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砷的發生對越南湄公三角洲之威脅

Threat of Arsenic Occurrence in the Vietnamese Mekong Delta

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摘 要

高溶解砷在地層孔隙水中移動已被世界廣泛報導，地下水中高濃度的砷對人類健康及地下水利用造成威脅。全球已有許多文獻針對含砷的地下水進行研究，但近年來，越南紅河及湄公河三角洲砷的發生才引起科學家的注意。本文主要目的在於回顧亞洲孟加拉及越南沖積扇平原砷的汙染，接著經由在越南湄公三角洲安江省安富縣的近期調查，於 2014 年乾濕季時收集 36 個地下水樣本及 2 個沉積物核心，說明湄公三角洲砷發生的證據。調查結果顯示，研究區域內砷的濃度介於 208 到 1,523μg/L 之間，遠高於世界衛生組織(WHO)及越南飲用水的標準(10μg/L)。此外，乾季時地下水砷的濃度比濕季時更高，且砷的濃度隨著沉積物深度增加而增加，在 25 公尺深時高達 201ppm。

關鍵詞：砷、地下水、湄公三角洲

Abstract

The occurrence of high dissolved Arsenic (As) levels mobilized from geogene sources in pore water has been reported worldwide. High elevations of Arsenic concentration in groundwater have posed threats to human's health and implication for groundwater use. Previous studies on As-rich groundwater around the world have been in the literature. Particularly, Arsenic occurrence in the Red River and Mekong River deltas of Vietnam has recently drawn much attention from scientists. The purpose of this paper is to review Arsenic contamination in alluvial plains of Asian deltas of Bangladesh and Vietnam. Further, an additional evidence of Arsenic occurrence in the Mekong Delta region will be

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reported through our recent survey in An Phu district, An Giang province of the Vietnamese Mekong Delta. Thirty six (36) groundwater samples and two (02) cores of sediment were collected in the rainy and dry seasons of 2014. The investigated results show that Arsenic concentration in groundwater resources in the study area is much higher than the WHO and Vietnam drinking water standard (10µg/L), ranging from 208 to 1,523µg/L. In addition, the arsenic concentration in groundwater in the dry season tends to be higher than in the rainy season. Arsenic concentration in sediment cores increases with depths, reaching to 210 ppm at 25m depth.

Keywords: Arsenic, groundwater, Mekong Delta.

Overview of arsenic around the world

Arsenic is a ubiquitous element in the Earth's crust, exists in the mineral gypsum, soil, water and organism (Smedley *et al.* 2002). Arsenic is ranked No.20 on the popularity and was one of the highly toxic substances on human health. Further, the International Agency for Research on Cancer classified Arsenic and its compounds as Group 1 – group of toxic carcinogenic to humans (IARC 1987).

Since the 1980s, Arsenic poisonous catastrophe was an alarming issue to humans in many parts of the world. There have been many reports on the quality of water resources, particularly in groundwater, with high concentration of arsenic which posed threats to communities in many countries such as Argentina, Bangladesh, Chile, USA, Canada, France, Taiwan, Mongolia, India, China, Japan and Vietnam. The geographical map of arsenic concentrations was illustrated by Smedley *et al*. (2002) with arsenic elevations greater than 50µg/L.

Arsenic contamination of groundwater is particularly severe in the Asian deltas. Ganges - Brahmaputra Delta (Bangladesh) and Bengal Delta (India) are most severely affected regions with an estimated 63 million people exposed to arsenic through the high arsenic concentration in groundwaters (Chowdhury *et al.* 2000).

In Bangladesh, the use of contaminated surface water by microbiology and toxic substances has resulted in diseases such as cholera intestinal, diarrhea and dysentery. The use of groundwater resources to serve the daily needs is a result of UNICEF’s policy to promote alternative water sources from 1970s to 1990s. The aim of this policy is to reduce the rate of child deaths due to diseases related to the intestines. This program has achieved remarkable success at reducing the mortality rate among children from 151 to 83/1000 children, in the period of 1960-1996 (Meharg 2005). However, the UNICEF’s program induced an unexpected effect in which about 27 million people have been estimated in using the groundwater with high concentration of arsenic (>50 µg/L) in 1998-1999. Currently, about 50 million people of Bangladesh (40% of the country’s population) are exposed to arsenic coming from wells (Smith *et al.* 2000; Hossain 2006; Ravenscroft *et al.* 2009).

Similarly, the occurrence of Arsenic in groundwater has been a serious issue for human’s health for communities in the two major deltas of Vietnam. In the Red River delta region, the maximum level of arsenic in groundwater was 845µg/L, the average value was 39µg/L (Berg *et al.* 2007). According to

Nguyen *et al.* (2009), the concentrations of arsenic in groundwater was 211-348 µg/L and more than 40% of people were estimated in facing the risk of arsenic poisoning.

In the Mekong delta region, the arsenic contamination in groundwater is also exacerbated. The average value of arsenic concentrations in groundwater is not as high as the Red River delta region (Pham *et al.* 2005). However, the highest arsenic concentration value was recorded in An Giang province of the Vietnamese Mekong delta, reached the peak at 1,351µg/L (Hoang *et al.* 2010), which is about one-hundred times higher than the WHO drinking water standard (10µg/L).

The Mekong River Delta

2.1 Geographical Setting

The Mekong River Delta, one of the largest deltas in Asia, is located in Southern Vietnam, and has been recognized as the most productive agricultural land and an important economic development zone in Vietnam. The Vietnamese Mekong Delta covers 13 provinces: Long An, Tien Giang, Dong Thap, Vinh Long, Tra Vinh, Hau Giang, Soc Trang, Ben Tre, An Giang, Kien Giang, Bac Lieu, Ca Mau and Can Tho City.

The Tien (Mekong) and Hau (Bassac) rivers, two major branches of the Mekong River, form a wedge shaped delta in Vietnam which is known as the Mekong Delta. These two branches subsequently divide into nine tributaries (so-called *Nine Dragons*) before entering the East Sea (MRC 2010). The Mekong Delta covers about 4 million hectares of arable land (12% of the country's area). This Delta is the home of nearly 18 million inhabitants, accounting for 22% of Vietnam's population (Tran *et al.* 2007).

2.2 Socio-economic conditions

Nearly one quarter of the Vietnamese population lives in this region contributing to 18% of the country's GDP, 90% of rice exports and 73% of farmed aquatic products (GSO 2011). About 2.6 million ha in the Mekong Delta are used for agricultural practices due to its mostly flat terrain and few forested areas, which is one fourth of Vietnam's total area (General Statistics Office 2012). The largest areas of agricultural land are Can Tho and Hau Giang, exceeding 80%, whilst Ca Mau and Bac Lieu provinces account for 32% and 42%, respectively. The region's land used for growing cereals makes up 47% of national products, more than northern and central Vietnam combined (GSO 2012).

Most importantly, the Mekong Delta is the "Rice bowl" of Vietnam which accounts for 80% to 85% of Vietnam's rice export (Vo *et al.* 2007, Nguyen *et al.* 2007). Rice output in 2011 was 23,186,000 tons, accounting for 54.8% of Vietnam's total output. The largest producers are Kien Giang, An Giang and Dong Thap provinces, producing almost 11 million tonnes together. Rice production in any two of these provinces is more than the entire Red River Delta (GSO 2012).

2.3 Geology

According to Ta *et al.* (2002), the Mekong Delta was formed by a deposition of sediment discharged from the Mekong River system in the late Holocene, around 3,000 years ago. The Cenozoic Mekong complex can be distinguished into eight hydro-geological units, namely Holocene (*qh*), Upper Pleistocene (qp_3), Upper- Middle Pleistocene ($qp_{2\text{-}3}$), Lower Pleistocene (qp_1), Middle Pliocene (n_2^2), Lower Pliocene (n_2^1), Upper Miocene (n_1^3) and Upper-Middle Miocene ($n_1^{2\text{–}3}$). Each hydrogeological unit of this delta has been divided into two parts. The upper part is composed of a low permeable silt, clay or silty clay. The lower part is a permeable one that consists of fine to coarse sand, gravel, and pebble with medium to high water yield. In generally, the yield of all aquifers varies from medium to high (1 to >5 L/s), exclusively the Holocene aquifer due to its more complex composition (Wagner *et al.* 2012).

2.4 Hydrology

Besides the vast river system, surface water and groundwater are the main water sources and decisive driver of socio-economic activities of the Mekong delta region. However, this delta has been facing with profound challenges in the use and management of water resources both quantity and quality, including water pollution, salt intrusion and acid sulfate soil (Le *et al.* 2007). The situation is exacerbated in the age of global change and climate risk events which pose negative effects on aquatic and terrestrial ecosystems of the delta region.

The Vietnamese Mekong delta is crisscrossed by an elaborate array of rivers belonging to the Mekong system and man-made canals which were created to enable irrigation and transport activities. Water dominates the landscape yet the region faces severe problems of freshwater supplies in some localities. These problems are linked to increasing periods of low flows during the dry season, pollution by the extensive use of agrochemicals and the release of industrial and household's contaminants and by salt intrusion from coastal areas. Surface water is therefore costly to clean in order to bring to acceptable drinking standards and many people and communities still drink polluted surface water throughout the region.

In the Mekong delta, groundwater has been long used for domestic, agricultural and industrial purposes. In coastal zones, groundwater is a crucial freshwater source for human daily basic needs as surface water has been facing with many serious problems such as acid sulfate soil, non-point pollutants during the wet weather and salt intrusion in the dry season (Tran Dang An *et al.* 2014). Further, the decline of water discharge due to the recent development of hydropower in the upstream part of the Mekong River basin lead to salt intrusion more severe in coastal areas of the Vietnamese Mekong delta. These factors make groundwater has become a more important freshwater resource for irrigation and domestic purposes of local residents.

Occurrence of Arsenic in groundwater in the Mekong Delta

3.1 Previous Surveys in the Mekong Delta

The contamination of Arsenic in groundwater in the Mekong Delta region was first reported by Stanger *et al*. (2005). Arsenic concentrations in groundwater at shallow depths (100-120 m) exceeded WHO standards (10 µg/L). Following studies indicated that As-rich groundwater was found in many provinces of the Vietnamese Mekong Delta, including Dong Thap and An Giang (Berg *et al.* 2007). In this study, 112 groundwater samples were analyzed for the purpose of overall Arsenic contamination assessment in these provinces. Results showed that the Arsenic concentration in groundwater ranged from <1 - 845 µg/L (average 39 µg/L). In another research, Shinkai et al. (2007) studied about the Arsenic occurrence in groundwater in Dong Thap and Tien Giang province. The Arsenic concentration of all groundwater samples from Tien Giang was below the WHO standards. Meanwhile, in Dong Thap, Arsenic concentration in 3 of 8 groundwater samples exceeded, the highest value was recorded at 321µg/L.

The occurrence of Arsenic in groundwater in Kandal province of Cambodia and areas bordering to Vietnam was also found (Berg *et al.* 2007). Remarkably, the Arsenic concentration in these areas ranged from <1 to 1610 µg/L (average 217 µg/L, n = 207). Furthermore, Buschmann *et al.* (2007) conducted 112 well samples in Southern Vietnam, 108 samples in the north and 132 in the south of Phnom Penh (Cambodia) to assess the occurrence of arsenic in groundwater. The study showed that Arsenic concentration in 37% of samples exceeded WHO standards (10 µg/L) in which 26% of them had arsenic concentrations over 50 µg/L. The average value of Arsenic concentration in groundwater was recorded at 92 µg/L (range from <1 to 1340 µg/L). Most importantly, the percentage of Arsenic-rich groundwater in Vietnam is less than in Cambodia, 22% in comparison to 44%, but the number of wells with high Arsenic levels is prevalent in Vietnam.

From 2003 to 2005, UNICEF has conducted a large-scale investigation on the occurrence of Arsenic in groundwater in the entire area of the Vietnamese Mekong Delta. The survey's results showed that most groundwater resources in the Mekong Delta region are contaminated by Arsenic with concentrations ranging from 10 to 50 µg/L. Accordingly, high elevation of Arsenic was found in Long An, Dong Thap and Kien Giang provinces with 56% (a total of 4,876 wells); 67% (of 2,960 wells) and 51% (of 3,000 wells) respectively. Particularly, Arsenic contamination was seriously in An Giang province, with 16% of the wells has concentration is below 50 µg/L and 40% is over 50 µg/L in total of 2,699 wells surveyed. Further, the Department of Science and Technology of An Giang carried out an additional survey for 6,293 wells throughout the province. It is importantly highlighted that Arsenic-contaminated groundwater is severely found in districts along the Bassac River, including An Phu, Phu Tan, Tan Chau and Cho Moi. Importantly, An Phu district was the worst arsenic contaminated area in comparison with others as depicted in Table 1.

Hoang *et al.* (2010) examined the extent of arsenic contamination in groundwater and sediments at four (04) provinces of the Mekong Delta, including Kien Giang, An Giang, Dong Thap and Long An. This study revealed that the average levels of Arsenic concentration were 110 µg/L and 57µg/L in An Giang and Dong Thap, respectively, which were much higher than WHO drinking water standard and

Vietnam National Technical Regulation on Drinking Water Quality standard (QCVN 01:2009/BYT).

Recently, high levels of Arsenic in groundwater were recorded in An Giang, Dong Thap, Can Tho, Tien Giang and Soc Trang provinces (Erban *et al.* 2013). The concentration of arsenic in groundwater tends to decrease gradually, from northwest to southeast of the Mekong Delta region. The high elevation of Arsenic in groundwater was found at depths less than 50 m in An Giang and over 200 m in Tien Giang. Similarly, high concentration of Arsenic in groundwater was also observed in the Red River Delta by Winkel *et al.* (2011). The highest levels of Arsenic concentration in the Red River and the Mekong Delta were 810 µg/L and 1500 µg/L, respectively.

Table 1 Arsenic concentration of groundwater in An Giang province (Tran *et al.* 2011)

No.	District/City	Number of well	Arsenic concentration (percentage)		
			< 10 µg/L	10 – 50 µg/L	> 50 µg/L
1.	An Phu	973	42 (4.3%)	144 (0.4%)	787 (80.9%)
2.	Chau Phu	210	142 (67.6%)	35 (32.2%)	33 (15.7%)
3.	Chau Thanh	652	644(98.8%)	7 (15.1%)	2 (0.3%)
4.	Cho Moi	964	613 (63.6%)	217 (6.6%)	134 (13.9%)
5.	Phu Tan	930	368 (39.6%)	206 (4.3%)	256 (27.5%)
6.	Tan Chau	490	325 (66.3%)	67 (13.5%)	98 (20%)
7.	Thoai Son	499	485 (97.2%)	11 (19.5%)	3 (0.6%)
8.	Tinh Bien	1,105	1,072 (97%)	33 (8.8%)	0 (0%)
9.	Tri Ton	2,829	2,812 (99.4%)	15 (3.5%)	2 (0.1%)
10.	Long Xuyen City	369	347 (94%)	18 (25.5%)	4 (1.1%)
11.	Chau Doc City	71	68 (95.8%)	3 (134.9%)	0 (0%)
Total		8,992	6,917 (76.9%)	756 (0.9%)	1,319 (14.7%)

3.2 Latest Survey on Arsenic Occurrence in An Phu district, An Giang province Study area

The study area is An Phu district of An Giang province which is close to the border with Cambodia and bends along the Bassac River (Fig. 1). About 88 % of existing wells in An Phu district are being used to serve for domestic (drinking and hygiene demands) and irrigation purposes. Groundwater is mainly extracted at Holocene aquifer with average depth of 25 m. Since 2010, total withdrawal of groundwater in An Phu was 3,829.5m^3/day (DONRE 2010). Therefore, the use of groundwater resources with high arsenic elevation might be at a considerable risk of arsenic poisoning for local people.

This investigation was conducted by our collaborative research group which was teamed by researchers from Ho Chi Minh City University of Technology (Vietnam) and EPFL at Lausanne (Switzerland). The purpose of this investigation, in associated with indicated elevation of As level from previous studies, is to examine changes in the concentrations of arsenic in groundwater by seasons in a specific location of the Mekong Delta.

3.2.1 Sampling and analyzing methods

Three sampling campaigns were done in 2014: January (dry season), May (ending of dry season – beginning of rainy season) and August (rainy season). The wells are located along the Bassac River and the Provincial Road No.956 and were clustered into 3 Areas 1, 2 and 3 (Fig. 2). The average distance from near-river wells to the river bank is 195 meters (range 88-275 m); and far-river wells is 621 meters away from the river (range 408-1014 m). Groundwater in the study area is primarily used for agricultural irrigation (corn, peppers, green beans, etc.) and domestic purposes.

Totally, 36 groundwater samples were collected from local farmers' wells at qh23 aquifer (Holocene) as a preferable aquifer for exploiting groundwater with depth of wells ranges from 13m to 37m under land surface. Well water is pumped and flushed in 10 – 15 minutes for oxygen stability before sampling. All samples were processed under anoxic conditions by using glove-bag or glove-box, and were filtered by 0.22 µm syringe filter (Hydrophilic Polypropylene, PALL 66 557). Hydrochloric acid 2M and Nitric acid 5M were added into filtered samples, and stored in cool conditions for analyzing. Two sets of core samples (at Area 1 and Area 2) were drilled at the depth of 35m (the common depth of local wells). Sediment samples were collected at different layers of the core's vertical dimension.

Geochemistry of groundwater including pH, ORP, DO and EC were on-site measured by Portable meter (HACH HQ40d Multi); total Iron (Fe) and total Arsenic (As) were analyzed by ICP-EOS (Shimadzu® ICPE-9000 Analyzer).

3.2.2 Results and discussion

Arsenic concentration in groundwater of surveyed wells ranged from 208 to 1,523 µg/L which are much higher than WHO guideline standard (10 µg/L) and the QCVN 01:2009/BYT (10 µg/L) and QCVN 09:2008/BTNMT – groundwater quality standard (50 µg/L). The variation of Arsenic concentration by seasons and locations (near-river and far-river wells) is shown in Fig. 3 and 4.

The concentration of Arsenic in January was much higher than in May and August. The difference of Arsenic concentration between May and August is not significant. Interestingly, 17 wells were sampled in January 2014, which were coincided with the surveyed wells by UNICEF in 2005, the concentration of arsenic tends to increase from 1.3 to 11.5 times.

There is not much variation of Arsenic level by seasons or the location of wells. Similar to the Ganges-Brahmaputra Delta (Bangladesh - India), geological characteristics and sediments of the Mekong River delta originate from the Himalayas (Kocar *et al.* 2008) which formed alluvial delta with low and flat terrain from the late Holocene (Ta *et al.* 2002). Furthermore, the recent study of Reid *et al.* (2014) also showed a positive correlation between the concentration of arsenic in sediment and depth in An Phu district of An Giang province (arsenic concentration increases with depth from 0-25 m). Similar results were also reported by Tran *et al.* (2011), arsenic concentration increases with depth of wells

(0-60 m). The high level of arsenic in sediments in An Giang is derived from alluvial sediment; geological characteristics and the correlation between arsenic and sediment are similar to the Brahmaputra river delta of Bangladesh as conducted by Halim *et al.* (2009).

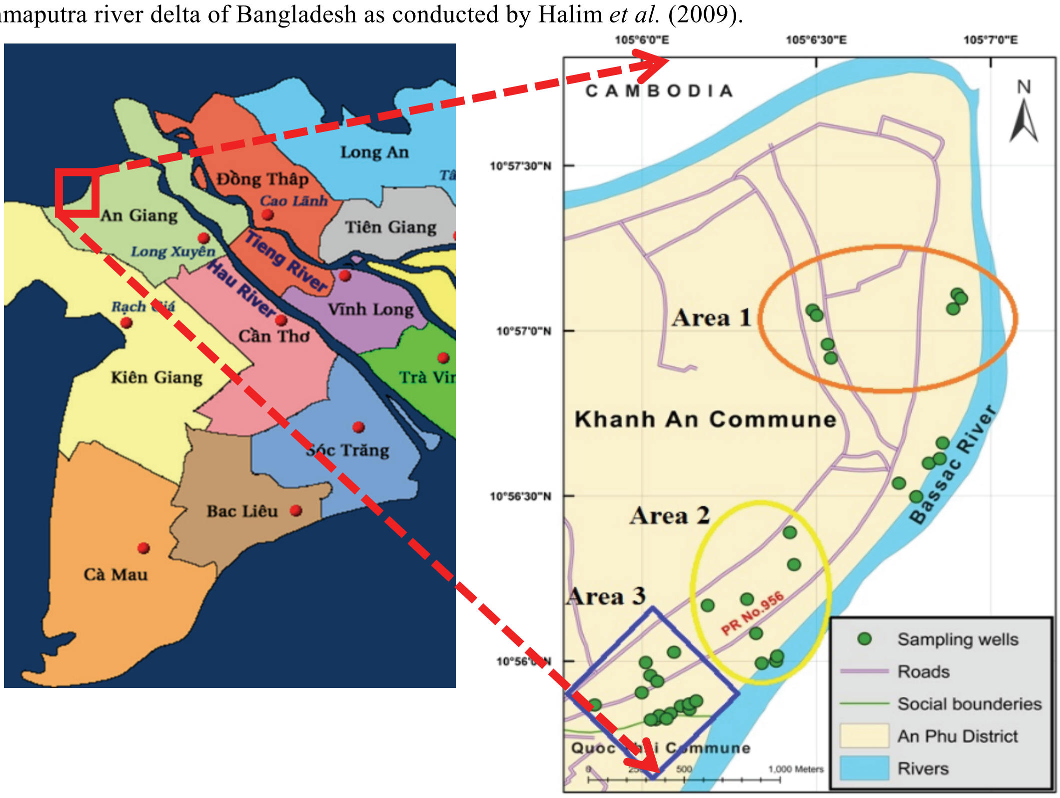

Fig. 1 Study area/ Sampling locations

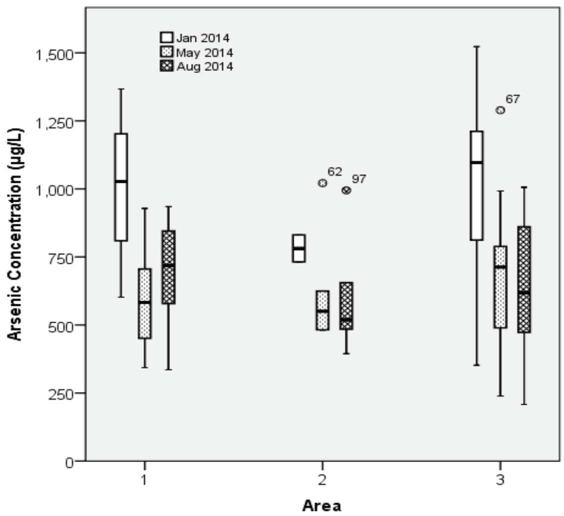

Fig. 2 Arsenic concentration in groundwater of An Phu district (by Seasons and Areas)

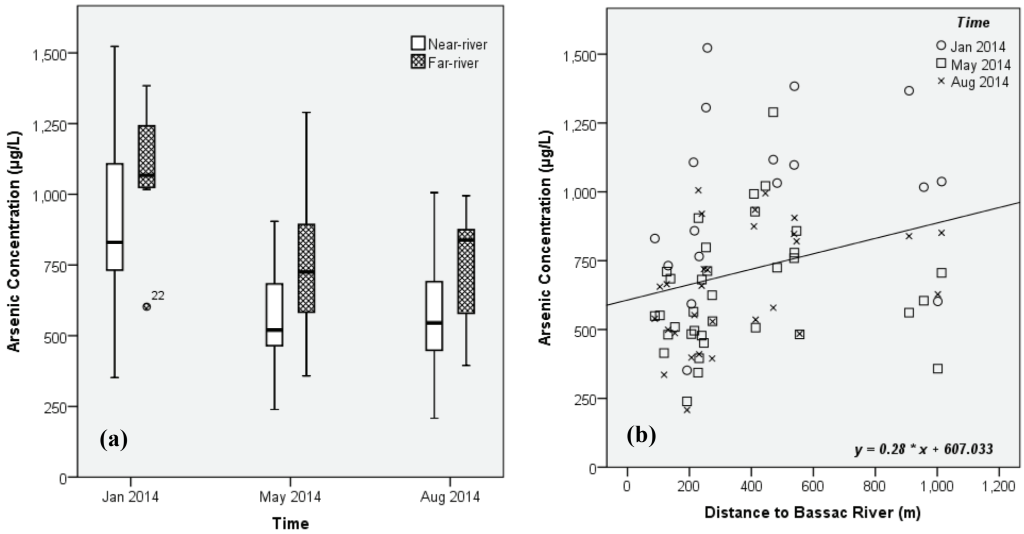

Fig. 3 (a) Spatial variation of As concentration in groundwater in An Phu district by seasons; (b) The correlation between As concentration and the location of wells

The concentrations of arsenic in groundwater are high in January (dry season), but there is a downward trend in the rainy season, except for the Area 1 with a slight increase in the rainy season (Fig. 2). The ranges of Arsenic concentrations in groundwater are vast in Area 1 and Area 3 (Fig. 2). Overall, far-river wells (the wells located far from the Bassac river) had high elevation of Arsenic concentrations than the near-river wells (Fig. 3a). Using the correlation analysis by bivariate analysis (Pearson's and Spearman's coefficient) with the Confidence Interval Percentage value was set at 95 %, the positive correlation was found between As versus distance (Fig. 3b). This difference of arsenic concentration between the far-river and near-river wells would pose implication for future groundwater management and use in An Phu district, An Giang province.

Core drilling conducted in the area of An Phu district to analyze the concentration of arsenic in the soil, the results showed that arsenic concentration in sediment samples increases from <1 to 210 ppm, corresponding to the depth from 1 to 25 m. Given the drilling core at the study area, it is highlighted that there is an increase in arsenic level from the peat layer and the highest level of arsenic in soil was recorded in the sandy layer, greater than 200ppm (Fig. 4).

With the respect of Arsenic occurrence in Holocene aquifer of the study area, it could be explained that arsenic in the sediments may be associated with iron oxyhydroxides and release to the groundwater by reductive dissolution of iron. The cause of arsenic occurrence groundwater resource of the Mekong Delta region may be similar to Bangladesh due to both the Mekong River and the Ganges-Brahmaputra river basin originates from the Himalaya (Kocar *et al.* 2008). In addition, the Mekong Delta was formed by a deposition of sediments from the Mekong River in the late Holocene, around 3,000 years ago (Ta *et al.* 2002). These foctors can be the attributes to the natural occurrence of arsenic in groundwaters in the

Mekong Delta region.

Previous studies on the process of Arsenic releasing from sediments into groundwater in West Bengal Delta and Bangladesh showed the Arsenic concentration in soil is directly proportional to the percentage of Iron in soil (Nickson *et al.* 2000). However, the results of our study showed the stark contrast in the Mekong Delta region. The concentrations of Arsenic and Iron in the soil were inversely proportional with the depth of sediment. These results are entirely different with the mechanisms of arsenic release from the soil into the groundwater comparing with the previous studies.

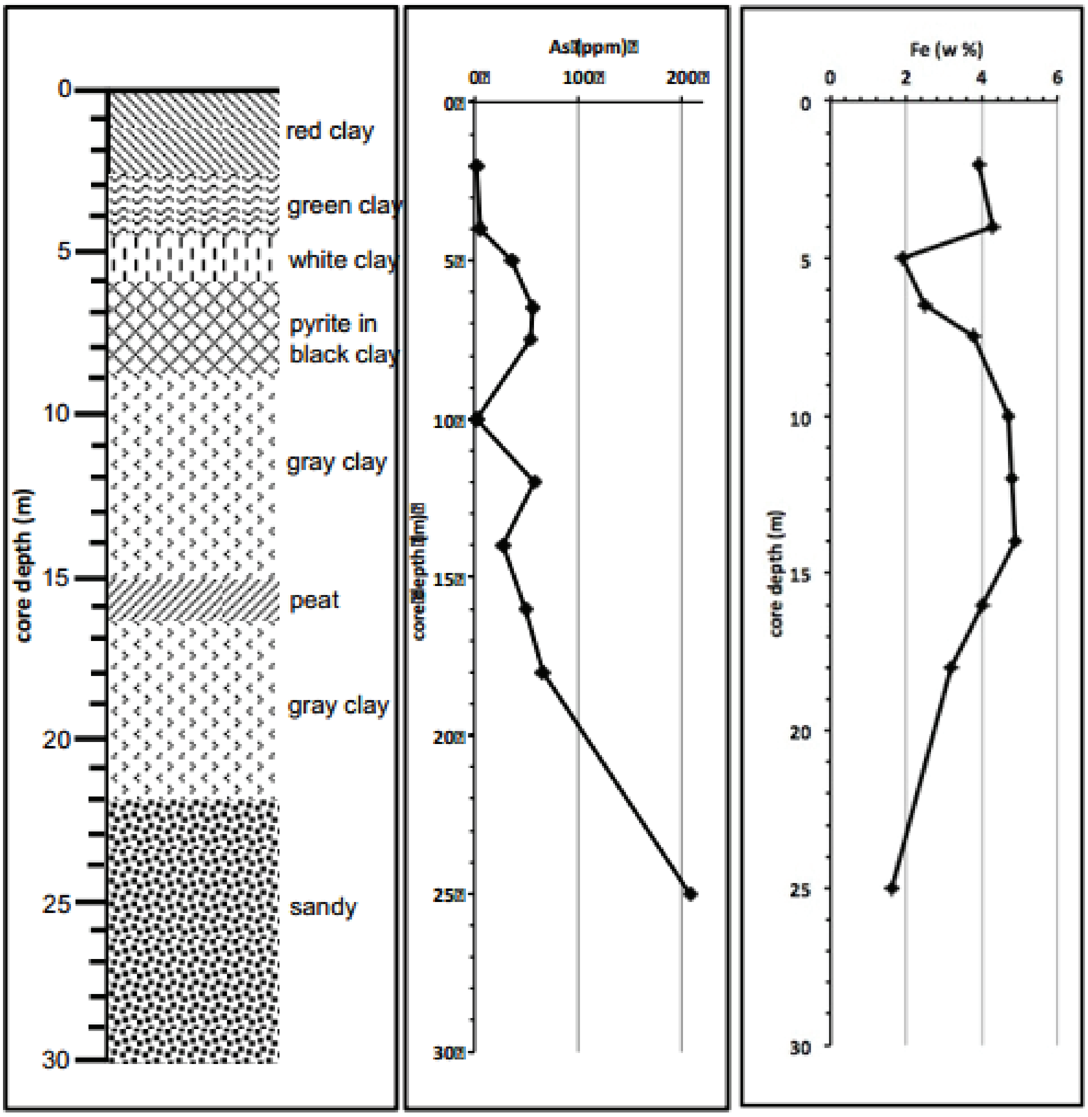

Fig. 4　Core composition, arsenic concentration and percentage of iron with depth

Additionally, the Vietnamese Mekong Delta is a lionshare of the country's agricultural products, which leads to the huge consumption of agro-chemical products and fertilizers. As a result, aresenic contamination in groundwaters can be also resulted from the overuse of fertilizers and pesticides in agricultural activities. Therefore, further studies are needed to identify point or non-point pollution sources of arsenic and the underline cause of arsenic contamination in groundwater and sediments in An Phu. Most importantly, in order to have a comprehensive understanding on the mechanism causing arsenic contamination in groundwater of the Vietnamese Mekong Delta, additional investigation on geo-hydrological characteristics and dynamics of groundwater are a vital of concern.

Conclusions

The occurrence of Arsenic in groundwater in the Mekong Delta region is at an alarming level, which is much higher than WHO and Vietnam drinking water standards. The high level of Arsenic concentrations would pose threats to human health of local communities consuming groundwater for agricultural practices and domestic use. The surveyed wells are shallow (Holocene), with depth ranges from 13 m to 37 m; and are located in agricultural areas along the Bassac River. The cause of Arsenic occurrence in groundwater and sediments in the Vietnamese Mekong Delta region is mainly attributed to natural factors of alluvial deposition forming the delta which could be similar to the West Bengal delta and Bangladesh of the Ganges – Brahmaputra river basin. Furthermore, the largest contribution of Vietnam's agricultural production in associated with huge consumption of agro-chemical products and fertilizers could be an additional factor causing Arsenic contamination for groundwater. However, to understand and clarify the mechanism caused high elevations of Arsenic in groundwater and soil, there is a need for future research which should be more specific and detailed to identify the underline mechanism of arsenic release in groundwater in the Vietnamese Mekong Delta.

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