Origin of Groundwater in Hanoi, Vietnam, revealed by environmental isotopes

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In 2015 and 2016, groundwater samples were collected in Hanoi to analyse the isotopic composition (δ^2H, $\delta^{18}O$ and ^3H) and elucidate the relationship between groundwater and surface water, as well as the origin of the groundwater. The values for $\delta^{18}O$ and δ^2H indicate that the groundwater originated from evaporated meteoric water and the isotopic enrichment is due to the evaporation of shallow groundwater. Evaporation is the primary process affecting stable isotope signatures. Water samples collected from both Holocene and Pleistocene aquifers are more depleted in the heavy isotopes ^{18}O and ^2H than the rainfall in the area. This indicates that part of the groundwater is palaeo-groundwater or may be caused by the altitude effect due to recharge at a higher elevation. The results also show the close interaction between two granular aquifers and the Red River. Furthermore, the contribution of modern groundwater could be observed by the appearance of tritium in both aquifers. The presence of tritium indicates that originally tritium-free groundwater from the margins of the basin has been diluted by young water. The results of this study might help managers to evaluate the origin and reserves of groundwater more accurately.

Keywords: deuterium excess; groundwater origin; Hanoi; hydrogen-2; hydrogen-3; isotope hydrology; oxygen-18; Red River

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1. Introduction

The industrialisation and modernisation that have taken place in Vietnam in recent decades have affected groundwater resources, especially in Hanoi. In addition, population growth remains unabated. Hanoi is the second most crowded city in Vietnam, with a population of about 7.5 million [1]. This puts pressure on the water supply for the whole city, as the surface water is more polluted, and there is a high risk of groundwater depletion.

Several solutions have been proposed to solve the water supply issues in Hanoi, such as exploiting surface water or removing some water supply plants from the central area [2]. These methods are not sustainable unless the origins of groundwater resources

are clarified. To the best of the authors' knowledge, there are no studies that address this research gap. Bui [3] and Vu [4] made early attempts to explain the prospects for applying the isotopic technique to groundwater in Vietnam and the Red River Delta with only a few samples. The origin of deep water might be related to the recharge from mountainous areas [5]. Close to the Red River in south Hanoi, Dang et al. [6] showed that the groundwater could partly come from the Red River. In the southern part of Hanoi, groundwater might be older than 100 years [7]. Therefore, it was an urgent requirement to determine the origin of groundwater for the whole of Hanoi.

In general, water isotopes (^3H, δ^2H, $\delta^{18}O$) are often used as an effective method to solve multiple problems in hydrogeology, such as origin of water and contribution of other sources to groundwater [8]. This is because the changes in oxygen and hydrogen isotopic composition depend on temperature, latitude, longitude, and altitude and are controlled by evaporation and condensation processes [9,10]. In the global mean of precipitation samples, there is a linear relationship between $\delta^{18}O$ and δ^2H, which is defined as the global meteoric water line (GMWL) [9]. Empirically, the GMWL was originally defined as $\delta^2H = 8 \times \delta^{18}O + 10$ [9]. However, wind systems and climatic conditions can locally deviate the isotopic composition of precipitation, resulting in an individual local meteoric water line (LMWL). In this way, $\delta^{18}O$ and δ^2H can be identified in precipitation 'traces' of locally recharged groundwater and groundwater of different origin [9]. The $\delta^{18}O$ and δ^2H values beneath the GMWL and LMWL commonly indicate groundwater from evaporated meteoric water. Such additional information can also help to assess the interaction between groundwater and surface water and to estimate the percentage contribution of each source in groundwater [11]. In the case of two contributing components, the percentage of each source can be extracted using a simple binary mixing model [12–15].

Furthermore, the use of deuterium excess (d) might also help to explain the origin of groundwater. The parameter d is defined as $d = \delta^2H - 8 \times \delta^{18}O$ [10,16] and reflects the degree of kinetic isotope fractionation, as occurs, for example, during the evaporation of water from the oceans [10]. Thus, d is also affected by humidity when air masses move over continents and water - which has already fallen due to precipitation - is recycled into them. It shows the deviation of a given sample from the GMWL. The deuterium excess can vary significantly from region to region and over time.

Tritium (^3H or T), the radioactive isotope of hydrogen with a half-life of 12.32 years, is also often used to characterise 'young' groundwater [15]. Due to

atmospheric thermonuclear bomb tests, the tritium content in precipitation rose between 1950 and 1963 and had been decreasing since then. It might be applied to detect the recent recharge during the last 60 years [8,16]. Between 2003 and 2007, the tritium content in the precipitation of Hanoi was about 8 TU maximum [17]. Based on this tritium input pattern, groundwater younger than about 60 years cannot be detected or even dated [8].

Considering the above, the aim of this work was to determine the origin of groundwater in Hanoi using tritium, $\delta^{18}O$, and $\delta^{2}H$. The clarification of groundwater sources can help managers to make decisions regarding the sustainable water supply.

2. Materials and methods

2.1. Region of interest

Hanoi is the capital of Vietnam and is located in the northern part of the country (Figure 1). It is situated on the Red River Plain. With 7.5 million inhabitants in 2017, Hanoi is the second-largest city by population in Vietnam. There are two main types of terrain: the mountainous area located in the northern and western parts and the plain area. In general, the topography shows a gentle reduction in altitude from the north to the south and from the west to the east. In the plain, the average height ranges from 5 to 20 m above mean sea level (m amsl). The main river flowing through the city of Hanoi is the Red River, the biggest river in the northern part of Vietnam. It originates from the northern part of Yunnan, a province in southern China. The main branch of the Red River is about 1140 km long. In the Vietnamese part, it is called Cai, Thao or Hong River (Red River). The Red River has two main tributaries, the Da and the Lo (the Black and Clear River). Downstream it is named the Red River and flows eastward to Gulf of Tonkin.

Hanoi is characterised by a warm, humid, subtropical climate with heavy rainfall during the monsoon season. According to data collected between 1961 and 2010, the average precipitation is around 1,700 mm/year, and 84 % of the annual precipitation occurs between May and October. During the winter season, Hanoi receives less rainfall of around 25 mm/month. The mean annual temperature is 23.6 °C, and the relative humidity is more than 80 % for the period from 1961 to 2010 [18].

2.2. Hydrogeological setting

Two porous aquifers exist in the region of interest: the uppermost Holocene aquifer (qh) and the lower Pleistocene aquifer (qp). The upper aquifer is distributed widely throughout the city of Hanoi with clay and sandy layers. The lower part of the Holocene aquifer is made up of fine to coarse sand mixed with cobbles. The Holocene aquifer is unconfined and sufficient for small-scale water supply. Groundwater from this aquifer is exploited via private dug wells and shallow wells with small diameters [19,20].

An aquiclude, distributed discontinuously, separates the upper and lower aquifers. The two aquifers partially overlap without aquiclude, creating 'hydraulic windows'. The Pleistocene aquifer consists of sand mixed with cobbles and pebbles [19]. In the suburban areas far from the Red River, Kuroda et al. [21] postulated that the major sources for both aquifers were surface water bodies such as ponds and irrigated farmlands. Due to the hydraulic windows, the Pleistocene aquifer can be classified as semi-confined. Groundwater from this aquifer has a significant potential and has been used for the water supply of Hanoi since the beginning of the last century [2,19]. Figure 2 (modified according to [22,23]) shows a sketch of hydrogeology in the investigated region.

The other, deeper aquifers in this area are fissured Neogene and Triassic aquifers [2,19]. Sandy conglomerate, sandstone, siltstone, and gritstone are the main rock types of the Neogene aquifer. The Triassic aquifers are locally distributed. Their lithology consists mainly of sandstone, siltstone, and shale (in Muong Trai, Nam Tham, and Tan Lac formations); sandstone and siltstone (Na Khuat formation); and limestone (Dong Giao formation) [2]. Only a small amount of groundwater from these aquifers is exploited for the water supply because the aquifers are not widely distributed.

2.3. Sampling and analysis

Groundwater samples were collected from the two upper aquifers, which date from the Holocene and Pleistocene, and from two lower aquifers (Neogene and Triassic aquifers). They were collected in September 2015 and from March to April 2016. In the first campaign in 2015, 28 samples were taken for $\delta^{18}O$ and $\delta^{2}H$ analysis (six samples from the Holocene aquifer, 19 samples from the Pleistocene aquifer, and three samples from lower aquifers), and 24 samples were taken for tritium analysis (six samples from the Holocene aquifer, 16 samples from the Pleistocene aquifer, and three samples from

lower aquifers).

In 2016, 56 samples were taken for $\delta^{18}O$ and $\delta^{2}H$ (15 samples from the Holocene aquifer, 36 samples from the Pleistocene aquifer, and five samples from lower aquifers), and 45 samples were taken for tritium analysis (15 samples from the Holocene aquifer, 25 samples from the Pleistocene aquifer, and five samples from lower aquifers). All locations from which samples were taken were either part of our groundwater monitoring network of the Red River Delta or wells in the water supply system of Hanoi (Figure 1). The groundwater samples were collected during pumping. At monitoring wells, the wells were purged with a pump after recording the water level. In general, three well volumes were pumped before samples were collected. In the field, the in-situ parameters of electrical conductivity (EC), water temperature (T), redox potential (Eh), and pH value were measured.

Water samples for the analysis of $\delta^{2}H$ and $\delta^{18}O$ were filled into 10-ml high-density polyethylene (HDPE) bottles, while 500-ml HDPE bottles were used for tritium samples. The bottles were pre-washed with water from the well inspected and then tightly sealed to protect the samples from exchange with atmospheric water vapour. The samples were stored in a cool box and later in a refrigerator at 4°C before being transported to the laboratory. All samples were analysed at the Helmholtz Centre for Environmental Research, UFZ, Germany. The values from $\delta^{18}O$ and $\delta^{2}H$ were determined for the first batch by high-temperature pyrolysis (Hekatech HTC with Thermo Conflow III to a Thermo Delta XLIRMS) and later with a laser-based system (Picarro L2120-i). Cross-check measurements with distilled water yield consistent results of both systems, HTC and laser, within uncertainties. Due to re-organisation of the lab we did not compare the systems with ‘real’ samples afterwards. The tritium concentration was measured via electrolytic enrichment (self-made system with 400 mL cell size; enrichment factor of about 20) and liquid scintillation counting (Quantulus 1220).

Stable isotopic compositions are indicated in δ units, as the deviations in per mil (‰) with respect to the Vienna Standard Mean Ocean Water (VSMOW), [8] and with 2σ uncertainty ranges of 0.25 ‰ ($\delta^{18}O$) and 0.8 ‰ ($\delta^{2}H$), respectively. Tritium values are expressed in tritium units (TU), where 1 TU corresponds to a frequency of one ^{3}H per 10^{18} hydrogen atoms [24].

3. Results and discussion

3.1. Stable isotopes in precipitation

Figure 3 shows the $\delta^{18}O$ and $\delta^{2}H$ values of the monthly precipitation samples collected in Hanoi between 2004 and 2007 [17]. Equation (1) represents the LMWL for Hanoi.

$$\delta\ ^{2}H = (7.9\ \pm 0.2) \times \delta^{18}O + (12.5\ \pm 1.6)\ ‰\ (R^2=0.98, n=38) \qquad (1)$$

Obviously the slope of the LMWL for Hanoi is nearly the same as the slope of the GMWL, so the two lines are almost parallel to the LMWL above the GMWL. Seasonal oscillations of stable isotopes are often observed as a result of changes in temperature and precipitation [25]. In general, the low values of stable isotopes in precipitation relate to the cold seasons, while higher values reflect the warm seasons [8]. However, stable isotopes in precipitation in Hanoi show enrichment in cold seasons and depletion in warm seasons (Figure 4c), similar to precipitation in Hong Kong and Bangkok [26,27]. Figure 4b,d shows that the low $\delta^{18}O$ values are not only due to the low temperatures but also to the high precipitation intensity due to the monsoon season. Hence, it seems most likely that the $\delta^{18}O$ values are caused by prior rainfall of the monsoonal clouds.

Figure 4a shows a significant inverse relationship between $\delta^{18}O$ and temperature. It indicates that stable isotope values decrease with increasing temperature (Spearman correlation $r = –0.751$; $p < 0.001$). This is in complete contrast to the general trend that rainwater becomes heavier at higher ambient temperatures, both globally and seasonally (Figure 4d). Figure 4b demonstrates the significant inverse correlation between $\delta^{18}O$ and precipitation at the Hanoi station (Spearman correlation $r = –0.811$; $p < 0.001$), showing that monsoon rain is characterised by slightly more negative stable isotope values. This must be carefully considered when discussing recharge. The data in the upper left corner refer to the dry seasons and point to the lower right corner to correspond to the warm seasons (Figure 4a–d). The annual distribution of $\delta^{18}O$ and $\delta^{2}H$ in Hanoi's precipitation shows more positive values in the dry seasons from October to April, as well as depleted values during the warm rainy season from May to September (Monsoon) (Figure 4c,d).

3.2. Stable isotopes in river water

Results from surface water samples taken from the Red River in Hanoi during the period 2003 to 2007 are given in [28]. The variations of $\delta^{18}O$ and $\delta^{2}H$ in the Red River water, ranging between –11.4 and –7.0 ‰ for $\delta^{18}O$ and from –71.1 to –43.7 ‰ for $\delta^{2}H$, are depicted in Figure 3 as surface water line (Hanoi). The mean values for $\delta^{18}O$ and $\delta^{2}H$ of the Red River are –8.7 and –58.9 ‰, respectively, as also shown in Figure 3. Applying a linear regression model, the relationship between $\delta^{2}H$ and $\delta^{18}O$ can be expressed as Equation (2).

$$\delta^{2}H = (5.6 \pm 0.4) \times \delta^{18}O - (9.1 \pm 3.7) \text{ ‰} \quad (R^2=0.79, n=50) \qquad (2)$$

In general, surface water reflects the isotopic composition of local meteoric water in the recharge areas. The slope for the Red River water regression line is less than that of the LMWL and closer to a typical evaporation line. The Red River has a catchment area of around 140,000 km^2. Kunming (in the Yunnan province in southern China), where the Red River originates, is situated in a mountainous area with high altitudes (up to 2400 m amsl) [29] and low temperatures (around 15.5 °C on average) [30]. The stable isotopes values in precipitation at Kunming station vary over –17.5 to +0.4 ‰ for $\delta^{18}O$ and –114.2 to +7.8 ‰ for $\delta^{2}H$ [30], with mean values of –7.8 and –54.2 ‰ for $\delta^{18}O$ and $\delta^{2}H$, respectively. Due to the altitude effect, the stable isotope values of the water from the mountains are more negative compared to waters from lower altitudes. The mean stable isotopes values in precipitation at Hanoi station are more positive than those at Kunming. Figure 3 shows the comparison of the isotopic compositions of surface water and precipitation in Hanoi. Mean values of stable isotopes in surface water of the Red River in Hanoi are between two weighted means of stable isotopes in precipitation in Hanoi and Kunming. This suggests that the isotopic compositions in the Red River in Hanoi depend much on the isotopic compositions in precipitation in Kunming (origin of the Red River) and Hanoi (local contributions). They also depend on the evaporation of river water and mixing with local rainfalls along the Red River from Kunming to Hanoi.

3.3. Stable isotopes and tritium in groundwater

The stable isotope and tritium data for groundwater in Hanoi are presented in Online Appendix 1. Table 1 presents basic statistics for stable isotopes and tritium in

precipitation, river water, and groundwater.

3.4. Origin of groundwater

The differences in $\delta^{18}O$ and $\delta^{2}H$ in groundwater in Hanoi are shown in Figure 5. The LMWL defined by Equation (1), surface water line described by Equation (2), and weighted means by the amount of monthly rainfall of stable isotopes in precipitation in Hanoi and Kunming are shown in the graph. Most groundwater values plot below the LMWL and a few above or on the surface water line. The groundwater lines of the Holocene and Pleistocene aquifers have a slope of around seven. These lines nearly cross the weighted mean value of precipitation at Kunming station. It supports the idea that the groundwater originates from the higher regions in the north-west of Hanoi and is mixed in the lower plain with local groundwater. Some of the groundwater samples are more enriched in heavier stable isotopes than the water of the Red River. It is likely that surface water is enriched by evaporation and infiltrates into the unsaturated zone to contribute to recharge. In the west of Hanoi there is a reservoir (Hoa Binh reservoir) where the surface water evaporates and there is likely a surface water intake and transport underground below the dam. Thus, the isotopic composition might be influenced by the high elevation of origin (depletion) and evaporation effects (enrichment). Dang et al [6] showed that groundwater in the southern part was recharged by local meteoric water.

In Figure 5, several groundwater values in the lower left corner are located below the surface water line (in rectangle). These wells in the Pleistocene aquifer are close to the Red River and are part of the water supply system (dash-circle in Figure 1). The *t*-test shows that there is a statistically significant difference between mean $\delta^{18}O$ values in those wells (mean = –7.8 ‰, SD = 1.1) and the other locations (mean = -6.6 ‰, SD = 1.3); $t(82) = 3.6$, $p < 0.05$). If it is considered that those wells close to the Red River are in a separated group, the source of groundwater for those wells is different from the other locations which plot between the surface water line and LMWL. Those wells close to the Red River and in the water supply system receive more water from the Red River than other locations. This suggests a hydraulic connection between the Pleistocene aquifer and the surface water. Correspondingly, Dang et al. [6] showed that for those wells close to the Red River in the south of Hanoi, a significant part of groundwater was from the Red River.

In many areas, stable isotopes in groundwater tend to have rather constant values without seasonal changes. Hanoi groundwater is behaving similarly. In the Holocene aquifer, the *t*-test confirms the lack of seasonal changes because there is no statistically significant difference between mean values of $\delta^{18}O$ in August 2015 (rainy season) (mean = –5.6 ‰, SD = 0.9) and in March/April 2016 (mean = –6.4 ‰, SD = 1.2); t(19) = 1.5, p > 0.05, and between $\delta^{2}H$ in 2015 (mean = –39.4 ‰, SD = 6.1); and in 2016 (dry season) (mean = –43.8 ‰, SD = 9.0); t(19) = 1.3, p > 0.05. For the Pleistocene aquifer, the null hypothesis is accepted because there is no statistically significant difference between mean values of $\delta^{18}O$ (mean = –7 ‰, SD = 1.4 in 2015, mean = –7.3 ‰, SD = 1.3 in 2016; t(53) = 0.8, p > 0.05) and $\delta^{2}H$ (mean = –48.6 ‰, SD = 9.3 in 2015, mean = –50.2 ‰, SD = 8.7 in 2016; t(53) = 0.6, p > 0.05) in 2015 and 2016. Though the samples were collected in only two seasons, the seasonal variation of stable isotopes in groundwater in Hanoi is suggested to not be significant.

Based on the assumption that groundwater originates from precipitation and river water and applying a mass balance analysis for the oxygen and hydrogen isotope values, the percentages of groundwater sources can be estimated [9,12,13]. The mixing between two distinct sources can be quantified by a simple linear algebraic Equation (3) [12,13].

$$C \times (V_A + V_B) = A \times V_A + B \times V_B \quad (3)$$

with A: isotope value in precipitation; B: isotope value in surface water; C: isotope value in groundwater; V_A: amount of precipitation; V_B: amount of river water. Therefore,

$$C = A \times (1 - x) + B \times x; 0 \leq x \leq 1 \quad (4)$$

and,

$$x = \frac{C-A}{B-A} \quad (5)$$

with x: recharge share of river water; (1-x): recharge share of precipitation. The amounts of river water and rainfall in groundwater were estimated based on the mean values of $\delta^{18}O$ in groundwater, precipitation, and surface water. The results show that the primary source for both aquifers is river water. Both aquifers receive more than

50 % from precipitation. The Pleistocene aquifer receives a slightly higher amount of river water than the Holocene aquifer (46.7 and 16.7 %, respectively) (Table 2). This result might be explained by the discontinuous distribution of the upper aquifer, while the lower aquifer is distributed over the whole area. Thus, the lower aquifer is a unified body. The Holocene aquifer might receive a higher percentage in areas close to the Red River, but the percentage might be lower than for the Pleistocene aquifer overall. The proportion also depends on the distance to the Red River [7]. The results also show that the Holocene aquifer receives a higher amount of rainfall than the Pleistocene does (83.3 versus 53.3 %, respectively). This also suggests that the rain water in the lower aquifer is more diluted by other sources. The relationship between the Pleistocene aquifer and the Red River can also be expressed by the relationship of stable isotopes in groundwater and in the Red River. Figure 6 shows the distribution of $\delta^{18}O$ in the Pleistocene aquifer in 2016, with more depleted $\delta^{18}O$ values towards the Red River. The $\delta^{18}O$ values in the Pleistocene aquifer are clearly impacted by the $\delta^{18}O$ values of the Red River, as shown before. Certain areas in the west and south of Hanoi which are far from the Red River show more positive values of $\delta^{2}H$ and $\delta^{18}O$, which is an indicator of recharge through rainwater that has experienced significant evaporation. In this area, the Tritium content is also higher than normal values (see below). Hence, comparably old water is a possible source for that groundwater.

3.5. Tritium

The occurrence of tritium indicates that groundwater was recharged by young water which recharged within the last ca. 60 years. Tritium in the atmosphere peaked in the Hanoi area in 1963 at 278 TU [3] and then declined slowly, as observed around the world. Nowadays, the mean of tritium in precipitation in Hanoi area can be estimated to be less than around at 2.5 TU, an average value in Hong Kong in 2009 according to [31]. Trinh et al. [7] showed that groundwater in Hanoi was recharged by young water based on high contents of tritium in groundwater (up to 5.8 TU) [7].

Tritium was detected in 44 out of a total of 70 samples from two aquifers with a detection limit of 0.5 TU (Online Appendix 1). In the Holocene aquifer, tritium was detected in 86 % of wells (18/21). In the Pleistocene aquifer, tritium was detected in 46 % (19/41). An aquiclude between aquifers distributing discontinuously [19,20] might cause this difference. Because the Pleistocene aquifer is partly confined, groundwater

recharge needs more time to reach it. However, tritium in the Pleistocene aquifer, varying from 0.6 to 4.7 TU, seems to be higher than tritium in the Holocene aquifer, ranging between 0.7 and 2.4 TU. The supporting information would be the higher contributions of water from the bomb peak era (ca. 60 years ago) in the Pleistocene aquifer, and younger water in the Holocene aquifer make it plausible. The mean values of tritium were 1.4 and 1.6 TU in the Holocene and Pleistocene aquifers, respectively. The difference between tritium in both aquifers was not significant (0.2 TU). Compared to tritium in river water and rainfall in the period of 2003–2007 [17,28], tritium content in both aquifers varies over only a small range (Figure 7a). The mean values of tritium in river water and rainfall are higher than those in Holocene and Pleistocene aquifers. Figure 7b also shows the concentration of tritium in the Pleistocene aquifer in 2016.

There are two areas with slightly elevated tritium values. The first one, in the north-east, is close to the Red River and a large lake (West Lake). Elevated tritium concentrations close to the Red River correlate with lower $\delta^{18}O$ values and are thus assumed as evidence for direct riverbank filtration. Another area with elevated tritium values is in the south-west of Hanoi where, in 2009–2010, Dang et al. [6] found tritium levels of up to 2.1 TU. Figure 7b shows a peak in the south of the region of interest which correlates with higher $\delta^{18}O$ values in this area (Figure 6). In general, higher tritium values indicate young water that is roughly between 10 and 60 years old, i.e. with contributions of higher amounts of so-called bomb tritium. The source of the elevated tritium content in the south may be surface water recharging quickly via poorly constructed wells. According to the inverse seasonal correlation between temperature and $\delta^{18}O$ because of the monsoon climate in this area (Figure 4), this inverse relationship is evidence for an area of low recharge rate. In other words, groundwater was not diluted so much and, thus, tritium has not declined too much, besides radioactive decay. In this case, tritium might relate to water from the 1960s. In addition, Trinh et al. [7] showed that groundwater might be old in the southern part of Hanoi [7]. Furthermore, elevated tritium values in this area show the highest $\delta^{18}O$ values combined with the lowest deuterium excess (at HP07, HP07-2, HP08, HP08-2, with *d* from 3.3-3.7). This combination commonly indicates evaporation. Actual evapotranspiration (AE) was calculated at around 750 mm/year [32]. AE was especially high in the south-west region of Hanoi, with an average of 2.3 mm/day (in May 2007) [32]. Figure 8 shows a cross-section SW–NE from the south-west to the north-east of the study area.

Tritium was detected in all wells of the Holocene aquifer in both seasons. In the Pleistocene aquifer, tritium was not detected in the first two wells of the cross-section, which are close to bedrock areas. This supports that groundwater is older than at least ca. 35 years. Tritium was detected from the third well in both campaigns. Based on groundwater pressure head, the groundwater flow direction can be determined. Groundwater flows from the bedrock area towards the centre and from the Red River towards the centre. The detection of tritium in the central part (close to the right side in the cross section) and the predicted flow line suggest that the central part may receive more water from recharging of the upper aquifer and surface water (from the Red River).

3.6. Deuterium excess

The calculated deuterium excess *d* of the rainfall varies in a range from 5.4 to 19.2 ‰, with an average of 12.9 ‰. In river water, the range is between 3.1 and 20.6 ‰, with an average of 11.5 ‰. The value of *d* in groundwater in both aquifers varies over a narrow range from 3.7 to 9.7 ‰, with an average of 6.7 ‰ in the Holocene aquifer. In the Pleistocene aquifer, *d* ranges between 3.3 and 10.1 ‰, with an average of 7.7 ‰. Groundwater samples show that *d* is lower than those of rainfall and surface water in the investigated area. This is evidence that a considerable amount of groundwater enters the area through the northern margin of the area. The *d* might be used to identify the vapour sources. [33,34]. The relative humidity is a major controlling factor of *d*. Lower relative humidity relates to higher *d* in precipitation and vice versa [34,35]. The wide range of *d* in rainfall and river water is a result of a long-distance travelling of moisture before falling as precipitation [33]. It might be due to the rainfall originating from the moisture of different regions or due to the exchange processes of moisture and surface water. Finally, in the case of passing air with low humidity, the evaporation modifies the δ^2H and δ^{18}O values in precipitation during rainfall [35].

$$d = (-1.15 \pm 0.1) \times \delta^{18}O + (0.5 \pm 0.6)\ ‰ \text{ (R2=0.73, n=76)} \qquad (6)$$

Comparing *d* to δ^{18}O in groundwater, there is a significant inverse relationship between them (Equation (6)). The negative relation between *d* and δ^{18}O in groundwater shows that the values of δ^{18}O increase gradually with a decrease in *d* (Figure 9). This supports the idea that evaporation is the main process affecting the isotopic signatures of oxygen

and hydrogen [36]. It also indicates that the current groundwater mixes with water from different rainfall events with different δ^2H and $\delta^{18}O$ values. Evidence of mixing of groundwater with rainfall is seen in the increase of *d* from the groundwater values towards the rainfall values.

3.7. Depth dependence of stable isotopes and tritium

Figure 10 shows the distribution of $\delta^{18}O$, δ^2H, and tritium values versus the depth of the Holocene and Pleistocene aquifers and deeper groundwater (Neogene and Triassic aquifers). In general, the stable isotopes and tritium correlate inversely with depth, especially for the Holocene and Pleistocene aquifers. In only two wells, which are in the south-west region of Hanoi, the stable isotopes are more enriched at a depth of more than 120 m. Tritium values in those wells are low, which supports that the groundwater in those wells of the lower aquifers may be older.

4. Conclusion

Isotope investigations of groundwater in Hanoi yielded insights concerning the origin of the groundwater. Based on the distributions and characteristics of δ^2H, $\delta^{18}O$, and tritium, and comparing them to GMWL and LMWL, the main findings are as summarised below.

- The $\delta^{18}O$ and δ^2H values in groundwater range from –8.8 to –4.0 ‰, and –61.0 to 28.5 ‰, respectively. Most groundwater samples plot below the LMWL and GMWL. This suggests that the groundwater originates at least partially from evaporated meteoric water.
- Both the Holocene and Pleistocene aquifers have partly hydraulic contact with the Red River. Some areas receive up to almost 60 % of their water from the Red River by riverbank filtration.
- Elevated tritium concentration in the vicinity of the Red River is close to today's atmospheric value and thus an indication of a dominant share of young water and riverbank filtration. On the contrary, the elevated concentration in the south of the region of interest may be seen as an indicator of groundwater from the 1960s, when tritium peaked in the atmosphere.

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Table 1. Statistical summary of $\delta^{18}O$, $\delta^{2}H$, and tritium in precipitation, surface water, and groundwater in Hanoi. Numbers (n) of samples, which were used to calculate mean values, are shown beside the mean values.

Types	**$\delta^{18}O$ (‰VSMOW)**			**$\delta^{2}H$ (‰VSMOW)**			**tritium (TU)**		
	Min	Mean (n)	Max	Min	Mean (n)	Max	Min	Mean (n)	Max
Precipitation (IAEA 2007a)	–14.1	–5.7 (38)	–0.9	–93.7	–33.0 (38)	6.8	0.5	3.6 (43)	7.8
River water (IAEA 2007b)	–11.4	–8.7 (50)	–7.0	–71.1	–58.9 (50)	–43.7	1.3	3.9 (21)	8.6
Holocene aquifer 2015	–6.5	–5.6 (6)	–4.2	–45.6	–39.3 (6)	–29.6	<0.5	1.2 (6)	1.6
Holocene aquifer 2016	–8.8	–6.4 (15)	–4.2	–60.8	–43.9 (15)	–29.1	<0.5	1.4 (15)	2.4
Pleistocene aquifer 2015	–8.8	–7.0 (19)	–4.1	–60.8	–48.6 (19)	–29.3	<0.5	2.0 (16)	4.7
Pleistocene aquifer 2016	–8.8	–7.3 (36)	–4.0	–61.0	–50.2 (36)	–28.5	<0.5	1.5 (25)	3.8

Table 2. Percentage rate that surface water and precipitation distributed to groundwater estimated from weighted means of stable isotopes. The means were weighted by precipitation (A: stable isotope value in precipitation; B: stable isotope value in surface water; C: stable isotope value in groundwater; x: recharge share of river water; (1–x): recharge share of precipitation).

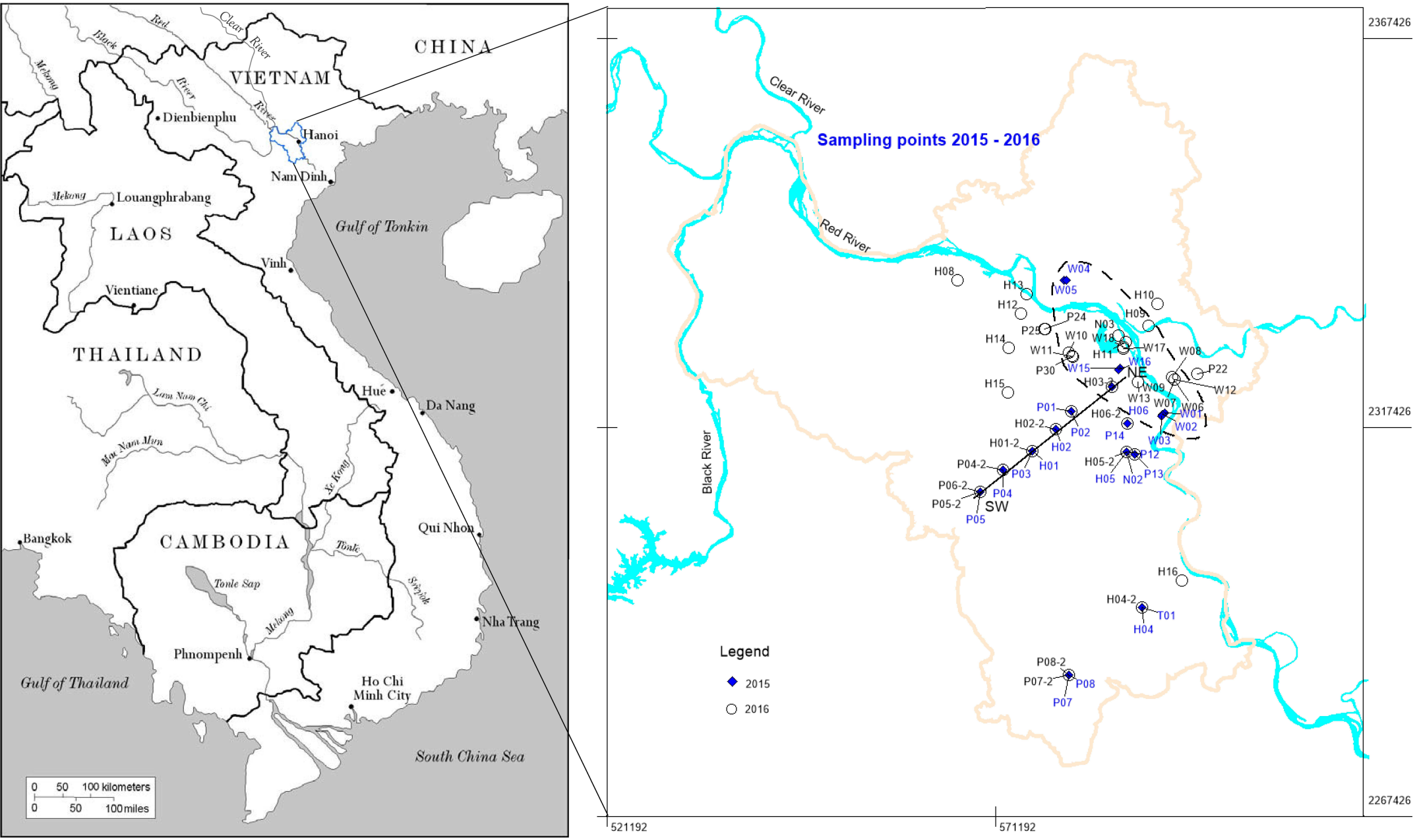
	Holocene aquifer	Pleistocene aquifer
C	–6.2	–7.1
A	–5.7	–5.7
B	–8.7	–8.7
x (%) (from river water)	16.7	46.7
1–x (%) (from precipitation)	83.3	53.3

Appendix 1. Results of $\delta^{18}O$, $\delta^{2}H$, and tritium in groundwater in Hanoi

IDs	Sampling date	Aquifers	$\delta^{18}O$ [‰$_{VSMOW}$]	$\delta^{2}H$ [‰$_{VSMOW}$]	^{3}H [TU]	± 2σ [TU]	*d* [‰$_{VSMOW}$]
H01	9/11/2015	Holocene	-6.03	-42.7	1.6	0.5	5.5
H01-2	3/22/2016	Holocene	-6.07	-42.7	1.7	0.5	5.9
H02	9/11/2015	Holocene	-5.17	-34.6	0.7	0.5	6.8
H02-2	3/22/2016	Holocene	-5.00	-32.7	0.9	0.5	7.3
H03	9/24/2015	Holocene	-6.35	-45.6	1.5	0.5	5.2
H03-2	4/22/2016	Holocene	-6.93	-48.0	1.4	0.5	7.4
H04	9/24/2015	Holocene	-4.16	-29.6	< 0.5		3.7
H04-2	4/13/2016	Holocene	-4.17	-29.1	< 0,5		4.3
H05	9/24/2015	Holocene	-5.64	-40.2	1.4	0.5	4.9
H05-2	4/22/2016	Holocene	-5.68	-40.5	1.5	0.5	4.9
H06	9/24/2015	Holocene	-6.47	-43.4	0.8	0.5	8.4
H06-2	4/22/2016	Holocene	-6.31	-42.8	0.9	0.5	7.7
H08	3/21/2016	Holocene	-8.04	-57.0	1.4	0.5	7.3
H09	3/23/2016	Holocene	-8.05	-57.3	1.7	0.5	7.1
H10	3/23/2016	Holocene	-5.40	-35.6	1.5	0.5	7.6
H11	3/25/2016	Holocene	-8.81	-60.8	2.4	0.5	9.7
H12	3/25/2016	Holocene	-6.62	-44.6	1.7	0.5	8.4
H13	3/25/2016	Holocene	-6.62	-44.7	< 0,5		8.3
H14	3/25/2016	Holocene	-6.56	-42.9	1.0	0.5	9.6
H15	3/25/2016	Holocene	-5.18	-37.3	1.5	0.5	4.1
H16	4/13/2016	Holocene	-6.04	-41.3	1.2	0.5	7.0
P01	9/11/2015	Pleistocene	-7.16	-50.4			6.9
P01-2	3/22/2016	Pleistocene	-6.58	-45.6	1.2	0.5	7.0
P02	9/11/2015	Pleistocene	-6.73	-47.3	1.3	0.5	6.6
P02-2	3/22/2016	Pleistocene	-6.51	-45.2	1.0	0.5	6.9
P03	9/11/2015	Pleistocene	-6.67	-47.3	1.0	0.5	6.1
P03-2	3/22/2016	Pleistocene	-6.71	-46.8	1.2	0.5	6.9
P04	9/11/2015	Pleistocene	-8.69	-59.9	< 0.5		9.6
P04-2	3/22/2016	Pleistocene	-8.75	-59.9	< 0,5		10.1
P05	9/11/2015	Pleistocene	-6.49	-43.3	< 0.5		8.6
P05-2	3/22/2016	Pleistocene	-6.41	-42.8	< 0,5		8.5
P06	9/11/2015	Pleistocene	-5.12	-35.4	< 0.5		5.6
P06-2	3/22/2016	Pleistocene	-5.10	-35.2	< 0,5		5.6
P07	9/11/2015	Pleistocene	-4.08	-29.3	2.7	0.5	3.3
P07-2	3/22/2016	Pleistocene	-4.04	-28.5	2.7	0.5	3.8
P08	9/11/2015	Pleistocene	-4.96	-36.2	4.7	0.6	3.5
P08-2	3/22/2016	Pleistocene	-4.53	-32.5	3.8	0.5	3.7

P09-2	3/22/2016	Pleistocene	-5.62	-39.1	0.9	0.5	5.9
P10	9/24/2015	Pleistocene	-6.41	-44.4	1.0	0.5	6.8
P10-2	4/22/2016	Pleistocene	-6.62	-45.4	1.0	0.5	7.6
P11-2	4/13/2016	Pleistocene	-7.65	-51.6	< 0,5		9.6
P12	9/24/2015	Pleistocene	-6.81	-46.8	< 0.5		7.7
P12-2	4/22/2016	Pleistocene	-6.66	-46.4	< 0,5		6.9
P13	9/24/2015	Pleistocene	-8.04	-55.5	< 0.5		8.8
P13-2	4/22/2016	Pleistocene	-7.86	-54.4	< 0,5		8.5
P14	9/24/2015	Pleistocene	-8.80	-60.8	1.5	0.5	9.6
P14-2	4/22/2016	Pleistocene	-8.79	-60.4	1.7	0.5	9.9
P20	3/21/2016	Pleistocene	-8.50	-60.0	0.7	0.5	8.0
P21	3/23/2016	Pleistocene	-8.38	-58.3	< 0,5		8.7
P22	3/23/2016	Pleistocene	-6.97	-47.6	< 0,5		8.2
P23	3/25/2016	Pleistocene	-8.70	-59.9	2.0	0.5	9.7
P24	3/25/2016	Pleistocene	-6.07	-40.8	< 0,5		7.8
P25	3/25/2016	Pleistocene	-8.46	-57.8	1.9	0.5	9.9
P26	3/25/2016	Pleistocene	-5.93	-40.7	1.3	0.5	6.7
P27	3/25/2016	Pleistocene	-7.14	-49.0	< 0,5		8.1
P28	3/25/2016	Pleistocene	-7.12	-49.1	0.6	0.5	7.9
P29	4/13/2016	Pleistocene	-7.60	-51.8	< 0,5		9.0
P30	4/22/2016	Pleistocene	-7.02	-48.2	< 0,5		8.0
W04	9/24/2015	Pleistocene	-6.52	-46.1	< 0.7		6.1
W06	4/21/2016	Pleistocene	-8.80	-60.6			9.8
W07	4/21/2016	Pleistocene	-8.56	-59.3			9.2
W05	9/24/2015	Pleistocene	-6.83	-47.7	< 0.5		7.0
W08	2/24/2016	Pleistocene	-8.71	-60.5			9.2
W09	2/24/2016	Pleistocene	-8.74	-60.2			9.7
W10	2/18/2016	Pleistocene	-6.87	-48.0			7.0
W11	2/18/2016	Pleistocene	-6.09	-42.5			6.2
W12	2/3/2016	Pleistocene	-8.82	-61.0			9.6
W01	9/24/2015	Pleistocene	-8.46	-59.4	1.8	0.5	8.4
W02	9/24/2015	Pleistocene	-7.88	-55.9	1.5	0.5	7.1
W03	9/24/2015	Pleistocene	-8.67	-60.2	2.2	0.5	9.2
W13	2/17/2016	Pleistocene	-6.66	-47.1			6.2
W14	4/3/2016	Pleistocene	-6.23	-44.5			5.3
W15	11/24/2015	Pleistocene	-8.22	-56.5			9.3
W16	11/24/2015	Pleistocene	-8.01	-56.7			7.4
W17	3/16/2016	Pleistocene	-8.50	-58.4			9.6
W18	3/16/2016	Pleistocene	-8.30	-56.5			9.9
T01	9/24/2015	Triassic	-8.82	-61.2	< 0.5		9.3

T01-2	4/13/2016	Triassic	-8.69	-61.0	< 0,5		8.5
N01	9/24/2015	Neogene	-8.78	-62.0	< 0.5		8.2
N02	9/24/2015	Neogene	-5.60	-39.0	0.8	0.5	5.8
N02-2	4/22/2016	Neogene	-5.64	-39.4	0.6	0.5	5.7
N03	3/25/2016	Neogene	-8.31	-56.4	1.7	0.5	10.1
N04	3/25/2016	Neogene	-8.61	-59.4	< 0,5		9.5
N05	4/13/2016	Neogene	-8.17	-57.3	< 0,5		8.1


CHINA
VIETNAM
LAOS
THAILAND
CAMBODIA
Gulf of Tonkin
Gulf of Thailand
South China Sea
Red River
Black River
Clear River
Mekong
Dienbienphu
Hanoi
Nam Dinh
Louangphrabang
Vinh
Vientiane
Hué
Da Nang
Qui Nhon
Nha Trang
Bangkok
Phnompenh
Ho Chi Minh City
Tonle Sap
Lam Nam Chi
Mae Nam Mun
Se Kong
Tonle
Sesipik
0 50 100 kilometers
0 50 100 miles
Sampling points 2015 - 2016
Legend
2015
2016
2367426
2317426
2267426
521192
571192
NE
SW
H08
H13
H12
H14
H15
H10
H09
H11
H16
W04
W05
W10
W11
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T01
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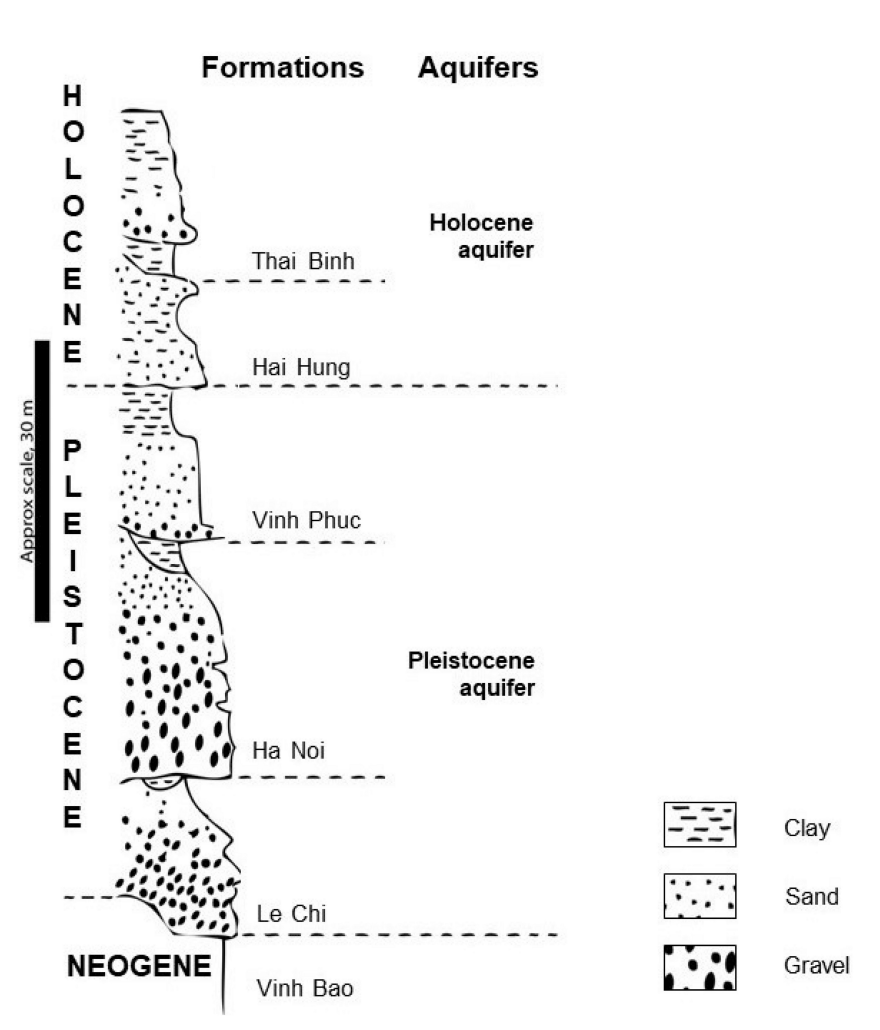
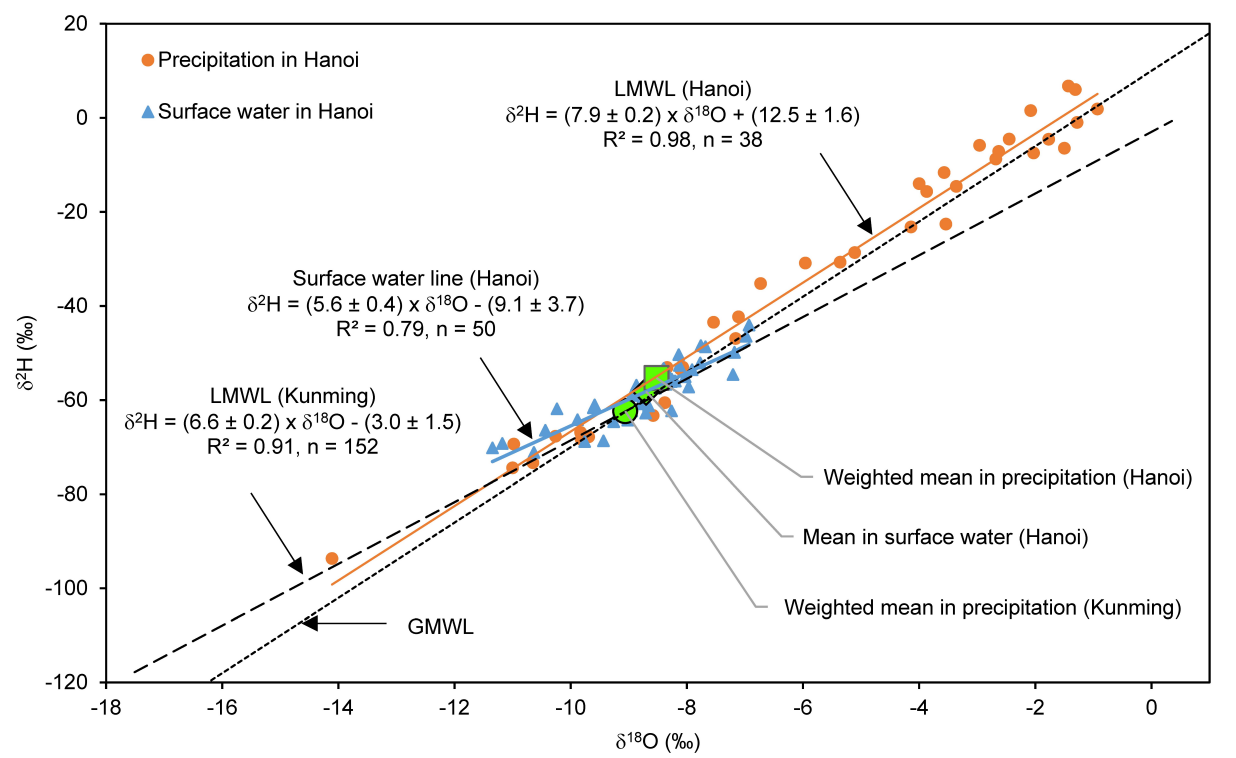
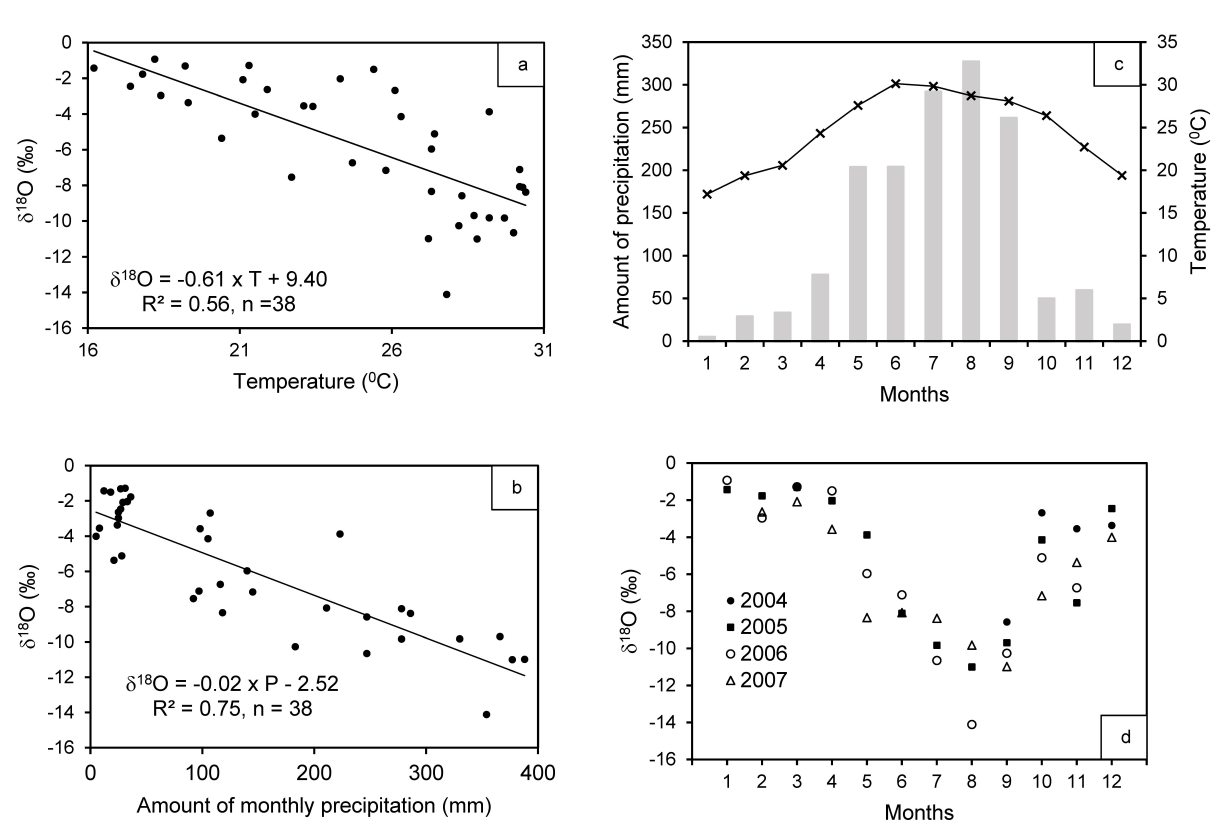
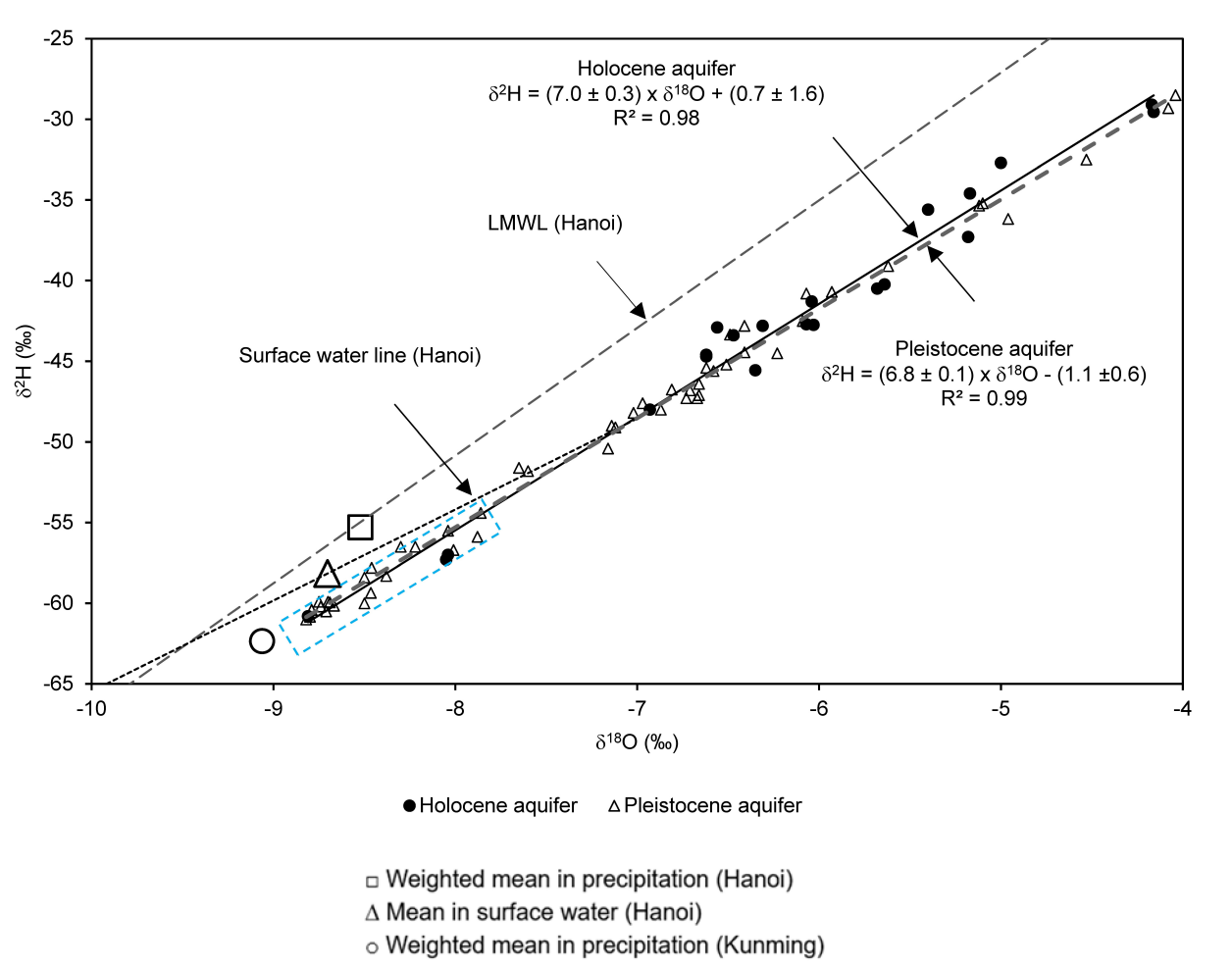
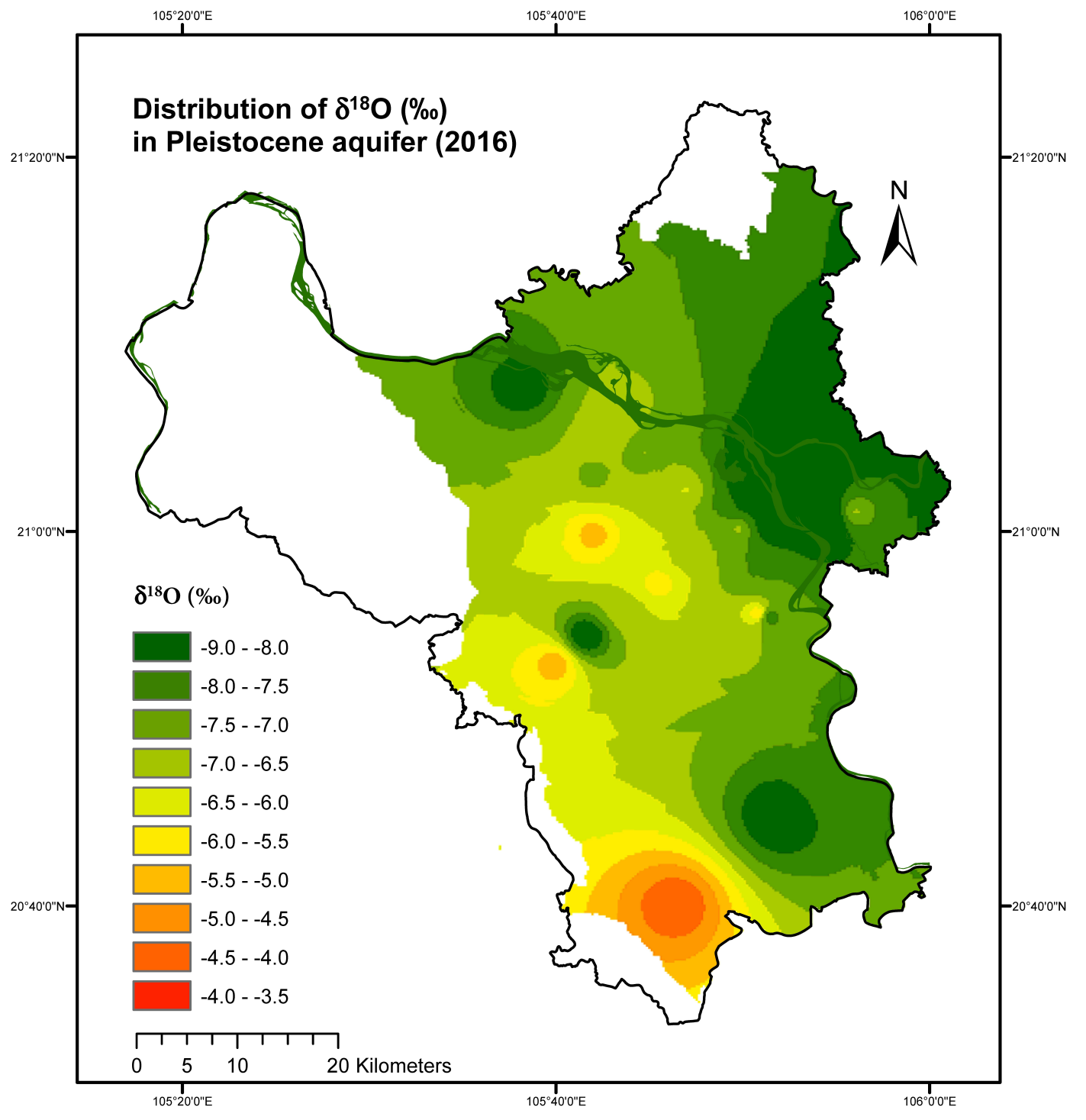
P07-2
P08
P07

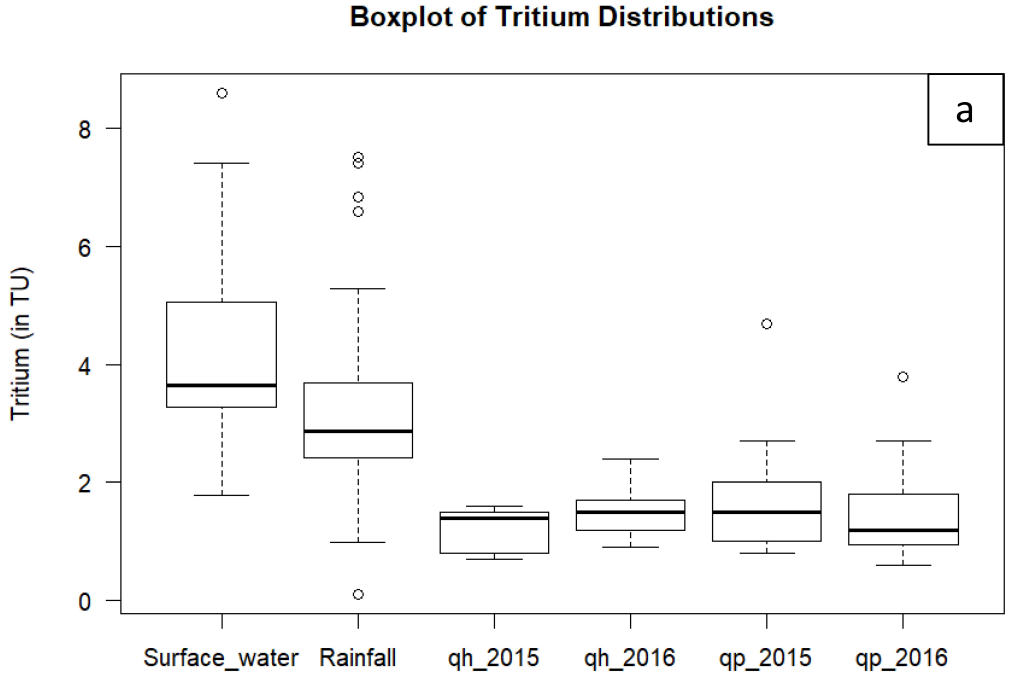

Formations
Aquifers
HOLOCENE
PLEISTOCENE
NEOGENE
Approx scale, 30 m
Holocene aquifer
Thai Binh
Hai Hung
Vinh Phuc
Pleistocene aquifer
Ha Noi
Le Chi
Vinh Bao
Clay
Sand
Gravel

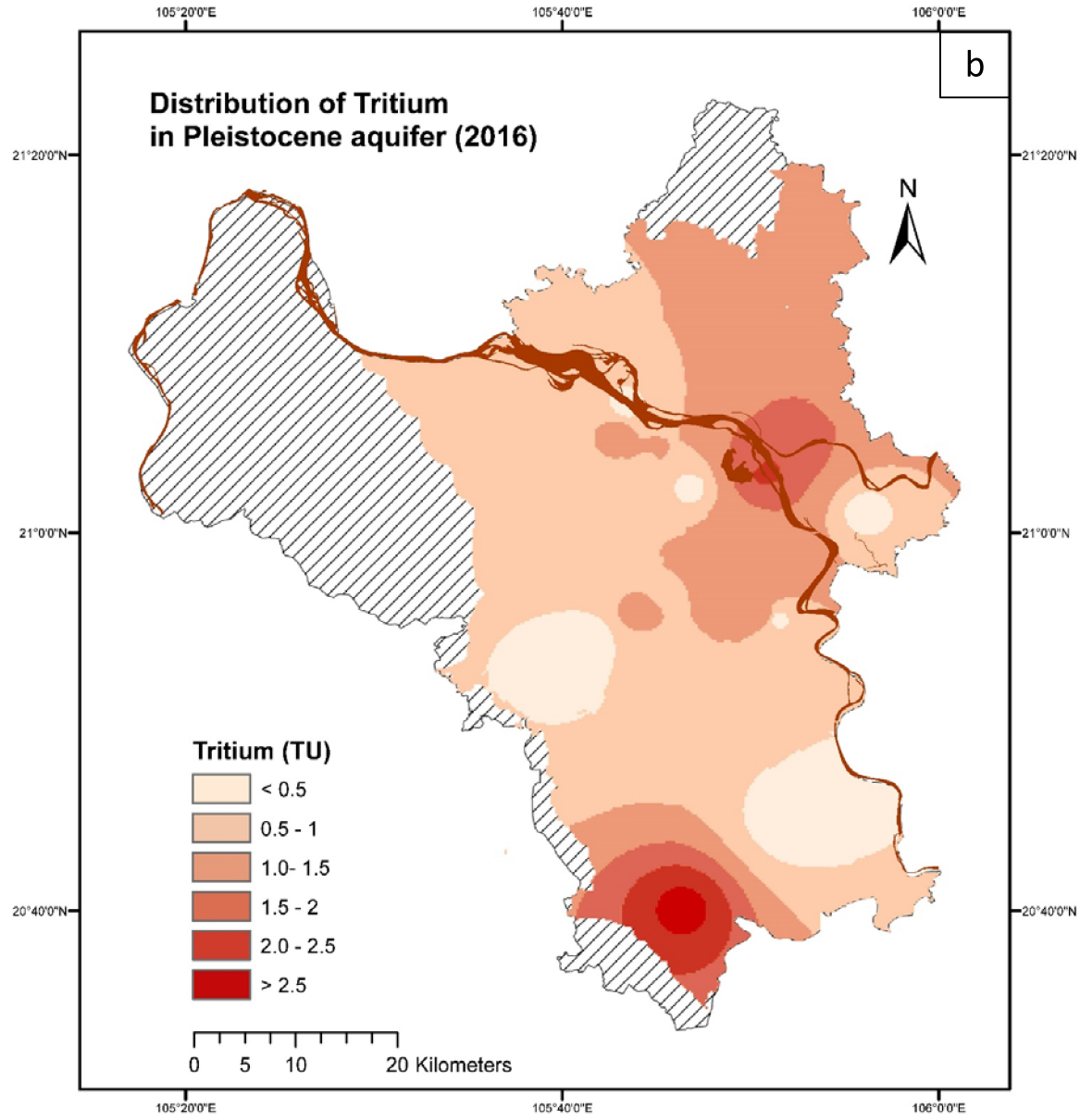

Precipitation in Hanoi
Surface water in Hanoi
LMWL (Hanoi)
δ²H = (7.9 ± 0.2) x δ¹⁸O + (12.5 ± 1.6)
R² = 0.98, n = 38
Surface water line (Hanoi)
δ²H = (5.6 ± 0.4) x δ¹⁸O - (9.1 ± 3.7)
R² = 0.79, n = 50
LMWL (Kunming)
δ²H = (6.6 ± 0.2) x δ¹⁸O - (3.0 ± 1.5)
R² = 0.91, n = 152
Weighted mean in precipitation (Hanoi)
Mean in surface water (Hanoi)
Weighted mean in precipitation (Kunming)
GMWL
δ²H (‰)
δ¹⁸O (‰)
20
0
-20
-40
-60
-80
-100
-120
-18
-16
-14
-12
-10
-8
-6
-4
-2
0

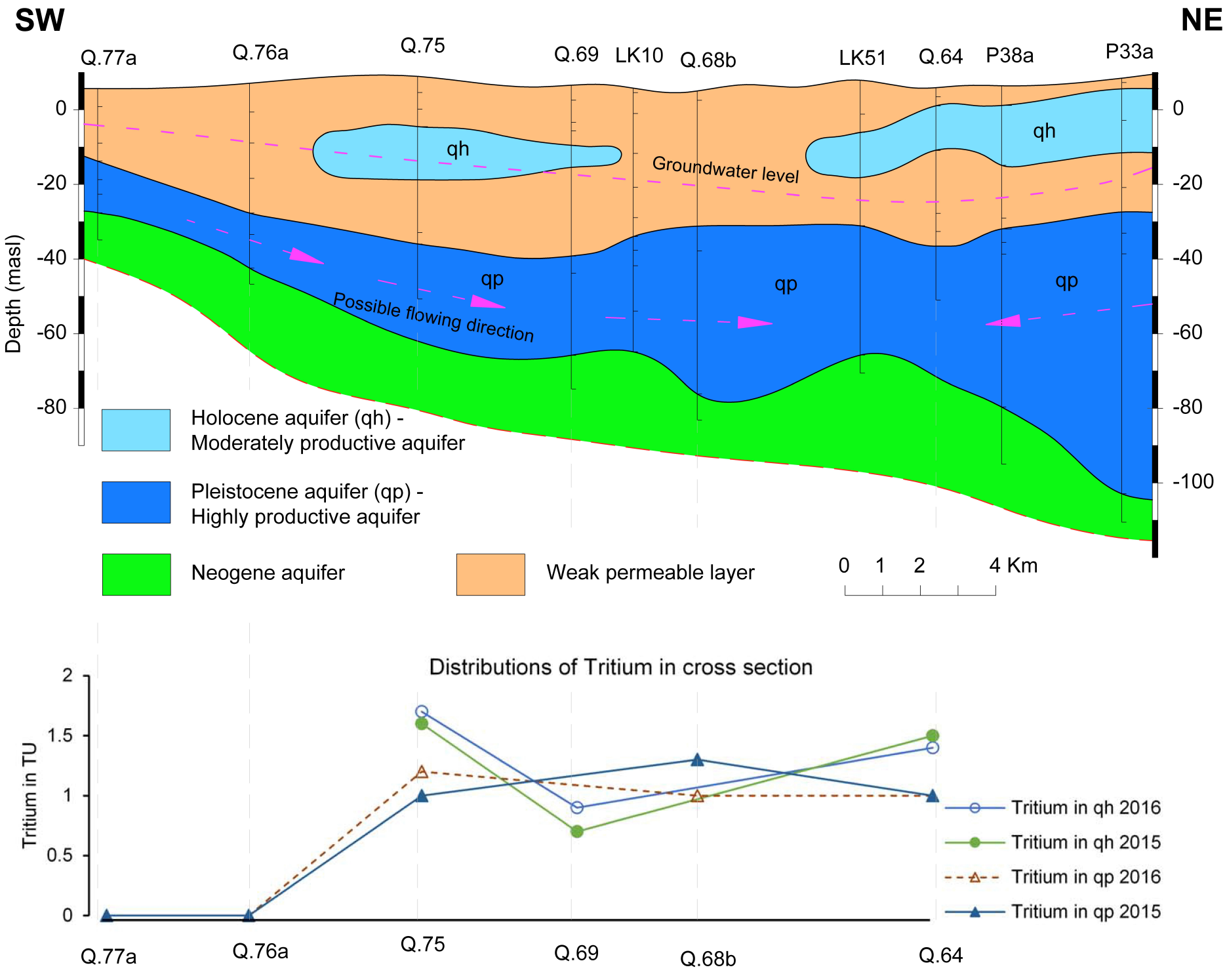

a
δ18O (‰)
δ18O = -0.61 x T + 9.40
R² = 0.56, n =38
Temperature (0C)
b
δ18O (‰)
δ18O = -0.02 x P - 2.52
R² = 0.75, n = 38
Amount of monthly precipitation (mm)
c
Amount of precipitation (mm)
Temperature (0C)
Months
d
δ18O (‰)
2004
2005
2006
2007
Months

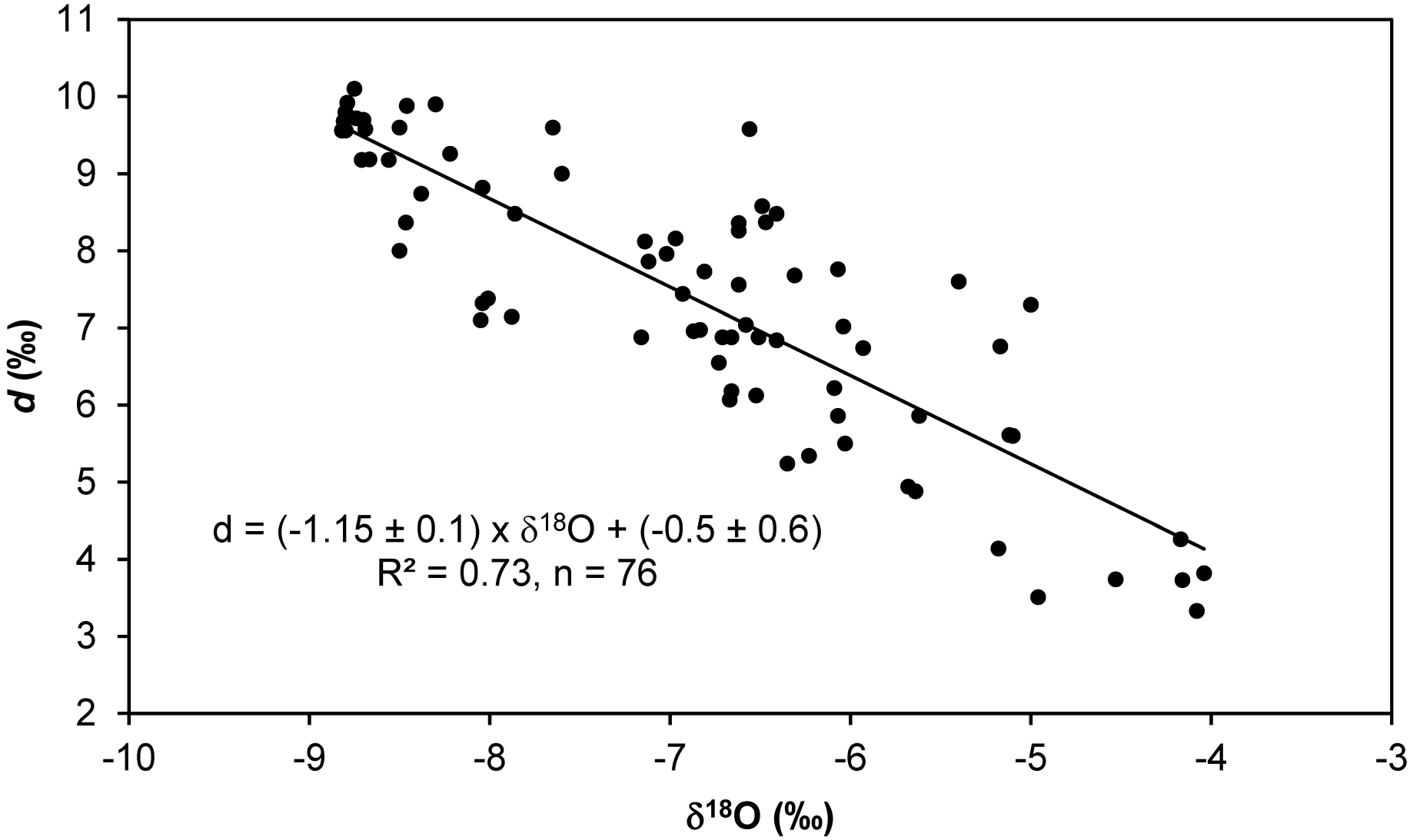

Holocene aquifer
$\delta^2H = (7.0 \pm 0.3) \times \delta^{18}O + (0.7 \pm 1.6)$
$R^2 = 0.98$
LMWL (Hanoi)
Surface water line (Hanoi)
Pleistocene aquifer
$\delta^2H = (6.8 \pm 0.1) \times \delta^{18}O - (1.1 \pm 0.6)$
$R^2 = 0.99$
δ^2H (‰)
$\delta^{18}O$ (‰)
-25
-30
-35
-40
-45
-50
-55
-60
-65
-10
-9
-8
-7
-6
-5
-4
Holocene aquifer
Pleistocene aquifer
Weighted mean in precipitation (Hanoi)
Mean in surface water (Hanoi)
Weighted mean in precipitation (Kunming)

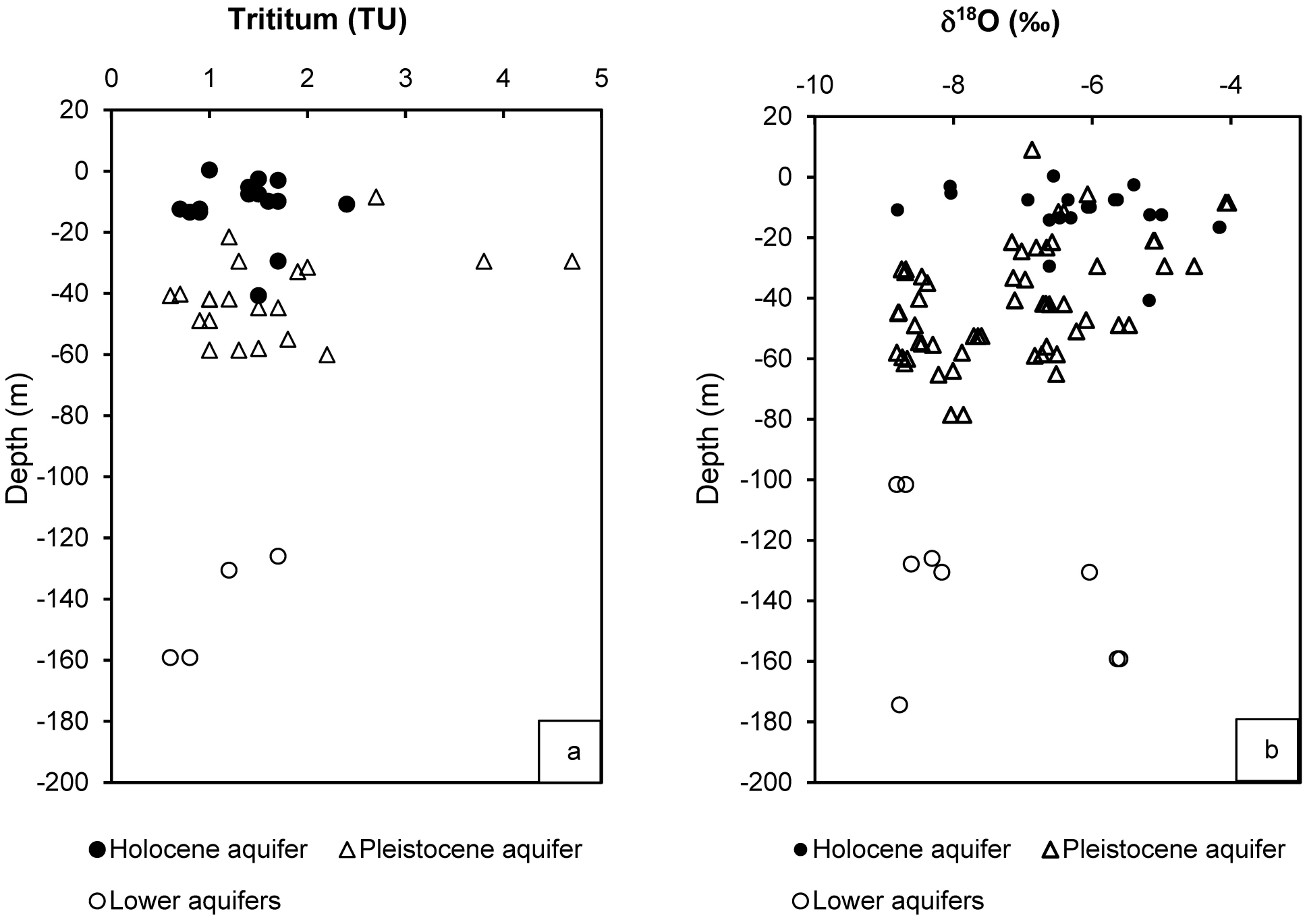

Distribution of δ18O (‰)
in Pleistocene aquifer (2016)
105°20'0"E
105°40'0"E
106°0'0"E
21°20'0"N
21°0'0"N
20°40'0"N
N
δ18O (‰)
-9.0 - -8.0
-8.0 - -7.5
-7.5 - -7.0
-7.0 - -6.5
-6.5 - -6.0
-6.0 - -5.5
-5.5 - -5.0
-5.0 - -4.5
-4.5 - -4.0
-4.0 - -3.5
0 5 10 20 Kilometers


Boxplot of Tritium Distributions
a
Tritium (in TU)
0
2
4
6
8
Surface_water
Rainfall
qh_2015
qh_2016
qp_2015
qp_2016


b
Distribution of Tritium
in Pleistocene aquifer (2016)
105°20'0"E
105°40'0"E
106°0'0"E
21°20'0"N
21°0'0"N
20°40'0"N
N
Tritium (TU)
< 0.5
0.5 - 1
1.0- 1.5
1.5 - 2
2.0 - 2.5
> 2.5
0 5 10 20 Kilometers


SW
NE
Q.77a
Q.76a
Q.75
Q.69
LK10
Q.68b
LK51
Q.64
P38a
P33a
0
-20
-40
-60
-80
-100
Depth (masl)
qh
qp
Groundwater level
Possible flowing direction
Holocene aquifer (qh) -
Moderately productive aquifer
Pleistocene aquifer (qp) -
Highly productive aquifer
Neogene aquifer
Weak permeable layer
0 1 2 4 Km
Distributions of Tritium in cross section
Tritium in TU
2
1.5
1
0.5
0
Tritium in qh 2016
Tritium in qh 2015
Tritium in qp 2016
Tritium in qp 2015


11
10
9
8
7
6
5
4
3
2
d (‰)
-10
-9
-8
-7
-6
-5
-4
-3
δ18O (‰)
d = (-1.15 ± 0.1) x δ18O + (-0.5 ± 0.6)
R² = 0.73, n = 76


Trititum (TU)
0
1
2
3
4
5
20
0
-20
-40
-60
-80
-100
-120
-140
-160
-180
-200
Depth (m)
a
Holocene aquifer
Pleistocene aquifer
Lower aquifers
δ18O (‰)
-10
-8
-6
-4
20
0
-20
-40
-60
-80
-100
-120
-140
-160
-180
-200
Depth (m)
b
Holocene aquifer
Pleistocene aquifer
Lower aquifers