2ND CAJG 2019



Stable isotope evidence on mechanisms and sources of groundwater recharge in quaternary aquifers of Kelantan, Malaysia

Mohammad Muqtada Ali Khan¹ · Kishan Raj¹ · Aweng A/L Eh Rak¹ · Hafzan Eva Mansor¹ · Roslanzairi Mostapa² · Kamarudin Samuding² · Zameer Ahmad Shah³

Received: 22 May 2020 / Accepted: 16 June 2021 © Saudi Society for Geosciences 2021

Abstract

The groundwater from shallow alluvial aquifers of northern Kelantan, Malaysia, was analysed for stable isotopes (δ^{18} O and δ^{2} H) during northeast and southwest season to assess the dynamics of the recharge processes. The groundwater shows quite a large variation in isotopic signatures (¹⁸O ranges from -7.5 to 0.62 ‰ for northeast (NE) and from -7.4 to -4.98 ‰ during southwest (SW)). These values reflect the contribution of different recharge sources in the aquifer. During NE season, the average isotopic signature of groundwater (δ^{18} O: -5.30‰) is similar to the rainfall (δ^{18} O: -4.13‰) indicating that rainfall is the main source of recharge compared to the other sources. The SW season shows an isotopically distinct signature due to contribution from surface water bodies (river sources) and enriched irrigation return-flow. Interestingly, the groundwater average isotope signature in SW season (δ^{18} O: -6.40‰) is very close to the surface water (δ^{18} O: -6.47‰), indicating significant contributions from surface water bodies of the area. Overall, the isotope data suggests that shallow groundwater is very dynamic during SW season whereas in NE the impact of surface manifestations is less. The study highlights the significance of further studies to accurately quantify the recharge volumes from different sources that will help to model and manage the aquifers both qualitatively and quantitatively. Moreover, the quantification of irrigation return-flow is very essential component keeping in view the contaminant transport from the agricultural areas.

Keywords Stable isotopes · Recharge · Shallow aquifers · Malaysia

Introduction

Groundwater is the main source of drinking water in the northern Kelantan area of Malaysia (Wan et al. 2014; Sefie et al. 2018). The groundwater is naturally recharged by meteoric water and streams that maintain the resource in the aquifers (Daud and Roslan 1981; Wan et al. 2012; Khairul Nizar et al. 2018). However, the groundwater is highly affected by

Responsible	Editor:	Broder	J. N	/lerkel	

This paper was selected from the 2nd Conference of the Arabian Journal of Geosciences (CAJG), Tunisia 2019

Mohammad Muqtada Ali Khan muqtada@umk.edu.my; muqtadakhan@gmail.com

- ¹ Faculty of Earth Science, Universiti Malaysia Kelantan, Campus Jeli, 17600 Jeli, Kelantan, Malaysia
- ² Malaysia Nuclear Agency, Kajang, 43000 Bangi, Malaysia
- ³ Geological Survey of India, Northern Region, UT: Jammu & Kashmir, Jammu 180006, India

anthropogenic factors leading to unsustainability in terms of water supply and food security as irrigation and domestic supply is largely dependent on groundwater in many parts of Malaysia (Brown 2009; Rasul 2016; Misra 2014; Narany et al. 2017). Rising population is also consistently putting pressure on the groundwater resources in the area. According to the review of the national water resources study (2000-2050) and formulation of national water resources policy (2011) for Kelantan, the groundwater demand is projected to be 210, 270, and 443 million liters per day (MLD) in years 2020, 2030, and 2050, respectively (NWRS 2011). Current groundwater abstraction is approximately increasing at 2.5% per year (Narany et al. 2017; Zamri et al. 2012; Sefie et al. 2018). Groundwater is also affected by land-use changes and contamination by various inorganic constituents. Over the past 25 years, the nitrate concentration in groundwater increased at 8% and 4% annually in agriculture and residential areas respectively (Narany et al. 2017). Therefore, one aspect of dealing with the groundwater problems is the proper assessment of the groundwater resources and recharge estimation in the Kelantan state.

Studies have been conducted on the stable isotope systematic in groundwater in many parts of the world, but such studies are not common in Malaysia. Based on major ion chemistry and isotopic analysis, Wan et al. (2012) concluded that meteoric water and streams recharge the groundwater in Kelantan River catchment and the isotope composition of shallow groundwater is influenced by evaporation. Wan et al. (2014), based on chloride mass balance (CMB), analyses of deuterium, oxygen 18, and tritium estimated that recharge rate ranges from 155 to 966 mm/year (average 484.3 mm/year or 19.4% of the total effective annual rainfall) and ranges from 11 to 1270 mm/y (average 261.5 mm/year or 10.5% of the total annual rainfall). Most of the groundwater isotopic investigations carried out in northern Kelantan had not been discussed in detail and were focused mainly on deeper aquifers. With this background, the present study was taken up to assess the mechanism and source of recharge in shallow aquifers keeping in view its qualitative and quantitative suitability for various purposes.

Materials and methods

Geography, geology, and hydrogeology of the study area

The study area is situated in northeastern part of Peninsular Malaysia (Fig. 1a) including Bachok, Tumpat, Pasir Mas, and Kota Bharu districts of Kelantan state (Fig. 1b) and covers an area of approximately 1400 km². The area lies between latitudes 5° 55" and 6° 15" north and between longitudes of 102° 4" and 102° 25" east (Fig. 1c). Topographically, the area is dominated by low-lying surfaces with an average elevation of less than 15 m above mean sea level (amsl). The main peaks in the area lie near Ketereh and Gunong where elevation reaches up to 248 m amsl (Fig. 1c). The area experiences a humid tropical climate, controlled by two monsoon seasons. The southwest Monsoon occurs between April and October often bringing less rainfall whereas the northeast monsoon occurs between November and March which frequently generates high rainfall intensity over the study area (Hayati et al. 2020). The average annual rainfall is 2788 mm. The average humidity is 70% while the average daily temperature is approximately 28°C. The area is mainly drained by the Kelantan River flowing across the area from south to north. Agriculture, especially rice cultivation is the most important economic activity in the area.

Geologically, the area is mainly covered by Quaternary sediments or alluvium of marine or fluviatile origin (i.e. gravel, sand, silt, and clay) underlain by granitic bed-rock (Zamri 2009; Muqtada et al. 2018). The geological map in Fig. 2 depicts the distribution of rocks from Tertiary, Permian, to Quaternary age. The Quaternary alluvium thickness in the north Kelantan region varies from a few meters near the mountainous areas and stretching up to 150 m towards the coast (Hayati 2011).

Based on the lithological data, the area consists of three aquifer layers (Sefie et al. 2018): shallow aquifer, intermediate aquifer, and deep aquifer (Fig. 3). The shallow aquifer with a thickness from a few meters to a maximum of about 15 m is mostly unconfined and semi-confined. The intermediate and deep aquifers lie at a depth of around 20 m to 50 m and > 50 m, respectively. The third aquifer has its thickness varying from place to place and generally increases towards the coast. Among the three, a shallow aquifer is suitable for abstraction purposes as it exhibits a significant recharge rate. The intermediate aquifer naturally has low thickness and stores a limited amount of groundwater.

Sampling and analytical procedures

Twenty-nine groundwater samples were collected uniformly over the entire study area each during January 2016 (referred to as northeast season) and June 2016(southwest season). Samples were collected in 500-mL glass bottles and subsamples in 7-mL vials were stored for isotope analysis. The sample bottles were thoroughly cleaned with distilled water prior to use. Vials were filled up to the brim and tightly sealed with double caps before storage and refrigeration. All the groundwater samples (P1 to P29) were collected from the first aquifer in which groundwater depth is very shallow (less than 8 meters). Three surface water samples (S1, S2, and S3) were also collected from the Kelantan River and its tributaries. Rainwater was collected at Kota Bharu station according to the standard guidelines of the International Atomic Energy Agency (IAEA) (Fig. 1c). Twelve rainwater samples were collected between July 2015 and June 2016 on monthly basis. The parameters analysed and the methods used are given in Table 1.

The isotopic composition is expressed as deviations in δ (pronounced as delta) values in parts per thousand units (‰ or per mille) notation relative to Vienna standard mean ocean water (VSMOW), shown as below:

$$\delta X = \frac{\left(R_{sample} - R_{standard}\right)}{R_{standard}} \times 1000\%$$
(1)

where the X is the measured value of hydrogen and oxygen; R_{sample} and $R_{standard}$ are the isotopic ratios of the samples and the VSMOW, respectively (Liu et al. 2018).

The δ^2 H and δ^{18} O are important tools for understanding recharge processes in watershed hydrology and can be used to trace precipitation of different origins through hydrological processes (Scholl and Murphy 2014; Yeh and Lee 2018; Oiro et al. 2018; Yang et al. 2019), assess groundwater recharge (Han et al. 2018; Yang et al. 2018), study the effects of evaporation of groundwater systems (Gonfiantini's 1986; Hendry 1988; Ding et al. 2013; Mahindawansha et al. 2019),

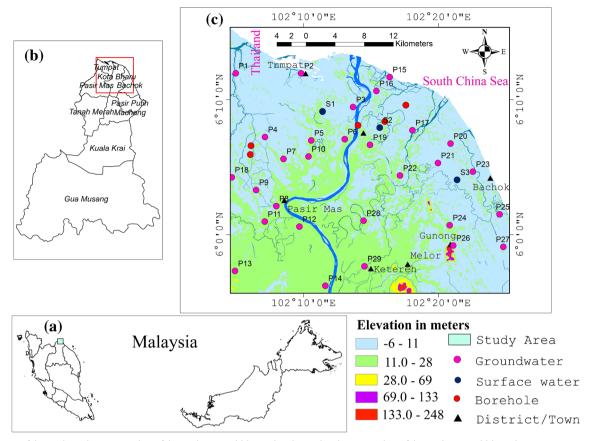


Fig. 1 Map of the study region: a Location of the study area within Peninsular Malaysia; b Location of the study area within Kelantan state; c Location of the water sampling sites imposed on ASTER DEM (30-m resolution)

and to study groundwater and surface water interactions (Krabbenhoft et al. 1990; Tweed et al. 2020). Previous isotope hydrology studies from the tropical areas include those from Hawaii (Scholl et al. 1996, 2002), Costa Rica (Rhodes et al.

2006; Lachniet et al. 2007), and Mexico (Munoz-Villers and McDonnell 2012). Any variation in δ^2 H and δ^{18} O in precipitation forms the primary data required for groundwater recharge investigations (Ingraham 1998; Gupta and

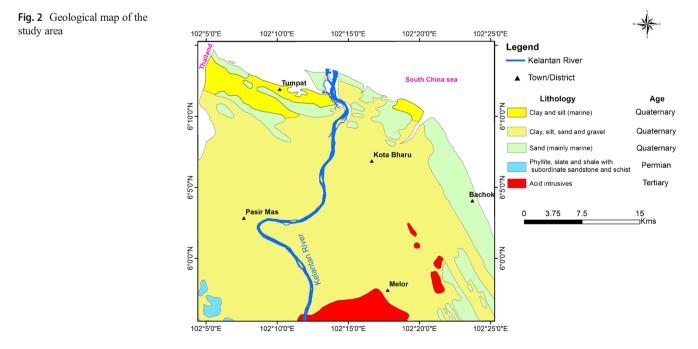
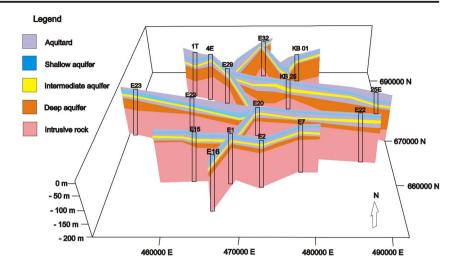


Fig. 3 Fence diagram showing aquifer disposition of the area (Sefie et al. 2018)



Deshpande 2003; Kortelainen 2009; Gat 2010; Voss et al. 2021). The δ^2 H and δ^{18} O of meteoric water vary from region to region and this variation is due to the varying intensity of isotopic fractionation. When meteoric water seeps into the ground to form groundwater, its isotopic variation is recorded in the groundwater. Hence, differences in isotope compositions can be used as a basis for the detection of groundwater sources (Yeh et al. 2011; Liu and Yamanaka 2012; Ding et al. 2013; Peng et al. 2016). During the entire hydrological cycle ranging from evaporation of water bodies to precipitation, the isotopes undergo continuous fractionation, which alters the isotopic composition in meteoric waters. As this process is based on isotopic fractionation during evaporation and condensation, there is a specific relationship between hydrogen and oxygen isotopic distributions in atmospheric precipitation. This relationship, first proposed by Craig (1961), is as follows:

$$\delta \mathbf{D} = 8\delta^{18}\mathbf{O} + 10\tag{2}$$

This relation defines the Global Meteoric Water Line (GMWL) and was obtained by using a linear regression method derived from the isotopic analysis of precipitation, snow water, and river water from all over the world; most of the precipitation in the world follows this relationship (Hsin-Fu et al. 2014). The slope of 8 and the intercept of 10 in the equation are useful in understanding the systematic isotopic

fractionation controlled by the hydrologic cycle. Global Meteoric water line (Craig's line) is global in the application and is actually an average of many local or regional meteoric water lines which differ from the local line due to varying climatic and geographic parameters. For investigations at local or regional scales, Regional Meteoric Water Lines (RMWL) or Local Meteoric Water Lines (LMWL) are generated from local precipitation isotopic values and are later used as a reference line. In the present investigation, LMWL: $\delta^2 H = 7.1$ δ^{18} O +7.75 was generated by the least square method from annual rainfall isotopic data. In addition, the Malaysian Meteoric Water Line (MMWL; $\delta^2 H = 8 \delta^{18} O + 13.25$; Shahid 2005) was also used. Keeping in view the proximity of Thailand to the study area, the Thailand's Meteoric Water Line (TMWL: $\delta^2 H = 7.35 \ \delta^{18}O + 6.11$, Kwansirikula et al. 2005) was also taken into consideration. The slope of line is very similar to that of Thailand's meteoric water line. All these lines (MMWL, TMWL, and LMWL) possess slope less than that of the Global Meteoric Water line, which indicated typical sub-tropical climate in this area.

Results

The isotopic data of the groundwater and surface water for the northeast and southwest season are presented in Table 2. Chloride concentrations show significant spatial variability

Table 1 Parameters and methods used

Parameters	Units	Instrument (method)	Lab
Oxygen-18 (¹⁸ O)	δ VSMOW ‰	SERCONGEO 20-20 (CF-IRMS).	Isotope Hydrology Laboratory of the Malaysian Nuclear Agency
Deuterium (² H)	δ VSMOW ‰	SERCONGEO 20-20 (CF-IRMS).	Isotope Hydrology Laboratory of the Malaysian Nuclear Agency
Chloride	Mg/l	Titration method	Geology lab, Universiti Malaysia Kelantan

Sample No.

P1

P2

P3

P4

P5

P6

P7

P8

P9

P10

P11

P12

P13

P14

P15

P16

P17

P18

P19

P20

P21

P22

P23

P24

P25

P26

P27

P28

P29

S1

S2

S3

Northeast Season

 $\delta^2 H$ (%)

-40.3

-39.04

-31.61

-33.84

-42.71

-1.63

-36.95

-41.14

-29.03

-40.94

-37.37

-37.43

-32

-32.53

-26.74

-32.25

7.2

-35.16

-29.69

-30.05

5.42

-30.76

-39.22

-23.91

-35.04

-35.92

5.02

-23.85

-26.36

-37.41

-26.88

-37.06

δ¹⁸O (%)

-6.75

-7.5

-6.38

-6.3

-7.15 -1.99

-6.36

-7.21

-5.03

-6.52

-5.8

-6.65

-5.63

-6.09

-6.49

-6.09

-0.83

-7.24

-5.47

-5.83

-1.64

-5.74

-6.2

-4.37

-5.32

-5.65

0.62

-3.77

-4.36

-6.9

-5.9

-6.63

61.84

45.04

36.38

31.01

42.76

24.52

24.63

18.4

23

42.56

50.05

43.14

32.59

39.45

38.34

20.27

52.54

42.6

32.57

29.46

27.95

32.62

31.1

26.04

 Table 2
 Analytical results of groundwater (P) and surface water (S) samples in northern Kelantan

		Southwest Season				
)	Cl (mg/l)	δ ² H (‰)	δ ¹⁸ O (‰)	Cl ⁻ (mg/l)		
	47.04	-35.52	-5.46	39.12		
	41.2	-40.94	-6.29	65.8		
	42.27	-33.71	-4.98	31.64		
	45.33	-42.29	-6.81	62.85		
	31.24	-33.71	-6.27	25.43		
	21.3	-37.53	-6.64	27.1		
	28.4	-40.26	-6.82	33.6		
	53.72	-35.41	-6.67	24.85		

-5.96

-6.27

-6.4

-6.88

-6.36

-7.38

-6.23

-6.9

-6.26

-6.25

-5.35

-7.4

-6.38

-6.95

-6.22

-6.62

-5.49

-6.3

-7.22

-6.83

-6.21

-5.63

-5.64

-5.7

-32.2

-35.96

-40.25

-38.49

-33.58

-41.05

-32.26

-44.67

-42.9

-45.02

-36.94

-46.69

-39.22

-44.99

-40.73

-38.36

-33.03

-38.03

-45.38

-45.12

-37.23

-38.45

-34.94

-33.17

with values ranging from 18.4 to 61.84 mg/L with an average value of 36.88 mg/L in NE season, whereas the values vary from 21.06 to 65.08 mg/L with an average of 35.84 mg/L in SW season. The δ^{18} O and δ^{2} H of the rainfall range from -6.2 to -2.37 ‰ and -43.52 to -8.35‰, respectively. The average values of δ^{18} O and δ^{2} H in the rainfall are -4.13 to -20.56 ‰, respectively. Values of δ^{2} H in groundwater range from -42.71 to 7.2‰ with an average of -28.55‰ during northeast and -46.69 to -32.20‰ with an average of -39.02‰ for the southwest season, respectively. For δ^{18} O, the values range from -7.24 to 0.62‰ with an average of -5.30‰ for northeast and -7.4 to -4.98‰ with an average of -6.40 for southwest, respectively (Table 3). Apparently, three groups of

groundwater samples can be identified during the northeast season. The first group includes those samples collected from the northeastern and southeastern parts of the area. For these samples, δ^{18} O and δ^{2} H values vary from -1.99 to 0.62% (average value of -0.96%) and from -1.63 to 7.2% (average value of 4.00%), respectively. The second group includes samples collected from the central and southern parts of the study area. In this group, δ^{18} O and δ^{2} H values vary from -4.37 to -3.77% (average value of -24.71%) and from -26.36 to -23.85 (average value of -24.71%), respectively. The third group shows δ^{18} O and δ^{2} H between -7.5 and -5.03% (average value of -6.25%) and -42.71 to -26.74% (average value of -34.99%), respectively.

53.76

39.41

42.75

28.1

39.6

47.25

31.86

26.4

39.6

31.47

46.25

40.87

21.06

24.63

32.57

23.82

44.3

35.17

21.32

22.08

36.67

17.42

23.51

17.45

Season	No. of samples	δ ² Η (‰)			δ ¹⁸ Ο (‰)		
		Range	Mean	Standard deviation	Range	Mean	Standard deviation
Northeast	29	7.2 to -42.71	-28.55	14.22	0.62 to -7.5	-5.30	2.00
Southwest	29	-32.2 to -46.69	-39.02	4.40	-4.98 to -7.4	-6.41	0.58

Table 3 The statistical isotopic characteristics of groundwater in northern Kelantan

Discussions

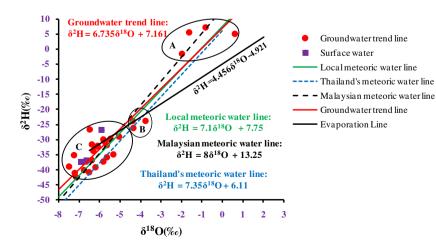
Sources and mechanisms of groundwater recharge

Stable isotopes (δ^2 H and δ^{18} O) of water carry attributes of their origin and thus can be used to trace the source and transport mechanism of water molecules (Clarke and Fritz 1997). Figure 4 is the plot of $\delta^2 H$ vs. $\delta^{18}O$ for groundwater and surface water samples of the northeast season. The samples lie most closely around local meteoric water line confirming that shallow aquifers in the area are recharged by present-day precipitation, though some samples (P15, P18, and P21) fall little away from the meteoric water line. This shift is due to the evaporation effect (Ebrahim et al. 2020; Hanafiaha et al. 2019), which is well evident from the groundwater trend line $(\delta^2 H = 6.73 \ \delta^{18} O + 7.16)$, with a slope of (6.73) lower than that of meteoric water lines. All these three samples fall in paddy fields where role of evaporation cannot be ruled out. The samples are apparently plotted in three separate groups. Group A (P6, P17, P21, and P27) includes those waters located in town areas in northern parts. The group mostly lies along MMWL and above LMWL in the upper right corner of the plot. The isotopic compositions of the samples are higher (more towards positive, with an average of δ^{18} O: -0.96%) than the average values (δ^{18} O: -4.13%) of local precipitation, implying the mixing effect through groundwater pumping, intrusion of waste water, and possibly evaporation effects when potential/actual evapotranspiration is high in the area.

Evapotranspiration has been acknowledged as a core process in the hydrological cycle in peninsular Malaysia (Yong et al. 2021). The overall impact of climate on water resources in the Kelantan river basin has already been assessed by many workers (Tan et al. 2017).

Keeping in view the shallow nature of the aquifer, the reevaporation from the water table is quite possible. This reevaporation from shallow water table has been estimated to be 24 mm/year (Ebrahim et al. 2020). The top clay layer as revealed in fence diagram (Fig. 3) also plays some role in impeding the downward movement of water, thus plays some role in the recharge process. Group B (P24, P28, and P29) contains the samples with mixed plantations (oil palm, rubber plantation, scrubs, etc.) in southeastern parts of the area. They lie at the center of the plot in Fig. 4 with the δ values being more depleted (δ^{18} O: -4.17%, δ^{2} H: -24.70%, than those of group A (δ^{18} O: -0.96‰, δ^{2} H: 4.00‰) and the average rainfall values (δ^2 H= -20.56% and δ^{18} O= -4.13%), which suggests that sources other than rainwater are also contributing to the groundwater system. Group C is dominant group (P1 to P5, P7 to P16, P18 to P20, P22, P23, P25, and P26; and three surface water samples) covering around 76 % of the groundwater samples, lying at the bottom left of the plot in Fig. 4. The average values of these samples are close (δ^{18} O: -6.24‰, δ^2 H: -34.98‰) but less than the mean annual δ^2 H and δ^{18} Ovalues of rainfall. This suggests that recharge sources of shallow aquifers are mainly from rainfall but not exclusively dominated by the monsoon season.

Fig. 4 $\delta 2H - \delta^{18}O$ plot for groundwater and surface water from the study area in January 2016 (northeast season)



In addition, the exposition of surface water bodies makes them more vulnerable to evaporation effects, which leads to the shifting of their δ^2 H and δ^{18} O values towards the positive side (Fig. 4). During evaporation, the water containing 18 O diffuses to the atmosphere more slowly than water molecules containing deuterium (Clarke and Fritz 1997). This differential behavior with respect to evaporation causes an increase in the ¹⁸O relative to the deuterium and resulting isotopic composition plots on a line with a lower slope than the meteoric water line. These evaporative trend lines typically exhibit slopes in the range of 3.9 to 5.2 (Gonfiantini's 1986). In the present study, from the average stable isotopic composition of northeast groundwater and surface water, the evaporation line $(\delta^2 H = 4.456 \ \delta^{18} O - 4.921)$ has a slope of 4.45 (the black solid line in Fig. 4). The original isotopic values of water prior to evaporation can be inferred by extrapolating the line along the LMWL (Clarke and Fritz 1997). The intersection point of LMWL and evaporation line represents the isotopic content of early recharge water that was not affected by evaporation (Zongyu et al. 2011). In the present case, the original values for δ^{18} O and δ^{2} H were -4.90 % and -26.75 %, respectively. These original isotopic values fall well within the δ^2 H and δ^{18} O range of annual rainfall and hence points to the fact that rainwater is the main source of groundwater recharge in most of the area. Moreover, the point of intersection of evaporation line with LMWL falls in group C, which indicates that the recharge of samples falling away from the meteoric water line in this group (P2, P15, and P18) mainly occurs from the surface water bodies which has analogous isotope values with this group.

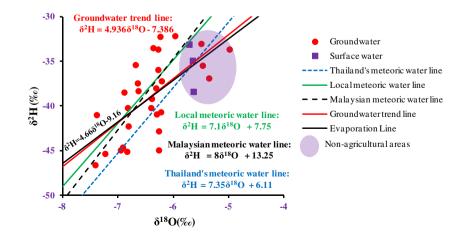
Figure 5 represents the plot of δ^2 H vs. δ^{18} O for southwest groundwater and surface water samples, which show significant deviation from LMWL. Around 62% of groundwater samples fall well below the LMWL, which suggests that enrichment has taken place through more than one mechanism. The best-fit regression equation for the southwest season (δ^2 H = 4.936 δ^{18} O - 7.386) exhibits a slope of 4.93, which is less than that of the LMWL (7.1) and northeast groundwater trend

line (6.73). Here the intensity of enrichment through evaporation is higher as compared to the northeast season and some data points show some significant deviation from average rainfall values, thus fall above LMWL and MMWL. These samples display significant moisture recycling which is well evident from average d-excess values of rainwater (d-excess 13%). The significant scattering of samples suggests that these waters were strongly affected by evaporation. Here the enrichment of groundwater in heavy isotopes through the mixing of irrigation return-flow water plays a key role in keeping in view the widespread rice fields in the area. The rice fields in the area are planted in the months of October and November. The large-scale pumping of groundwater for irrigation purposes may result in the mixing between enriched irrigation return-flow water and more depleted groundwater (Basu et al. 2002; Zheng et al. 2005).

In the study area, the farmers traditionally plan their crop production primarily on the basis of expected rainfall. In such areas, a significant portion of water used for rice production returns to the atmosphere in the form of evaporation and transpiration. Both these processes cause enrichment of residue water in heavier isotopes (Clarke and Fritz 1997). According to pan-evaporation data in Pasir Mas area (1969–1979), the monthly average evapotranspiration of a paddy crop is about 155 mm. The average evapotranspiration during the dry months is 5.5 mm per day and maximum daily values reach up to 9 mm (Kaishoven et al. 1984). The rainfall is at the lowest in the area during dry periods from January to May (average monthly rainfall of 72.5 mm from January to May 2016, Malaysian Meteorological Department 2016). During these months, the rainfall volume is insufficient to meet the crop water requirements. Under such circumstances, the dependency on groundwater for irrigation purposes is inevitable.

In the southwest season, 28% of the samples plot close to the LMWL, which suggests groundwater recharge at different stages and this recharge is typically representative of southwest monsoon season. The evaporation trend line

Fig. 5 $\delta^2 H - \delta^{18} O$ plot for groundwater and surface water from the study area in June 2016 (southwest season)



 $(\delta^2 \text{H}=4.66\delta^{18}\text{O} - 9.16, \text{ Fig. 5})$ for SW season shows a slope of 4.66. The original pre-evaporation isotopic values (-6.92 for $\delta^{18}\text{O}\%_0$ and -41.41 $\%_0$ for $\delta^2\text{H}$) does not fall within the isotopic range of annual rainfall. This clearly indicates significant mixing of enriched irrigation return-flow with pre-existing depleted water. However, samples encircled in purple color at the top right corner of the plot (three surface water and four groundwater, Fig. 5) show original pre-evaporation isotopic values of -6.18 $\%_0$ and -35.98 $\%_0$, which falls well within the isotopic range of annual rainfall and close to the surface water range. Remarkably, all these seven samples fall in non-agricultural areas where possibilities of irrigation return-flow are negligible. This also suggests the interconnectivity of surface water and the underlying shallow aquifer in the area.

Isotopic evidence of evaporation:

In order to assess more precisely the recharge mechanism and impact of evaporation on isotopic signatures, chloride concentrations were plotted against the δ^{18} O signature of the groundwater (Fig. 6a and b). Such relationships are very useful while

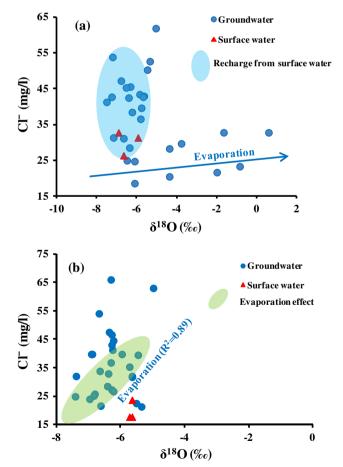


Fig. 6 Relationship between δ^{18} O and Cl⁻ concentrations in northeast season (a) and southwest season (b)

identifying the mechanism of groundwater recharge (Shao 1989; Chen et al. 2006). Any increase in salt concentrations due to evaporation should be reflected by enrichment in δ^{18} O signatures along a linear relationship (Clarke and Fritz 1997; Négrel et al. 2011). By contrast, the dissolution of salts leads to salinity *increase* but not to the fractionation of water molecules (Lorenzen et al. 2012). The main inferences, which can be made out of δ^{18} O-Cl⁻ plots, are as follows:

- The role of evaporation in enriching isotopic signatures is clear from δ¹⁸O-Cl⁻ plot of NE-season (Fig. 6a). The samples (P6, P17, P21, P27, P28, and P29) in the right corner of the plot show enriched isotopic signatures with increasing Cl⁻ concentrations, which can be well attributed to the evaporation effect.
- During NE-season, the Cl⁻ concentration of some samples (P1 to P4, P8 to P10, P13, P18 to P20, P25, and P26) encircled in light blue color (Fig. 6a) show an increasing trend, while the δ¹⁸O values show lower variations. Interestingly, the δ¹⁸O composition of these samples was depleted than in rainwater samples and *verges* more towards those of surface water, indicating that they are not entirely recharged by rainfall, but partially by Kelantan river and its tributaries mostly in the northwestern and northeastern parts of the area.
- In SW-season, 50% of the samples show a positive correlation (R²=0.89) between Cl⁻ and δ¹⁸O where isotopes show enrichment with increasing Cl⁻ concentrations (samples encircled in light green color, Fig. 6b). These samples (P1 to P3, P5 to P8, P10 to P13, P18, P22 to P24, P26, P27, and P29) reflect the recharge through irrigation return-flow when the area experiences very scanty rainfall.
- The samples (P21 and P28) in the bottom of the plot (including three surface water samples with Cl⁻ concentrations less than 25 mg/L and δ¹⁸O values less than -5%_o, Fig. 6b) are interpreted as fresh groundwater through seepage from surface water bodies. Moreover, the source of this fresh groundwater recharge is surface water which exhibits similar trends of Cl⁻ and δ¹⁸O.

Conclusions and suggestions

The demarcation of groundwater recharge sources and the related processes are decisive for executing better policies for modeling and management of aquifers. From the environmental isotope data of the groundwater collected in the NE season, it can be inferred that shallow zones get recharge mainly through rainfall. However, the partial recharge from other sources is quite clear keeping in view the clustering of surface water samples with groundwater. The role of irrigation return-flow as a recharge source is visible during the SW season. The groundwater from the irrigated areas shows more evaporated signature and quantifying the return-flow is required to model the contaminant transport from agricultural fields in the area. The proximity of average groundwater isotope values during SW with those of surface water clearly manifests the significant contributions from surface water bodies. For long-term groundwater management, the command over potential contamination from surface water bodies could be of huge scope as well as the potential of water purification of infiltrated river water for drinking purposes. In SW season, the recharge is not rapid as the original preevaporation isotope signature is altered before reaching the aquifer. The outcome of the present investigation shows that monitoring groundwater isotopes in shallow aquifers could be a very helpful tool for early localization of changes in water sources.

Future work should include detailed age determination of the groundwater in the area to develop management strategies for shallow groundwater systems that contain mostly young groundwater. The qualitative aspect must be evaluated from time to time keeping in view the overexploitation for largescale rice cultivation and subsequent irrigation return-flow in the area. This is particularly important for poor sections of the population, which are totally dependent on shallow groundwater resources. The shallow aquifers show the signatures of contamination that could leak into the lower aquifers. Quantifying the leakage and contaminant transport will help to manage the aquifers at an early stage.

Acknowledgements The study was carried out at the Universiti Malaysia Kelantan, Campus Jeli. The authors are grateful to the faculty of Earth Science, Universiti Malaysia Kelantan, Campus Jeli, for endowing with basic facilities to carry out the present investigation. Dr. Wan Zakaria from Nuclear Agency Malaysia is warmly thanked for providing vital inputs throughout this study. We also thank the anonymous reviewers for providing critical comments that improved this manuscript.

Funding The financial assistance is received from Fundamental Research Grants (R/FRGS/A08.00/00644A/002/2015/000228 and R/FRGS/A0800/00644A/003/2018/00556) is highly acknowledged.

Declarations Funding

Conflict of interest The authors declare that they have no competing interests.

References

Basu AR, Jacobsen SB, Poreda RJ, Dowling CB, Aggarwal PK (2002) Groundwater flow in the Ganges delta. Science 296(1563):5573

- Brown L (2009) Chapter 9, Feeding Eight Billion People Well. In: Plan B 4.0: Mobilizing to save civilization. Earth Policy, New York
- Chen ZY, Wan L, Nie ZL, Shem JM, Chen JS (2006) Identification of groundwater recharge in the Heihe basin Using environmental isotopes. HyrdogeolEngGeol 33(6):9–14
- Clarke I, Fritz P (1997) Environmental Isotopes in Hydrogeology. Lewis Publishers, pp 36–42
- Craig H (1961) Isotopic variations in meteoric waters. Science 133:1702– 1703
- Daud M and Roslan MA (1981) Prelimenary study of the environmental isotope in hydrology in the lower Kelantan river basin Kelantan, Malaysia, Submitted to the International Atomic Energy Agency (IAEA)- and Australian-Atomic Energy Commission.
- Ding Z, Ma J, Zhao W, Jiang Y, Love A (2013) Profiles of geochemical and isotopic signatures from the Helan Mountains to the eastern Tengger Desert, northwestern China. J Arid Environ 90:77–87
- Ebrahim N, Mohamed AMZ, Rowshon MK, Nurulhuda K (2020) Evaluation of groundwater recharge based on climate change: a case study at Baung's Watershed, Kota Bharu, Kelantan. SainsMalaysiana 49(11):2649–2658. https://doi.org/10.17576/jsm-2020-4911-04
- Gat JR (2010) Isotope hydrology: a study of the water cycle, Series on environmental science and management, vol 6. Imperial College Press, London
- Gonfiantini's R (1986) Environmental isotopes in lake studies. In: Fritz P, Fontes JC (eds) Hand-book of Environmental Isotope Geochemistry, vol 2. Elsevier, New York, pp 113–168
- Gupta SK, Deshpande RD (2003) Synoptic hydrology of India from the data of isotopes in precipitation. Curr Sci 85:1591–1595
- Han G, Lv P, Tang Y, Song Z (2018) Spatial and temporal variation of H and O isotopic compositions of the Xijiang River system, Southwest China. Isot Environ Health Stud 2018(54):137–146
- Hanafiaha MM, Nor FG, Siti NH, Hayder SA, Mahmood JAH, Mohd KAK (2019) Assessing water scarcity in Malaysia: a case study of rice production. Desalin Water Treat 149:274–287. https://doi.org/ 10.5004/dwt.2019.23841
- Hayati NH (2011) Hydrogeochemical Study and Iron Removal of Groundwater in North Kelantan Master dissertation. Department of Geology, University of Malaya
- Hayati NH, Ismail Y, May R (2020) Comparison of applications to evaluate groundwater recharge at lower Kelantan River Basin, Malaysia. Geosciences 2020(10):289. https://doi.org/10.3390/ geosciences10080289
- Hendry MJ (1988) Hydrogeology of clay till in Ana prairie region in Canada. Ground Water 26:607–614
- Hsin-Fu Y, Hung-I L, Cheng-Haw L, Kuo-Chin H, Chi-Shin W (2014) Identifying seasonal groundwater recharge using environmental stable isotopes. Water 6:2849–2861
- Ingraham NL (1998) isotopic variations in precipitation. In: Kendall C, McDonell J (eds) Isotope tracers in catchment hydrology. Elsevier, Amsterdam, pp 87–118
- Kaishoven G, Daane JRV, Fredericks LJ, Van den Steen van Ommeren F, Van Tilburg A (1984) Paddy farmers, irrigation and agricultural services in Malaysia: a case study in the Kemubu Scheme, pp 69–71
- Khairul Nizar MS, Wan NAS, Mohammad FR, Faradiella MK, Kamarudin S (2018) Assessments of seasonal groundwater recharge and discharge using environmental stable isotopes at Lower Muda River Basin, Malaysia. Appl Water Sci 8:120
- Kortelainen N (2009) Isotopic composition of atmospheric precipitation and shallow groundwater in Olkiluoto: O-18, H-2 and H-3. Working report 2009-06, Plosives.
- Krabbenhoft DP, Carl JB, Mary PA, John WV (1990) Estimating groundwater exchange with lakes: the stable isotope mass balance method. Water Resour Res 26:2445–2453

- Kwansirikula K, Fongsaward SS, Itsuro K, Isao T (2005) Hydrochemical and isotopic characteristics of groundwater in the Lampang Basin, Northern Thailand. Sci Asia 31:77–86
- Lachniet MS, Patterson WP, Burns ASY, Polyak V (2007) Caribbean and Pacific moisture sources on the Isthmus of Panama revealed from stalagmite and surface water d180 gradients. Geophys Res Lett 34: L01708. https://doi.org/10.1029/2006GL028469
- Liu Y, Yamanaka T (2012) Tracing groundwater recharge sources in a mountain-plain transitional area using stable isotopes and hydrochemistry. J Hydrol 464–465:116–126
- Liu Z, Xinxiao Y, Guodong J, Dandan W (2018) Oxygen and hydrogen isotopes of precipitation in a rocky mountainous area of Beijing to distinguish and estimate spring recharge. Water 10:705. https://doi. org/10.3390/w10060705
- Lorenzen G, Sprenger C, Pekdeger A (2012) Origin and dynamics of groundwater salinity in the alluvial plains of western Delhi and adjacent territories of Haryana State, India. Hydrol Process 26: 2333–2345
- Mahindawansha A, Christoph K, Philipp K, Lutz B (2019) Estimating water flux and evaporation losses using stable isotopes of soil water from irrigated agricultural crops in tropical humid regions. Hydrol Earth Syst Sci J
- Malaysian Meteorological Department (2016) Monthly rainfall data collected from Malaysian Meteorological Department. Malaysian Meteorological Department, Kuala Lumpur
- Misra AK (2014) Climate change and challenges of water and food security. Int J Sustain Built Environ 3(1):153–165
- Munoz-Villers L, McDonnell JJ (2012) Runoff generation in a steep, tropical montane cloud forest catchment on permeable volcanic substrate. Water Resour Res 48:W09528. https://doi.org/10.1029/ 2011WR011316
- Muqtada AK, Kishan RP, Aina M, Zakiyah AK, Hafzan EM (2018) Evaluating the suitability of shallow aquifer for irrigational purposes in some parts of Kelantan, Malaysia. Bull Geol Soc Malay 66:45–53
- Narany TS, Ahmad ZA, Anuar S, Saskia K (2017) Detecting and predicting the impact of land use changes on groundwater quality, a case study in Northern Kelantan. Malay Sci Total Environ 599– 600:844–853. https://doi.org/10.1016/j.scitotenv.2017.04.171
- National Water Resources Study (2011) Review of the National water resources study (2000-2050) and formulation of national water resources policy, Final report, volume 10, Kelantan August 2011.
- Négrel P, Hélène P, Benoît D, Jean-Marie G, Cédric M, Mohamed-Salem A (2011) Understanding groundwater systems and their functioning through the study of stable water isotopes in a hard-rock aquifer (Maheshwaram watershed, India). J Hydrol 397(1-2):55–70. https://doi.org/10.1016/j.jhydrol.2010.11.033
- Oiro S, Jean-Christophe C, Chris S, Kristine W (2018) Using stable water isotopes to identify spatiotemporal controls on groundwater recharge in two contrasting East African aquifer systems. Hydrol Sci J 63(6):862–877
- Peng TR, Huang CC, Zhan WJ, Wang CH (2016) Assessing groundwater sources and their association with reservoir water using stable hydrogen and oxygen isotopes: a case study of the Taipei Basin, northern Taiwan. Environ Earth Sci 75:1–13
- Rasul G (2016) Managing the food, water, and energy nexus for achieving the Sustainable Development Goals in South Asia. Environ Develop 18:14–25
- Rhodes AL, Guswa AJ, Newell SE (2006) Seasonal variation in the stable isotopic composition of precipitation in the tropical montane forests of Montverde, Costa Rica. Water Resour Res 42:W11402. https:// doi.org/10.1029/2005WR004535
- Scholl MA, Murphy SF (2014) Precipitation isotopes link regional climate patterns to water supply in a tropical mountain forest, eastern

Puerto Rico. Water Resour Res 50:4305–4322. https://doi.org/10. 1002/2013WR014413

- Scholl MA, Ingebritsen SE, Janik CJ, Kauahikaua JP (1996) Use of precipitation and groundwater isotopes to interpret regional hydrology on a tropical volcanic island: Kilauea volcano area, Hawaii. Water Resour Res 32(12):3525–3537
- Scholl MA, Gingerich SB, Tribble GW (2002) The influence of microclimates and fog on stable isotope signatures used in interpretation of regional hydrology: East Maui, Hawaii. J Hydrol 264:170–184
- Sefie A, Zaharin AA, Firuz MR, Tahoora SN, Khairul Nizar MS, Syaiful BS, Munirah AZ (2018) Hydrogeochemistry and groundwater quality assessment of the multilayered aquifer in Lower Kelantan Basin, Kelantan, Malaysia. Environ Earth Sci 77:397. https://doi.org/10. 1007/s12665-018-7561-9
- Shahid MA (2005) Malaysian meteoric water line. Institute for Nuclear Technology Research (MINT), Kajang
- Shao YS (1989) Environmental isotope geochemistry of groundwater in Hohhot basin, Inner Mongolia. Geotech Investig Surv 4:41–43
- Tan ML, Ab Latif I, Zulkifli Y, Vivien PC, Ngai WC (2017) Climate change impacts under CMIP5 RCP scenarios on water resources of the Kelantan River Basin, Malaysia. Atmos Res 189:1–10. https:// doi.org/10.1016/j.atmosres.2017.01.008
- Tweed S, MarcLeblanc I, Cartwright A, Bass Y, Travi V, Marc T, Nguyen B, Dang DN, Massuel S, Saravana KU (2020) In: Maurice PA (ed) Stable isotopes of water in hydrogeology, encyclopedia of water: science, technology, and society. John Wiley & Sons, Inc
- Voss KA, BodoBookhagen DS, Oliver AC (2021) Variation of deuterium excess in surface waters across a 5000-m elevation gradient in the east-central Himalaya. Hydrol Earth Syst Sci 586:124804. https:// doi.org/10.5194/hess-2018-534
- Wan ZWMT, Johari AL, Kamaruzaman M, Rohaimah D, Seyed RS, Ismail Y, Zainudin O (2012) Preliminary assessment of groundwater recharge using environmental isotopes investigation (¹⁸O, ²H, ¹³C) to support groundwater management in the north Kelantan River Basin. Conference Proceeding, on 2nd International Conference on Water Resources (ICWR 2012), Langkawi, Malaysia.
- Wan ZWMT, Nur HH, Ismail Y, Kamaruzaman M, Johari AL, Rohaimah D (2014) Integrated assessment of groundwater recharge in the north Kelantan river basin using environmental water stable isotopes, tritium and chloride data, Research and Development Seminar 2014; Bangi (Malaysia); 14-16 Oct 2014.
- Yang K, Han G, Liu M, Li X, Liu J, Zhang Q (2018) Spatial and Seasonal Variation of O and H Isotopes in the Jiulong River, Southeast China. Water 10:1677
- Yang K, Guilin H, Chao S, Peng Z (2019) Stable H-O isotopic composition and water quality assessment of surface water and groundwater: a case study in the Dabie Mountains, Central China. Int J Environ Res Public Health 2019(16):4076. https://doi.org/10.3390/ ijerph16214076
- Yeh HF, Lee JH (2018) Stable hydrogen and oxygen isotopes for groundwater sources of Penghu Islands, Taiwan. Geosciences 8:84. https:// doi.org/10.3390/geosciences8030084
- Yeh HF, Lee CH, Hsu KC (2011) Oxygen and hydrogen isotopes for the characteristics of groundwater recharge: a case study from the Chih-Ben Creek basin, Taiwan. Environ Earth Sci 62:393– 402
- Yong SLS, Ng JL, Huang YF, Ang CK (2021) Trend analysis of potential evapotranspiration in peninsular Malaysia, IOP Conf. Ser. Mater Sci Eng 1101:012008. https://doi.org/10.1088/1757-899X/1101/1/ 012008
- Zamri WM (2009) Groundwater for domestic needs in Kelantan. Water Malay 20:6–10

- Zamri WM, Ismail Y, Bahaa-eldin EAR (2012) Simulation of horizontal well performance using visual MODFLOW. Environ Earth Sci 68(4):1119–1126
- Zheng Y, Datta S, Stute M, Dhar R, Hoque MA, Rahman MW, Ahmed KM, Schlosser P, Van GA (2005) Stable isotopes (¹⁸O, ²H) and arsenic distribution in the shallow aquifers in Araihazar,

Bangladesh. EOS Transactions American Geophysical Union, 86 (52), Fall Meeting Supplement, Abstract H31B-1305.

Zongyu C, Miao J, Liu WW, Wang Y, Chen J (2011) Tracing sources of recharge to groundwater in the Northern Henan plain using environmental isotopes. In: 2011 International Symposium on Water Resource and Environmental Protection, Xi'an, China, pp 282– 285. https://doi.org/10.1109/ISWREP.2011.5893000