench whereas the isotopes of $\delta^2\text{H}$, and $\delta^{18}\text{O}$ were analysed using an Isoprime Dual Inlet isotope ratio mass spectrometer. These three values represent the departure of the isotopic ratio for a sample as compared to the Vienna Standard Mean Ocean Water (VSMOW) for hydrogen, and oxygen and to Vienna Pee Dee Belemnite for carbon. The local meteoric water line for the site was derived from the closest IAEA Global Network of Isotopes in precipitation monitoring stations, Woomera $\delta^2\text{H} = 6.6 \delta^{18}\text{O} + 6.0$ (Crosbie et al. 2012). The Global meteoric water line is $\delta^2\text{H} = 8 \delta^{18}\text{O} + 10$ (Craig 1961). The raw data collected and used throughout this paper is displayed in Figure 3. The groupings in this Figure relate directly to the observation of how that water emission was expressing from the mine at the time and were detailed in section 2.1.

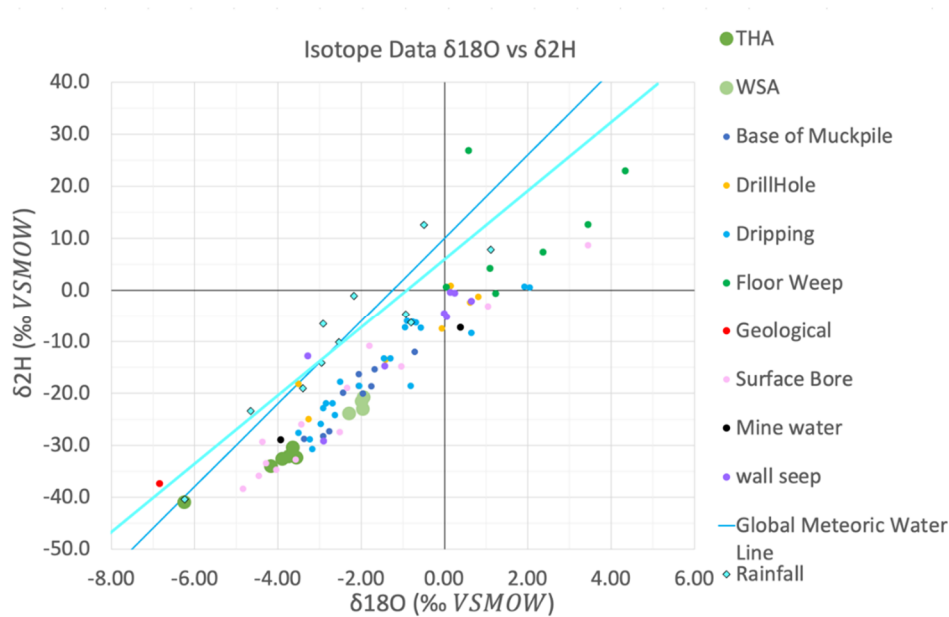


Figure 3 $\delta^{18}\text{O}$ versus $\delta^2\text{H}$, entire raw isotopic dataset, historical data as big circles, modern data as small dots

4 Discussion

4.1 Water grouping and classification of the assets water

The classifications of the hydrochemical and isotopic water data and the interpretations for the signatures of water groupings is discussed in this section. These are referenced throughout the remainder of the paper. The classified data falls into the groupings outlined:

- The Whyalla Sandstone Aquifer (WSA grouped)
- The Tent Hill Aquifer (THA grouped)
- The Basal Aquifer (basal)
- Active hydrothermal water (hydrothermal)
- A mixture of WSA and native hydrothermal water (WSA/hydrothermal mix)
- An additional source of water that is rarely observed (Unknown).
- A mixture of the Tent Hill and Whyalla Sandstone Aquifers (THA/WSA mix).

4.1.1 Aquifer waters

Establishing the isotopic and hydrochemical signatures of the known aquifers; THA and WSA, was completed by overlaying the modern surface bore isotopic data with the historical isotopic dataset and performing a proximity analysis of the two groups. This identified potential modern-day samples of similar isotopic composition that were then used to establish the baseline hydrochemical fingerprint of the water for the THA and WSA. This process was necessary, as no hydrochemical data could be found for the historical isotopic dataset.

Similarly, the 'Dripping' dataset was overlaid on the historical dataset as it was suspected that some of the dripping expressions from the mines intersection with Whyalla Sandstone unit would yield the best proxy for the WSA water where they were an isotopic match. These two modern-day sample groupings then formed the basis of the fingerprints of the aquifer water isotopically and hydrochemically. The raw points selected through this process are shown circled in Figure 4 on the isotopic plot and again circled in Figure 5, the hydrochemical plot for magnesium.

The remainder of the aquifer points were identified using a recursive proximity check across the hydrochemical and isotopic datasets, whereby points from all recent sample groupings were analysed for their proximity to this fingerprint in carbon isotopes and hydrochemical results.

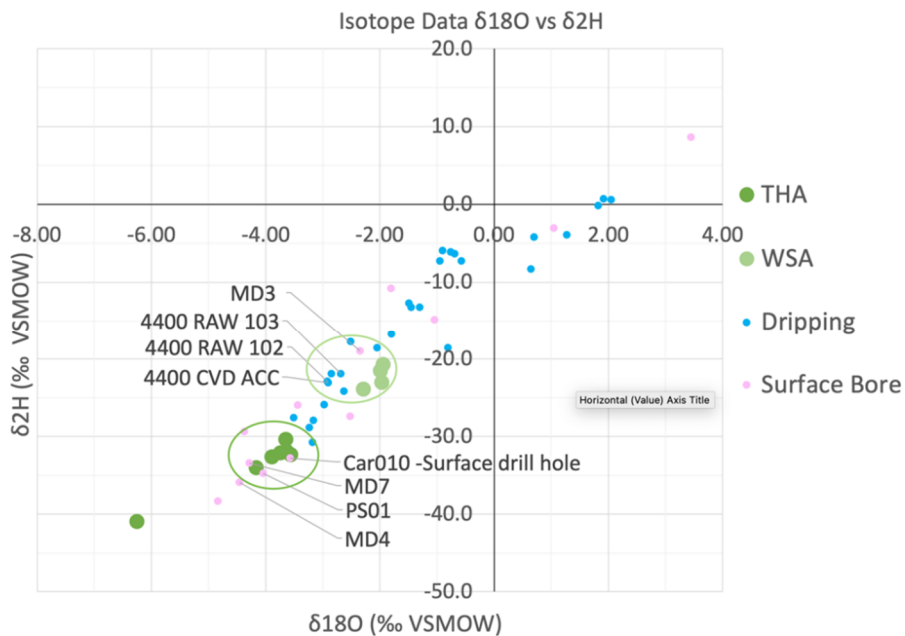


Figure 4 Isotopic dataset of dripping, surface bore and historical Whyalla Sandstone Aquifer and Tent Hill Aquifer data, selected data for fingerprints is labelled and circled

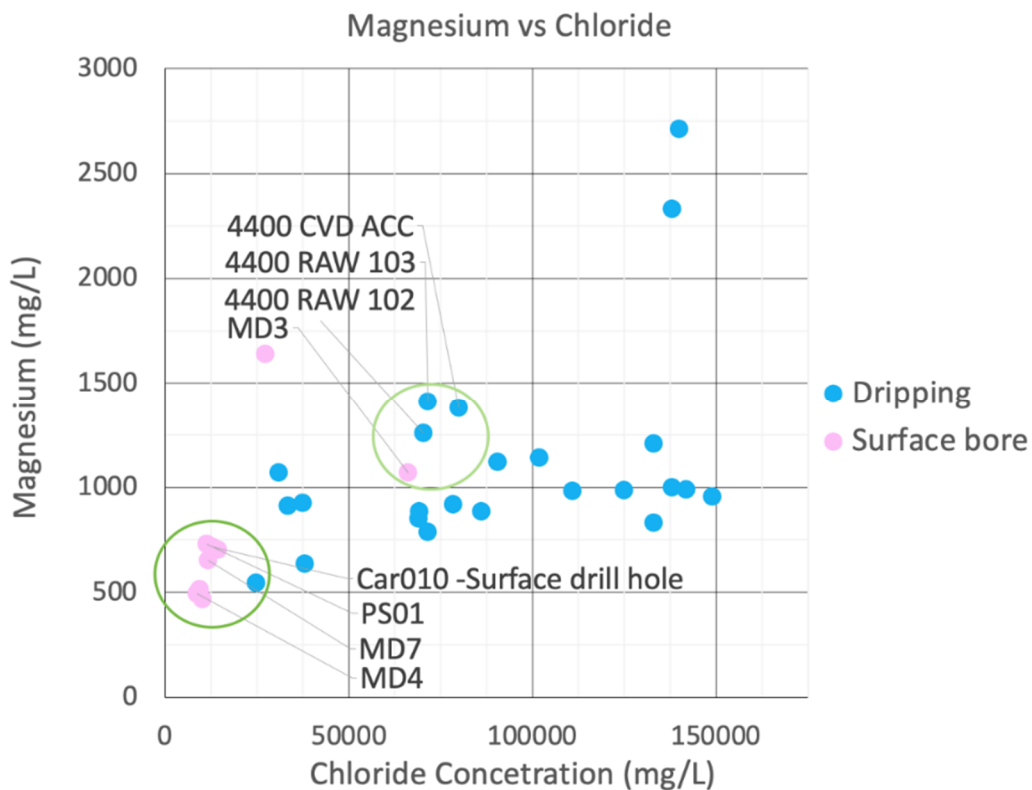


Figure 5 Magnesium versus chloride hydrochemical plots for dripping and surface bore data, selected data for fingerprints is labelled and circled

4.1.2 Upwelling fluids

The spread of data beyond the THA and WSA fingerprints in Figures 4 and 5 indicated that there was at least one more fluid within the sampled dataset. Infield observations corroborated this, as when within the mine and below the level of the aquifers, water was found expressing through the floor as hot springs and upwelling in intersected drill holes. Furthermore, radon gas was detected emitting from these upwellings, not previously observed in aquifer intercepts. The data was replotted on the $\delta^{13}\text{C}$ -DIC versus $\delta^{18}\text{O}$ isotopic graph in order to identify whether this spread on the $\delta^{18}\text{O}$ versus $\delta^2\text{H}$ plot was caused by one or more water sources. Figure 6 displays the raw data overlaid with the THA and WSA groupings and the two additional water sources highlighted in light blue and orange.

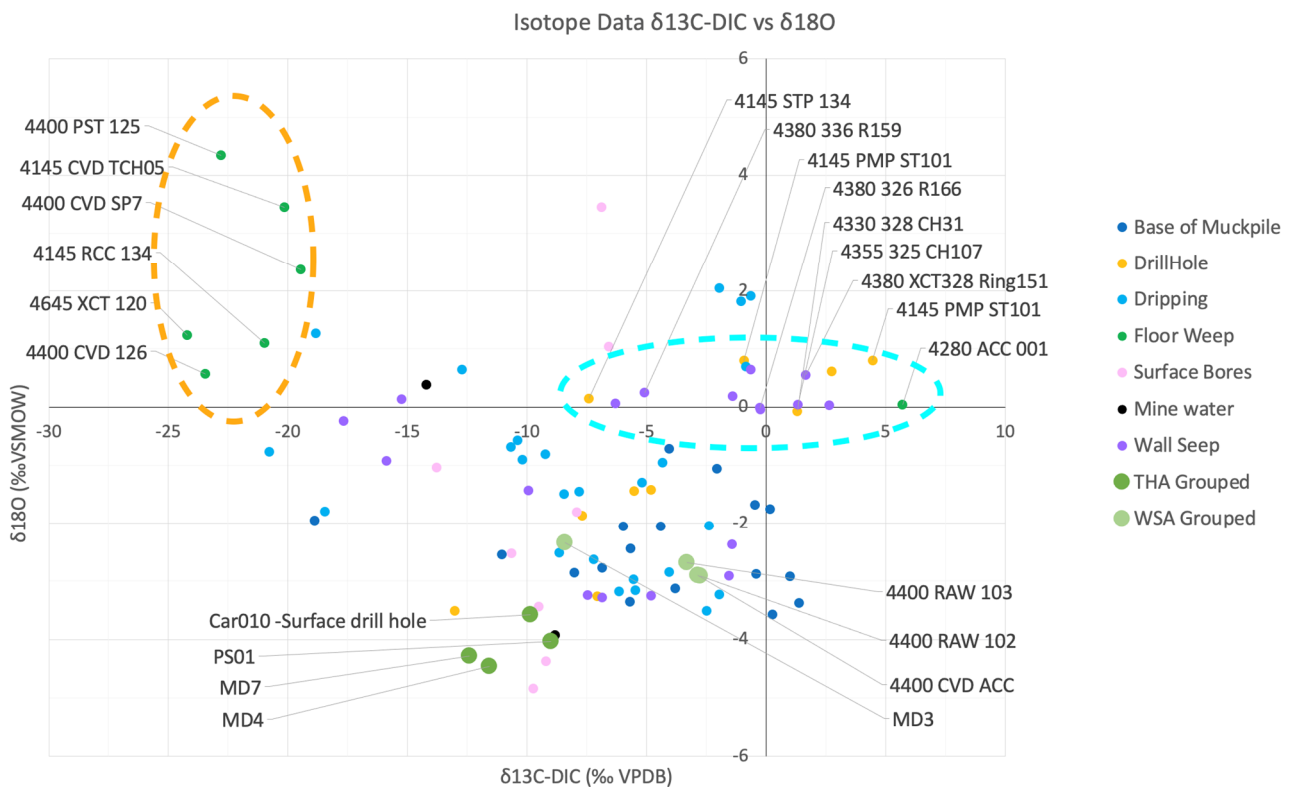


Figure 6 $\delta^{13}\text{C}$ - DIC versus $\delta^{18}\text{O}$ Isotope plot of raw data with WSA and THA groups overlaid, and additional suspected deep sources highlighted and labelled in light blue and orange

These two circled groups were then analysed for hydrochemical similarities. It was established that these two groups were two separate water sources that have their own unique hydrochemical fingerprints, this is shown in Figures 7 and 8, the calcium and magnesium plots for the grouped populations. The groups have been labelled basal and hydrothermal water to reflect their perceived origins. 'Basal' (light blue) was selected for the water exhibiting enriched $\delta^{13}\text{C}$ -DIC (0,0) because of its association with drill holes that intersect the 'Basal Fault'. It is believed this fault is the reservoir for this water. 'Hydrothermal' (orange) was selected for the other grouping as the chemical and more depleted $\delta^{13}\text{C}$ -DIC isotopic composition of this water indicates an active hydrothermal system. However, an assessment of potential geochemical interactions affecting $\delta^{13}\text{C}$ -DIC in both end members is warranted, and this will be discussed further in section 4.1.4.

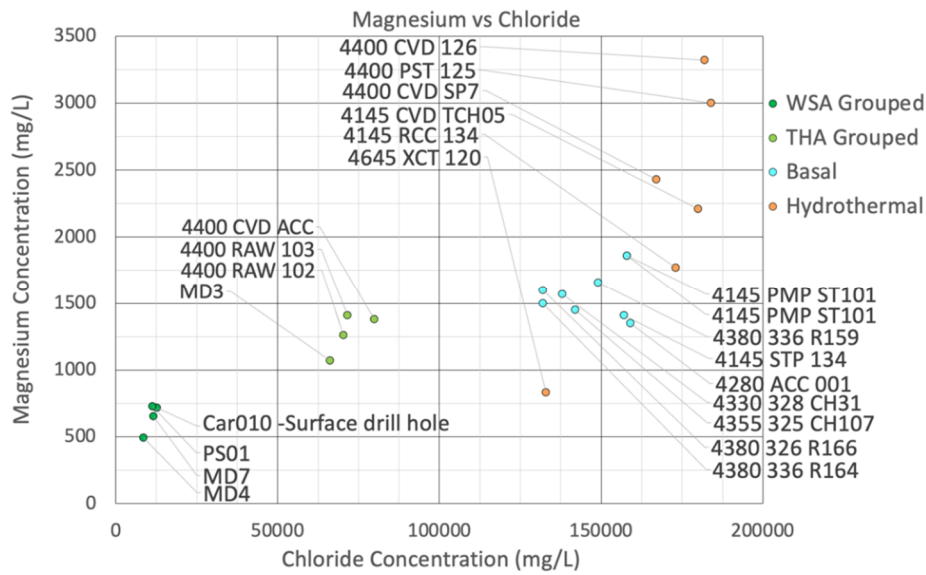


Figure 7 Magnesium versus Chloride for the four asset water groupings

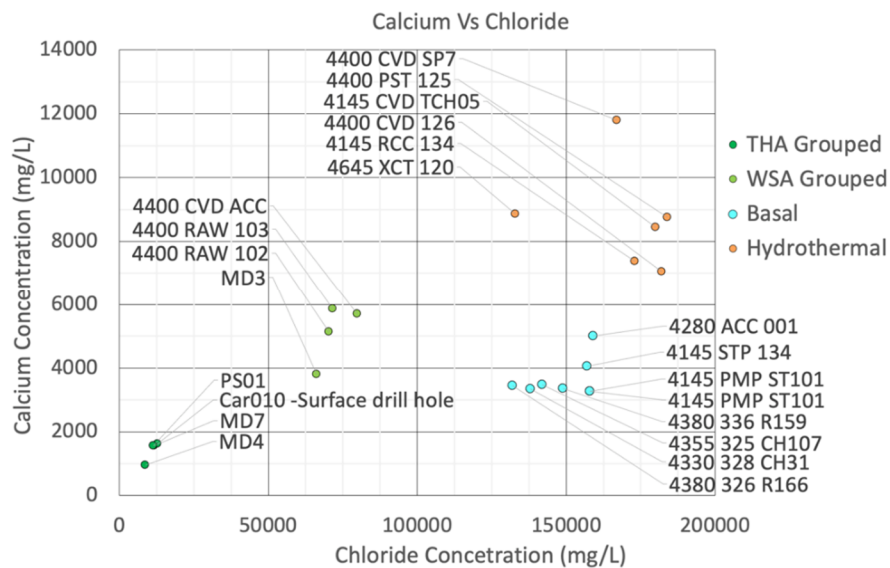


Figure 8 Calcium versus Chloride plot for the four asset water groupings

4.1.3 Mixes

The remaining water samples plotted within the isotopic and hydrochemical bounds of the four water sources defined in the previous section. As such, they were considered to be a mixture of these previously defined water sources.

Two distinct mixtures are defined: The THA/WSA mix and WSA/hydrothermal mix. The THA/WSA mix isotopically and hydrochemically plots between the THA and WSA fingerprints. The WSA/hydrothermal mix hydrochemically and isotopically plots between the active hydrothermal and WSA fingerprints. It should be noted that the WSA/hydrothermal mix group is better described as ‘native water’, as it primarily expresses when the orebody is first developed through. It is likely residual active hydrothermal water that has been pervaded by the overlying WSA over time and remained trapped within the joint network of the orebody, only to release upon development.

The basal water is not found to mix with the WSA water. The hydrochemical deviation of the basal group from the rest of the population, specifically in the magnesium and calcium data, is not replicated by the remaining unclassified hydrochemical data points, thus mixing of the basal group with others is unlikely.

The full set of classified data for magnesium, calcium, sodium, potassium, lithium and bicarbonate are displayed in Figures 9, 10 and 11, they demonstrate the spread and classifications of the data. The full set of classified data for the $\delta^{18}O$ versus $\delta^{2}H$ and $\delta^{13}C-DIC$ vs $\delta^{18}O$ isotope plots is also shown in Figures 12 and 13, respectively.

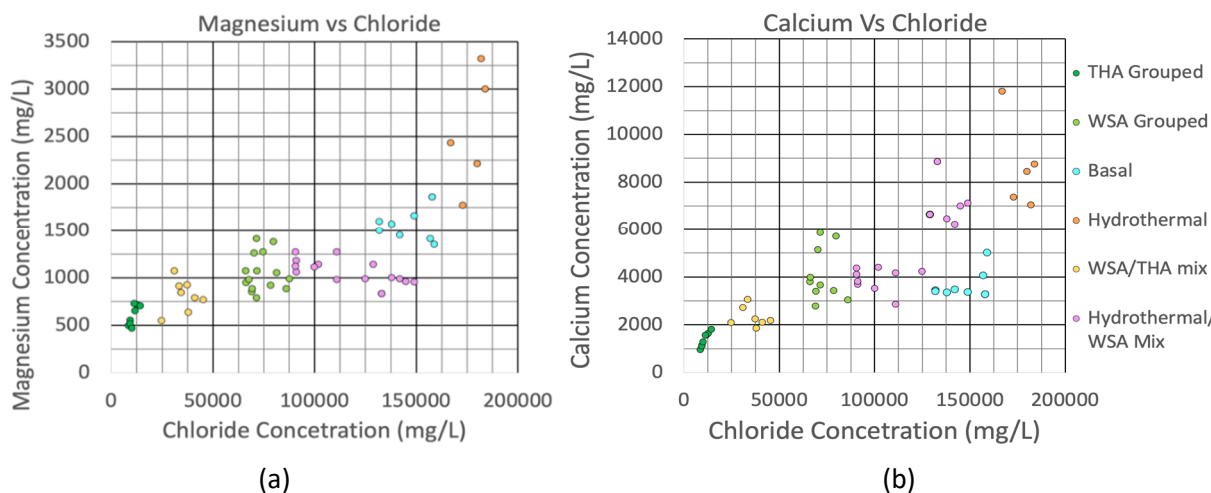


Figure 9 (a) Magnesium versus chloride plot and (b) calcium versus chloride plot for the classified dataset

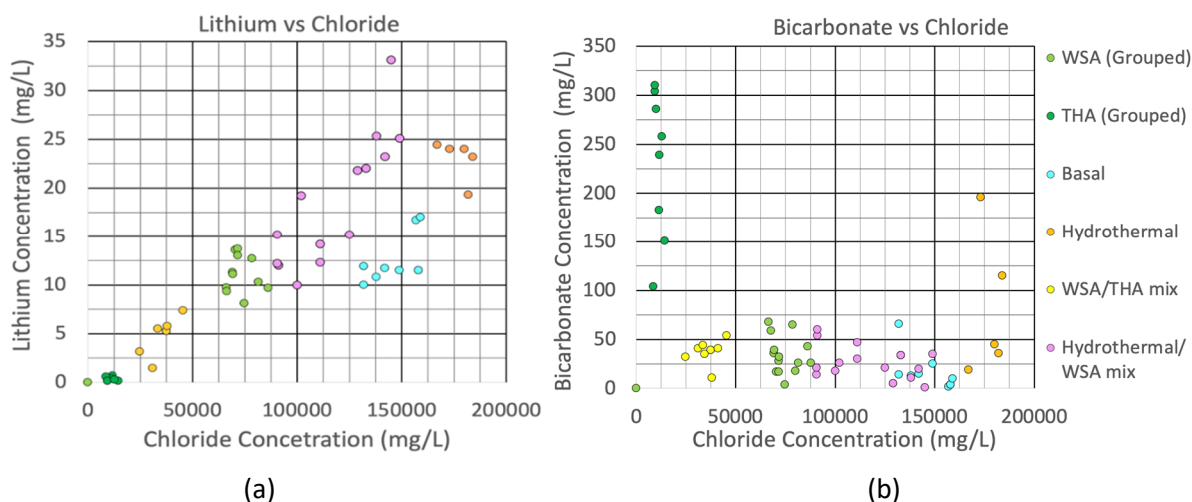


Figure 10 (a) Potassium versus chloride plot and (b) sodium versus chloride plot for the classified dataset

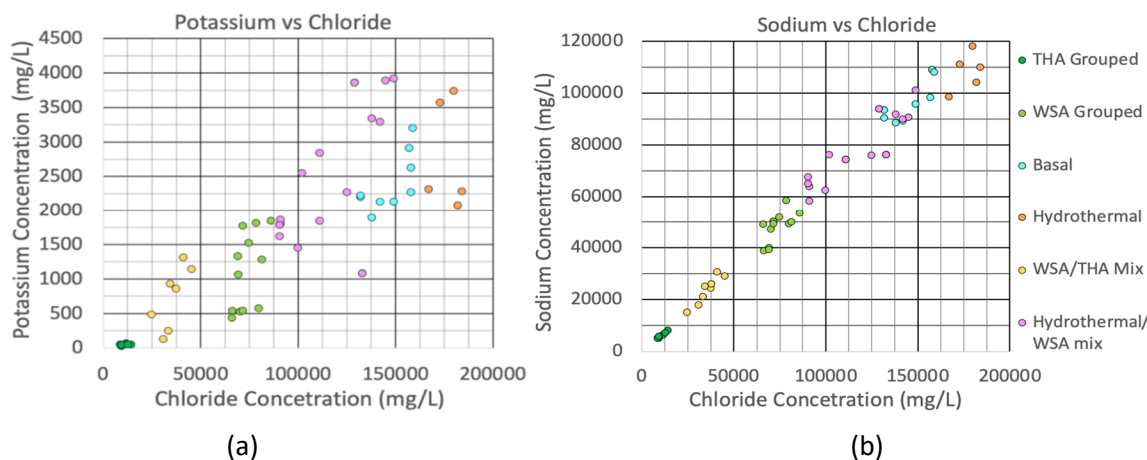


Figure 11 (a) Lithium versus chloride plot and (b) bicarbonate versus chloride plot for the classified dataset

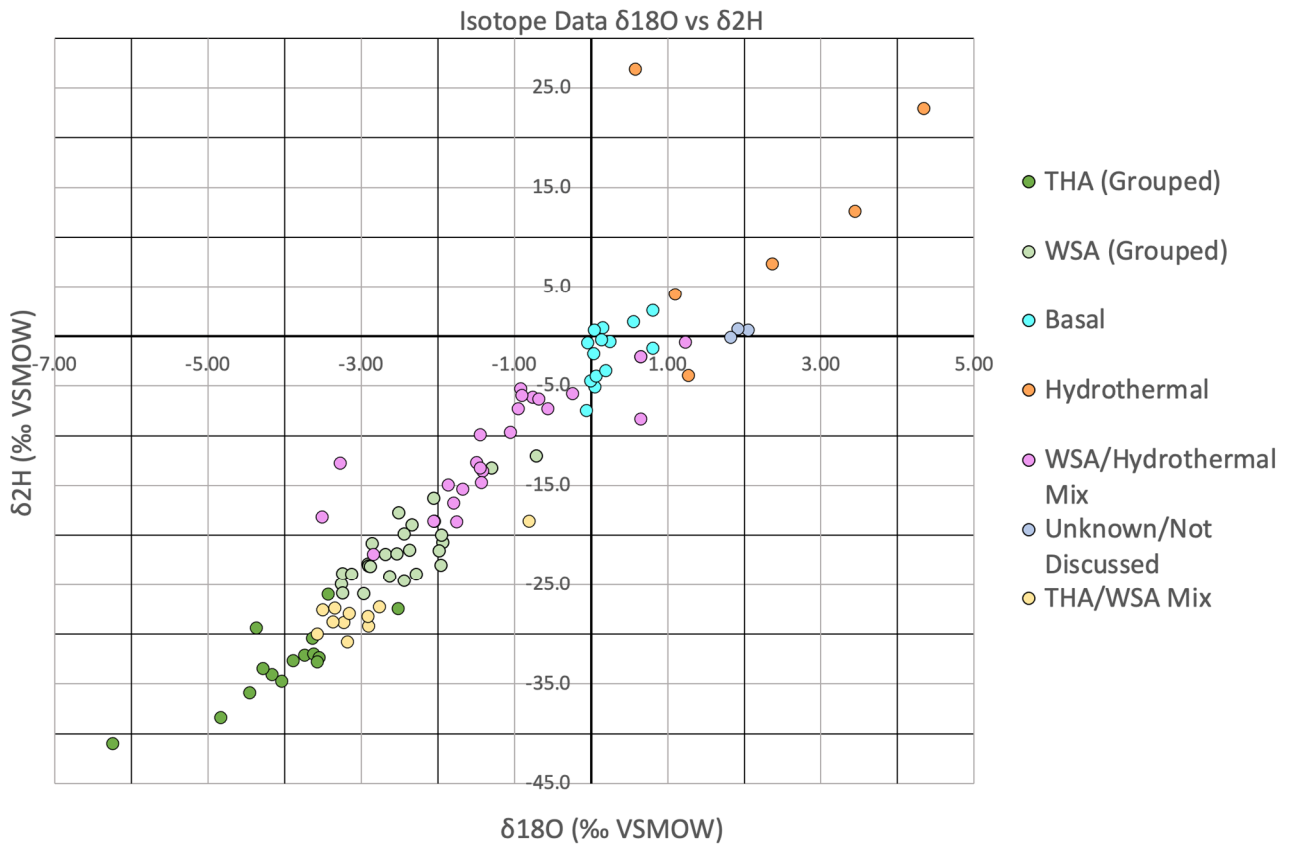


Figure 12 $\delta^{18}\text{O}$ versus $\delta^2\text{H}$ isotope plot, classified for the entire asset dataset

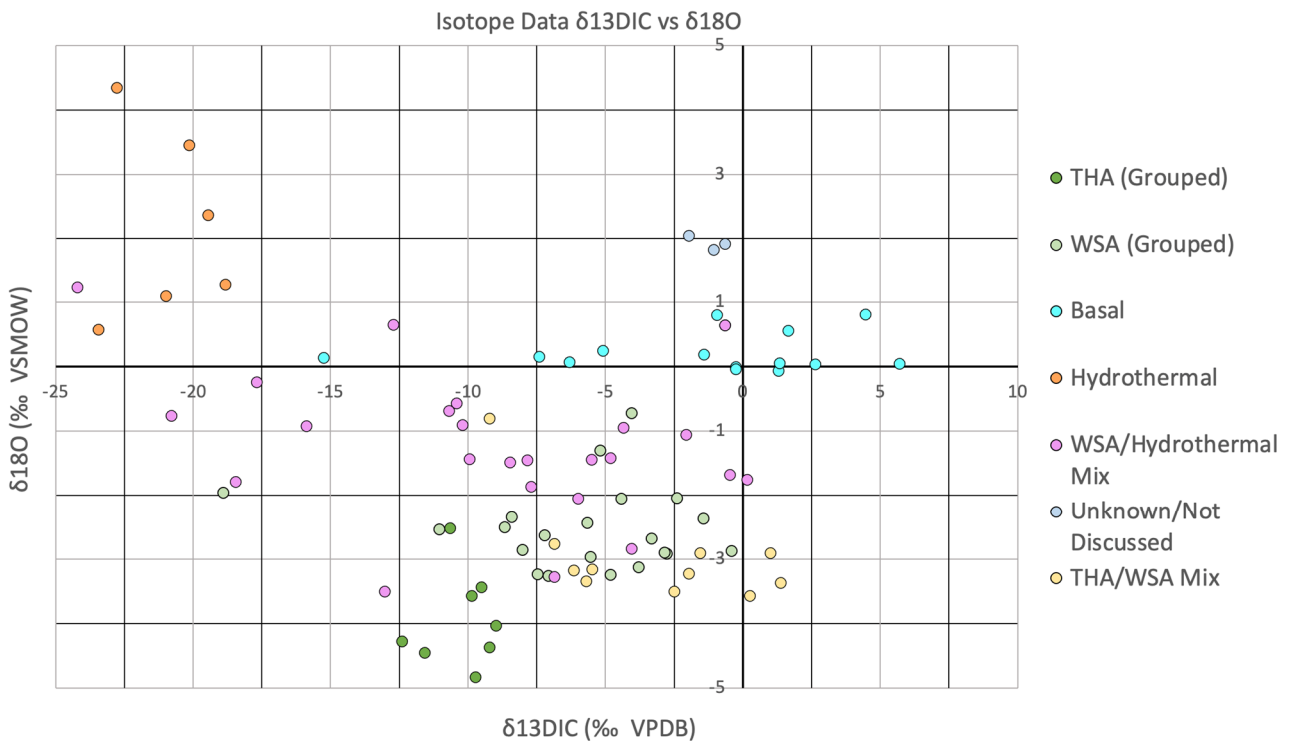


Figure 13 $\delta^{13}\text{C}$ - DIC versus $\delta^{18}\text{O}$ isotope plot, classified for the entire asset dataset

4.1.4 *Hydrochemical and isotopic classification of waters as per the literature.*

The most interesting hydrochemical groupings are that of the Hydrothermal and Basal. This section is a justification for referring to them as active hydrothermal systems; not just because they present hot upwelling fluid, but because they are chemically enriched when compared to standard subsurface brines. The literature on subsurface water is primarily derived from deep aqueous fluids within sedimentary basins intersected during oilfield production and exploration (Kharaka & Hanor 2003) with some fluid analysis conducted on crystalline basement fluids from around the globe (Frape et al. 2003).

Despite residing within a crystalline host rock (Granite) these two fluid groupings do not seem to present as such in their isotopic compositions (Frape et al. 2003; Kharaka & Hanor 2003). Figure 1 shows a clear shift to the right of the Global Meteoric water line for the entire dataset, indicating evaporative enrichment commonly observed in sedimentary brines (Frape et al. 2003) or geothermal alterations taking precedence (Clark & Fritz 1997). Furthermore, isotopically heavy (positive) values for crystalline basement water are extremely uncommon (Frape et al. 2003). Isotopically, the water of the asset appears more as a meteoric fluid involved in the dissolution of halite or as a deep ocean brine derived from the subaerial evaporation of sea water (Posey & Kyle 1988; Kharaka & Thorsden 1992).

Certain trends have been established for these meteoric/ocean waters and a departure from these often indicates that either the water was not derived of a traditionally studied source, or that there has been alteration to the fluid, whereby some diagenetic, metamorphic or hydrothermal interaction has imparted influence (Kharaka & Hanor 2003).

Trend departure is observed in both the hydrothermal and basal groupings, primarily in the calcium, magnesium and lithium concentrations and the isotopic groupings of $\delta^{18}\text{O}$, $\delta^2\text{H}$ and $\delta^{13}\text{C-DIC}$.

Trends commonly observed in the literature for deep sedimentary brines of Northern American oilfields at depths up to 5,000 m are:

- Decreasing magnesium concentration with a hotter (deeper) source (Kharaka & Hanor 2003).
- Decreasing pH with increasing chloride content (Kharaka & Hanor 2003).
- A pH of between 6-8 (Kharaka & Hanor 2003).
- A log-log plot of potassium to chloride slope of 1:1 (Kharaka & Hanor 2003).
- A log-log plot sodium to chloride slope of 1:1 (Kharaka & Hanor 2003).
- A log-log plot of calcium to chloride of 2:1 (Kharaka & Hanor 2003).
- A log-log plot of magnesium to chloride that is 2:1 (Kharaka & Hanor 2003).
- A lithium content of between 0–5 mg/L (Kharaka & Thorsden 1992).
- A magnesium content of between 200–1,400 mg/L (Kharaka & Thorsden 1992).
- A calcium content of between 1,000 – 4,000 mg/L (Kharaka & Thorsden 1992).
- $\delta^{18}\text{O}$ versus $\delta^2\text{H}$ values that are isotopically lighter (negative) than that of the VSMOW.
- $\delta^{13}\text{C-DIC}$ values that are +/- 4‰ where no organic methane is presumed to have been produced but marine carbonates were expected (Kharaka & Hanor 2003).

The following is a series of observations of the assets water drawn against the above criteria:

- Figures 9 through 11 show the water constituent of the basal and the hydrothermal groupings either reside outside of the expected hydrochemical thresholds or on the edge of the range for deep, unaltered sedimentary fluids. This indicates that the fluids have been enriched and involved in some diagenetic, metamorphic or hydrothermal process. (Kharaka & Hanor 2003; Holser 1979).

- Figure 12 shows the water constituent of the basal and hydrothermal groupings reside outside the isotopic ranges expected for entrapped meteoric water (isotopically lighter than VSMOW). Whereby, both the basal and hydrothermal groupings plot as evaporated sea water as opposed to entrained meteoric water (heavier or equal to VSMOW) (Kharaka & Hanor 2003; Holser 1979).
- Figure 13 shows the water constituent of the basal grouping is centred around (0,0) with a spread of $\pm 5\text{‰}$ for the $\delta^{13}\text{C}\text{-DIC}$ values, indicating that there is likely a marine carbonate component influencing the composition of the water and that it is ocean water (Kharaka & Hanor 2003).
- Figure 13 shows that the water constituent of the hydrothermal grouping ranges from -18 to -25‰ $\delta^{13}\text{C}\text{-DIC}$. The literature indicates this could be a result of entrained thermogenic methane (-20 to -50‰) mixed with CO_2 from mantle outgassing (-4 to -7‰) (Kharaka & Hanor 2003). Similarly, dissolution of carbonate minerals and oxidation of organic carbon can produce a very large range of between -60 to 10‰ depending on the source of the carbon and the fractionation factors accompanying the bicarbonate ions (Robinson 1993). $\delta^{13}\text{C}$ composition of freshwater carbonate are much wider ranging between -30 to 10‰ whereas the marine carbonates exhibit more enriched values (-5 to 5‰) (Clark & Fritz 1997). Considering that the hydrothermal dataset is particularly enriched in bicarbonate as compared to the rest of the samples taken underground, shown in Figure 11, it is presumed that dissolution of carbonate minerals is more likely than thermogenic methane entrainment. Methane gas is not detected in the mine; however, further assessment of geochemical interaction is warranted.
- In regard to plotting slopes on log-log charts, the populations for each dataset are not sufficiently populous to accurately infer ratios. When plotted as a whole, they reflect the trends described in the literature with each subset deviating slightly in its own unique way. This slight deviation indicates that the water is likely composed of lightly diagenetically, metamorphically, or hydrothermally altered evaporated sea water.
- The basal and hydrothermal groupings also show a departure from the literature in terms of pH. The basal group plots below the expected range for deep brines and the hydrothermal group plots too high for a brine consistent with the chloride content. It is likely that the dissolution of carbonate material is causing the buffering of the pH in this particular water. The pH versus cations graph for all classified waters is shown in Figure 14.

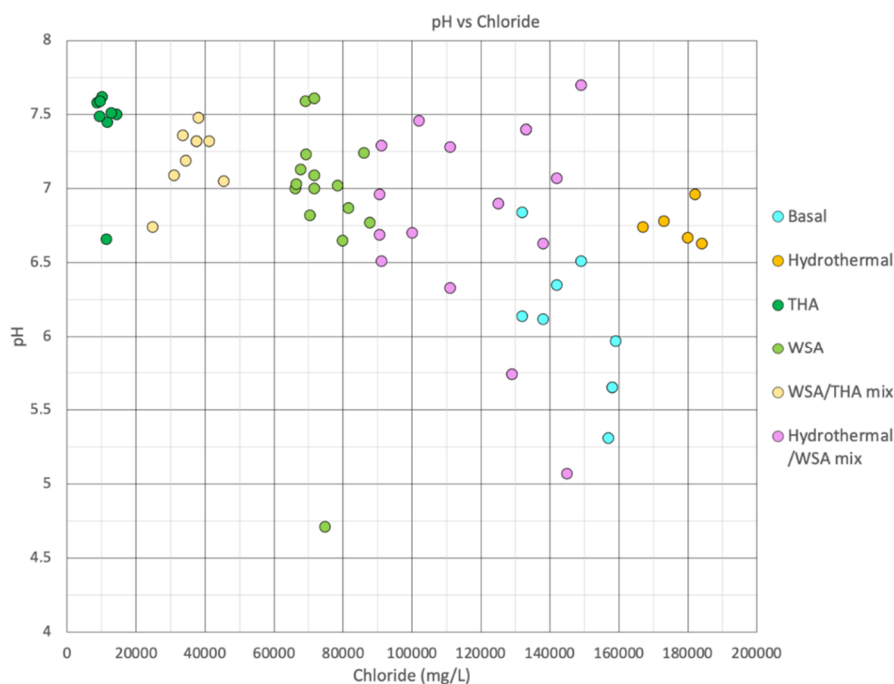


Figure 14 pH versus chloride for all classified data

As a whole, the comparison to the literature reveals that the basal and hydrothermal waters are more enriched in magnesium, calcium and lithium than an expected deep sedimentary brine. They are also not of expectant pH values for their chloride content.

In a hydrochemical sense, they plot as a fluid that has resided within a crystalline basement and in the specific case of the hydrothermal grouping, the water has undergone the dissolution of carbonates. It is likely that the diagenesis has been brought on by the heat of the fluid.

In an isotopic sense, both of the basal and hydrothermal waters plot as evaporated sea water as opposed to meteoric origin.

Thus, the basal water, plotting about (0,0,0) for isotopic compositions of $\delta^{18}\text{O}$, $\delta^2\text{H}$ and $\delta^{13}\text{C-DIC}$ with enriched, acidic hydrochemical properties is defined as slightly evaporated sea water that has undergone a diagenetic process in the presence of a heat source.

Likewise, the hydrothermal water, plotting as isotopically enriched water in $\delta^{18}\text{O}$ and $\delta^2\text{H}$ and depleted in $\delta^{13}\text{C-DIC}$ with even greater enriched hydrochemical properties is also defined as more evaporated, possibly older, sea water that has undergone dissolution and diagenesis in the presence of a heat source.

Radiogenic heat is the most likely source of the thermal influence. It is well documented that the Proterozoic Terranes which host the iron oxide, copper, gold (IOCG) asset have anomalously high crustal heat flow due to radioactive decay. (Pollett et al. 2003.) Hence, both fluids are classified as active, radiogenic hydrothermal water sources.

5 Conclusion

Through isotopic analysis of $\delta^{18}\text{O}$, $\delta^2\text{H}$ and $\delta^{13}\text{C-DIC}$ and the complementary analysis of hydrochemical data; two new, previously uncharacterised, and unknown water sources were identified, fingerprinted and distinguished from the previously quantified, overlying sedimentary aquifers. A literature review was able to characterise these two sources as active hydrothermal systems, both created by radiogenic geothermal heating of evaporated sea water, one of which is likely modern, plotting about (0,0,0) isotopically and the other which is unknown in age. The sampling campaign was successful in categorising the water of the SLC, allowing for future investigations into the flow of water throughout the SLC and approaching block cave and most importantly distinguished the upwelling basal fluid as non-mixing, isolating it such that its significance and contribution to inrush hazard can be further investigated and defined through a water mapping campaign. The establishment of fluid fingerprints for the SLC now allows for the determination of fluid source in the event of increased fluid flow or inrush event. Future investigations incorporating age tracer dating would help verify the residence times of water.

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