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Assessment of Groundwater Recharge Sources And Their Dynamics Using Environmental Isotope And Hydrochemical Approaches In Coastal Aquifers of Peninsular Malaysia

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The aims of this study are to investigate the interconnection and the dynamics between groundwater and surface water in the coastal area in the eastern part of Peninsular Malaysia using isotope technique integrated with the hydrochemical and hydrogeological approaches. Using stable isotopic composition ($\delta^{2}H$ and $\delta^{18}O$), it was shown that there is no relationship between the mid–deep and deep groundwater indicating that they came from different aquifers. Based on the two–component mass balance for oxygen, the source of groundwater salinity was determined. The results showed that 12% of the groundwater is recharged by the seawater while 88% is from both rainwater and river water. This reveals that one of the deep groundwater (UMPB) was affected by seawater intrusion. However, the deep groundwater in the southern part of the study area was not influenced by saline water because of the geological conditions. As a result of the seawater intrusion, UMPB is showing the characteristics of shallow groundwater due to mixing with modern saline water which was also supported by the natural radioactive isotope (^{3}H and ^{14}C) and hydrochemical results.

Keywords: Groundwater; coastal aquifer; hydrogen and oxygen stable isotope; tritium; carbon–14

I. INTRODUCTION

Globally, groundwater resources are facing various threats both quantitatively and qualitatively. The massive increase in population has boosted water demand and stress on the groundwater resources. Moreover, due to the changes in climatic and land–use patterns, the quality and quantity of drinking water has declined throughout the world (Pant *et al.,* 2021a). According to Manap *et al.* (2014), groundwater has become the main source for drinking and household activities, especially during the drought period. In comparison, a large part of the rural population is still dependent on groundwater from dug wells and shallow tube wells to fulfil their daily needs (Razak & Karim, 2009).

Groundwater plays an important role in supplying domestic, industrial and agricultural water needs in Malaysia. Being located in the tropical region with high rainfall precipitation throughout the year, Peninsular Malaysia suffered from water crises brought about by the El–Nino phenomenon in 1991 and 2002 (Rahim *et al.*, 2010). Also, some states reported water shortages in 2014, caused by the dry period. The lack of knowledge of hydrogeological characteristics could hinder groundwater resources management. Studies on groundwater quality in Malaysia have not been in–depth in terms of its origin, relationship to the different types of aquifers, interrelationship with surface water, contamination possibilities as well as the intrusion of saline water into the aquifers (Karim *et al.*, 2014). Poor management of groundwater resources could lead to the decline in groundwater levels which causes uneconomic pumping lifts, land subsidence, and seawater intrusion in coastal aquifers (Singhal & Gupta, 1999).

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Groundwater aquifers especially that located near the coastline are vulnerable to seawater intrusion. Contamination of 2–3% of saltwater into coastal aquifer either under the influences of natural events or anthropogenic activities will increase the salinity of the water and result in water quality and soil degradation (Hairoma *et al.*, 2016). Seawater intrusion threatens socio–economic activities and sustainability including that of residents and agriculture in coastal areas. Baharuddin *et al.* (2013) and Al Mabrok *et al.* (2018) stated that seawater intrusion is the most prominent threat and a serious groundwater contamination issue which leads to the depletion of fresh groundwater resources besides affecting the crop in both west coast and east coast aquifers of Peninsular Malaysia.

To address this problem, this study aimed to understand various components of the water cycle and groundwater evolution. This includes the investigation of source, origin of groundwater and surface water, and finally explains the ground and surface interactions and its dynamics using isotopic and hydrochemical approaches. Stable isotope tracers of water, i.e. deuterium (^{2}H) and oxygen–18 (^{18}O) can achieve the purpose due to their unique 'fingerprint' of sources that are often preserved within the subsurface (Pant *et al.,* 2021a). Precipitation process has a geographically specific isotopic fingerprint, which is inherited by the local groundwater (Kortelainen, 2011). This fact leads to the use of the environmental isotopes technique in this study. By collecting information and establishing a database on stable isotopes of oxygen and hydrogen of meteoric water, and analysing the groundwater and surface water together with the hydrogeological information, an evaluation can be made to define the recharge, sources of groundwater salinity and the mass balance in the study area (Yeh *et al.*, 2009). In the meantime, the natural radioactive isotopes of tritium (^{3}H) and carbon–14 (^{14}C) provide a timescale for the subsurface flow. Integration of isotope techniques with conventional techniques, i.e. the hydrochemistry technique gives excellent results particularly in understanding the interaction of groundwater and surface water. This study will be useful to comprehend the groundwater dynamics in coastal aquifers for better groundwater utilisation in the future.

A. Site Description

The study area is situated in Pekan District, Pahang (Figure 1). Pahang occupied the huge Pahang River Basin (latitude N 2° 48' 45"– N 3° 40' 24" and longitude E 101° 16' 31"– E 103° 29' 34"). This area is bordered by the South China Sea to the east, the Rompin River Basin in the south and the highlands (Titiwangsa Range) from the high north to the west. The sampling sites are in the surroundings of Pekan and Nenasi. The study area was chosen due to its laterally and vertically extensive alluvial aquifers and considered to have the highest potential for groundwater. This is in contrast with the West Coast aquifers which contain high proportion of clay and the groundwater is found to be saline (Chong & Tan, 1986). Therefore any impact on the groundwater system for example, the rising sea levels from climate change can be studied.

The climate of Peninsular Malaysia is tropical and the humidity is high (approximately 85%) all year round, with the temperatures ranging from 21°C to 32°C. Average monthly annual rainfall in the study area each year is between 2500 and 3500 mm. Evaporation is approximately 1800 mm per year (Baharuddin, 1992). In general, Malaysia can be distinguished into two monsoon systems, namely, the northeast monsoon from November to March (wet season) and southwest monsoon from May to September which is a relatively dry season. There are two shorter periods of inter–monsoon between these two monsoons. Over the east coast, the northeast monsoon bringing the bulk of the precipitation from November to January (with the highest being in December), while June and July are the driest months in the most area (MMD, 2017).

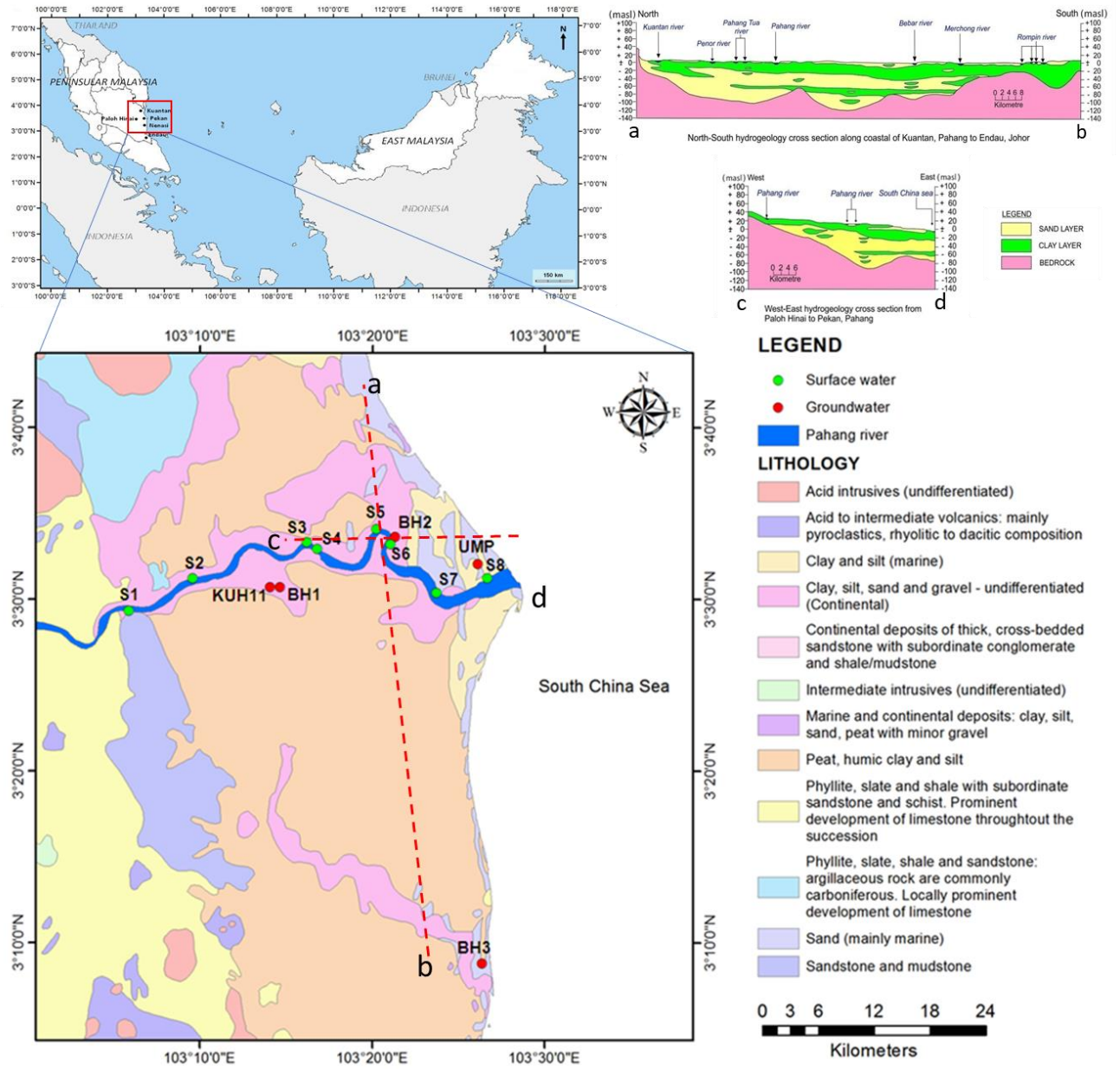

Figure 1. Location of the study area representing geological map of the study area and sampling locations. The hydrogeological cross–section of the study area represent the sub–surface aquifer conditions around section "a–b" and "c–d" (Baharuddin, 1992).

The geological map of the study area and the sampling locations are shown in Figure 1. The study area is covered by the Quarternary alluvium. The area covered by this alluvial deposit is approximately 2,000 km². These alluvium sediments form lowland areas, where sand ridges, floodplains, and swamps can be found in the area. The alluvium consists of a layer of sand layers, gravel, silt, and clay. Generally, Pre–Quarternary rock outcrops such as sedimentary rock or metasediments can be found around the study area except in the east. The outcrops formed a medium height topography. Volcanic and granite rock bodies also can be found around the study area, especially in the north and south, respectively (Department of Minerals and Geoscience, 2012).

Generally, the geomorphology of this study area is characterised by a flat topography and has a topographic height of lower than 50 meters above sea level. More than half of the area consists of swampy forests. Most of the forests have not yet been explored, especially in inland. The main river flowing in this area is the Pahang River. In addition, there are several other large rivers flowing across the region from west to east such as Kuantan River, Penur

River to the north and Bebar River, Merchong River and Rompin River to the south (Figure 1). The hydrogeological cross–sections from north to south and west to east are also shown in this figure.

II. MATERIALS AND METHOD

A total of 61 water samples were collected from three sampling campaigns in January, June and November 2014. However, samplings for ^{3}H and ^{14}C were performed only in January and November 2014. Samples for ^{3}H analysis were taken at all the sampling points while for ^{14}C the water was sampled from only the deep groundwater as it generally contains only old water which can only be measured via ^{14}C. The water samples were taken from the Pahang river (S1 – S8), shallow groundwater (BH1 – BH3, UMPA) and deep groundwater which are BH3A, BH3B, KUH11A, KUH11B and UMPB. The surface waters were collected using a grab sampler from the river. The monitoring wells were purged with three well–casing volumes to remove stagnant water in the well so that the water being sampled would be stabilised; representative groundwater was thus obtained prior to sampling (Zhang, 2007). Boreholes with the same name (for example, BH3, BH3A, and BH3B) are located at the same location where the last letter in the boreholes name (A or B) represent its depth (shallow or deep, respectively). In this study, the wells are classified as deep groundwater when the depth is more than 40 meters (>120 feet). However, KUH11A and KUH11B are further sub–categorised as the mid–deep groundwater since the depth is only approximately 44 – 53 m.

Rainwater samples for 2013 and 2014 were collected on a monthly basis from the nearest International Atomic Energy Agency (IAEA) Global Network of Isotopes in Precipitation (GNIP) rainfall station which is in Kuantan, Pahang. The rainwater from the station is collected using a rain gauge which is permanently fixed by Malaysian Nuclear Agency in the GNIP rainfall station. The setup of the rain water collection apparatus is using Tube–dip–in–water collector with pressure equilibration and known diameter funnel. A stainless steel table tennis ball and debris screen is placed in the collection funnel to help in sealing the collector bottle against evaporation and debris, respectively. The rain gauge will be placed at the sampling area on the first day of every month and the monthly composite samples of precipitation will be emptied at the end of the month.

The sampling for hydrochemistry included the in–situ measurement of pH, salinity, temperature, Total Dissolved Solids (TDS), Dissolved Oxygen (DO) and Electrical Conductivity (EC) using portable YSI 556 Multi Probe System. The Multi Probe System was calibrated with an appropriate buffer solution (pH 7) before every sampling program started. For each sampling point, two water samples were collected in 1 litre HDPE bottle. Meanwhile, the ^{2}H and ^{18}O sampling, a 100 ml HDPE bottles were fully filled with a water sample to ensure that there were no air bubbles and the bottles were then tightly capped. All bottles were rinsed with distilled water before sampling and these bottles were rinsed again two to three times using sampled water in the field. Water samples were filtered immediately on–site through 0.45μm syringe filters. Reagent grade HNO_3 was used for the preservation process during this sampling for cation elements. Laboratory analyses were performed at Department of Mineral and Geoscience, Malaysia in Perak.

Stable isotope analyses were performed using SERCON GEO 20–20 Continuous Flow Isotope Ratio Mass Spectrometer (CF–IRMS) at Malaysian Nuclear Agency. The continuous flow mass spectrometry offers online sample preparation, smaller sample size, faster and simpler analysis and cost–effective compared to Dual Inlet Isotope Ratio Mass Spectrometer (DI–IRMS) according to Benson *et al.* (2006).

Samples for $\delta^{2}H$ and $\delta^{18}O$ analyses were treated in the SERCON Water Equilibration System (WES) prior to analysis by the IRMS. A total of 0.5ml of water samples were used in the vials during analysis. The $\delta^{18}O$ value was measured via equilibration of the water samples with CO_2 gas at 50°C for 8 hours, and $\delta^{2}H$ value was measured via equilibration of the water samples with H_2 gas and its reaction with the platinum stick catalyst at 50°C for 1 hour (SERCON, 2007). In the $\delta^{2}H$ analysis, a platinum catalyst stick was used to accelerate the reaction, and gas exchange equilibrium took place between the introduced pure H_2 gas and water vapour. This gas exchange equilibrium caused the water vapour to emit a signature to the introduced pure

H_2gas, which represents the isotopic composition of the water before the H_2 gas is analysed by the IRMS.

Stable isotope compositions are generally reported as δ values and denoted as ‰ or permil/per mill which is relative to a standard of known composition (e.g., Vienna Standard Mean Ocean Water, VSMOW for ^2H and ^{18}O analyses). The δ values are calculated by using Equation (1), in this case, for deuterium or ^2H:

$$\delta^2 H(‰) = \frac{R_{Sample} - R_{Standard}}{R_{Standard}} x\ 1000 \quad (1)$$

where R represents the ratio of heavy to light isotope ($^2H/^1H$), and R_{Sample} and $R_{Standard}$ are the isotope ratios in the sample and the standard, respectively. The sample is described as depleted (more negative) if the δ values are lower and enriched (more positive) if the δ values are higher with respect to a reference, i.e., the VSMOW (IAEA, 1983; Clark & Fritz, 1997; Kendall, 1998).

For ^3H analysis, 1 litre of a water sample is needed. ^3H values were measured using Liquid Scintillation Counter (LSC) after being distilled and electrolysed.^{14}C was measured also by LSC on CO_2 absorption method which is synthesised from $BaCO_3$ precipitated in the field from 150 litres of water samples.

A. Groundwater Isotopic Compositions Mass–Balance

In water budget studies, it is crucial to evaluate mass–balance of the groundwater and the components that actually recharge it. Therefore, the mixing between two distinct recharge sources can be quantified by the equation below which is adopted from Clark and Fritz (1997) and Kendall (1998). Subscripts represent the samples while V and δ are the volume and isotopic composition of the corresponding components.

$V_{UMPB} = V_{S8} + s_{eawater}$

$V_{UMPB}\delta_{UMPB} = V_{S8}\delta_{S8} + V_{Seawater}\delta_{Seawater}$

$V_{Seawater}\delta_{Seawater} = V_{UMPB}\delta_{UMPB} - V_{S8}\delta_{S8}$

Substitute $V_{S8} = V_{UMPB} - V_{Seawater}$

$V_{Seawater}\delta_{Seawater} = V_{UMPB}\delta_{UMPB} - (V_{UMPB} - V_{Seawater})\delta_{S8}$

$V_{Seawater}\delta_{Seawater} = V_{UMPB}\delta_{UMPB} - V_{UMPB}\delta_{S8} + V_{Seawater}\delta_{S8}$

$V_{Seawater}\delta_{Seawater} - V_{Seawater}\delta_{S8} = V_{UMPB}\delta_{UMPB} - V_{UMPB}\delta_{S8}$

$V_{Seawater}(\delta_{Seawater} - \delta_{S8}) = V_{UMPB}\ (\delta_{UMPB} - \delta_{S8})$

$V_{Seawater}/V_{UMPB} = (\delta_{UMPB} - \delta_{S8})/(\delta_{Seawater} - \delta_{S8})$ x 100 (2)

Where $V_{Seawater}/V_{UMPB}$or *X*(in %)is the recharge proportion of seawater in the groundwater, i.e. UMPB (or the fraction of seawater intrusion), δ_{UMPB} is the $\delta^{18}O$ value for UMPB, δ_{S8} is the $\delta^{18}O$ value for S8 and $\delta_{Seawater}$ is the $\delta^{18}O$ value for seawater. Nevertheless, the $\delta^{18}O$ composition (‰) may be substituted for δ^2H in Equation (2).

III. RESULTS AND DISCUSSION

A. Hydrochemical Results

Results for field data and hydrochemical analyses for all the water samples are presented in Table 1 and Table 2, respectively. The water samples (surface water, shallow and deep groundwater) from the study area are fairly acidic to slightly above neutral with pH value ranging from 5.98–8.31 with an average of 6.91. The TDS value for surface water and groundwater range from 26–17778 mg/l and 68–5548 mg/l, respectively. EC for all the water samples varies from 52 – 35556 µS/cm. The surface and groundwater samples (S8 and UMPB) comprises of highest TDS (17778 and 5548 mg/l) and EC (35556 µS/cm and 11099 µS/cm)of, respectively. The TDS and EC results were in accordance with each other and as in general, the TDS and EC relationship is given by equation TDS = (0.55 to 0.7) x EC (Thirumalini, 2009; Tchobanoglous *et al.*, 2003). Based on the sampling for surface water (S1 – S8) during low tide and high tide (AM/morning and PM/afternoon, respectively) suggested that the seawater reaches up until S7 during high tide where its EC value is slightly high (approximately three folds) compared to upstream or background (S1 – S6). This finding was also supported by the salinity result. As for the DO, the value in surface water is higher compared to the groundwater due to the interface with the atmosphere at the surface layer. However, the DO in groundwater is still detectable as the vadose zone is porous and oxygen distribution decreases with depth (Rose, 1988).

The results show that surface water illustrates a high concentration of Cl^- between 2 and 11718 mg/l with a mean value of 600.9 mg/l. As for the cations, Na^+ is dominant element with a value between 2.6 and 462 mg/l and mean value of 55.7 mg/l. Sample S8 reported high concentrations of Na^+ and Cl^-.

Similarly, the groundwater shows a high concentration of Cl^- ranging from 1 – 3125 mg/l with an average of 515 mg/l for anion and the Na^+ value ranges from 4.2 – 1647 mg/l with an average of 335.3 mg/l. Sample UMPB demonstrates high values of Na^+ and Cl^- with the values of 1647 and 3125 mg/l, respectively. The high concentration of the cation and anion concentration in the UMPB and S8 sample indicates the presence of the saltwater intrusion in the aquifer system. The order of dominance of major anion and cation in the water samples are given in Table 3.

1. Hydrochemical facies

To determine the main hydrochemical facies in the surface and groundwater, the major cations and anions were plotted in the conventional Piper diagram. Detailed classification of water types using Piper diagram illustrates that the water samples can be divided into 3 main groups (Figure 2). Group 1 is a mixed Ca^{2+}–Mg^{2+}–Cl^- water type consists of UMPA, BH3A and BH3B. Group 2 which consists of S8 and UMPB are of Na^+–Cl^- water type. The rest of the water samples formed Group 3 which is Ca^{2+}–HCO_3^- water type.

Table 1. Summary of field data results for water samples from the study area in January, June and November 2014

Sample	Depth (m)	pH	Sal (ppt)	T (°C)	TDS (mg/L)	DO (mg/L)	EC (µS/cm)
S1	–	7.03–7.41 (7.17)	0.02–0.05 (0.04)	26.26–31.99 (28.33)	27.00–60.00 (48.23)	1.29–5.14 (3.85)	54.0–120.0 (96.3)
S2	–	7.02–7.44 (7.19)	0.02–0.05 (0.04)	26.54–32.50 (28.57)	27.00–59.00 (47.67)	1.61–4.97 (3.79)	55.0–118.0 (95.7)
S3	–	6.05–6.96 (6.53)	0.02–0.05 (0.04)	26.20–32.96 (28.50)	26.00–58.00 (43.85)	1.38–5.14 (3.83)	52.0–117.0 (88.0)
S4	–	6.92–7.32 (7.09)	0.02–0.05 (0.04)	26.33–31.74 (28.17)	26.00–59.00 (47.00)	1.64–6.63 (4.57)	53.0–119.0 (94.9)
S5	–	6.58–6.95 (6.79)	0.03–0.05 (0.04)	26.28–30.96 (27.86)	28.00–60.00 (48.00)	1.64–5.88 (4.04)	57.0–119.0 (96.0)
S6 (AM)	–	6.98–7.43 (7.21)	0.05–0.05 (0.05)	26.12–31.36 (28.74)	56.40–59.00 (57.70)	4.75–5.01 (4.88)	113.0–118.0 (115.5)
S6 (PM)	–	6.92–8.30 (7.44)	0.03–0.06 (0.05)	26.67–33.06 (29.12)	28.00–59.95 (48.98)	3.04–5.70 (4.49)	57.0–119.7 (98.6)
S7 (AM)	–	7.01–7.47 (7.22)	0.02–0.34 (0.14)	25.98–31.49 (27.85)	28.00–356.00 (146.67)	2.27–4.59 (3.78)	55.0–712.0 (293.3)
S7 (PM)	–	6.71–7.42	0.06–0.08	27.20–32.03	61.00–92.00 (76.50)	4.31–5.08	122.0–184.0 (153.0)

		(7.07)	(0.07)	(29.62)		(4.70)	
S8 (AM)	–	6.89–8.31 (7.47)	0.06–22.26 (7.87)	26.06–30.68 (27.85)	68.00–17778.50 (6369.50)	2.14–5.56 (3.92)	136.0–35556.5 (12738.8)
S8 (PM)	–	7.31–7.97 (7.64)	0.43–2.96 (1.70)	28.02–32.44 (30.23)	437.00–2777.70 (1607.35)	4.56–4.78 (4.67)	873.0–5555.4 (3214.2)
KUH11A[b]	53.4	6.30–6.45 (6.38)	0.06–0.09 (0.08)	26.44–27.05 (26.70)	68.00–93.00 (84.67)	0.01–2.04 (0.97)	137.0–186.0 (169.7)
KUH11B[b]	44.1	6.26–6.48 (6.38)	0.06–0.09 (0.08)	26.46–26.97 (26.79)	68.00–93.00 (84.67)	0.05–1.14 (0.66)	136.0–186.0 (169.3)
UMPA[a] (AM)	17.5	6.10–6.88 (6.36)	0.16–0.24 (0.21)	27.05–31.13 (29.19)	173.00–253.00 (220.92)	0.12–4.51 (1.71)	346.0–507.0 (428.0)
UMPA[a] (PM)	17.5	5.98–6.22 (6.10)	0.23–0.23 (0.23)	30.58–30.75 (30.66)	238.85–240.75 (239.80)	0.85–1.90 (1.37)	477.0–480.9 (479.0)
UMPB[b] (AM)	80.0	6.03–7.26 (6.74)	0.16–6.25 (4.03)	29.53–30.18 (29.92)	169.00–5548.20 (3601.80)	0–1.37 (0.55)	339.0–11098.5 (7205.8)
UMPB[b] (PM)	80.0	6.22–7.28 (6.75)	0.29–5.46 (2.87)	30.09–30.43 (30.26)	299.00–4893.75 (2596.38)	0.29–1.76 (1.02)	598.0–9787.6 (5192.8)
BH1[a]	14.2	6.29–6.62 (6.44)	0.09–0.11 (0.10)	27.45–28.17 (27.88)	94.00–117.00 (108.67)	0–1.16 (0.66)	187.6–234.0 (217.5)
BH2[a]	15.3	6.74–7.37 (7.06)	0.26–0.32 (0.29)	28.55–29.22 (28.89)	272.00–330.00 (301.00)	0–0.64 (0.32)	186.0–660.0 (463.1)
BH3[a]	11.1	7.05–7.31 (7.16)	0.71–0.75 (0.73)	29.54–30.78 (30.11)	717.10–756.20 (737.75)	0.30–1.40 (0.93)	1435.0–1656.0 (1523.5)
BH3A[b]	83.2	6.86–7.15 (7.01)	0.45–0.50 (0.47)	30.20–30.50 (30.33)	459.00–508.95 (477.43)	1.17–3.98 (2.72)	918.0–1017.0 (954.7)
BH3B[b]	104.2	6.84–7.10 (6.96)	0.77–0.92 (0.86)	29.68–30.64 (30.05)	770.55–913.36 (860.64)	0.84–2.73 (1.84)	1540.9–1826.7 (1721.0)

Range of concentrations in time and averages (in parenthesis); [a]Shallow groundwater; [b]Deep groundwater

Table 2. Summary of hydrochemical concentrations for water samples from the study area in January, June and November 2014

Sample	**Depth (m)**	**Na^+**	**K^+**	**Ca^{2+}**	**Mg^{2+}**	**HCO_3^-**	**SO_4^{2-}**	**Cl^-**	**NO_3^-**
S1	–	3.2	4.2	5.7–6.8 (6.3)	1.6	24–30 (27)	nd	2–4 (3)	1.0–1.6 (1.3)
S2	–	2.8	4.2	5.9–6.4 (6.1)	1.5	22–29 (25.5)	nd	4–4 (4)	0.9–1.4 (1.2)
S3	–	2.6	4.0	4.7–5.1 (4.9)	1.2	17–30 (23.5)	nd	3–3 (3)	0.9–1.2 (1.1)
S4	–	2.8	4.5	5.6–6.3 (5.9)	1.5	24–29 (26.5)	nd	3–3 (3)	1.0–1.4 (1.2)
S5	–	2.7	4.2	5.4–6.0 (5.7)	1.4	23–31 (27)	nd	2–2 (2)	1.2–1.5 (1.4)
S6 (AM)	–	2.9	4.4	5.5–6.6 (6.1)	1.6	23–29 (26)	38–38 (38)	2–4 (3)	1.0–1.4 (1.2)

S6 (PM)	–	3.5	4.5	5.8–6.7 (6.3)	1.7	24–29 (26.5)	nd	4–4 (4)	1.0–1.3 (1.2)
S7 (AM)	–	2.9	4.3	6.5–8.8 (7.7)	1.5	25–29 (27)	24–24 (24)	3–153 (78)	1.4–1.4 (1.4)
S7 (PM)	–	3.7	4.3	6.1–6.8 (6.4)	1.7	24–29 (26.5)	nd	4–25 (14.5)	1.1–1.5 (1.3)
S8 (AM)	–	462.0	21.0	24.0–157.7 (90.9)	55.0	27–78 (52.5)	86–997 (541.5)	740–11718 (6229)	1.6–1.7 (1.7)
S8 (PM)	–	124.0	8.5	11.0–34.4 (22.7)	16.0	24–41 (32.5)	38–166 (102)	194–338 (266)	1.2–1.5 (1.4)
KUH11A[b]	53.4	4.2	1.2	1.4–1.7 (1.6)	1.3	19–22 (20.5)	nd	3–3 (3)	nd
KUH11B[b]	44.1	4.4	1.3	1.6–2.0 (1.8)	1.4	22–29 (25.5)	nd	2–3 (2.5)	nd
UMPA[a] (AM)	17.5	22.0	4.3	37.1–39.0 (38.0)	8.0	36–40 (38)	71–89 (80)	20–34 (27)	1.9–2.5 (2.2)
UMPA[a] (PM)	17.5	48.0	4.8	37.0–46.0 (41.5)	9.9	31–36 (33.5)	75–88 (81.5)	26–102 (64)	2.1–2.2 (2.2)
UMPB[b] (AM)	80.0	1647.0	63.0	68.6–69.0 (68.8)	133.0	372–422 (397)	159–167 (163)	3050–3125 (3087.5)	4.2–4.8 (4.5)
UMPB[b] (PM)	80.0	1541.0	59.0	66.0–69.0 (67.5)	121.0	66–487 (276.5)	76–122 (99)	230–2655 (1442.5)	3.8–6.1 (5.0)
BH1[a]	14.2	6.9	3.5	2.1–3.0 (2.6)	2.4	55–57 (56)	nd	2–2 (2)	nd
BH2[a]	15.3	67.0	7.7	25.4–30.0 (27.7)	23.0	161–167 (164)	9–10 (9.5)	82–88 (85)	0.8–0.8 (0.8)
BH3[a]	11.1	168.0	16.0	53.2–59.0 (56.1)	42.0	206–218 (212)	nd	156–300 (228)	2.4–5.5 (4.0)
BH3A[b]	83.2	573.0	12.0	54.2–64.0 (59.1)	39.0	161–170 (165.5)	nd	160–187 (173.5)	1–5.3.0 (3.2)
BH3B[b]	104.2	207.0	17.0	96.4–145.0 (120.7)	78.0	154–170 (162)	nd	451–651 (551)	1.9–2.2 (2.1)

Range of concentrations in time and averages (in parenthesis); n=3 except for Na^+, K^+ and Mg^{2+} (n=1); All major ions are expressed in mg/L; [a]Shallow groundwater; [b]Deep groundwater; nd = Not detected (Below detection limit).

Table 3. Order of dominance of major anion and cation in water samples (values are in percentage)

Type of water	**Anion**	**Cation**
Surface water	Cl^-> HCO_3^-> SO_4^{2-}> NO_3^-> F^-	Na^+> Ca^{2+}> Mg^{2+}> K^+
	69.21 > 18.43 > 10.83 > 1.13 > 0.39	71.48 > 10.70 > 9.88 > 7.94
Groundwater (Shallow)	HCO_3^->Cl^-> SO_4^{2-}> NO_3^-> F^-	Na^+> Ca^{2+}> Mg^{2+}> K^+
	42.33 > 32.36 > 24.23 > 0.76 > 0.30	44.20 > 36.25 > 13.30 > 6.23
Groundwater (Deep)	Cl^-> HCO_3^-> SO_4^{2-}> NO_3^-> F^-	Na^+> Mg^{2+}> Ca^{2+}>K^+
	81.86 > 14.14 > 3.79 > 0.17 > 0.26	81.82 > 8.02 > 6.78 > 3.37

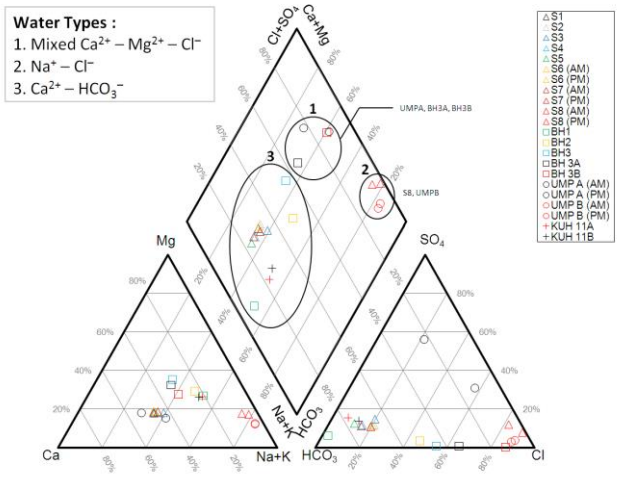

Figure 2. Piper diagram for the water samples

2. Water–rock interaction

Water–rock interaction influenced the chemistry of water and hence it is very important to identify the geochemical processes occurred in the area (Pant *et al.,* 2021a). The Piper plot is very useful in defining the relations between different dissolved components and water type classification. Gibbs (1970; 1971) differentiated water quality based on evaporation or seawater interaction, water–rock interaction, and precipitation, and is widely used to measure functional sources of dissolved chemical constituents. Gibbs plot (Figure 3) shows that there are three processes that are affecting the water chemistry for the water samples. While most of the water samples indicate water–rock interaction, S8 and UMPB exhibit high possibility of interaction with seawater compared to BH3, BH3A and BH3B which seem to be moderately influenced by it. BH1, KUH11A and KUH11B illustrate a slight signal of rainfall dominated water.

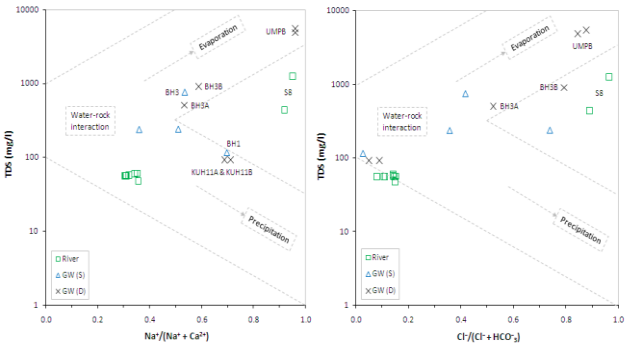

Figure 3. Gibbs plot for the water samples

B. Isotope Results

1. Precipitation

Precipitation is the source of groundwater hence it provides an important input signal of ^{2}H and ^{18}O to trace the groundwater recharge. The local meteoric water line will present a baseline for characterising stable isotope distribution in the water bodies (Ayub, 2005; Kendall, 1998). Therefore, knowledge of the isotopic composition of the water samples in this study is very important. The $\delta^{2}H$ and $\delta^{18}O$ values for rainfall range from –70.79 to –13.85‰ and –10.25 to –2.41‰, respectively with the amount–weighted mean annual of –44.63 and –6.93‰, respectively (Table 4). Amount–weighted means for dry season (July to April) and wet season (December) were also distinguished. Deuterium excess (d–excess) values range from 0.9 to 18.8‰ with an average of 10.3‰. The d–excess is defined as $d = \delta^{2}H – 8\delta^{18}O$ and the value of the d–excess indicates the origin of the air moisture which is related to variations in humidity (Dansgaard, 1964).

Table 4. Isotopic composition and weighted mean value of precipitation

		$\delta^{18}O$ (‰)	$\delta^{2}H$ (‰)	d–excess	Rainfall (mm)
2013	Jan	na	na	na	232.8
	Feb	na	na	na	449.0
	Mar	na	na	na	20.4
	Apr	–7.67	–50.87	10.5	135.8
	May	–6.08	–38.93	9.7	167.8
	Jun	–4.47	–31.40	4.4	129.2
	July	–7.14	–45.87	11.3	271.0
	Aug	–5.58	–38.31	6.3	164.8
	Sept	–3.61	–28.00	0.9	413.0
	Oct	–6.08	–37.20	11.4	238.2
	Nov	–3.06	–15.54	8.9	195.0
	Dec	–10.25	–70.79	11.2	1075.2
2014	Jan	na	na	na	277.2
	Feb	na	na	na	22.6
	Mar	na	na	na	31.2
	Apr	–3.31	–14.28	12.2	77.4
	May	–6.83	–35.88	18.8	288.2
	Jun	–6.63	–40.34	12.7	178.2
	July	–4.22	–25.81	8.0	182.0
	Aug	–5.29	–30.10	12.2	206.8

Sept	−2.41	−13.85	5.5	70.4
Oct	−8.90	−57.58	13.6	347.2
Nov	−4.56	−19.88	16.6	288.8
Dec	−7.36	−47.71	11.1	1806.0
Weighted means value				
Annual	−6.93	−44.63		
Wet season	−8.44	−56.32		
Dry season	−6.12	−37.26		

na = Not available

2. Surface water and groundwater

Results for isotopic analyses for all the water samples are presented in Table 5 including the age of the groundwater. The $\delta^{2}H$ and $\delta^{18}O$ values for surface water range from −52.64 to −13.52‰ and −7.78 to −2.56‰, respectively with S8 is the most enriched (−13.52 and −2.56‰, respectively). The groundwater values vary in the range of −47.75 to −35.14‰ for $\delta^{2}H$ and −7.50 to −5.07‰ for $\delta^{18}O$. The d−excess ranges from +2.86 to +20.39‰ with an average of +11.17‰.

The ^{3}H value is found to be in the range of 0.95 – 2.13 TU which suggests that these water samples are a mixture of between submodern water which prior to 1952 and recent recharge is less than 10 years (Clark & Fritz, 1997; Mook 2000). The tritium concentration of <1TU, suggesting that the water is older than 50 years (Pant *et al.,* 2021b). Moreover, the ^{14}C values for the collected water samples ranges between 22.42 and 83.42 pmc with average age ranges from 1664–11047 years. BH3A and BH3B are the oldest deep groundwater with values of 11047 and 9975 years, respectively. Whereas the shallow groundwater ranges from 1664 – 6571 years. This result is in accordance with the ^{3}H result as it indicates the combination of young and old water. The inverse relationship between ^{3}H and ^{14}C values is shown in Figure 4 which illustrates the ^{3}H values decrease as the water gets deeper and older. The age of the groundwater was calculated based on Vogel model with initial ^{14}C content, A_o= 85 ± 5% pmc as the study area was mainly on alluvium (Clark & Fritz, 1997; Geyh, 2000; Mook, 2000).

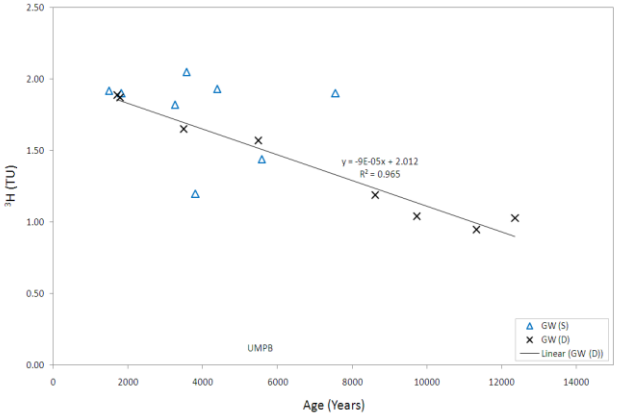

Figure 4. ^{3}H versus Age

3. Surface water and groundwater interconnections

The interaction of surface water and groundwater in coastal aquifer is shown in Figure 5 and defined by the $\delta^{18}O$ and $\delta^{2}H$ relationship. The Global Meteoric Water Line (GMWL) is the one according to Craig (1961) and defined as $\delta^{2}H = 8\delta^{18}O + 10$. Local Meteoric Water Line (LMWL) was established and is given by the equation $\delta^{2}H = 7.10\delta^{18}O + 5.09$. The isotopic composition shows the enrichment in the surface water and groundwater compared to the precipitation isotopic signatures (weighted annual mean). This is anticipated as the study area is located near to the sea and subjected to coastal effect. In general, all the surface water and groundwater fall quite uniformly on the GMWL and LMWL. However, the deep groundwater was separately clustered into two groups. Group 1 comprises of KUH11A and KUH11B of the mid–deep level (44.1 m and 53.4 m) was more enriched compared to Group 2 which is from the deeper groundwater (80 – 104.2 m). This indicates that both groundwaters came from different aquifers. UMPB demonstrated slightly enriched values of $\delta^{2}H$ and $\delta^{18}O$ and thus indicate the possibility of mixing with seawater.

Table 5. Summary of isotope results for water samples from the study area in January, June and November 2014

Sample	Depth (m)	$\delta^{18}O$ (‰)	$\delta^{2}H$ (‰)	d–excess	^{3}H (TU)	^{14}C (pmc)	Age (years)
S1	–	−7.55–−7.08 (−7.35)	−47.7–−36.3 (−43.6)	11.5–20.4 (15.2)	1.94–2.03 (1.99)	–	–
S2	–	−7.38–−7.01 (−7.23)	−49.3–−42.4 (−45.3)	9.8–16.1 (12.6)	1.91–2.00 (1.96)	–	–
S3	–	−7.68–−7.18 (−7.39)	−51.3–−40.8 (−45.0)	10.2–17.7 (14.1)	1.91–1.97 (1.94)	–	–
S4	–	−7.34–−6.88 (−7.08)	−47.8–−37.1 (−42.3)	10.9–18.0 (14.4)	2.02–2.15 (2.09)	–	–
S5	–	−7.56–−6.48 (−7.05)	−49.6–−38.6 (−44.4)	6.7–18.3 (12.0)	1.99–2.05 (2.02)	–	–
S6 (AM)	–	−6.88–−6.66 (−6.77)	−44.1–−39.0 (−41.5)	11.0–14.3 (12.6)	1.92–1.92 (1.92)	–	–
S6 (PM)	–	−7.78–−6.65 (−7.04)	−48.7–−39.0 (−43.3)	11.4–14.2 (13.0)	1.94–1.94 (1.94)	–	–
S7 (AM)	–	−7.30–−6.08 (−6.58)	−49.5–−41.7 (−44.8)	7.0–8.9 (7.8)	1.87–1.96 (1.92)	–	–
S7 (PM)	–	−6.90–−6.39 (−6.65)	−48.3–−36.9 (−42.6)	2.9–18.3 (10.6)	na	–	–
S8 (AM)	–	−7.51–−2.56 (−5.40)	−52.6–−13.5 (−36.9)	4.7–7.4 (6.4)	1.95–2.05 (2.00)	–	–
S8 (PM)	–	−6.24–−5.68 (−5.96)	−42.6–−35.6 (−39.1)	7.3–9.8 (8.6)	na	–	–
KUH11A[b]	53.4	−6.31–−5.76 (−6.09)	−38.9–−37.4 (−38.1)	8.2–13.1 (10.7)	1.87–1.89 (1.88)	80.48–81.34 (80.91)	1751 ± 100
KUH11B[b]	44.1	−6.89–−5.79 (−6.45)	−38.1–−35.1 (−36.5)	11.2–17.0 (15.0)	1.99–2.13 (2.06)	na	na
UMPA[a] (AM)	17.5	−6.51–−6.04 (−6.33)	−45.3–−41.0 (−43.3)	4.7–11.1 (7.3)	1.90–1.92 (1.91)	80.17–83.42 (81.80)	1664 ± 93
UMPA[a] (PM)	17.5	−6.91–−6.53 (−6.72)	−46.3–−41.6 (−44.0)	9.0–10.6 (9.8)	na	na	na
UMPB[b] (AM)	80.0	−6.06–−5.07 (−5.49)	−38.7–−36.1 (−37.5)	4.5–9.7 (6.4)	1.57–1.65 (1.61)	51.47–65.59 (58.53)	4489 ± 250
UMPB[b] (PM)	80.0	−6.46–−5.34 (−5.90)	−37.7–−37.4 (−37.6)	5.0–14.3 (9.6)	na	na	na

BH1[a]	14.2	−6.81−−6.50 (−6.62)	−40.4−−36.1 (−38.9)	11.9–16.3 (14.1)	1.20–1.93 (1.57)	58.75–63.07 (60.91)	4104 ± 233
BH2[a]	15.3	−6.46−−5.77 (−6.21)	−41.1−−35.5 (−37.8)	10.6–14.4 (11.9)	1.82–2.05 (1.94)	64.88–67.40 (66.14)	3419 ± 195
BH3[a]	11.1	−7.50−−6.42 (−6.80)	−46.5−−41.3 (−43.9)	5.4–16.2 (10.5)	1.44–1.90 (1.67)	40.07–50.90 (45.49)	6571 ± 355
BH3A[b]	83.2	−7.38−−6.86 (−7.18)	−47.8−−44.5 (−46.1)	10.4–12.9 (11.4)	1.03–1.04 (1.04)	22.42–30.81 (26.62)	11047 ± 598
BH3B[b]	104.2	−7.17−−6.92 (−7.08)	−46.7−−45.7 (−46.2)	9.1–11.7 (10.4)	0.95–1.19 (1.07)	25.41–35.24 (30.33)	9975 ± 558

Range of isotope results in time and averages (in parenthesis); n=3 except for ^{3}H and ^{14}C (n=2); [a]Shallow groundwater; [b]Deep groundwater; na = Not analysed.

As for the surface water samples, S8 has the most enriched values of $\delta^{2}H$ and $\delta^{18}O$ due to its location in the estuaries which was directly affected by the tidal effect during the high tide.

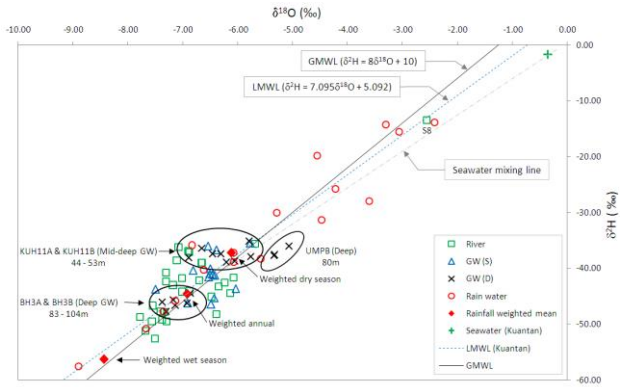

Figure 5. Relationship between the $\delta^{18}O$ and $\delta^{2}H$ of all the water samples in the study area

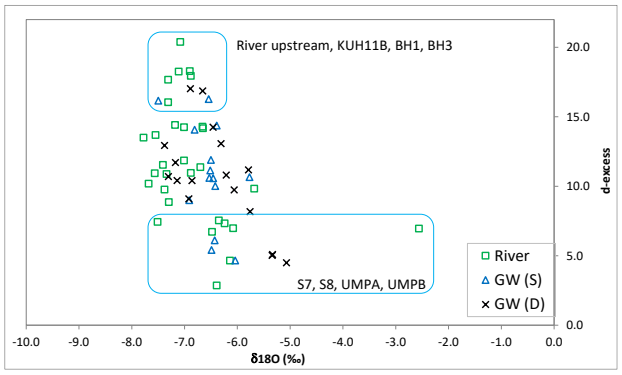

Figure 6. Cross plot between $\delta^{18}O$ and d−excess

The surface water and groundwater samples are close to the meteoric water lines (LMWL), indicating the meteoric origin of the groundwater (Pant *et al.,* 2021b). This means that the source of the groundwater in the study area is mainly from the rain water. However, the low d−excess value with respect to $\delta^{18}O$ shows that groundwater samples near to the sea coast (UMP) are evaporative in nature (Figure 6).The findings suggest that the aquifer is occupied by recent water demonstrating that the source of water recharge for groundwater is mainly originated from river. This could be due to the high rainfall distribution throughout the year at this area.

Basically, the coastal aquifers or groundwater, is recharged from the rainfalls and river water drained from mountain watersheds. Another possible source of groundwater recharge can be the seawater intrusion.Vice versa, the river basin discharge which is mainly a mixture of river flow and precipitation can also be recharged by the groundwater along the way before it reaches the sea. However, since the recharge zones for precipitation originated from the mountain watershed in the far western part of the study area, the hydrologic budget can be simplified to include only one term for source of freshwater recharge which is the river water (S8) at the endpoint, i.e., the estuaries after going through a mixing process along the river. The seawater influence over the groundwater can be clearly highlighted in the cross plot between $\delta^{18}O$ and Cl/HCO_3 ratio, where the UMPA and BH3B are on the border of seawater influence zone (Figure 7).

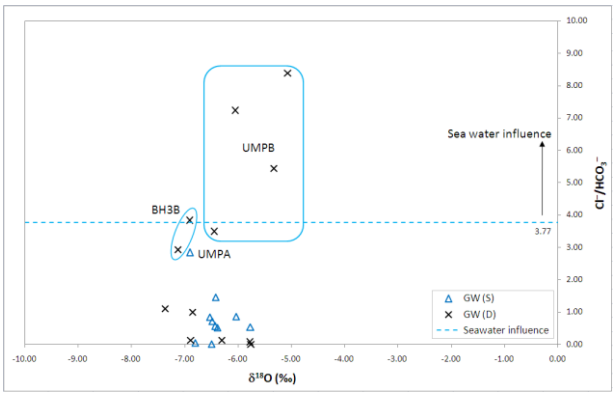

Figure 7. Cross plot representing $\delta^{18}O$ vs Cl/HCO_3relationship

Based on the mass balance analysis for the oxygen isotopic composition (Equation (2)), the result shows that the most affected deep groundwater is UMPB which is being recharged by 12% of the seawater. This conforms to all the hydrochemical and isotope results above as shown clearly in Figures 5, 6 and 7. The hydrogeological cross–section in Figure 1 also verifies this finding where the aquifer is up to 80m below sea level on the east side towards the coastal in Pekan. However, as the study area moves south, the area is dominated by clay and bedrock and this explains why BH3 was not affected by seawater intrusion although there is a sign of slight influence of seawater.

IV. CONCLUSION

The results showed that both the meteoric water and river water (88%) are the main source of recharge for one of the deep groundwater (UMPB) which is near to the coast. Seawater contributes 12% to this particular groundwater recharge. Other deep groundwaters located at the southern part of the study area (BH3A and BH3B) and shallow groundwater, including near to the coast (UMPA and BH3) showed slight influence of seawater intrusion. The stable isotopic compositions clearly demonstrated that there is no interaction between the mid–deep and deep groundwater and thus indicate that they came from different aquifers. However, due to the seawater intrusion, it has shifted UMPB isotopic signature close to the mid–deep groundwater and making it more enriched compared to other deep groundwater. These are in accordance with natural radioactive isotopes result where UMPB is delineating the characteristics of a shallow groundwater (relatively high ^3H and younger age) which means that it is affected by the modern saline water. The hydrochemical result also conforms to all the environmental isotopes findings.

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