

Hydrogeochemical assessment of groundwater quality during dry and rainy seasons for the two main aquifers in Hanoi, Vietnam

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Abstract Groundwater from the Holocene unconfined aquifer (HUA) and Pleistocene confined aquifer (PCA) is the major source of drinking and domestic water in Hanoi, Vietnam. A clear understanding of the groundwater hydrogeochemical properties, particularly their changes during the dry and rainy seasons, is invaluable for the management and protection of this important water resource. In this study, the changes in the hydrogeochemical properties of groundwater during the dry and rainy seasons were investigated by analyzing the major ions (Ca^{2+} , Mg^{2+} , Na^+ , K^+ , HCO_3^- , SO_4^{2-} , Cl^-) that were recently obtained in 2011 from 13 conjunctive sampling wells for HUA and PCA in Hanoi. The hydrogeochemical assessment was carried out using the Piper and Gibbs diagrams. Based on the results, at least 30 % of the sampling wells in both aquifers exhibited changes in the hydrogeochemical facies during the dry and rainy seasons. Most of the changes occurred for the cation-type facies, while the anion type remained unchanged. Moreover, the hydrogeochemical facies of HUA was found to be different from that of PCA by approximately 50 % of the sampling wells for both the dry and rainy seasons. In addition, the Gibbs diagram showed apparent differences in the weight ratios $\text{Cl}/(\text{Cl} + \text{HCO}_3)$

between PCA and HUA. The results revealed that rock weathering is the main process involved in the evolution of chemical composition of groundwater, but agricultural activities and salty paleowater are the other important factors that affect the groundwater chemistry in some parts of the study area.

Keywords Major ions · Holocene unconfined aquifer · Pleistocene confined aquifer · Dry and rainy seasons · Piper diagram · Hanoi

Introduction

Many people in highly urbanized areas and key cities around the world depend on groundwater as a main source for drinking and domestic water for daily use. In Hanoi, Vietnam's capital, its entire population solely depends on groundwater for the daily water consumption. Recently, the rapid rate in urbanization and industrialization, as well as the drastic increase in the population of Hanoi has led to the unmitigated decline of groundwater levels (Bui et al. 2012a) and the deterioration of water quality (Duong et al. 2003; Montangero et al. 2007). Sustainable management of groundwater is thus necessary to secure its future availability and ecological value.

Pollutants, such as heavy metals and organic compounds, generally interact with the ions present in groundwater (William 1997). A clear view of the predominant ions can help understand the interaction and mechanisms of groundwater contamination. Hydrogeochemical facies, an important diagnostic chemical aspect of groundwater, is commonly examined in the assessment of groundwater quality. Hydrogeochemical facies analysis provides information on the distinct zones of cations and

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anions along different layers of aquifers (Daughney and Reeves 2005).

A study by Bui et al. (2012a) revealed that subsidence is occurring in several parts of Hanoi, which is primarily due to overextraction of its groundwater. In terms of water quality, the groundwater in Hanoi was found to contain arsenic (Berg et al. 2008) but the cause and mode of its contamination, however, are not yet clearly understood. The presence of deleterious substances in the aquifers of Hanoi thus should be of immediate concern for the safety of the population. For these reasons, it is imperative to understand the hydrogeochemical processes occurring in the groundwater of Hanoi so as to aid the formulation of appropriate groundwater management plans. There is, however, little available information regarding the groundwater hydrogeochemical properties in Vietnam, including Hanoi.

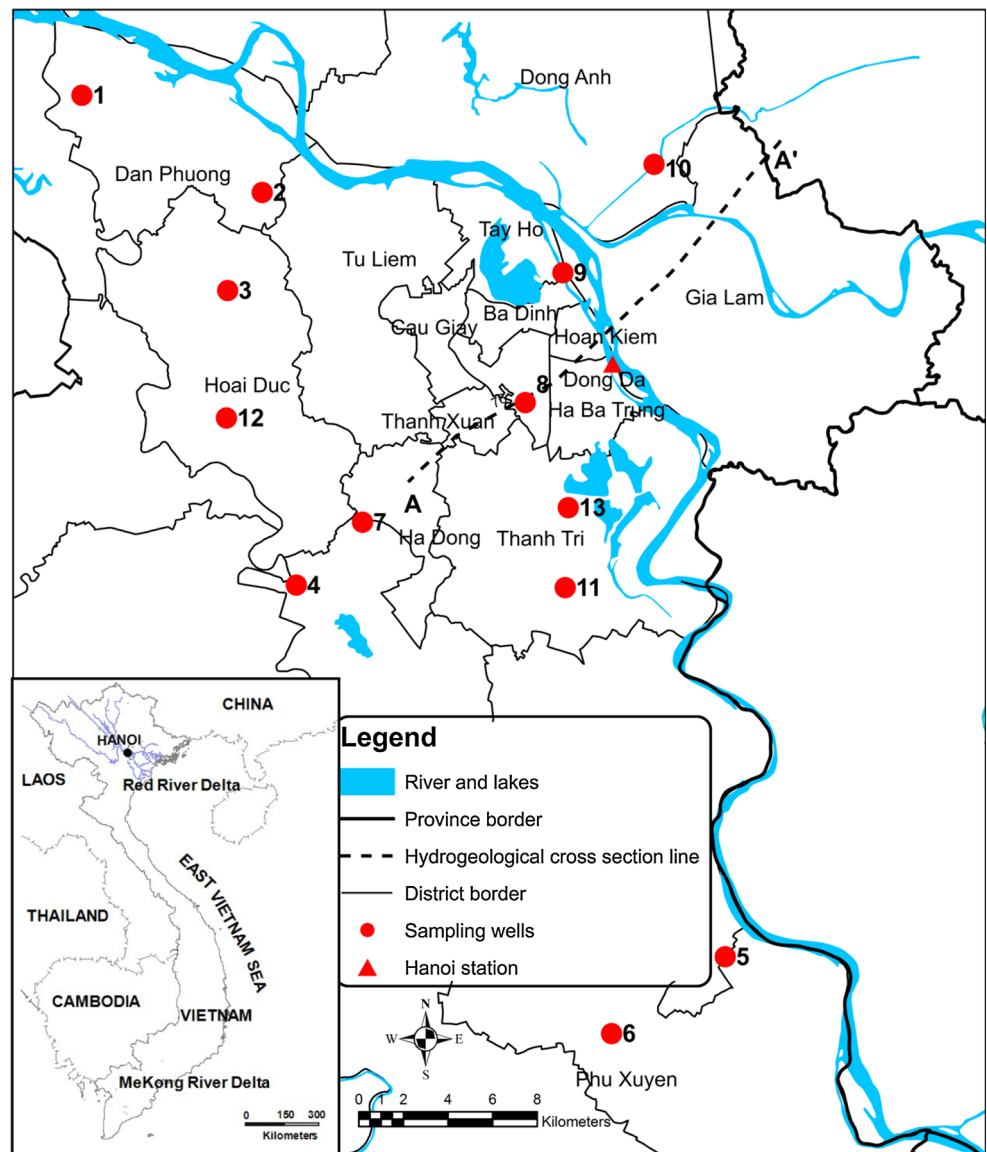
Around the world, many studies have been carried out to investigate and evaluate the hydrogeochemistry of groundwater. For example, Ahmed et al. (2013) investigated the main factors and mechanisms that control groundwater salinization and hydrogeochemical processes in the Eastern Nile Delta, Egypt. Li et al. (2013) assessed groundwater quality for irrigation purposes and identified the hydrogeochemical evolution mechanisms in Pengyang County, China by using the Piper and Gibbs diagrams. Baghvand et al. (2010) studied the groundwater quality of the Kashan Basin, central Iran, and characterized the groundwater species by using the Piper diagram. However, there are few references regarding the changes in the hydrogeochemical properties during the dry and rainy seasons. Groundwater interacts with surface hydrologic systems, such as rivers, lakes, and oceans, and is indirectly influenced by seasonal changes through groundwater recharge from rainfall and surface water in the rainy season and groundwater discharge to rivers in the dry season. Thus, the change in seasons can potentially affect the hydrogeochemical properties of groundwater, especially in areas that have two distinct dry and rainy seasons, like Vietnam. Other studies have focused on investigating the hydrogeochemical properties of shallow (unconfined) aquifers that have high probability of being contaminated from domestic, agricultural and industrial sources (Subba Rao et al. 2012; Marghade et al. 2012; Umar and Alam 2012; Magesh et al. 2013; Yidana et al. 2010). In Hanoi, groundwater from the Holocene unconfined aquifer (HUA) and Pleistocene confined aquifer (PCA) is the major source of water supply. High groundwater abstraction from PCA causes vertical percolation of water from HUA, leading to changes in groundwater chemistry. Therefore, the investigation of the different hydrogeochemical characteristics between the two aquifers, HUA and PCA, is important in understanding the interaction between two aquifers.

Hydrogeochemical evaluation of groundwater systems is usually based on the availability of a large amount of information concerning groundwater chemistry (Hussein 2004). In Vietnam, however, observation data on groundwater chemistry were scarce, which has been an obstacle in the implementation of even the most basic studies on groundwater quality. Therefore, a time-consuming and costly project named the “Nation Hydrogeological Data Project” was initiated under the support and nomination of the Department of Geology and Minerals of Vietnam to construct the GIS-based hydrogeological database. Through this project, groundwater quality data of the unconfined and confined aquifers in Hanoi were collected during dry and rainy seasons of 2011. To take advantage of this unique database, this paper is the first attempt to investigate the hydrogeochemical characteristics of groundwater in HUA and PCA in Hanoi. Especially, this study aims to detect the changes in the hydrogeochemical properties during dry and rainy seasons. The Piper diagram was used to investigate the hydrogeochemical facies. Decades of studies have already proven the efficacy and robustness of the Piper diagram method in classifying the ions into various hydrogeochemical types (Jamshidzadeh and Mirbagheri 2011; Joshi and Seth 2011; Arumugan and Elangovan 2009; Tatawat and Chandel 2008; Raji and Alagbe 1997). Gibbs (1970) proposed chemical diagrams for assessing the functional source of the dissolved chemical constituents, which also show the mechanism controlling the chemistry of surface water. Various studies have already demonstrated the usefulness of these diagrams for groundwater of shallow (unconfined) aquifers (Marghade et al. 2012; Raju et al. 2011; Magesh et al. 2013; Yidana et al. 2010). In this study, the Gibbs diagram was used as reference to determine the factors that govern the composition of groundwater not only in the unconfined aquifer, but also in the confined aquifer.

Study area

Figure 1 shows the geographical locations of Hanoi and the 13 conjunctive sampling wells for both HUA and PCA. Hanoi is located at the north-eastern part of Vietnam. It covers a total area of about 3400 km² and has a population of about 7 million (in 2011), which comprises approximately 7.5 % of the total population of Vietnam. Hanoi is situated in the tropical monsoonal region with two distinct dry and rainy seasons. The rainy season starts in May and ends in October, while the dry season lasts from November until April. The annual average rainfall is about 1,600 mm, but in 2011, it was measured at 1,795 mm. The annual average humidity is about 80 % and the average temperature is around 24 °C. The annual evaporation average is around

Fig. 1 Study area and distribution of sampling points



900 mm. The river network is quite extensive, with a network density of about 0.7 km/km². Hanoi has more than 100 lakes with a total surface area of more than 21.8 km². In 2011, the recorded average discharge of the Red River at the Hanoi station, shown by the triangle in Fig. 1, was 2,182 m³/s during the flood season and 927 m³/s during the dry season; both are lower than the average discharge in the past, 3,970 and 1,160 m³/s, respectively, based on data from 1956 to 2011 (MONRE 1956–2011). High concentration of suspended solids is always present in the Red River. The lakes, ponds and canals in Hanoi are highly polluted because of untreated domestic and industrial wastewater. Since groundwater is relatively cleaner and remains generally unaffected by the surface environmental problems, it has become the most trusted water source (Bui et al. 2012b).

In terms of regional geology, Hanoi is underlain by the Pleistocene and Holocene sediments, with the latter being partly derived from postglacial marine transgressions and tectonic activities. Figure 2 shows the hydrogeological cross section along the A–A' line shown in Fig. 1. From this figure, there are two main aquifers, i.e., HUA and PCA, lying over the Neogene water bearing layer (NWL) and sandwiching the Holocene–Pleistocene aquitard. This aquitard is mainly composed of slightly permeable or impermeable materials like silty clay, clay sand and clays. It has low permeability, less than 0.1 m/day. However, in some minor places in Hanoi, this aquitard is completely missing, which create hydrogeological windows, as indicated in Fig. 2, resulting in total connectivity between the two aquifer systems. Therefore, groundwater levels of the two aquifers are highly interconnected. From our previous

Fig. 2 Hydrogeological cross section along A–A' line as shown in Fig. 1

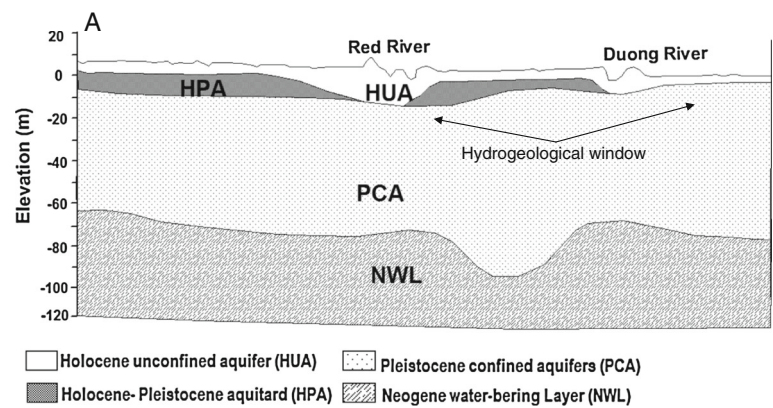
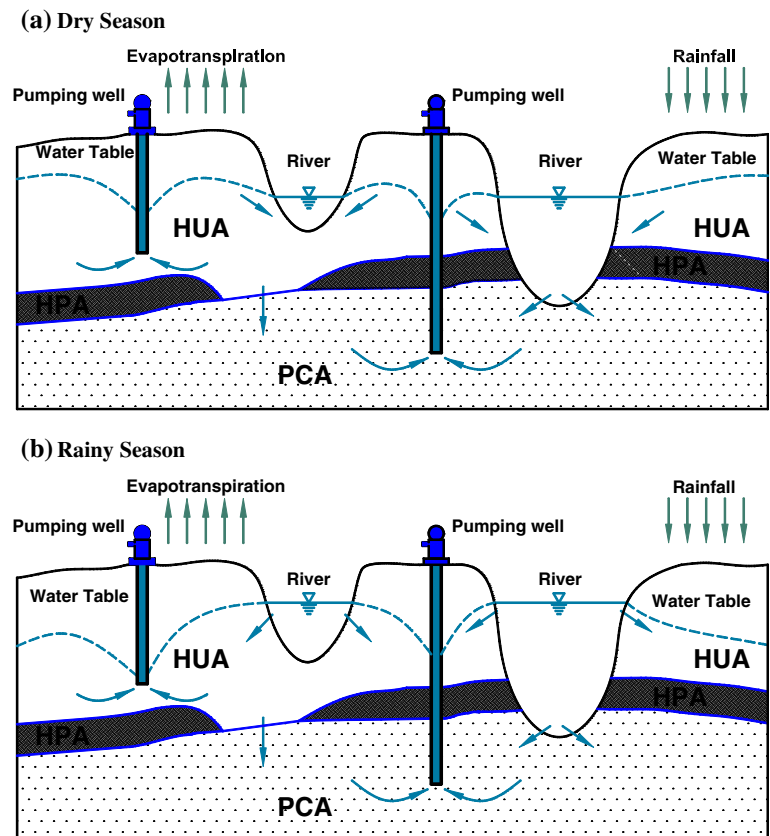


Fig. 3 A schematic diagram showing the inflow and outflow to the groundwater system in Hanoi: **a** for dry season, **b** for rainy season



studies (Bui et al. 2012b), HUA mainly consists of silty clay and various type of sand mixed with gravel. The thickness of this layer varies greatly up to more than 35 m with an average of about 15 m. The transmissivity in HUA is from 20 to 1,788 m²/day. PCA consists of sand mixed with cobbles and pebbles, and is situated lower in the stratigraphic sequence. The thickness of PCA fluctuates over a large range with an average of about 35 m, and gradually increases from north to south. The transmissivity ranges from 700 to 2,900 m²/day and indicates very high potential of groundwater resources. NWL is mainly formed

by geological cracks, weather erosion and unconsolidated sediments. The materials of NWL include: cemented gravel, cemented clay, arkosic sandstone, argillite, and clay carbon. The NWL has very limited groundwater potential with existence of cleft and karst water. The Red River is an important natural recharge source for groundwater storage in Hanoi because it runs across HUA and in some places across PCA due to stream bed erosion. The seasonality in groundwater levels for both HUA and PCA is closely associated with the annual cycles of river water levels (Bui et al. 2011).

Fig. 4 The annual average groundwater levels and flow directions for HUA

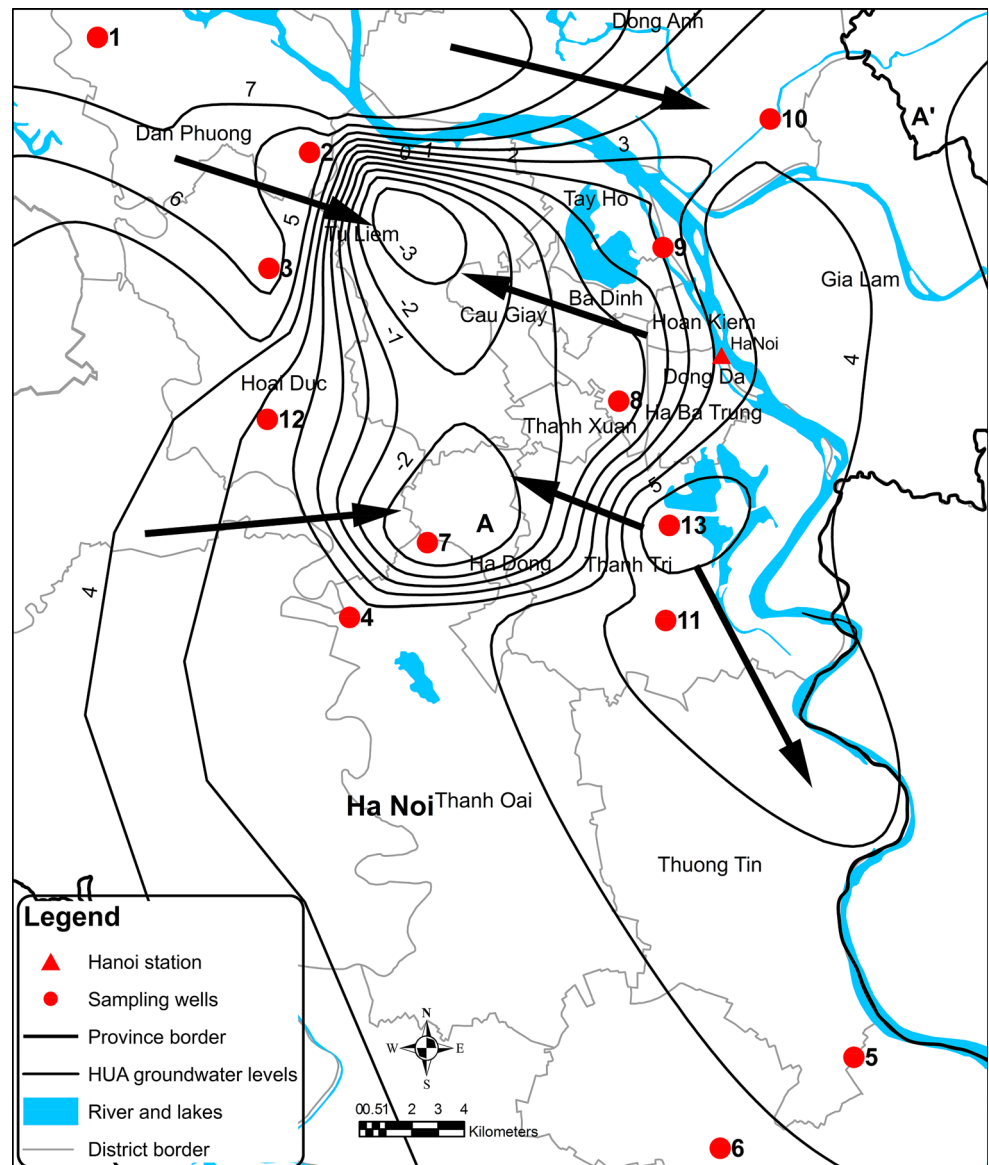
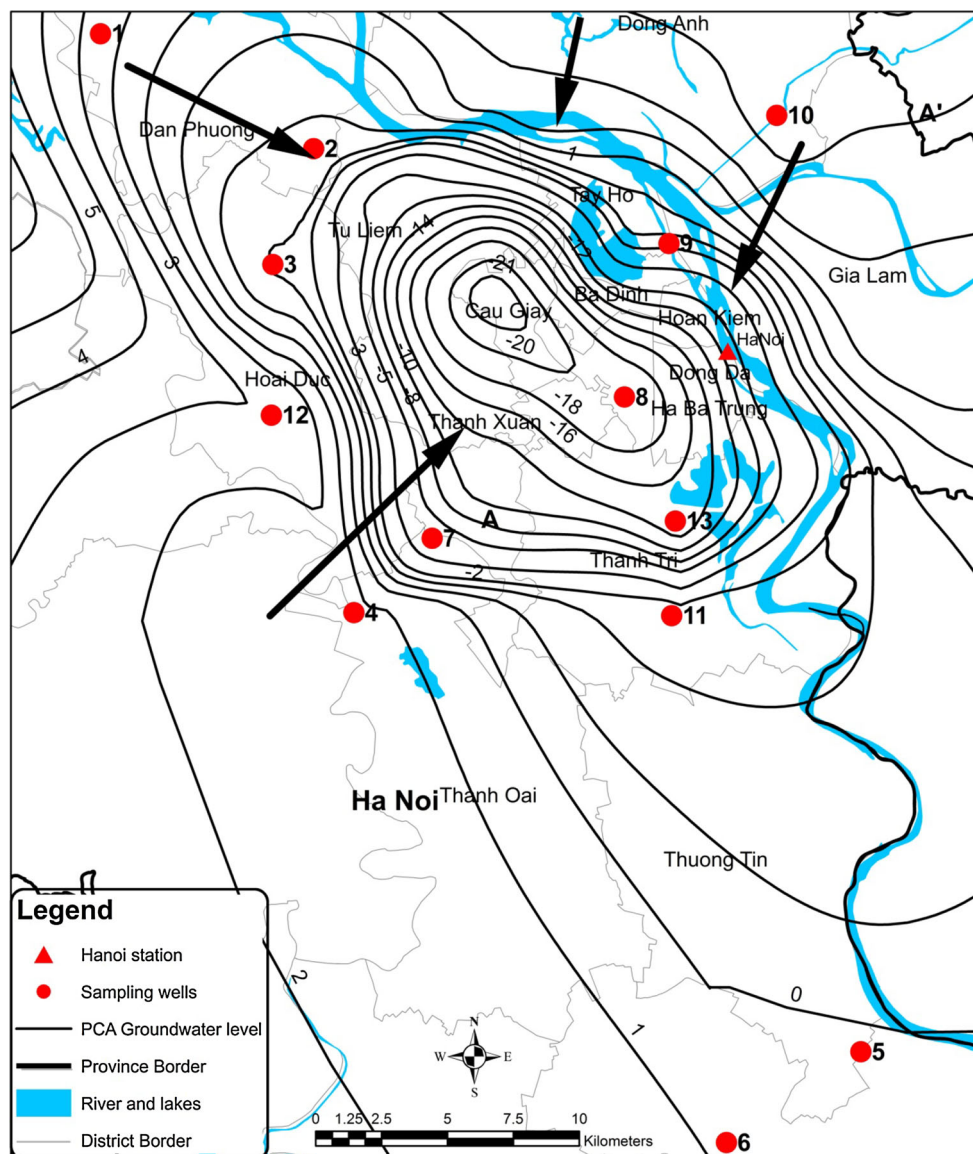


Figure 3 is a schematic diagram showing the inflow and outflow to the groundwater system in Hanoi (Fig. 3a for the dry season and Fig. 3b for the rainy season). As shown in this figure, the main sources contributing the inflows to HUA are rainfall infiltration and groundwater recharge from river (in the rainy season), while groundwater abstraction, groundwater discharge to the river (in the dry season), groundwater infiltration to the deeper aquifer, and evapotranspiration are the outflows of this aquifer. The inflows to PCA are infiltration from HUA through hydro-geological windows, groundwater recharge from the river in some places where the river bed is directly connected with PCA, and the main outflow is groundwater abstraction.

Figures 4 and 5 show the annual average groundwater levels and flow directions for HUA and PCA, respectively.

As shown in these figures, under the present situation, groundwater flow in both aquifers is mainly directed toward the center of Hanoi due to the constant groundwater abstraction. However, in general the groundwater moves from northwest to southeast (from inland to the sea) following the general topography of the study area (Bui et al. 2012a). According to our previous study (Bui et al. 2011), HUA groundwater levels are usually situated within 4 m under the ground surface. Their annual cycle was strongly governed by those of rainfall and river water level with average amplitude of about 2 m. On the other hand, PCA groundwater levels showed rapid decline trends over the area with a speed of about 0.2 m/year. Groundwater in Hanoi receives mostly recharges from surface water because shallow aquifers adjacent to river mostly experience great groundwater recharge. However, large cones of depression

Fig. 5 The annual average groundwater levels and flow directions for PCA



have formed in the center of Hanoi due to excessive groundwater exploitation as shown in Figs. 4 and 5.

Data used and methods

Data used

In Vietnam, hydrogeochemical data are sparse, seldom systematically organized and accessible to a very limited number of users. These primary data sets come from various sources, such as: the Vietnamese geological survey departments, local or national environmental agencies, public and private institutions, consultant firms and many others. A time-consuming and costly project named the “National Hydrogeological Database Project” was

therefore initiated under the Ministerial Decision which was part of the Prime Minister’s own decisions, in which one of the authors was nominated as project leader to construct the GIS-based hydrogeological database. The groundwater monitoring network was installed for groundwater management purposes and, therefore, the recorded data are not for research purposes. Details about this project and database were described in the project report (Tong 2004). Hanoi has the densest hydrogeological data with a large number of data owners in Vietnam. Therefore, the implementation of the database project in Hanoi was much more difficult and valuable than any of the others. The valuable data sets maintain a vital role for further groundwater studies in Hanoi, but so far they are not yet open to the public and only internally accessible to those who are involved in the implementation of the database project.

Table 1 Hydrogeochemical data of groundwater in HUA

Well Nos	Season	TDS (mg/L)	pH	Ca (mg/L)	Mg (mg/L)	Na (mg/L)	K (mg/L)	HCO ₃ (mg/L)	SO ₄ (mg/L)	Cl (mg/L)	NH ₄ (mg/L)	NO ₂ (mg/L)	NO ₃ (mg/L)
1	Dry	659	7.17	126.25	33.44	58.40	19.00	540.03	33.62	102.81	0.56	0.1	0
	Rainy	678	7.45	137.27	31.01	70.00	11.40	610.20	19.21	101.03	5.72	0.07	0
2	Dry	485	6.73	84.17	27.43	55.20	1.35	326.46	1.44	134.71	30	0.3	0
	Rainy	546	7.14	62.23	20.79	73.80	0.99	322.18	1.44	110.61	28.6	0.06	0
3	Dry	1510	8	162.83	92.72	215.00	22.50	610.20	308.21	333.23	20	5	1
	Rainy	1245	7.65	133.27	60.19	218.00	23.80	808.52	94.88	280.06	23	0.1	0
4	Dry	418	6.95	105.81	20.47	23.65	1.85	433.24	2.40	37.22	10	0.02	0
	Rainy	358	7.36	83.77	23.41	14.30	1.75	382.90	6.21	35.45	10.6	0.03	0
5	Dry	362	6.71	56.11	32.83	24.50	3.95	366.12	2.40	46.97	1	0.02	0
	Rainy	338	7.44	54.11	34.05	25.00	3.55	338.66	1.78	46.09	1.14	0.23	0
6	Dry	395	6.74	72.14	19.94	10.20	3.64	219.68	39.63	31.91	6.5	0.75	2.56
	Rainy	399	7.39	89.18	27.36	14.75	4.15	442.09	23.78	5.58	11.25	0	0
7	Dry	660	7.4	102.41	39.16	67.20	7.70	715.92	6.84	23.58	80	0.02	0
	Rainy	594	7.17	96.19	36.12	53.40	4.98	617.37	5.70	28.62	53.6	0.05	0
8	Dry	319	6.74	46.90	27.61	50.40	1.50	318.84	2.42	60.45	2.5	0	0
	Rainy	165	7.39	20.79	9.73	25.25	0.60	143.40	3.00	25.70	2.94	2	0
9	Dry	116	6.98	31.14	4.59	3.50	1.43	125.09	5.28	6.20	0	0.32	0
	Rainy	133	7.85	36.08	7.60	3.80	1.05	149.50	9.61	7.09	0	0.02	0
10	Dry	393	6.72	88.18	21.89	18.50	1.80	387.48	21.01	26.59	0	12	4
	Rainy	398	8	90.18	27.97	20.00	2.20	411.89	14.41	30.13	0	0.09	3.6
11	Dry	270	7.52	37.07	21.28	26.80	3.05	213.57	16.81	44.31	0.7	0	0
	Rainy	242	7.85	42.08	17.63	23.10	2.60	207.47	14.23	33.68	0	0.9	0
12	Dry	228	6.74	24.07	22.74	15.40	0.80	197.70	9.61	18.61	0	0	0
	Rainy	205	7.63	33.07	19.94	14.05	0.84	219.67	2.40	21.27	0.61	0.14	0
13	Dry	531	7.76	72.94	33.20	52.00	8.26	521.72	4.32	28.72	60	1.1	0.4
	Rainy	619	7.4	56.11	68.40	42.50	5.55	645.29	1.20	29.91	36	9.2	0.2

To investigate hydrogeochemical characteristics of groundwater in Hanoi, we used the most recent groundwater chemical data from our National Hydrogeological Database Projects, which were collected from 13 conjunctive observation wells for HUA and PCA. These observation wells belong to the National Groundwater Monitoring Network, which were set up by the Vietnamese government. The samples were collected in February (dry season) and August (rainy season) of 2011 and analyzed according to ISO standard test methods for the following physico-chemical parameters: TDS, pH, major cations (Ca²⁺, Mg²⁺, Na⁺, K⁺), major anions (HCO₃⁻, Cl⁻ and SO₄²⁻), NH₄⁺, NO₂⁻, and NO₃⁻. The carbonate ion (CO₃²⁻) was calculated from the observed bicarbonate (HCO₃⁻) and pH data (James 1982). The hydrogeochemical data of 26 samples from 13 conjunctive HUA and PCA wells were presented in Tables 1 and 2, respectively. This study is the first to utilize the groundwater chemical data from this unique database.

Method

Groundwater samples were collected throughout Hanoi from observation wells. Sampling was done in accordance to the guidance on the sampling, preservation and handling of groundwater samples of Ministry of Natural Resources and Environment (MONRE 2008). All samples were filtered with 0.45 μm filter membranes and collected in clean and dry Polyethylene or Polytetrafluoroethylene plastic bottles filled to the top and capped tightly to avoid evaporation and exchanges of sample water with atmospheric materials. To take account of any physico-chemical change that might take place, all field-based water parameters such as temperature and pH were measured in situ.

Chemical analyses were undertaken at the laboratory of Analytical Chemistry Department, Vietnam Academy of Science and Technology, following the national technical regulation on underground water quality (MONRE 2008). This regulation was based on ISO standards. Major cation concentrations were analyzed by the EDTA titrimetric

Table 2 Hydrogeochemical data of groundwater in PCA

Well Nos	Season	TDS (mg/L)	pH	Ca (mg/L)	Mg (mg/L)	Na (mg/L)	K (mg/L)	HCO ₃ (mg/L)	SO ₄ (mg/L)	Cl (mg/L)	NH ₄ (mg/L)	NO ₂ (mg/L)	NO ₃ (mg/L)
1	Dry	659	7.17	78.25	24.08	11.00	3.00	390.53	16.81	10.64	3	0.02	0
	Rainy	678	7.45	101.20	32.83	10.50	1.90	434.77	16.81	47.65	5	0	0
2	Dry	485	6.73	44.09	13.38	26.68	3.19	268.49	1.80	11.52	3.6	0.02	0
	Rainy	546	7.14	41.08	15.81	25.00	3.30	271.54	1.80	13.29	4	3.8	0.28
3	Dry	1510	8	43.09	26.14	19.50	3.60	288.32	4.32	22.34	23	2.4	1.68
	Rainy	1245	7.65	56.21	22.74	41.25	3.74	384.43	10.81	22.34	20.67	0.02	0
4	Dry	418	6.95	96.20	20.06	21.50	1.78	433.24	2.40	35.45	23	0	0
	Rainy	358	7.36	87.17	13.38	18.50	1.10	347.81	6.21	21.27	3.44	0	0
5	Dry	362	6.71	55.11	33.44	30.00	3.95	381.38	3.36	37.22	1	0	0
	Rainy	338	7.44	62.12	32.23	28.25	4.15	384.43	6.02	42.54	0	0.19	0
6	Dry	395	6.74	60.70	41.91	299.50	8.90	479.01	30.02	467.94	16.55	0	0
	Rainy	399	7.39	47.60	41.04	340.00	8.30	390.53	36.02	538.84	15	0	0
7	Dry	660	7.4	93.69	35.66	58.20	1.92	651.70	9.61	19.14	80	0	0
	Rainy	594	7.17	51.21	25.75	34.41	3.78	300.30	36.02	3.24	70.4	2.25	0
8	Dry	319	6.74	46.29	20.42	41.48	2.47	284.11	2.30	60.40	8.5	0.02	0
	Rainy	165	7.39	55.11	18.06	46.20	2.04	302.05	2.20	65.58	12.1		0
9	Dry	116	6.98	33.65	10.18	4.00	1.25	167.81	1.60	7.09	0	0.12	0.2
	Rainy	133	7.85	43.09	9.73	5.25	1.05	186.11	1.20	8.86	0.34	0	0
10	Dry	393	6.72	41.89	11.36	11.00	4.13	210.52	4.20	8.86	2.4	0.07	0
	Rainy	398	8	48.24	13.53	9.74	5.51	222.72	14.41	9.60	0.98	0	3.01
11	Dry	270	7.52	47.24	19.58	70.15	4.37	425.31	14.29	15.82	40	0	0
	Rainy	242	7.85	48.10	52.29	73.20	4.02	619.35	11.38	21.27	27.4	0.02	0
12	Dry	228	6.74	32.06	13.74	11.50	1.35	194.20	2.28	9.26	1.5	0.16	2
	Rainy	205	7.63	28.06	13.95	10.25	0.95	180.01	7.20	7.98	1.67	0	0
13	Dry	531	7.76	24.65	15.65	29.66	4.94	238.90	2.16	11.65	10	0	0
	Rainy	619	7.4	30.87	12.03	29.04	4.68	222.42	2.16	13.56	11.15	0.08	0

method for Ca²⁺ and Mg²⁺ (ISO 6058 ISO 1984a; ISO 6059 1984b) and the flame emission spectrometry method for Na⁺ and K⁺ (ISO 9964-3 1993). Calibrations for cations analyses were carried out by appropriate standards. Both laboratory and international reference materials were used to check the accuracy of the chemical analyses. The concentrations of Cl⁻, HCO₃⁻, and SO₄²⁻ anions were determined using Silver nitrate titration on chromate indicator (Mohr's method), ion chromatography and continuous flow analysis (CFA) methods, respectively (ISO 9297 1989; ISO 22743 2006). To determine the concentrations of NH₄⁺, NO₂⁻, and NO₃⁻, the dimethylphenol spectrometric method, molecular absorption spectrometric method and spectrometric method using sulfosalicylic acid were applied, respectively (ISO 7150-2 1986; ISO 6777 1984c; ISO 7890-3 1988). The analytical precision for measurement of ions was determined by calculating the ionic balance error, which was within 5 %.

The term “hydrogeochemical facies” is used to describe the occurrence modes of groundwater in an aquifer with

respect to chemical composition. To determine the hydrogeochemical facies of groundwater, the percentages of the equivalents of each physico-chemical parameter are plotted on a Piper diagram (Piper 1944). This diagram is then used to identify the dominant cation and anion in each well by using the left and right ternary diagrams, respectively. The left ternary diagram is divided into three cationic classification regions, namely the [Ca²⁺], [Mg²⁺], and [Na⁺] types, whereas the right ternary diagram is divided into three anionic classification regions, namely the [HCO₃⁻], [Cl⁻], and [SO₄²⁻] types. Each observation has a dominant cation and anion type. The combination of these predominant ion types is the hydrogeochemical facies of the aquifer at a specific observation well. After plotting the data, the hydrogeochemical facies of each well was investigated for changes related to the dry and rainy seasons by comparing the dominant ions.

A chemical diagram of the major natural mechanisms controlling the composition of the dissolved solids in surface water was proposed by Gibbs (1970). The

Table 3 Ranges of chemical parameters in HUA and their comparison with the Vietnamese and WHO standards for drinking water

Chemical parameter	Concentrations of ions				WHO standard value (maximum limit)	Vietnamese standard value (maximum limit)	Sample numbers exceeding maximum limit value	
	Dry season		Wet season				Dry season	Wet season
	Range	Mean	Range	Mean				
pH	6.7–8.0	7.1	7.1–8.0	7.5	–	6.5–8.5	None	None
TDS (mg/L)	116–1510	488	133–1245	455	–	1000	3	3
Ca ²⁺ (mg/L)	24.07–162.83	77.7	20.79–137.27	71.9	–	–		
Mg ²⁺ (mg/L)	4.59–92.72	30.6	7.6–68.4	29.6	–	–		
Na ⁺ (mg/L)	3.5–215	47.8	3.8–218	45.9	200	200	3	3
K ⁺ (mg/L)	0.8–22.5	5.9	0.6–23.8	4.9	–	–		
HCO ₃ [–] (mg/L)	125.09–753.6	383	143.4–808.52	407.6	–	–		
SO ₄ ^{2–} (mg/L)	1.44–308.21	34.9	1.2–94.88	15.2	500	250	3	None
Cl [–] (mg/L)	6.2–333.23	69	6.2–280.06	58.1	250	250	3	3
CO ₃ ^{2–} (mg/L)	0.000–0.003	0.0006	0.000–0.002	0.0008	–	–		
NH ₄ ⁺ (mg/L)	0–80	16.3	0–53.6	13.3	–	1.5	2, 3, 4, 6, 7, 8, 13	1, 2, 3, 4, 6, 7, 8, 13
NO ₂ [–] (mg/L)	0–5	1.5	0–9.2	1	50/total nitrogen	3	3	13
NO ₃ [–] (mg/L)	0–4	0.6	0–3.6	0.3	–	50	None	None
TH (mg/L)	90–788	299	92–580	296	–	300	1, 3, 4, 6, 10	1, 3, 6, 7, 10

weight ratios Na/(Na + Ca) and Cl/(Cl + HCO₃) against the total dissolved solids (TDS) on a logarithmic axis were plotted separately in two diagrams, a Gibbs cation diagram and a Gibbs anion diagram. Although the Gibbs diagram was originally applied to surface water, it is widely used to assess the functional sources of dissolved chemical constituents in groundwater in shallow (unconfined) aquifers, which have high potential for being influenced by surface water (Marghade et al. 2012; Raju et al. 2011; Nagarajan et al. 2010). According to Gibbs (1970), the three major natural mechanisms controlling surface water chemistry are precipitation dominance, rock dominance, and evaporation dominance. Evaporation of surface water and moisture in the unsaturated zone has been found as the most influential process in the development of the chemical composition of shallow groundwater (Richter and Kreitler 1993). Evaporation concentrates the remaining water and leads to precipitation and deposition of evaporites that are eventually leached into the deeper aquifer (Zhu et al. 2008). In addition, as mentioned above, groundwater levels of two aquifers (HUA and PCA) are highly interconnected in some parts of the study area. Therefore, Gibbs diagram will be a useful tool to analyze geochemical processes of groundwater in both unconfined and confined aquifers in the study area. This study is the first attempt to use the Gibbs diagram as reference for assessing the factors governing groundwater chemistry in a confined aquifer as well as an unconfined aquifer in the study area.

Results and discussions

Chemical quality of groundwater

The range of hydrogeochemical parameters in HUA and PCA and their comparison with the WHO and Vietnamese drinking water standards are listed in Tables 3 and 4, respectively, and the suitability of groundwater for drinking and domestic use is evaluated. In general, the concentrations of most ions in HUA are higher than those in PCA throughout the year. This may be due to evaporation of water in the unsaturated zone, which increases the concentration of the ions that eventually leach into the HUA groundwater.

The results in Tables 3 and 4 clearly indicate that the change from the dry to the rainy season causes changes in the mean values of the groundwater quality indices. The TDS, Ca²⁺, K⁺, SO₄^{2–}, Cl[–], NO₂[–] values decrease from the dry to the rainy season in HUA, but increase in PCA. The dilution of groundwater ions because of rainfall may be the reason for the downward trend in HUA, whereas the upward trend in PCA may be due to the leaching of ions from overlying materials by infiltrating recharge waters to PCA during the rainy season through hydrogeological windows, as described above.

As shown in Tables 3 and 4, out of 26 dry and rainy season samples, 15 in HUA and 21 in PCA have ammonium concentrations higher than Vietnamese drinking standard. Although ammonium is not directly related to health problems, it may be used as an indicator of

Table 4 Ranges of chemical parameters in PCA and their comparison with the Vietnamese and WHO standards for drinking water

Chemical parameter	Concentrations of ions				WHO standard value (maximum limit)	Vietnamese standard value (maximum limit)	Sample numbers exceeding maximum limit value	
	Dry season		Wet season				Dry season	Wet season
	Range	Mean	Range	Mean				
pH	6.5–8.2	7.2	7–7.9	7.5	–	6.5–8.5	None	None
TDS (mg/L)	153–1145	390	162–1224	398	–	1,000	None	None
Ca ²⁺ (mg/L)	24.65–96.2	53.7	28.06–101.2	53.8	–	–		
Mg ²⁺ (mg/L)	10.18–41.91	22	9.73–52.29	23.33	–	–		
Na ⁺ (mg/L)	4–299.5	48.78	5.25–340	51.67	200	200	6	6
K ⁺ (mg/L)	1.25–8.9	3.4	0.95–8.3	3.4	–	–		
HCO ₃ [–] (mg/L)	167.81–814.62	339.5	100.1–619.35	326.7	–	–		
SO ₄ ^{2–} (mg/L)	1.6–30.02	7.32	1.2–36.02	11.71	500	250	None	None
Cl [–] (mg/L)	7.09–467.94	55.18	3.24–538.84	62.77	250	250	6	6
CO ₃ ^{2–} (mg/L)	0.000–0.0026	0.0006	0.000–0.001	0.0005	–	–		
NH ₄ ⁺ (mg/L)	0–80	16.35	0–70.4	13.17	–	1.5	1, 2, 3, 4, 6, 7, 8, 10, 11, 12, 13	1, 2, 3, 4, 6, 7, 8, 11, 12, 13
NO ₂ [–] (mg/L)	0–2.4	0.22	0–3.8	0.53	50/total nitrogen	3	None	2
NO ₃ [–] (mg/L)	0–2	0.3	0–3.01	0.25	–	50	None	None
TH (mg/L)	120–340	212	115–388	231	–	300	4, 6, 7	1, 7, 11

groundwater contamination resulting from urbanization and agricultural activities (Keith 2002). Tables 3 and 4 also show that the vast majority of the groundwater samples have concentrations of nitrite and nitrate less than the WHO and Vietnamese standard values. However, most nitrogenous materials in natural waters tend to convert to nitrate; therefore, all sources of combined nitrogen, particularly ammonium and nitrite, should be considered as potential nitrate sources.

Schoeller (1962) devised a semi-logarithmic diagram (Schoeller diagram) of the concentration of the main ionic constituents in water in meq/L. Concentrations of each ion in each sample are represented by points on six equally spaced lines and points are connected by a straight line. This diagram gives the absolute concentration of each ion and, in addition, the concentration differences among various groundwater samples. The line also gives the ratio between two ions in the same sample. Figure 6 shows the Schoeller diagrams: (a) for HUA and (b) PCA, respectively, to illustrate major constituents of groundwater in the study area. The dashed and solid lines indicate the dry and rainy seasons, respectively. From this figure, the relative concentrations of the ions occur in the following order: Ca²⁺ > Mg²⁺ > Na⁺+K⁺ and HCO₃[–] + CO₃^{2–} > Cl[–] > SO₄^{2–}, except for HUA well No. 3 and PCA well No. 6. Comparison of Fig. 6a and b shows that groundwater in HUA has a wider range of ion concentrations than in PCA. This implies that the spatial distribution of the chemical composition in groundwater of PCA is more homogeneous than that of HUA.

Hydrogeochemical facies and water types

The Piper diagram has been widely used to study the similarities and differences in the composition of waters and to classify them into certain chemical types (Chadha 1999). The usefulness of the Piper diagram in the representation of some geochemical processes and understanding the hydrogeochemical regime of a study area has been proven by many researchers (e.g., Jalali 2011; Parasanna et al. 2011; Subba Rao et al. 2012). In this study, Piper diagram was used to help in classifying groundwater types and identifying hydrogeochemical facies in Hanoi, Vietnam. Figures 7 and 8 show the Piper diagram plots for HUA and PCA, respectively. The purpose of these figures is to visually show how much, in general, HUA is similar to (or different from) PCA in terms of water types. The numbers in these figures correspond to the locations of the observation wells in Fig. 1. The white and black colors indicate the dry and rainy seasons, respectively. As indicated in the left ternary diagram of Fig. 7, the water samples identified as the [Ca²⁺], [Na⁺], and [Mg²⁺] types are 19, 4 and 3, respectively. The right ternary diagram shows all water samples to be of the [HCO₃][–] type. Thus, HUA is mostly of the [Ca²⁺–HCO₃[–]] type (calcium ion-bicarbonate ion type). On the other hand, water types in PCA, as shown in Fig. 8, have the same number of the [Ca²⁺], [Na⁺], and [Mg²⁺] types. PCA also has 24 out of the 26 samples that are dominated by the [HCO₃[–]]

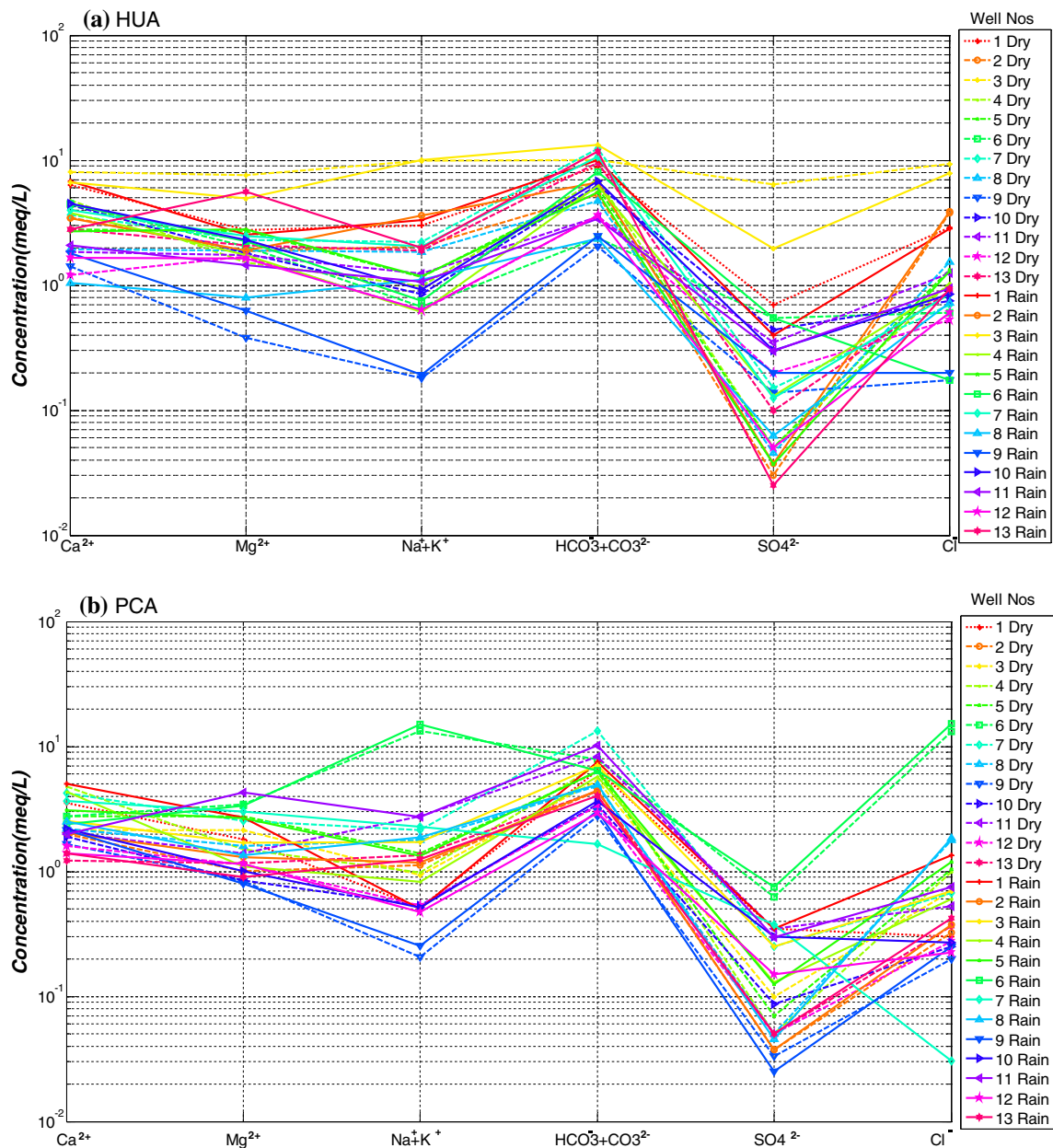


Fig. 6 Schoeller diagram showing the hydrogeochemical composition of groundwater: **a** for HUA, **b** for PCA

type, thus, generally speaking, PCA is highly similar to HUA in terms of the hydrogeochemical facies.

To compare further the water types of HUA with PCA by means of the location of their sampling wells, the water types of each well for both aquifers, during the dry and rainy seasons, are summarized in Table 5. The well numbers in Table 5 correspond to the well numbers in Fig. 1. As can be seen in Table 5, changes in the hydrogeochemical facies during the dry and rainy seasons can be observed. For instance, in well H13 of HUA, the water type during the dry season was $[Ca^{2+}-HCO_3^-]$, but became $[Mg^{2+}-HCO_3^-]$ during the rainy season. In P11 of PCA, it

was of the $[Na^+-HCO_3^-]$ type during the dry season and changed to the $[Mg^{2+}-HCO_3^-]$ type during the rainy season. In HUA, the hydrogeochemical facies of 5 out of the 13 observation wells exhibited seasonal changes, particularly of the cation type. H5 and H13 changed from the $[Ca^{2+}]$ to $[Mg^{2+}]$ type and H12 changed from the $[Mg^{2+}]$ to $[Ca^{2+}]$ type. This may be due to recharge from surface water such as rainfall, lake, or river. H2 and H8 changed from the $[Ca^{2+}]$ to $[Na^+]$ type, which suggests the influence from surface water to groundwater in HUA during the rainy season. Regarding PCA, 4 out of the 13 observation wells showed changes in the cation type during the dry and

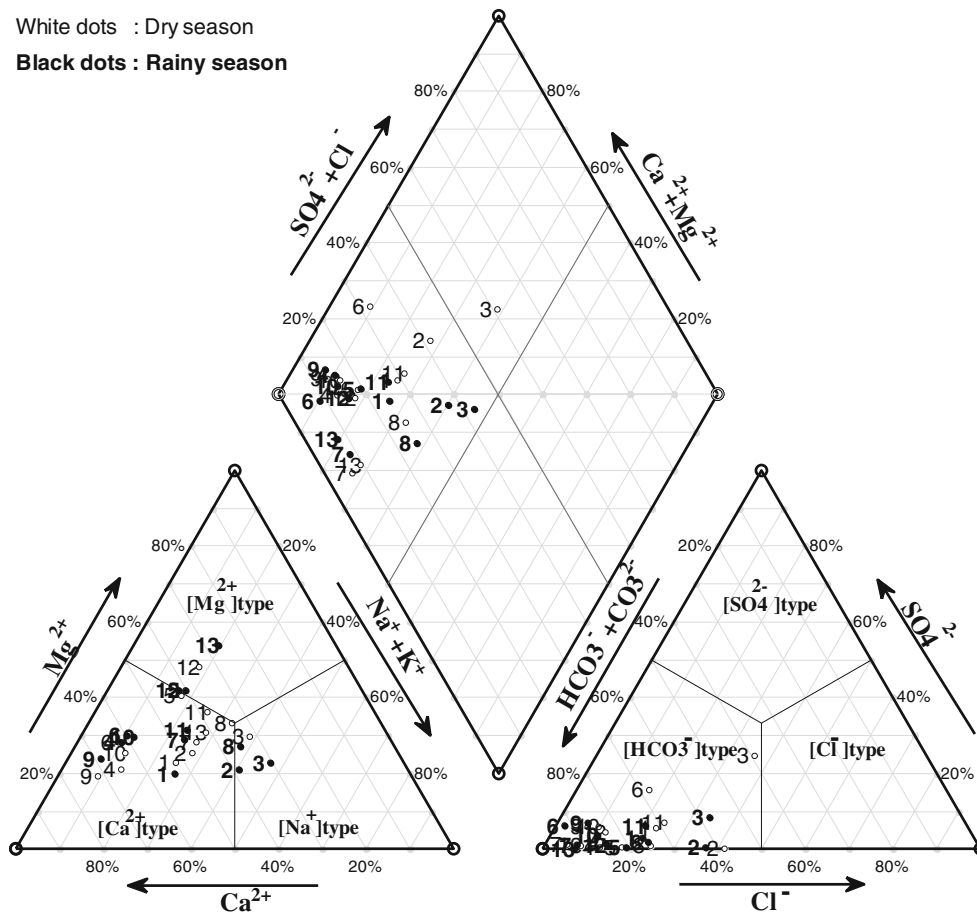


Fig. 7 Piper diagram for HUA groundwater in Hanoi

rainy seasons. P3 and P5 changed from the $[Mg^{2+}]$ to $[Ca^{2+}]$ type, P11 changed from the $[Na^+]$ to $[Mg^{2+}]$ type, and P13 changed from the $[Na^+]$ to $[Ca^{2+}]$ type. This implies that water infiltration from HUA may affect the concentrations of chemical constituents of the PCA groundwater during the rainy season through hydrogeological windows, as mentioned above.

It is worth to note that the differences in the hydrogeochemical facies between the two aquifers are also observed in Table 5. For instance, the water in H3 of HUA was of $[Na^+ - HCO_3^-]$ type during the dry season, but concurrently of the $[Mg^{2+} - HCO_3^-]$ type in P3 of PCA. Interestingly, during the rainy season, the water in H2 of HUA was of the $[Na^+ - HCO_3^-]$ type, but of the $[Ca^{2+} - HCO_3^-]$ type in P2 of PCA. In total, 6 out of the 13 observation wells during the dry season and 7 out of the 13 during the rainy season exhibited differences in the water type between the two aquifers. These differences are possibly influenced by the materials in the two aquifers, as well as interaction with surface water and between the two aquifers, which affect the groundwater chemical characteristics and flow paths.

Factors governing water chemistry

Gibbs diagram is an important tool to analyze geochemical processes. It is widely employed to assess the functional sources of dissolved chemical constituents such as precipitation dominance, rock dominance and evaporation dominance (Ahmed et al. 2013). Figure 9 shows the Gibbs diagram for groundwater in HUA. Gibbs (1970) found that the composition of most of the world's surface water falls in the boundaries, which the authors expressed in this study as boundary G^+ on the Gibbs cation diagram (Fig. 9a) and boundary G^- on the Gibbs anion diagram (Fig. 9b). The area inside these boundaries was subdivided into three domains (precipitation dominance, rock dominance, and evaporation dominance) by Gibbs (1970) on the basis of analytical chemistry data, which represent the three major natural mechanisms controlling surface water chemistry as indicated in Fig. 9. To make these domains clear, (Kumar et al. 2009) delineated the boundaries of the rock-water interaction dominance field, which were also adopted by other researchers (Ravikumar et al. 2011; Raju et al. 2011; Gurugnanam et al. 2009). For clarity in this study, these

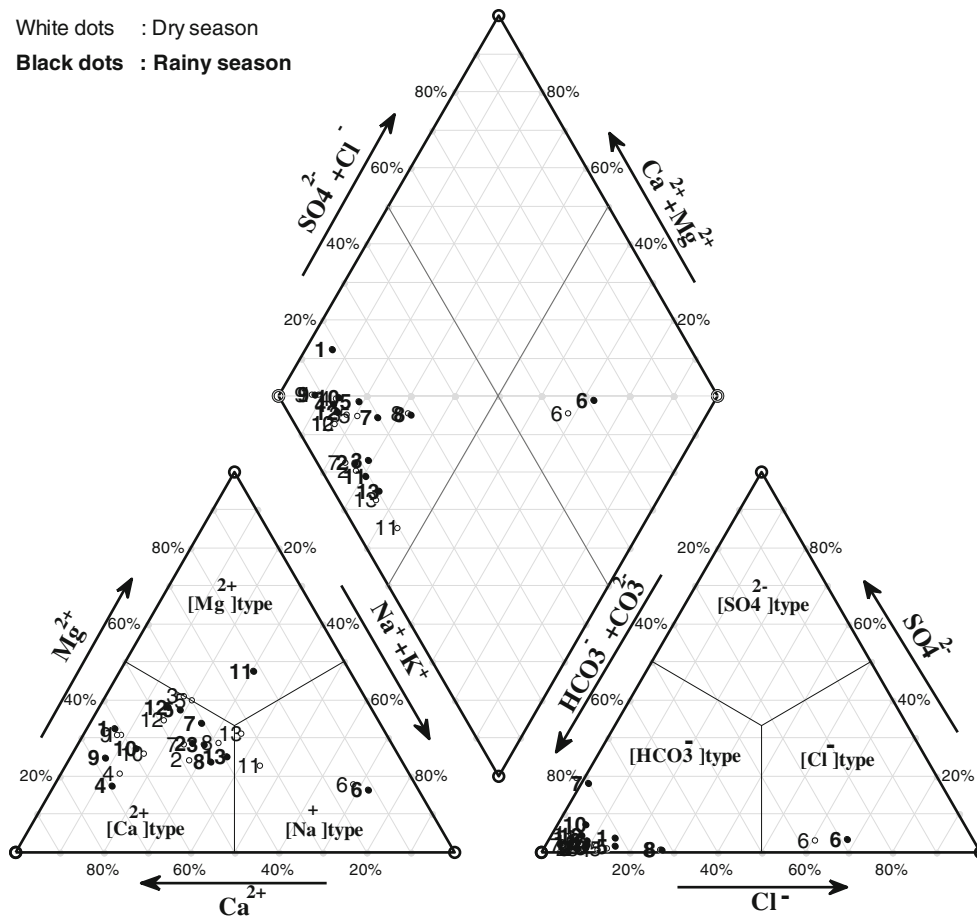


Fig. 8 Piper diagram for PCA groundwater in Hanoi

boundaries are presented by the boundaries K^+ on the Gibbs cation diagram (Fig. 9a) and K^- on the Gibbs anion diagram (Fig. 9b). As in the Piper diagram, the number symbols in the figures correspond to the locations of the observation wells in Fig. 1. The white and black dots indicate dry and rainy season data, respectively.

As shown in Fig. 9, 12 out of the 13 groundwater wells fall inside not only G^+ , G^- but also K^+ , K^- boundaries, which suggests that rock–water interaction is the major source for dissolved ions in HUA. In general, there are no significant changes in the weight ratios $Na/(Na + Ca)$ and $Cl/(Cl + HCO_3)$ between the dry and rainy seasons as well as TDS in HUA. Note that two groundwater samples from well H3 show the highest TDS and high weight ratios $Na/(Na + Ca)$ and $Cl/(Cl + HCO_3)$ among the HUA wells for both seasons, and thus fall outside the K^+ and K^- boundaries. Upon the closer inspection of land use, this well is located in an agricultural area of intensive irrigation (Dijk et al. 2012). In addition, the irrigation and drainage systems have problems such as inadequate capacity, inadequate grade, and absence of (or ineffective) water control structures, accompanied by rapid increases in canal seepage (Asia Development Bank 2000). Therefore, it is

reasonable to infer that agricultural activities cause the groundwater samples of H3 to fall outside the domain of rock dominance.

To determine the functional source of the dissolved chemical components in the PCA groundwater, the Gibbs diagram was also created as shown in Fig. 10. This figure indicates that there are no significant differences between the dry and rainy seasons for the groundwater samples in PCA, which is similar to the result in HUA. However, from Figs. 9b and 10b, it is interesting to note that the weight ratios $Cl/(Cl + HCO_3)$ of the PCA groundwater samples are obviously different from those in HUA. The ratios in the PCA samples are extremely low, less than 0.1 in 11 out of the 13 sampling wells, whereas the same ratios in the HUA samples reach values up to 0.4. This is consistent with the Piper diagram for PCA (Fig. 8), where the $[HCO_3^-]$ type is exceedingly dominant in the PCA groundwater (except for sampling well P6 as explained later). This observation implies that the PCA groundwater chemistry is controlled by the dissolution of carbonate minerals, whereas the HUA groundwater composition is affected by the dissolution of carbonate minerals and surface water. Figure 10 also shows that the PCA groundwater

Table 5 Water types of groundwater samples in HUA and PCA

Sampling wells	HUA		PCA	
	Dry season	Rainy season	Dry season	Rainy season
1	[Ca ²⁺ –HCO ₃ [−]]	[Ca ²⁺ –HCO ₃ [−]]	[Ca ²⁺ –HCO ₃ [−]]	[Ca ²⁺ –HCO ₃ [−]]
2	[Ca ²⁺ –HCO ₃ [−]]	[Na ⁺ –HCO ₃ [−]]	[Ca ²⁺ –HCO ₃ [−]]	[Ca ²⁺ –HCO ₃ [−]]
3	[Na ⁺ –HCO ₃ [−]]	[Na ⁺ –HCO ₃ [−]]	[Mg ²⁺ –HCO ₃ [−]]	[Ca ²⁺ –HCO ₃ [−]]
4	[Ca ²⁺ –HCO ₃ [−]]	[Ca ²⁺ –HCO ₃ [−]]	[Ca ²⁺ –HCO ₃ [−]]	[Ca ²⁺ –HCO ₃ [−]]
5	[Ca ²⁺ –HCO ₃ [−]]	[Mg ²⁺ –HCO ₃ [−]]	[Mg ²⁺ –HCO ₃ [−]]	[Ca ²⁺ –HCO ₃ [−]]
6	[Ca ²⁺ –HCO ₃ [−]]	[Ca ²⁺ –HCO ₃ [−]]	[Na ⁺ –Cl [−]]	[Na ⁺ –Cl [−]]
7	[Ca ²⁺ –HCO ₃ [−]]	[Ca ²⁺ –HCO ₃ [−]] ₃	[Ca ²⁺ –HCO ₃ [−]]	[Ca ²⁺ –HCO ₃ [−]]
8	[Ca ²⁺ –HCO ₃ [−]]	[Na ⁺ –HCO ₃ [−]]	[Ca ²⁺ –HCO ₃ [−]]	[Ca ²⁺ –HCO ₃ [−]]
9	[Ca ²⁺ –HCO ₃ [−]]	[Ca ²⁺ –HCO ₃ [−]]	[Ca ²⁺ –HCO ₃ [−]]	[Ca ²⁺ –HCO ₃ [−]]
10	[Ca ²⁺ –HCO ₃ [−]]	[Ca ²⁺ –HCO ₃ [−]]	[Ca ²⁺ –HCO ₃ [−]]	[Ca ²⁺ –HCO ₃ [−]]
11	[Ca ²⁺ –HCO ₃ [−]]	[Ca ²⁺ –HCO ₃ [−]]	[Na ⁺ –HCO ₃ [−]]	[Mg ²⁺ –HCO ₃ [−]]
12	[Mg ²⁺ –HCO ₃ [−]]	[Ca ²⁺ –HCO ₃ [−]]	[Ca ²⁺ –HCO ₃ [−]]	[Ca ²⁺ –HCO ₃ [−]]
13	[Ca ²⁺ –HCO ₃ [−]]	[Mg ²⁺ –HCO ₃ [−]]	[Na ⁺ –HCO ₃ [−]]	[Ca ²⁺ –HCO ₃ [−]]

samples were plotted mostly inside the G⁺, G[−], K⁺, and K[−] boundaries. This suggests that rock–water interaction is the main factor controlling the chemical composition of the PCA groundwater. However, two groundwater samples from well P6 clearly deviate from the others by falling outside the boundaries G⁺ and K⁺ in Fig. 10a and outside boundary K[−] but inside G[−] in Fig. 10b. On the other hand, the Piper diagram for PCA (Fig. 8) shows that the [Na⁺] and [Cl[−]] types dominated in this well in both seasons. Another noteworthy point emerging from the Gibbs diagram for HUA (Fig. 9) is that the HUA groundwater samples at the same location (well H6) have low weight ratios Na/(Na + Ca) and Cl/(Cl + HCO₃). According to Tanabe et al. (2003), during the Holocene, the sea transgressed the flood plain as far inland as the present location of Hanoi. The transgression during the Holocene, induced by sea level rise, must have caused an intrusion of seawater into the underlying high-permeability Pleistocene sediments. Therefore, it is presumable that in the area of the P6 well, salty paleowater affects the deeper aquifer (PCA), whereas the HUA groundwater is affected by rainfall and surface water. Earlier research also indicated that the groundwater in the south of Hanoi is more salty than the other parts (Tong 2007).

Conclusion

The main objectives of this study are to investigate the changes during dry and rain seasons in hydrogeochemical facies and to determine factors governing water chemistry of groundwater not only in HUA but also in PCA. In this paper, taking advantage of the unique database, hydrogeochemical parameters from 13 conjunctive sampling wells

for HUA and PCA in Hanoi acquired during dry and rainy season in 2011 were comprehensively analyzed. Interpretation of hydrogeochemical analysis revealed that the concentrations of most ions are higher in HUA than in PCA throughout the year. Moreover, TDS, Ca²⁺, K⁺, SO₄^{2−}, Cl[−], and NO₂[−] values decrease from the dry season to the rainy season in HUA, but increase in PCA.

In terms of hydrogeochemical facies, from analysis of the Piper diagrams for HUA and PCA, the following generalizations were obtained as the groundwater properties in Hanoi in 2011: the [Ca²⁺] type groundwater is quite abundant in both aquifers; almost all groundwater in the 13 conjunctive observation wells is of the [HCO₃[−]] type during the dry and rainy seasons in both aquifers. A change in the hydrogeochemical facies was detected from the dry to the rainy season in at least 30 % of the sampling wells in both aquifers. The change particularly occurs in the cation type (i.e., [Ca²⁺] to [Mg²⁺] or [Na⁺], [Mg²⁺] to [Ca²⁺], [Na⁺] to [Ca²⁺] or [Mg²⁺]), whereas the anion type remains unchanged. Differences in the hydrogeochemical facies between HUA and PCA were also observed in the majority of the observation wells (6 out of the 13 observation wells during the dry season and 7 out of 13 during the rainy season).

The Gibbs diagram suggests that rock dissolution is apparently the dominant process affecting the groundwater composition in both aquifers. The results from the Gibbs diagram show no significant changes in the weight ratios Na/(Na + Ca) and Cl/(Cl + HCO₃) as well as the TDS of the groundwater samples in both seasons and both aquifers. The results also indicate that the PCA weight ratios Cl/(Cl + HCO₃) are obviously different from those in HUA. The weight ratios Cl/(Cl + HCO₃) are extremely low in PCA, less than 0.1 in most of the samples, whereas in the

Fig. 9 Gibbs diagram for HUA groundwater in Hanoi: **a** Gibbs cation diagram, **b** Gibbs anion diagram

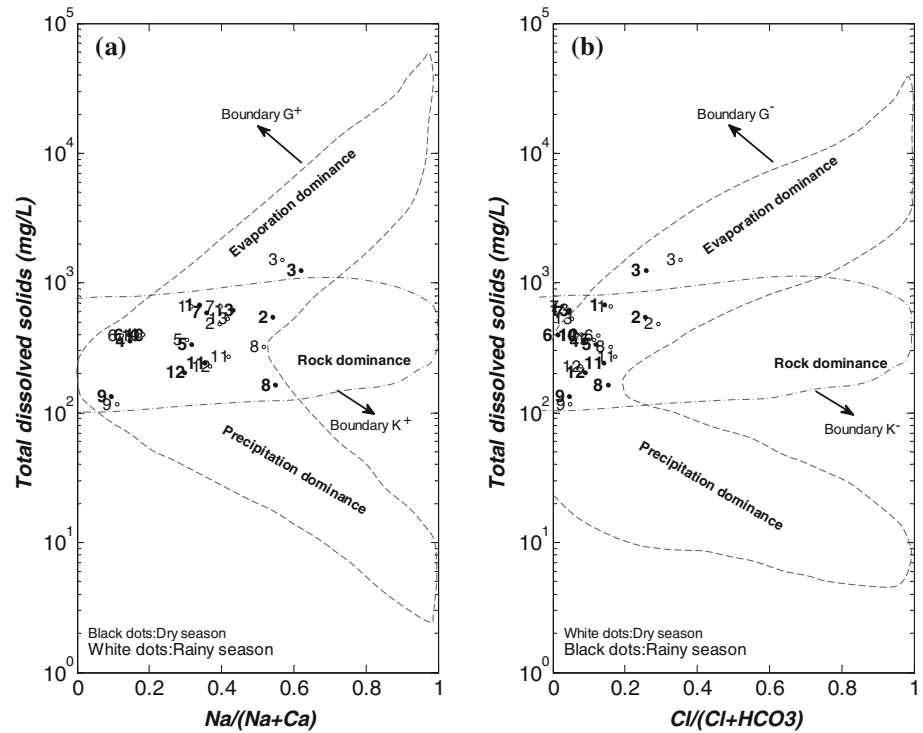
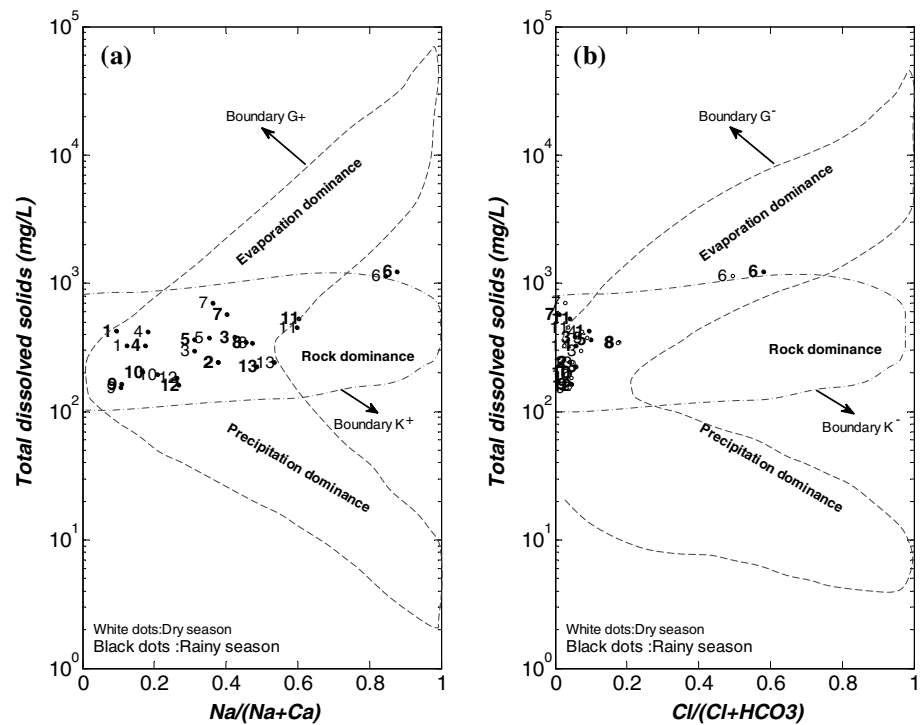


Fig. 10 Gibbs diagram for PCA groundwater in Hanoi: **a** Gibbs cation diagram, **b** Gibbs anion diagram



HUA samples, they were scattered within the zone of rock dominance. Furthermore, it is reasonable to argue that agricultural activities and salty paleowater are the additional factors that cause some samples to fall outside the zone of rock dominance. The findings of this study provide

valuable information regarding the groundwater hydrogeochemical properties and hydrodynamics at present in Hanoi, Vietnam.

In this study, seasonal changes in groundwater hydrogeochemistry in Hanoi were detected by using major ion

data of one year (2011). Although it is not enough for providing a general conclusion about seasonal changes in hydrogeochemical properties of groundwater in Hanoi in terms of a long-term period, the results of this study provided a quite detailed view of hydrogeochemical characteristics at present (2011). This study is the first attempt to investigate the seasonal changes in hydrogeochemical properties in the study area; hence the findings of this study will be the first step for further investigation of seasonal changes in groundwater hydrogeochemistry in the study area. It is recommended that more observation data in a long-term period in the future should be analyzed to confirm the seasonal hydrogeochemical characteristics of groundwater in the study area.

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