USING ISOTOPE TECHNIQUE TO ESTIMATE GROUNDWATER RECHARGE IN THE RED RIVER DELTA PLAIN

Le Viet Hung[1], Pham Quy Nhan[1*], Tran Thanh Le[1], Dang Duc Nhan[2]

Abstract: *The Red river delta plain is the second largest delta in Vietnam and is located in the North of the country with an area of 14,860 km² and residing more than 22.5 million inhabitants. Groundwater is mainly exploited in Quaternary sedimentary aquifers with a total discharge of about 3 million m^3/day. Some localities have shown signs of over-exploitation such as in Hanoi and in Nam Dinh, which may lead to related problems such as depletion, subsidence, saltwater intrusion, and water pollution. In order to be able to sustainably exploit groundwater, the groundwater recharges need to be estimated. There have been many studies referring to different methods of estimating the groundwater recharge in which the most effective one is the isotope technique. Field trip and water sampling for chemical compositions, stable isotopes ^{18}O, ^{2}H, and radioactive ^{3}H analysis were also implemented. Ground recharge rate in the range from 77 to 440 mm/year was estimated by using isotope analysis and interpretation*

Keywords: Groundwater recharge, Red river delta plain, stable isotopes ^{18}O, ^{2}H, radioactive isotope ^{3}H.

1. INTRODUCTION

The groundwater recharge of a certain area is a fundamental component of the balance of a certain catchment, which contributes to the sustainable exploitation of water resources in general and groundwater in particular. It is hard to measure directly so Lerner et al. (1990), Scanlon et al. (2002) reviewed numerous methods, ranging widely in complexity and cost, have been used to estimate groundwater recharge. However, due to large uncertainties involved in the measurement of individual parameters of each method, a common recommendation is that recharge should be estimated by the use of multiple methods and the results compared (Scanlon B.R et al., 2002).

The Red river delta plain (RRDP) is the second-largest delta in Vietnam with an area of over 14,860 km^2, residing 22.5 million people and the place with the largest population density in the country (MPI, 2020). Groundwater is a valuable resource that has been exploited quite a lot in this region with total exploitation discharges of about 3,000,000 m^3/day (MONRE, 2015). Many problems such as ground subsidence, pollution, depletion, and saltwater intrusion related to groundwater have also occurred in this area (Q.N. Pham et al., 2019). There have been a number of studies to assess the amount of groundwater recharge in this area. However, either the reliability of the research results is limited, or the research results are still local. Q.N. Pham (2000) estimated the groundwater recharge in RRDP by using a modeling method. Due to the lack of input data to build the model, the evaluation results have some uncertainty. T.L Tran et al. (2011) determined the recharge and the interaction between aquifers in Quaternary sediments in Thach That - Dan Phuong area, Hanoi by using stable isotope ^{18}O, ^{2}H, and radioactive isotope ^{3}H based on determining the average travel time in

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the aquifer. Through the stable isotope signature $\delta^{18}O$ and $\delta^{2}H$, it shows that the Holocene aquifer is related to the Pleistocene aquifer with the supply of 19.4%. Q.N. Pham et al. (2019) conducted isotope sampling to evaluate the interaction between river and groundwater. Postma et al. (2017) determined the recharge from the Red River to aquifers by Tritium/Helium dating in Nam Du area. Larsen et al. (2008) using isotopes and modeling to determine the recharge from Red river and rainwater to Quaternary aquifers in Dan Phuong area. In short, although there have been several studies to evaluate groundwater recharge as mentioned above, either the reliability of the research results is limited, or the research results are still local. Therefore, the objectives of this study are to apply the isotope technique to estimate the groundwater recharge rate in the RRDP. In this study, we only focus on diffuse (direct) recharge derived from precipitation or irrigation, etc. that occurs fairly uniformly over large areas.

The RRDP is located between latitude 21°34′ to 19°5′N and longitude 105°17′ to 107°7′10'E as its extremities. The overall area of the plain is approximately 14,860 km² (Figure 1) (Q.N. Pham, 2019). The delta gradually lowers from the Northwest to the Southeast, from the ancient alluvial shelves with an elevation of 10-15m down to the alluvial flats of 2-4m in the center where the tidal flats are still flooded every day. In the center of the delta terrain elevation varies from 8-10m, flat terrain. In the North, the delta is limited by the Tam Dao - Yen Tu mountain range, the South is limited by the Ba Vi - Vien Nam mountain range, in the East is limited by the coastline. In the middle of the plain, there are round top hills with gentle slopes and elevations from 25-45 to 100m. The rainy season is from May to October and the dry season is from November to April next year. The average annual rainfall for the whole delta is from 1033.1mm to 2338.7mm and the amount of evaporation is from 828.2mm to 1057.1mm. RRDP has a dense network of rivers with an average density of 0.7 to 1 km/km^2. The whole region has two main river systems: the Red river system and the Thai Binh river system. Due to the impact of waves, tides and river systems, the surface water has been significantly saline intrusion in the estuaries. This saline intrusion in the river system not only affects the coastal ecosystem, irrigation water supply etc. but also affects the shallow aquifers in the vicinity (Nielsen LH et al., 1999).

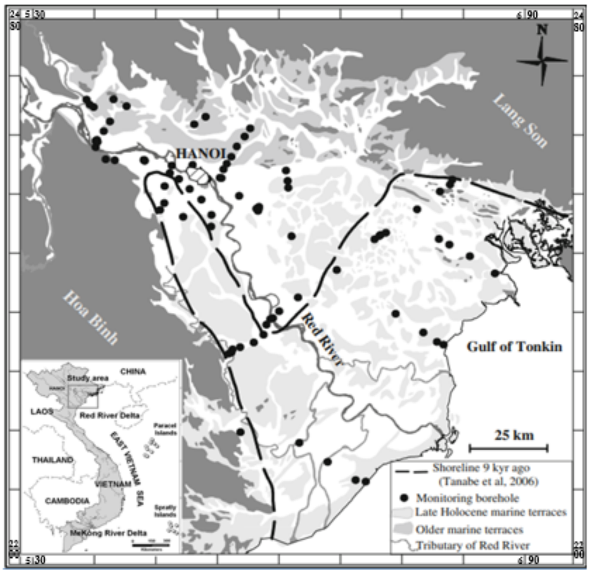

Figure 1. Location of study area. Modified from Tran Thi Luu et al. (2018)

2. METHODS AND MATERIALS

2.1. Data collection

The data on the groundwater monitoring network in the RRDP were collected, including data on the structure of the monitoring boreholes, the arrangement of the screen at the aquifer. Monitoring data on rainfall and evaporation at 6 meteorological stations, 7 rivers, water levels at hydrological stations and groundwater level data at 128 groundwater monitoring boreholes were also collected.

2.2. Water sampling and analysis

The Geo-hydrogeology of the study area in the

RRDP makes up the Northwest part of the Red River sedimentary basin; a basin filled with Paleocene, Neogene, and Quaternary deposits (Nielsen LH et al., 1999). The RRDP is surrounded by mountain ranges composed of crystalline rocks from Paleozoic and Mesozoic sedimentary rocks (Mathers SJ et al., 1996, 1999). According to Tran N et al. (1991), the sediments were deposited in five fining-upward sedimentation cycles. Hydraulic gradients in the range of 0.05-0.15% are typical and groundwater flow velocities in the Holocene aquifers are a few tens of meters per year (Larsen F et al., 2008).

The samples were taken once pH and electrical conductivity (EC) in water became unchanged. For chemical analyses, around 100 ml of groundwater samples were first filtered through 0.45 mm mesh filters to remove the suspended matters and then they were split into two parts. One part was acidified with 2-3 drops of HNO_3 (65%, PA grade, Merck, Germany) to make pH of the samples 1-2. The stable isotopic signatures of hydrogen and oxygen (δ^2H, $\delta^{18}O$) in water were analyzed at HUNRE on a Picarro's cavity ring-down spectrometer, CRDS L2130-I, which works based on the principle of absorption spectroscopy (Picarro's Operation Manual, 2016). The precision of the method was as high as 1.5‰ and 0.15‰ for δ^2H and $\delta^{18}O$, respectively. For ^3H analysis, the water samples were first subjected to distillation to remove the minerals dissolved until the electric conductivity was less than 10 mS/cm. Around 500 ml of the distilled water samples were then subjected to the electrolytic enrichment for tritium at 4°C till around 10 ml was attained (IAEA, 2002). This sample was analyzed at the Institute of Nuclear Science and Technology, Hanoi (INST) by means of electrolytic enrichment following ^3H activity measurement on a liquid scintillation counter (LSC). This dating method is currently recommended by IAEA and the INST has got assistance from IAEA with a system of the ^3H enrichment as well as LSC within a Technical Cooperation Project VIE8.016 in 2011-2013. To estimate the age of groundwater in a certain aquifer, the lumped-parameter model should be used with the ^3H input function (A_0)/time series of the ^3H in fallout which has been consecutively monitored at the IAEA Bangkok and/or Hong Kong stations since 1960. 16 (sixteen) ^3H radioisotope samples, as well as major ion samples, were collected from the national monitoring network of groundwater with different screen depths in the Holocene aquifer where is the upper aquifer of the Quaternary system in the RRDP. For Tritium dating, we will use the method of electrolytic enrichment following ^3H activity measurement on a liquid scintillation counter (LSC).

2.3. Estimation of groundwater recharge using isotope technique

The contribution ratio or surface water relationship to groundwater is described by a simple binary mixing model. It is the dilution or mixing of old water containing isotopes with new water containing different isotope content. The content of an isotope X (a substance X) in a sample is described by the following equation:

$$(X) = (X_b \cdot f + (X_p)(1 - f) \quad (1)$$

Where: X_p is the isotope content of the new water; X_b is the isotope content of the old water

f ($0 \le f \le 1$) is the fraction of the old water in the mixture (Widory, 2005). Using isotopes as an environmental tracer, the following system of equations can be obtained (Widory, 2005):

$$\left[\begin{array}{c} (X) = (X_b) \cdot f + (X_p) \cdot (1 - f) \\ (X) \cdot \delta X - (X_b) \cdot f \cdot \delta X_b + (X_p) \cdot (1 - f) \cdot \delta X_p \end{array}\right. \quad (2)$$

From equations 1 and 2 above, it is possible to estimate the mixing ratio of isotopes if the content of isotopes can be determined. Alternatively, it can be used to determine mixing conditions between water from different pollution sources. In this study, the above mixing (dilution) model was used to construct relationship curves to see how the ^{18}O and ^2H contents were mixed together from other sources.

On the basis of using two isotopes ($\delta^{18}O$) and deuterium (δ^2H), the balanced equations are established from the specific formulas (3,4):

$$\delta^{18}O_{gw} = X_i * \delta^{18}O_{rain} + (1 - X_i) * \delta^{18}O_{riv} \quad (3)$$

$$\delta^2H_{gw} = Y_i * \delta^2H_{rain} + (1 - Y_i) * \delta^2H_{riv} \quad (4)$$

Where:

$\delta^{18}O_{rain}$, δ^2H_{rain}: Content of ^{18}O and ^2H in rainwater, respectively (‰)

$\delta^{18}O_{riv}$, δ^2H_{riv}: Content of ^{18}O and ^2H in river water, respectively (‰)

$\delta^{18}O_{gw}$, δ^2H_{gw}: Content of ^{18}O and ^2H in groundwater, respectively (‰)

X_i, Y_i: The percentage of contribution of rainwater to the groundwater by using ^{18}O and ^2H, respectively, at the time i (%)

From the above equations, we determine the values of X_1, Y_1 as follows:

$$X_i = [\frac{\delta^{18}O_{gw} - \delta^{18}O_{riv}}{\delta^{18}O_{rain} - \delta^{18}O_{riv}}]*100 \quad (\%) \qquad Y_i = [\frac{\delta^2H_{gw} - \delta^2H_{riv}}{\delta^2H_{rain} - \delta^2H_{riv}}]*100 \quad (\%) \quad (5)$$

According to David J.Toth, 1995 the groundwater recharge calculated on the basis of radioactive isotope ^3H is determined by the following formula:

$$W \geq \frac{CD - (EL - WT)}{A} n \quad (6)$$

Where:

-W: Groundwater recharge rate (mm/year)

-CD: Depth of borehole's screen, isotope sampling position (m)

-EL: Elevation of borehole top (m)

-WT: Elevation of the water table (m)

-A: The age of the water in the borehole is determined by measuring Tritium ($T/^3He$).

-n: Formation porosity (%).

3. RESULTS AND DISCUSSION

At a campaign of field trip in the last dry season in 2021, we have measured groundwater levels and taken 128 stable isotope samples in Quaternary aquifers and in surface water bodies, and 16 radioactive isotope ^3H samples which were out of 128 monitoring boreholes in the RRDP. Rainwater samples were collected according to IAEA guidelines and the sampling equipment was located on the roof of the building of HUNRE for nearly 3 years. The results of the stable isotope sample analysis are presented in Table 1 and Figure 2. The vertical groundwater velocities determined according to the ^3H analytical result at different depths are presented in Table 2 and Figure 3. The regional meteoric water line (RMWL) is determined on the basis of analysis of rainwater samples and modifications from the study of Nhan et al. (2013).

3.1 Determination of groundwater recharge contribution from stable isotopes

Figure 2 presents water lines of groundwater and surface water samples taken for this study in the dry season of 2020-2021. In this figure, the regional meteoric water line (RMWL) characterized for the RRDP is also presented as a dotted line and described by the model (Nhan et al., 2013):

$$\delta^2H = (8.04 \pm 0.07).\ \delta^{18}O + (12.96 \pm 0.15),\ ‰\ vs.\ VSMOW \quad (7)$$

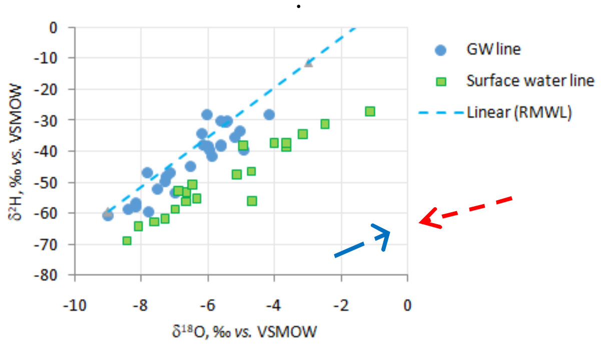

Figure 2. Isotopic compositions of ground water (GW line), surface water (surface water line) and regional meteoric water line (RMWL) of RRDP showing groundwater in the region is recharged by surface (red arrow) and local precipitation (blue arrow)

As seen from Figure 2 groundwater in the study area (the blue dots) was recharged from local precipitation and surface water as indicated by the blue and red arrows, respectively. In this case, surface water implies water from rivers or reservoirs existing around the sampling sites. The contribution of river and precipitation including irrigation and wastewater to groundwater was estimated following Equation (5) using $\delta^{18}O$ values of respective water samples. As sampling was conducted in the dry season so the $\delta^{18}O_{Prec}$ used for the calculation was taken as an average value of oxygen-18 composition in the precipitation of the previous rainy season to be -8.38‰ which was deduced from the RMWL of the area. Results of the calculation are presented in Table 1.

Table 1. Contribution of river water to groundwater in the study area

No	Borehole ID	Sampling Location	$\delta^{18}O_{SW}$	$\delta^{18}O_{GW}$	$\delta^{18}O_{Prec}$	X_{SW}	Y_{Prec}
1	Q83	Phu Ly, Ha Nam	-4.65	-6.14	-8.38	0.60	0.40
2	Q33	Dong Anh, Ha Noi	-3.12	-9.02		1.00	0
3	Q115	Song Ho, Bac Ninh	-6.35	-5.99		0.94	0.06
4	Q158	Hoa River bridge, Thai Binh	-6.65	-5.89		0.89	0.11
6	Q147	Tu Ky, Hai Duong	-4.71	-8.18		1.00	0
7	Q32	Đong Tru bridge, Ha Noi	-8.10	-8.18		1.00	0
8	Q144	Kim Thanh, Hai Duong	-7.31	-5.96		0.82	0.18
9	Q131	Kim Thanh, Hai Duong	-3.62	-8.18		1.00	0
10	Q146	Thanh Ha, Hai Duong	-4.03	-4.91		1.00	0
11	Q62	Tay Tuu, Ha Noi	-5.12	-5.47		1.00	0
12	Q55	Dan Phuong, Ha Noi	-8.45	-6.19		0.73	0.27
13	Q129	Lam Sơn, Hung Yen	-6.87	-4.15		0.60	0.40
14	Q130	Tien Lu, Hung Yen	-2.51	-5.20		1.00	0
15	Q143	Phuc Son bridge, Hai Phong	-7.58	-6.52		0.86	0.14
16	Q167	Ng. Truong To bridge, Hai Phong	-6.64	-5.62		0.85	0.15
17	Q168	An Hoa, Hai Phong	-7.00	-8.78		1.00	0
18	Q15	Dong Anh, Ha Noi	-3.62	-6.04		1.00	0
19	Q116	Gia Dong, Bac Ninh	-4.93	-7.53		1.00	0
20	Q35	Dong Anh, Ha Noi	-1.10	-5.61		1.00	0

As seen from Table 1 groundwater in the lower regions of the RRDP, e.g. in the region of boreholes Q130, Q131, Q146, Q147 in Hai Duong, or Q168 in Hai Phong groundwater was completely recharged by surface water.

3.2 Results from radioactive isotopes

Table 2 showed results of ^{3}H-age of the groundwater samples under this study and these results were separated into 3 groups which attributed to 3 zones (Le Viet Hung et al., 2021)

Table 2. ^{3}H-age of groundwater samples taken from boreholes with different water table elevation H (mbs: meter below the ground surface)

Zone I: Low recharge			Zone II: Moderate recharge			Zone III: High recharge		
Borehole ID	H, mbs	^{3}H-Age, y	Borehole ID	H, mbs	^{3}H-Age, y	Borehole ID	H, mbs	^{3}H-Age, y
Q68a	-8	2.8	Q83	-7.7	0.0	Q108	-12	11.0
Q1	-9	6.4	Q89	-8.4	1.4	Q109	-9	5.5

Zone I: Low recharge			Zone II: Moderate recharge			Zone III: High recharge		
Borehole ID	H, mbs	^{3}H-Age, y	Borehole ID	H, mbs	^{3}H-Age, y	Borehole ID	H, mbs	^{3}H-Age, y
Q67	-7	0.4	Q115	-14	22.0	Q110	-9	3.5
Q33	-8.94	20.0				Q159a	-7.5	3.0
Q66	-12	27.0				Q158	-7.4	1.7
						Q164	-9	3.5
						Q145	-9	3.5
						Q147	-9	2.5

With results in Table 4 potential recharge rates for the three expected zones in the RRDP were estimated based on the relationship between the ^{3}H-age of groundwater samples and the elevation of the water table in boreholes from where the samples were taken in this study. Figure 4 presents this relationship.

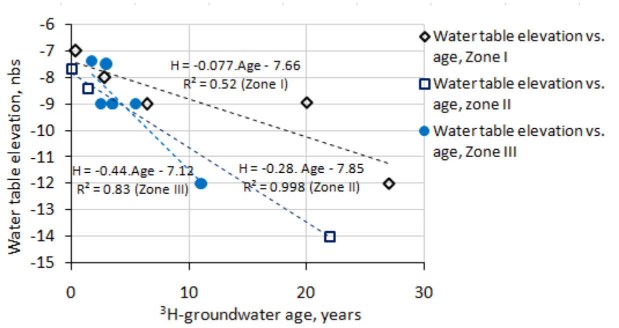

Figure 3. A graph showing a relationship between groundwater table elevation and ^{3}H age of groundwater samples taken from 3 zones with different potential recharge rates; mbs: meter below the surface; ^{3}H-age: groundwater age estimated by the ^{3}H method

As seen from Fig 4 in zone I, II, and III the relationships between groundwater table elevation and 3H-age followed three models as:

Zone I: H = -0.077.Age - 7.66 (m) (R^2=0.52) (8)

Zone II: H = -0.28.Age - 7.85 (m) (R^2= 0,998) (9)

Zone III: H = -0.44.Age - 7.12 (m) (R^2 = 0.83) (10)

This means that in the zone I, II, and III the potential recharge rates could be as high as 77 mm/year, 280 mm/year, and 440 mm/year, respectively. The depth of the unsaturated zone in the RRDP was as deep as from 7 to 8 m below the ground surface. These groundwater recharge rates imply that the classification by GIS and remote sensing as above mentioned and weight and rating which were assigned to each factor are reasonable. Postma et al. (2007) estimated the recharge from the Red River to aquifers by Tritium/Helium dating in Dan Phuong where it is located in North Hanoi and in-between zone I and zone II. Water samples for Tritium/Helium dating of the groundwater were taken from screens placed at different depths in the distance range from 64 to 75 m. The results suggest the groundwater to be less than 40 years old and a downward groundwater velocity of 0.5 m/yr. If formation porosity is 30% groundwater recharge rate could be about 150mm/year. T.L Tran (2011) used a water balance site with 05 boreholes and groundwater level measurements in the period 2008-2011 in Dan Phuong - Thach That area where is margin of the delta. Groundwater recharge rates were estimated as 175mm/year. This result also proved that the groundwater recharge rate in the RRDP which was estimated by using the isotope technique is acceptable.

4. CONCLUSIONS

The Quaternary aquifer system in the RRDP is recharged mainly from rainwater, river water, and surrounding bedrocks. The topmost Holocene aquifer is an unconfined and semi-unconfined aquifer and recharged mainly from rainwater and surface water bodies. In the dry season, stable isotope signature showed that in the region of Hai Duong, Hai Phong groundwater was completely recharged by surface water. Using isotope technique to estimate groundwater recharge rate in the RRDP on the basis of ^{3}H radioisotope analysis the groundwater recharge rate from high, moderate, and low potential zone were roughly estimated at 77; 280; and 440 mm/year, respectively. However, isotope samples are quite few and have only been conducted in the dry season, so they need to continue to be followed up in the near future.

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Tóm tắt:

ỨNG DỤNG PHƯƠNG PHÁP ĐỒNG VỊ XÁC ĐỊNH LƯỢNG BỔ CẬP CHO NƯỚC DƯỚI ĐẤT VÙNG ĐỒNG BẰNG SÔNG HỒNG

Đồng bằng châu thổ sông Hồng là đồng bằng lớn thứ hai của Việt Nam nằm ở phía Bắc đất nước với diện tích 14.860 km² và dân số hơn 22,5 triệu người. Nước dưới đất được khai thác chủ yếu ở các tầng chứa nước trầm tích Đệ tứ với tổng lưu lượng khoảng 3 triệu m^3*/ngày. Một số địa phương có dấu hiệu khai thác quá mức như Hà Nội, Nam Định có thể dẫn đến các vấn đề liên quan như cạn kiệt, sụt lún, xâm nhập mặn, ô nhiễm nguồn nước. Để có thể khai thác bền vững, cần phải xác định lượng bổ cập cho nước dưới đất. Đã có nhiều nghiên cứu đề cập đến các phương pháp ước tính lượng bổ cập nước dưới đất khác nhau, trong đó hiệu quả nhất là ứng dụng kỹ thuật đồng vị. Lấy mẫu nước phân tích thành phần hóa học, đồng vị bền* ^{18}O, ^{2}H, *và đồng vị phóng xạ* ^{3}H *đã được thực hiện trên toàn đồng bằng. Bằng cách sử dụng phân tích và giải đoán thành phần đồng vị trong nước, lượng bổ cập cho nước dưới đất đã được xác định biến đổi từ 77 đến 440 mm/năm.*

Từ khóa: Bổ cập nước dưới đất, đồng bằng sông Hồng, đồng vị bền ^{18}O, ^{2}H, đồng vị phóng xạ ^{3}H.

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