

# Using Environmental Isotopes in a Coal Mine and a Gold Mine to Determine Groundwater Interaction

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**Abstract** The use of stable isotope ratios of deuterium ( $\delta D$ ) and oxygen ( $\delta^{18}O$ ) together with major ion data can indicate the origins and movement of surface water and groundwater. During this investigation, hydrochemical and stable environmental isotope sampling were used to determine the interaction between the Karoo aquifer and the Witwatersrand aquifer. It was evident that shallower groundwater samples taken from the Karoo aquifer were affected by annual precipitation and had a short residence time, since their isotopic signature was very close to that of rainfall. By contrast, rainfall events had not recharged the Witwatersrand aquifer and had not significantly contributed to the isotopic fingerprint of the deeper aquifer samples. The Karoo aquifer is generally enriched in both  $\delta^{18}O$  and  $\delta D$ ; the deeper Witwatersrand aquifer is more depleted in  $\delta^{18}O$  and  $\delta D$ . This is explained by their different intake histories and travelling periods. The results also indicate that there is interaction between the Karoo and Witwatersrand aquifer, both where coal mining is present and where it is not.

**Keywords** Deuterium ( $\delta D$ ) · Environmental isotopes · Karoo aquifer · Oxygen ( $\delta^{18}O$ ) · Witwatersrand aquifer

## Introduction

Oxygen-18 ( $^{18}O$ ) and deuterium ( $^2H$  or  $D$ ), which are stable isotopes, are useful in hydrogeological investigations. Isotopes act as naturally occurring tracers in groundwater (environmental tracers) and can provide valuable information to the hydrogeologist about aquifer characteristics and groundwater flow paths that would otherwise be difficult, if not impossible, to establish (Verhagen et al. 1991). Tracking changes of  $^{18}O/^{16}O$  and  $^2H/^1H$  concentrations along groundwater flow paths are an effective way to determine the altitude of groundwater recharge, estimate mixing proportions of different sources or component flows, and determine the relationships between groundwater and surface water (Gat 1996).

The Highveld Coal Field, which is situated in South Africa's Mpumalanga Province, partially overlies the deeper gold mining operations of the gold-bearing Kimberley reef. The study area is located in the Area H coal deposits, where the No. 4L coal seam will be extracted when the existing colliery is expanded into the Area H reserves to the west and north of its current operations. Refer to Vermeulen et al. (2013), elsewhere in this issue, for a detailed description of the study area, mining methods, and aquifer information. This study aimed to characterise the origin of groundwater within the various aquifers, determine the interaction between the Karoo aquifer and the Witwatersrand aquifer through hydrochemical and stable environmental isotope sampling, and interpret and “fingerprint” the origin of each sample using hydrochemical and isotope analysis.

In order to characterise the origin of the groundwater within the various aquifers, X–Y column plots were used to fingerprint groundwater types with different flow paths.

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The positions of the sampling points for which ground-water chemistry data are available are shown in Fig. 1 in Vermeulen et al. (2013). It is important to note from the layout of the mines and the positions of the samples that only gold mine samples H6, H7, and H10 might be affected by coal mining, as mining has not yet reached the position of the other gold mine samples (Vermeulen et al. 2013).

## Sampling

To determine the origin and composition of groundwater in the gold and coal mine, sampling was done at strategic positions in each mine. Samples were taken from surface boreholes as well as underground at different depths within in each mine where water was found. A summary of sampling positions is displayed in Table 1 and in Table 2 of Vermeulen et al. (2013), elsewhere in this issue.

The same sampling protocol was applied during each sampling survey. Macro elements, as well as the stable isotopes  $^{18}\text{O}$  and  $^2\text{H}$ , were analysed.

Hydrochemical constituents were sampled using new 250 mL plastic sampling bottles. To avoid contamination, the sample bottles were rinsed before each sample was taken, first with sterilised water and then with water from the sampling point. Sample bottles were sealed and labelled appropriately. After sampling was completed, the samples were stored in a cool ( $<10^\circ\text{C}$ ), dry storage place until delivered to the laboratory.

Water samples were collected for  $\delta\text{D}$  and  $\delta^{18}\text{O}$  analyses in new 250 mL amber glass bottles to avoid contamination. The bottles were first rinsed with the sample water, then completely filled and sealed. The water was not treated in any way before or after sampling.

## Methods

The electrical conductivity (EC) and pH values were measured in the laboratory using a portable conductivity- and pH meter respectively. In the laboratory, the water samples were filtered through  $0.45\ \mu\text{m}$  Millipore membrane filters to separate suspended particles. Acid titration was used to determine the concentration of bicarbonate in the mine water samples.

Concentrations of major cations (Na) were determined using inductively coupled plasma atomic emission spectroscopy (ICP-AES), also referred to as inductively coupled plasma optical emission spectrometry (ICP-OES). The analytical precision was maintained by running a known standard after 15 samples. The overall precision (expressed as percentage relative standard deviation, RSD) was below 5 % for all samples. Major anions ( $\text{SO}_4^{2-}$  and  $\text{Cl}^-$ ) were analyzed with an ion-exchange chromatography (or ion chromatography) using AS12A/AG12 columns coupled to an anion self-regenerating suppressor (ASRS) in recycle mode.

## Hydro-chemical Discussion

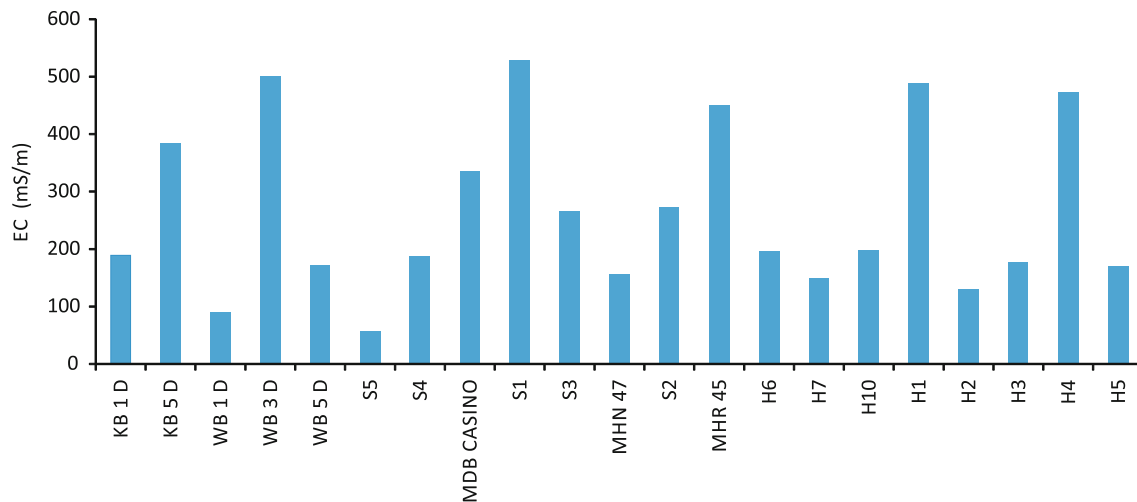
In order to characterise the origin and composition of the groundwater within the various aquifers, X–Y column plots were used to fingerprint groundwater types with different flow paths. Groundwater quality information was obtained for a total of 21 sampling points.

### Electrical Conductivity (EC)

EC is an indication of the dissolved mineral content of groundwater. The EC values of the samples ranged between 56.2 mS/m (S5) and 529 mS/m (S1), with an

**Table 1** Summary of samples taken from boreholes on surface of the study area

Gold mine	Coordinates	Coordinates	Elevation (m.a.m.s.l)	Sampling depth below surface (m)	Borehole depth below surface (m)	Depth below surface (m)
Borehole	Y	X	Z			
KB 1 D	–2,929,816.932	7,611.298	1,591.324	28	30	3.5
KB 5 D	–2,929,401.146	8,886.292	1,615.099	28	30	1.02
WB 1 D	–2,932,446.352	10,378.745	1,589.468	28	30	1.6
WB3D	–2,933,838.364	10,023.214	1,599.229	28	30	1.3
WB 5 D	–2,933,225.999	11,669.846	1,621.612	28	30	22
Coal mine	Y	X	Z			
MHN 47	–2,935,823.037	11,478.324	1,578.937	110	In mine	108.19
MHR 45	–2,935,046.371	10,963.433	1,600.137	130	In mine	129.44
MDB CASINO	–2,933,683.916	15,952.46	1,581.346	100	In mine	97.67



**Fig. 1** EC values (millisiemens per meter (mS/m)) of samples collected in this study, from shallow (*left*) to deep (*right*)

average value of 265 mS/m. The samples are plotted shallow to deep, from left to right, in Fig. 1.

Groundwater EC does not appear to increase from the shallower Karoo aquifer samples (KB1D to MHR45) towards the deeper Witwatersrand aquifer samples (H1–H10) shown in Fig. 1, since KB1D (shallowest at 189 mS/m) and H5 (deepest at 169 mS/m) share the same order of magnitude. This unexpected finding is attributed to geological structures, such as sills and dykes, which created separate groundwater compartments by displacing the coal seams during dolerite intrusion. These groundwater compartments may receive less or more recharge than the surrounding aquifer (Vermeulen and Usher 2006a), which increases or decreases the groundwater salinity (Hodgson and Krantz 1995). This can be seen in samples MDB CASINO (336 mS/m), MHN47 (156 mS/m), and MHR45 (449 mS/m), all of which were taken from the already water-filled underground coal mine workings. The range of 293 mS/m between MHR45 (449 mS/m) and MHN47 (156 mS/m) indicates not only chemical reactions specific to compartments, but also a difference in recharge potential (Hodgson and Krantz 1995).

Variation in recharge potentials and chemical reactions may be specific to aquifer and sampling depth. This can be seen by comparing sample S5 (56.2 mS/m), taken from the shallow Karoo aquifer, to sample H1 (488 mS/m), from the deeper Witwatersrand aquifer. The difference in EC value is 431.8 mS/m between H1 (Witwatersrand) and S5 (Karoo).

Tailing storage facilities and other contamination sources can also affect the EC of the samples, especially the shallow Karoo aquifer samples taken down-gradient or on preferred pathways near the gold mine TSF. This is illustrated in samples WB1D (89.1 mS/m), which is situated

down-gradient and in close proximity to slimes dam no 3, and WB3D (500 mS/m), which is unaffected by the slimes dam.

The EC values of the gold mine samples H1 (488 mS/m) and H4 (472 mS/m) are much higher than the other gold mine samples. Both of these samples were taken from faults intersecting the gold mine workings, suggesting that their origin is the base of the Karoo or Transvaal Supergroup, as the faulting pre-dates the deposition of the Transvaal and hence the Karoo Supergroup (Tweedie 1986). They were taken at 1,380 m (H1) and 1,830 m below the surface (H4). The Karoo Supergroup is approximately 210 m thick. This shows that these waters had to travel approximately 1,170 m (H1) and 1,620 m (H4). Migration and dissolution along the fault plane causes higher EC values.

H3 (177 mS/m) was taken from an exploration borehole intersecting the gold mine workings. Its EC value is much lower, which suggests that it has been mixed with precipitation. The borehole is directly linked to both the surface and the Karoo aquifer, and the lower EC values presumably reflect a relatively short residence time.

If the shallower Karoo aquifer were well-connected with the deeper Witwatersrand aquifer, one would expect to see a significant increase of EC from the Karoo aquifer samples to the deeper Witwatersrand aquifer, which is not the case.

## pH

Groundwater pH of all the samples in the study area is near-neutral to slightly alkaline, ranging from 6.33 (MHR 45) to 8.8 (H2), with an average pH of 7.73. This circumneutral pH is due to the naturally occurring marine carbonate minerals that are present in the Karoo Supergroup in the study area

(Vermeulen and Usher 2006b). The lack of oxygen in the deeper samples could also contribute to a more alkaline pH because oxidation reactions tend to be acid forming (Appelo and Postma 2005). Most metals are immobile in the marginally alkaline regime in the study area. Groundwater pH does not appear to increase or decrease from the shallower Karoo aquifer [samples KB1D (7.4) towards the deeper Witwatersrand aquifer samples H5 (8)].

### Sodium (Na)

The deeper the groundwater moves in the study area, the more enriched it becomes in Na as more dissolution and cation-exchange takes place. The Na concentration ranged from 91.3 mg/L (S5) to 816.1 mg/L (H1), with an average of 394.5 mg/L (Fig. 2). This indicates that groundwater in the deep Witwatersrand aquifer is either derived from groundwater moving slowly through the sub-surface and reacting with Na-rich minerals through dissolution and cation exchange, or it is paleo-meteoric water that was trapped during the deposition of the Witwatersrand sediments. The latter should have a high Na concentration due to the deposition environment and relatively long residence times (Onstott et al. 2006).

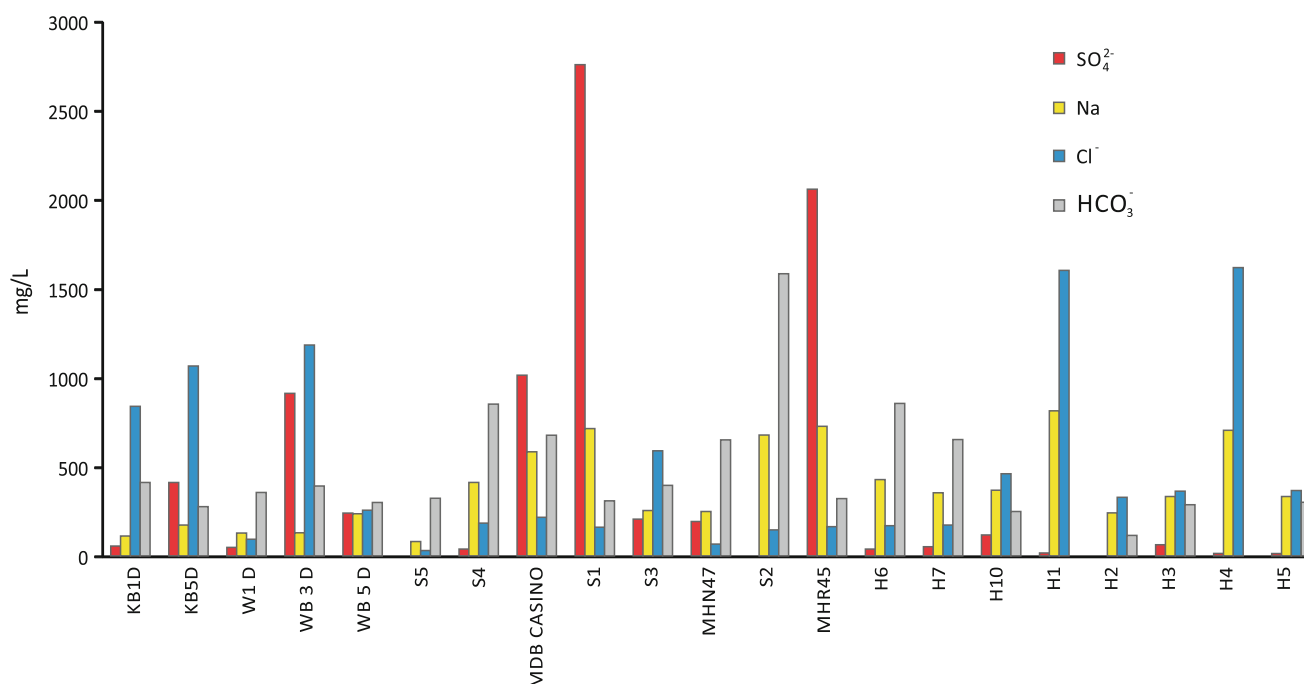
H1 (816 mg/L) and H4 (717 mg/L) (water from the fault, Table 2 of Vermeulen et al. 2013) are higher in Na than the other gold mine samples (see also EC). A preferred conduit and enhance comminution of rock fragments along the fault plane promote dissolution and cation exchange.

The lower Na concentration in H3 (347 mg/L), an exploration borehole (Table 2 of Vermeulen et al. 2013) suggest a relatively short residence time (greater transmissivity) from the Karoo to the Witwatersrand aquifer.

MDB CASINO (595 mg/L), S1 (729 mg/L), S2 (691 mg/L), and MHR45 (740 mg/L), which were all collected from within the water-filled coal mine, are all elevated in Na. Surface water that drains towards and into the collapsed columns above high extraction areas undergoes base exchange reactions; Ca and Mg are adsorbed onto clays within the sediments in exchange for Na (Eby 2004). Interstitial release of Na from the shale occurs as water flows down the fractured columns (Hodgson and Krantz 1995). The degree to which cation exchange has affected water chemistry is a function of the distance that the water has travelled and the composition of the rock through which it has moved. The resultant water that ends up on the coal seam horizon ranges from water almost totally devoid of Ca and Mg to water that may be slightly enriched in Na (Hodgson and Krantz 1995).

### Sulphate ( $\text{SO}_4^{2-}$ )

The  $\text{SO}_4^{2-}$  concentration ranges from 4 mg/L (H2) to 2,759 mg/L (S1), with an average value of 397.8 mg/L (Fig. 2). Most of the shallower samples taken from the Karoo aquifer (KB5D, WB3D, MDB CASINO, S1, S3, MHN47, MHR45) have a higher  $\text{SO}_4^{2-}$  concentration than the deeper samples (H1–H10) taken from the



**Fig. 2** Sodium, sulphate, chloride, and bicarbonate concentrations for the samples collected in this study, from shallow (*left*) to deep (*right*)

Witwatersrand aquifer. The samples taken from the Witwatersrand aquifer (H1–H10) were either water dripping from the mine roof, fault, or from an exploration borehole. They have a much lower  $\text{SO}_4^{2-}$  levels than the shallower Karoo aquifer samples. This can be attributed to a reducing environment (lack of oxygen). The varying  $\text{SO}_4^{2-}$  concentration could also be attributed to the varying geology and compartmentalisation, which in some places may or may not contain sulphate-rich minerals with more or less water and oxygen.

Samples WB3D, MDB CASINO, S1, and MHR45, all taken from the Karoo aquifer, have relatively high concentrations of  $\text{SO}_4^{2-}$  compared to other samples taken in the Karoo aquifer. WB3D's elevated concentration can be attributed to its close downstream proximity to slimes dam B, whereas elevated concentrations in samples MDB CASINO, S1, and MHR45 can be attributed to the oxidation of sulphate minerals within the coal mine (Vermeulen and Usher 2006b).

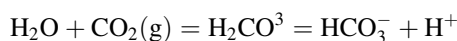
The elevated sulphate concentrations in samples MDB CASINO (1,024 mg/L) and S1 (2,749 mg/L) indicate that the coal seam has a high acid-generating potential (Hodgson and Krantz 1995). However, the alkaline pH level of the samples suggests that the coal seam also has sufficient base potential to neutralise the generated acid at this point in time. The sulphate concentrations in the gold mine samples (H1–H10) are much lower than those of the shallow Karoo aquifer samples (Fig. 2), despite the presence of sulphate-generating minerals, such as pyrite, in the gold mine.

### Chloride ( $\text{Cl}^-$ )

The chloride concentrations range from 24.8 mg/L (S5) to 1,612 (H4) mg/L, with an average value of 476.9 mg/L, and increase with depth (Fig. 2). Samples KB1D, KB5D, and WB3D are elevated in chloride; this is attributed to the chloride-rich shales found in the Eccia formation of the Karoo Supergroup. The elevated  $\text{Cl}^-$  concentrations of H1 and H4 were undoubtedly enhanced by interaction with gouge in the fault zone.

### Bicarbonate ( $\text{HCO}_3^-$ )

The relative concentrations of inorganic carbon-containing species ( $\text{H}_2\text{CO}_3$ ,  $\text{HCO}_3^-$ , and  $\text{CO}_3^{2-}$ ) depend on the pH.  $\text{CO}_2$  is enriched in the soil because of decomposition of organic carbon and carbonate mineral dissolution. As recharge water moves through the unsaturated zone,  $\text{CO}_2$  dissolves to form carbonic acid ( $\text{H}_2\text{CO}_3$ ).



As the pH of the recharge increases, because of neutralization reactions with soil and aquifer minerals,

carbonic acid converts to bicarbonate ( $\text{HCO}_3^-$ ). Bicarbonate predominates within the pH range from 6.36 to 10.25 in fresh water (Appelo and Postma 2005; Drever 1997), so it would be expected to be the primary inorganic carbon species in the water sampled.

The decreasing trend in  $\text{HCO}_3^-$  concentrations in samples taken from the Karoo aquifer (Fig. 2) reflects the decreasing effect of recharge precipitation. This indicates that recharge for the Witwatersrand aquifer is via the Karoo aquifer, where H1 has 0 mg/L and S2 has 1,585 mg/L  $\text{HCO}_3^-$  in solution.

KB1D, KB5D, W1D, WB3D, WB5D, S5, S4, MDB CASINO, S1, S3, MHN47, S2, MHR45, H6, and H7 all have relatively high concentrations of  $\text{HCO}_3^-$ , suggesting that they are recharged by precipitation or water in the shallow unconfined Karoo aquifer that freely reacts with the atmosphere. Although H6 and H7 were not taken directly from the Karoo aquifer, their  $\text{HCO}_3^-$  concentration indicates that they are of Karoo aquifer origin.

H2, H3, H5, and H10 have higher  $\text{HCO}_3^-$  concentrations than the other gold mine samples if H6 and H7 are excluded. H3 is from an exploration borehole intersecting the gold mine, and therefore its  $\text{HCO}_3^-$  concentration is expected to be a mixture of precipitation and flux from confined aquifers. H2, H5, and H10 represent an influx of confined aquifer water taken from fractures intersecting the gold mine. Their lower  $\text{HCO}_3^-$  concentrations, compared to H3, are attributed to a lack of  $\text{CO}_2$  present (confined aquifer), resulting in the slow loss of  $\text{HCO}_3^-$  (Freeze and Cherry 1979). This explains the lower  $\text{HCO}_3^-$  concentrations in H2 and H5. It indicates that these waters are recharged by the shallower Karoo aquifer, but that it has had a long residence time in the sub-surface.

H1 and H4 were both taken from faults, and their  $\text{HCO}_3^-$  concentrations suggest that they are recharged by influx from a confined aquifer below the No. 4L coal seam, presumably the deep fractured Karoo aquifer, or the Transvaal dolomites.

### Stable Environmental Isotopes

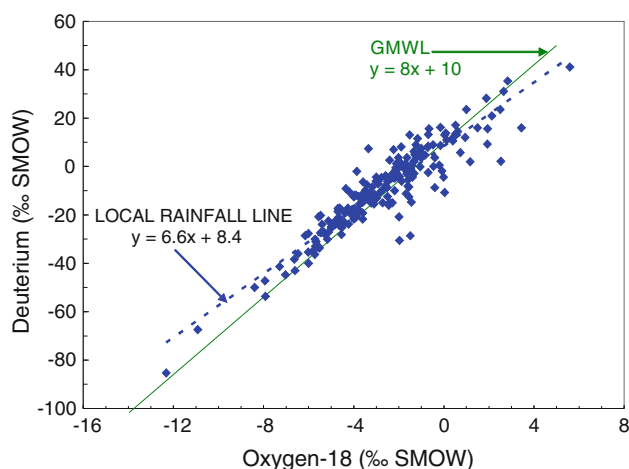
Deuterium and oxygen-18, which are stable isotopes, are commonly used to determine the average elevation of recharge for a particular mass of groundwater. The ratios of deuterium ( $\delta\text{D}$ ) and oxygen ( $\delta^{18}\text{O}$ ) in the water molecule undergo small changes through fractionation processes such as evaporation and condensation (Fritz et al. 1980). The universal reference standard is the standard mean ocean water (SMOW). On a graph of  $\delta\text{D}$  against  $\delta^{18}\text{O}$ , with respect to SMOW, all current precipitation (meteoric water) worldwide plots along a linear regression trend called the Global Meteoric Water Line (GMWL) or the

universal meteoric water line (UMWL). The position of any precipitation sample on the line will be determined by geographic, climatologic, and physiographic factors. Isotopic ratios in groundwater are conservative in the sense that they are not readily altered by processes in closed systems and in the saturated zone. Even where subsequent changes in hydrochemistry might occur, the isotopic composition remains stable.

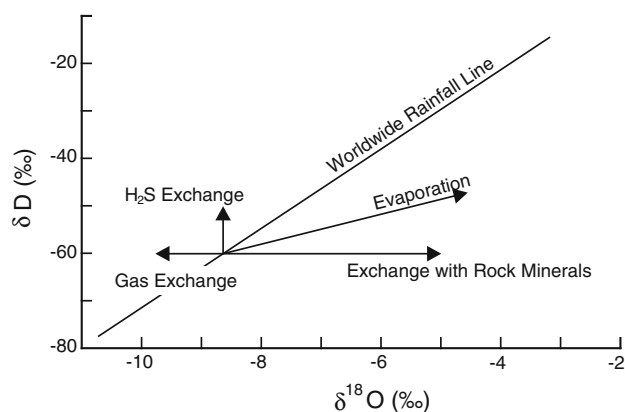
Oxygen and hydrogen are in some ways the perfect tracers of water because their concentrations are not subjected to changes by interactions with the aquifer material (UNESCO 1973). Once underground and removed from zones where fractionation through evaporation and condensation can occur, the isotope ratios are conservative and can only be affected by mixing. When precipitation infiltrates to recharge groundwater, mixing in the unsaturated zone smoothes the isotopic variations, meaning that water in the saturated zone has a composition that corresponds to the mean isotopic composition of infiltration in the area. This may differ slightly from the mean isotopic composition of precipitation, since not all precipitation during the year infiltrates in the same proportion (UNESCO 1973).

Analyses of  $\delta D$  and  $\delta^{18}O$  can be used to identify the probable source of underground water. If the isotopic composition of underground water plots close to the meteoric water line in a position similar to that of present-day precipitation in the same region, the water is almost certainly meteoric (Fig. 3), as indicated for the Pretoria rainfall region. Although the isotopic composition of precipitation at a particular location is approximately constant, it varies from season to season and from one rainstorm to another. These variations can be used to identify the season in which most of the recharge took place (Drever 1997).

Light isotopes are preferentially transferred to the vapour phase. The resulting surficial layer, which is



**Fig. 3** The relationship between  $\delta^{18}O$  and  $\delta D$  for Pretoria rainfall



**Fig. 4** Generalised  $\delta D$  versus  $\delta^{18}O$  plot showing the world rainfall (meteoric) line and processes commonly associated with deviation from the rainfall line (Fritz et al. 1980)

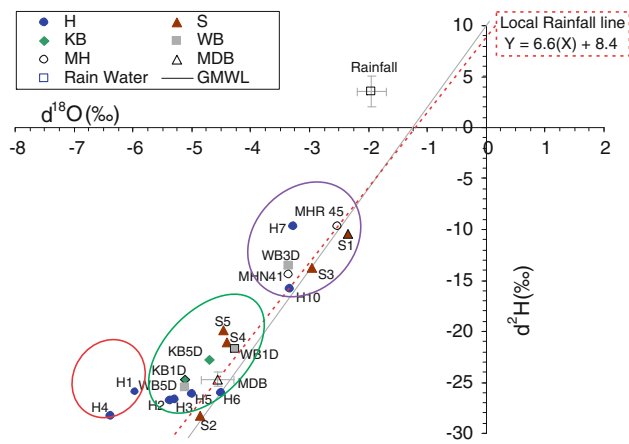
enriched in the heavy isotopes, is then readily mixed into the bulk of the water body through convective processes (Fig. 4). This evolutionary enrichment produces  $\delta D$  and  $\delta^{18}O$  values that lie to the right of the meteoric water line, and plot on an evaporation line of lesser slopes (usually 4–5) and lower D than the GMWL. Groundwater derived through infiltration from such water bodies will carry this distinctive evaporative isotopic signal (Gat 1996).

Regional groundwater flow in the study area follows a complex system of interconnected, structurally-controlled flow paths, incorporating multiple lithologies with both fracture and matrix-dominated flow. The use of oxygen and hydrogen isotopic composition proved to be a useful tool to fingerprint the origin of groundwater found in the study area and, more specifically, the origin of the deeper groundwater found in the gold mining areas. The isotopes  $\delta D$  and  $\delta^{18}O$  assisted in determining the type of water, as well as whether any groundwater flux exists between the mines. The thought process behind this was that if there is hydrologic interaction between Area H of the coal mine and the gold mine, then groundwater in both mines would have the same relative isotopic composition, and if the contrary is true and there is no groundwater interaction between the mines, then the relative isotopic composition would differ.

#### Stable Environmental Isotope Data

From the data plotted in Fig. 5, the following samples can be fingerprinted:

- The deeper Witwatersrand aquifer samples (H1, H2, H3, H4 and H5) are more depleted in  $\delta^{18}O$  and  $\delta D$  than the shallower Karoo aquifer (WB1D, WB3D, MHR45, MHN47, S1, S3, S4, and S5). The relative enrichment in both  $\delta^{18}O$  and  $\delta D$  in the Karoo aquifer (less negative) relative to the Witwatersrand aquifer



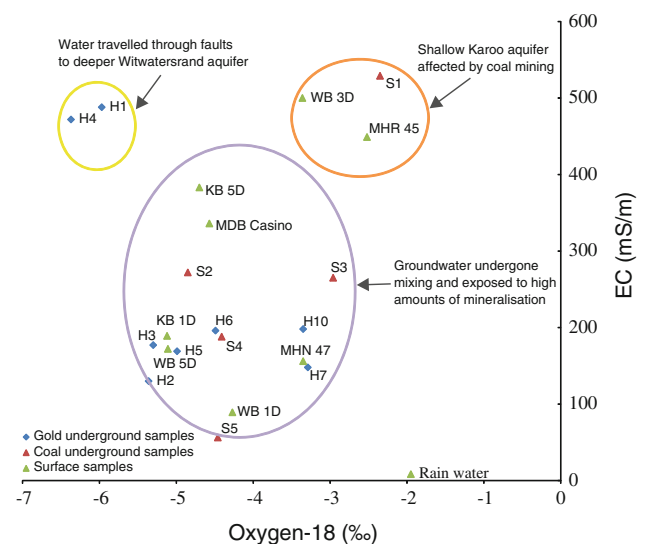
**Fig. 5**  $\delta^{18}\text{O}$  and  $\delta\text{D}$  relationship isotope plot for the samples collected in this study

(Fig. 3) can be explained by current precipitation recharge.

- H1 and H4 are more depleted in  $\delta^{18}\text{O}$  and  $\delta\text{D}$  than the other gold mine samples (Fig. 3). H1 and H4 are taken from faults intersecting the gold mine workings (Table 2 of Vermeulen et al. (2013), elsewhere in this issue). The outlying values for H1 and H4 are rationalized as a fault-related exchange process.
- H7 and H10 are water dripping out of the roof of the gold mine. Its isotopic “signature” is unrelated to the other gold mine samples. It plots with samples WB1D, WB3D, MHR45, MHN47, S1, S3, S4, and S5, which are samples from the coal mine. This indicates that the H7 and H10 water likely originated in the Karoo aquifer, most likely where it has been affected by coal mining.
- It was difficult to distinguish between the other samples (KB1D, KB5D, S2, and MDB CASINO) as they plot in the same area on the  $\delta^{18}\text{O}$  and  $\delta\text{D}$  relationship isotope plot (Fig. 5), with no unique isotopic signature. They all plot close to the local rainfall line (Fig. 5), suggesting that they have been affected in some way by precipitation, directly or indirectly (mixing), over a period of time.

### Major Ion Chemistry and Stable Environmental Isotopes

Scatter plots of ionic ratios and/or environmental isotopes were used to identify the groundwater origins in the aquifers. From Fig. 6, it is possible to distinguish between samples, and although no direct relationship between  $\delta^{18}\text{O}$  and EC exists, the combination of the information obtained independently from  $\delta^{18}\text{O}$  and EC is significant. The  $\delta^{18}\text{O}$  signature provides information on the recharge, whereas EC provides information on dissolution processes in the



**Fig. 6**  $\delta^{18}\text{O}$  versus electrical conductivity plot

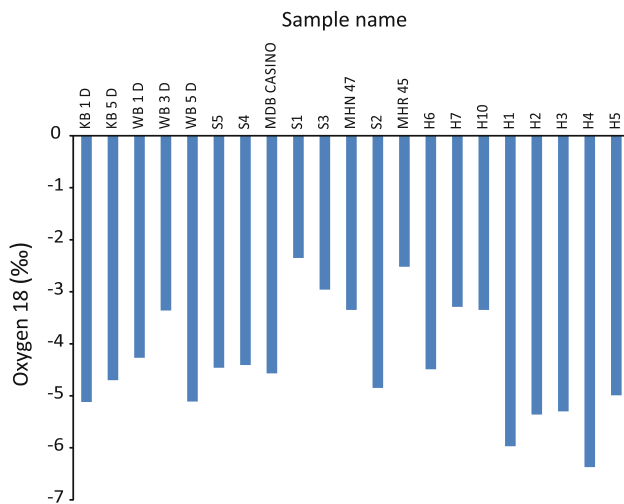
aquifer, flow process, and path. The Witwatersrand aquifer groundwater clusters around  $\delta^{18}\text{O}$  and EC values between  $-4.5$  to  $-6.4$  ‰ and 200–500 mS/m, respectively, whereas the Karoo aquifer sample clusters around  $\delta^{18}\text{O}$  and EC values between  $-2.3$  to  $-5.15$  ‰ and 89–500 mS/m, respectively (Fig. 6).

From Fig. 6, it is possible to distinguish three different groups of groundwater:

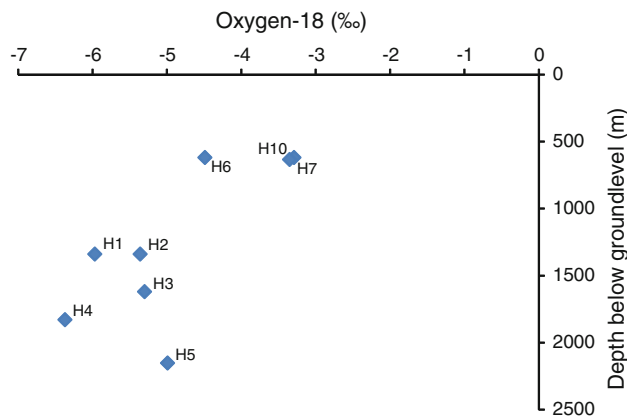
- S1, WB 3D, and MHR45—groundwater originating from the Karoo aquifer, which is recharged by rainfall on a fairly constant basis with a relatively short residence time (Fritz et al. 1980).
- H1 and H4—groundwater originating from the faults with a relative long travel time and exchange with rock minerals, as seen in Fig. 4 (Fritz et al. 1980).
- H2, H3, H5, H6, H7, H10, S2, S3, S4, S5, KB1D, KB5D, WB1D, WB5D, and MHN47—groundwater that has undergone a fair amount of mixing and mineralization. It is not possible to determine which aquifer these samples originate from using isotopic compositions alone. They all plot close to the local rainfall line (Fig. 5), suggesting that they have been affected in some way by precipitation, directly or indirectly (mixing), over a period of time.

### Depth and Stable Environmental Isotopes

- In Fig. 7, the samples were plotted from shallowest (KB1D) at 28 m, to deepest (H5) at 2,152 m below the surface. Figure 8 shows the depleted  $\delta^{18}\text{O}$  signature of the deeper gold mine samples, suggesting that water was recharged at a much earlier time with a much longer flow path.



**Fig. 7**  $\delta^{18}\text{O}$  versus depth for samples in the study area

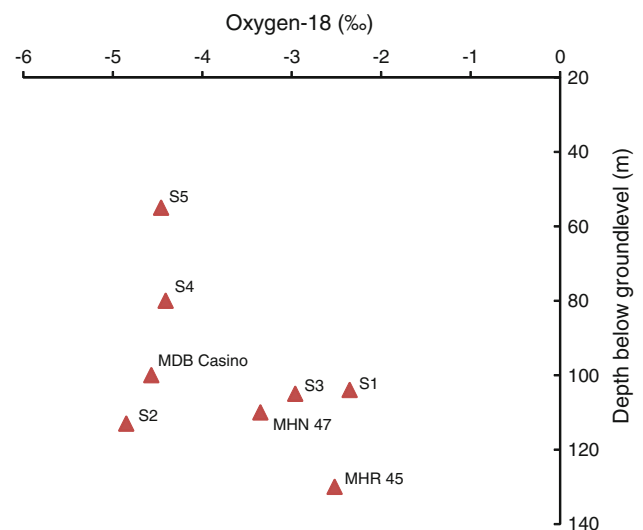


**Fig. 8**  $\delta^{18}\text{O}$  versus depth for underground samples originating in the gold mine

- Figures 7 and 8 show the depleted  $\delta^{18}\text{O}$  signature of the gold mine samples (H1, H2, H3, H4, H5, and H10) with depth. This indicates their long residence and flow paths to reach the gold mine.
- Figures 7 and 8 show that the  $\delta^{18}\text{O}$  signature for H6 and H7 is in the same order of magnitude as that of the coal mine samples (Fig. 9). They are of Karoo aquifer origin and they are the closest samples taken to the floor of the No. 4L coal seam. H1 and H4 are also believed to be of Karoo aquifer origin, but are much more depleted than samples H6 and H7.

## Discussion of Isotope Results

A rainfall signature prevails in the shallower groundwater samples (KB1D, KB5D, WB1D, WB3D, WB5D, MDB CASINO, S1, S2, S3, S4, and S5) taken from the Karoo



**Fig. 9**  $\delta^{18}\text{O}$  versus depth for underground samples originating in the coal mine

aquifer in the study area (Fig. 5). These samples are affected by a relatively large annual precipitation and apparently short residence time, because their isotopic signature is close to that of rainfall (Fig. 5). Therefore, most of the Karoo aquifer samples probably reflect the character of the largest rainfall events during the previous year. By contrast, the deep groundwater samples generally have a signature that differs from rainfall (Fig. 5). It is apparent that rainfall events have not recharged the deep aquifer over a short period of time because they have not made a significant contribution to the isotopic fingerprint of the samples found in the Witwatersrand aquifer.

Thus, the Karoo aquifer is generally enriched in both  $\delta^{18}\text{O}$  and  $\delta\text{D}$ , as shown in Fig. 5 (less negative). The deeper Witwatersrand aquifer (see samples H1–H5 and H10 in Fig. 8) is more depleted in  $\delta^{18}\text{O}$  and  $\delta\text{D}$  than the shallower Karoo aquifer (samples WB1D, WB3D, MHR45, MHN 47, S1, S3, S4, S5, and H7 in Fig. 9). This is explained by different intake events and residence times, with direct recharge from meteoric precipitation into the shallower Karoo aquifer, and lengthy indirect infiltration into the Witwatersrand aquifer.

H1 and H4 are the most depleted samples in  $\delta^{18}\text{O}$  and  $\delta\text{D}$  (Fig. 5). Their association with faults suggests reactions with gouge (comminuted rock), which played a part in their depleted isotopic composition (Fig. 4).

## Conclusions and Recommendations

The 21 samples collected, which represent a depth range between 28 and 2,152 m, contain the following three distinct types of sub-surface water regimes: (a) shallow Karoo

aquifer, (b) deep Witwatersrand Supergroup aquifer, and (c) a fault zone aquifer. Stable isotope ratios (hydrogen and oxygen), pH, EC, and solute concentrations (sodium, sulphate, chlorine, bicarbonate), were used successfully to discriminate and characterize these regimes (Vermeulen et al. 2013).

The source of water in samples H1 and H4 is from faults intersecting the gold mine. The origin of this water is from the base of the Karoo aquifer where the Transvaal Supergroup (aquiclude) is absent and where the faults extend through the Ventersdorp Supergroup (aquiclude) and into the Witwatersrand aquifer. Groundwater will move along these faults until it intersects the gold mine workings. Faults intersect the gold mine workings at 1,340 and 1,830 m in the No. 8 shaft (Table 2 of Vermeulen et al. 2013). No anthropogenic impact is anticipated because coal mining is unlikely to extend to the trace of these faults.

Sample site H2 is in fractures associated with a dyke, 1,340 m below the surface in the No. 8 shaft of the gold mine. These fractures may or may not be directly connected to the surface (Cook 2003), and therefore recharge may occur in the sub-surface at different depths through the influx of overlying aquifers or directly by precipitation from surface. No anthropogenic impact is anticipated because coal mining is unlikely to extend to the trace of these faults.

The source of water dripping (samples H5, H6, H7, and H10) from the mine roof is inferred to be seepage from overlying aquifers. Their locations are shown in Vermeulen et al. 2013, and could be affected by coal mining. The exception is H5, which is at the gold mine's No. 8 shaft and will remain unaffected by coal mining as no coal mining will occur around that shaft (Vermeulen et al. 2013). H6 and H7 were sampled near current coal mining activities and show a fingerprint that is characteristic of the Karoo aquifer affected by coal mining. Therefore, the only current interaction between the coal mine and the gold mine is at the No. 9 shaft.

Interaction between aquifers is apparent in samples H1, H3, H4, H6, and H7, probably along fracture-controlled conduits. The absence of an anthropogenic influence in the source region of the H1, H3, and H4 samples is noteworthy.

The exploration boreholes intersecting the gold mine workings at 1,620 m below the surface in the No. 8 shaft should be sealed off with non-reactive material to prevent the drainage of groundwater from the coal mine to the gold

mine, and thus responsibility and/or liability of polluting the deeper gold mining areas. It is recommended that all exploration holes be sealed to prevent leakage and contamination between aquifers. Once this is done, no anthropogenic impact is anticipated because coal mining is unlikely to extend to the trace of these faults.

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