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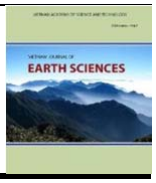

High arsenic concentration in groundwater related to sedimentary facies in the Mekong River Delta, Vietnam

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ABSTRACT

Arsenic (As) contamination in groundwater has been recognized in the Mekong River Delta (MRD) since 2001, especially in Dong Thap and An Giang provinces. The investigations were carried out during 2008-2009 and from 2012 to present. The investigations of As distribution in groundwater and sediments were based upon the field, and laboratory analyses using field kit (Hironaka, 1998) and AOAC laboratory - Vietnamese standard 6626-2000 (Vilas). Results show that the As concentration ranging from 15 to 1,650 µg/l is found in 520 private tubewells at depth between 15 and 90 m, while being rare or very low at depth between 200 and 300 m. High As content is usually found in 15-70m tubewells in which approximately 70 - 80% of tubewells having arsenic levels of >500 µg/l are located at some areas in Dong Thap and An Giang provinces in the upper MRD plain. Bore core drilling records show a probable relationship between As concentration in groundwater and sedimentary facies in the MRD. The highest As content is found along the Mekong River valleys in the Late Pleistocene and Holocene aquifers.

Keywords: Mekong River Delta; Arsenic contamination; Groundwater; Sedimentary facies.

1. Introduction

The Mekong Delta, one of the largest deltas in Southeast Asia, is located at the mouth of the Mekong River in Southern Vietnam. The river has its headwaters in the Tibetan mountains, drains a catchment area of 800×10^3 km^2, discharge 470 km^3 of water and 160 million tons of sediment per year into the South China Sea. The area of the delta is approximately 62,520 km^2. Regional rainfall of 1,000-2,000 mm/yr is mainly concentrated in wet season between May and November. In dry season, groundwater becomes a drinking water source even around the Mekong River channels owing to considerably polluted river water. Thus numerous wells are undertaken to solve living water requirement, especially in drinking.

Mekong River Delta is a vast flat plain with main agricultural productions such as rice, shrimp, fish and so on. Its population is about 18-20 million people mainly living in the rural areas. In dry season, living water is limited in the rural areas. Around 1980s, UNICEF assisted project for groundwater use in the rural areas, and then people had dug wells by themselves. The exploitation of groundwater to replace the use of surface water has been made and become an important contribution to reducing the

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incidence of water-borne diseases in the rural areas. Most living water of rural people is obtained from groundwater through several ten to hundred meter wells. A large quantity of groundwater has been exploited at the depth from 15-90 m in Holocene and Pleistocene aquifers. However, the improvement in water quality has been negatively affected by the presence of As in groundwater, which is now widely recognized as a threat to public health. Since 2001, we have investigated As contamination in shallow groundwater and discovered a mediate-high level of As contamination of 15-90 m wells in the upper delta plain, especially in Dong Thap and An Giang provinces. Two local small research projects have been carried out and As contaminated distributions are recognized. Moreover, As contamination of groundwater is also found in Kien Giang, Can Tho, Vinh Long and Tien Giang provinces. Recent discovery and report on elevated hazardous concentrations of As in shallow aquifers in the MRD (Nguyen et al., 2009) raise the specter of future deleterious health impacts on population that particularly in rural areas, extensively use groundwater for drinking and irrigation purposes. A combination of As concentration in groundwater and sedimentary facies from the bore cores in the MRD gives a good opportunity to investigate deposition facies related to sea-level changes in the Holocene. Moreover, the results of distribution of As concentration in groundwater and sedimentary facies changes from bore cores in Cambodia are useful to this study.

2. Geological setting

A high sediment yield caused from high precipitation due to the monsoonal climate, in combination with slightly falling sea level over the last 6ky, have resulted in the 62,520 km^2 delta plain in which 52,100 km^2 in Vietnam (Nguyen et al., 2000). Geomorphologically, the sub-aerial delta plain can be divided into an upper delta plain dominated by fluvial processes, and a lower delta plain characterized by a well-developed beach-ridge system and mainly influenced by marine processes. A sea-level curve for the last 15 ky has been established in the MRD (Ta et al., 2002). Estimation of the Last Glacial lowstand of sea-level is about -120 m in Southeast Asia at around 18 to 20 ^{14}C ky BP. The drop in sea level led to the lowering of base level of the Mekong River, and formation of an incised valley system of over -60 to -70 m. The Late Pleistocene sediments were mainly composed of stiff, slightly oxidized, yellowish gray silt, fine- medium sand bearing scattered quartz pebbles and laterite and dated 43.4 and 50.4 ^{14}Cky.BP. The following Holocene deposits unconformity covered the Late Pleistocene sediments with a sharp contact in lithology and color. The Holocene transgressive incised-valley filling succession is 40-45 m thick estuarine marine facies dated 15-7 ky.BP., and finally overlain by 10-13m thick open bay mud facies dated 6.0-5.5 ky.BP. During the sea-level highstand and the subsequent slight fall in the last 6-5 ky, delta prograded over 200 km from Cambodia - Vietnam border to the South China Sea. In general, delta presents a complex sequence of depositional facies ranging from coarse to fine sand, mud, peat with rapid facies variation both vertically and laterally. In the north, Plain of Reeds and Long Xuyen quadrangle, an individual sub-to intertidal mud facies has been preserved and dated 6-4.5 ky.BP. It is characterized by 2-5 m thick sub-to intertidal flat mud facies and common mangrove peat and peaty clay. Moreover, among Mekong and Bassac river areas, shift channel deposits are mainly characterized by channel, channel bar sand and sandy silt facies. In the south-east part, active delta, it is composed of delta progradation sequences made of an overall coarsening upward succession of prodelta, delta front, sub-to intertidal flat facies, related to slightly falling sea-level since 5-4.5 ky. In the southern part, Ca Mau deltaic margin, it is characterized by 15 to 20 m thick delta/shelf mud facies with well-developed mangrove marsh on the subaerial delta plain.

3. Data collection and analytical methods

This study is based on the following data: Investigation of data on As contamination in groundwater in Dong Thap province was carried out from 2008-2009 by project of Department of Science and Technology. On the base of this result, more detailed study has been taken since 2012 by field survey and analysis in laboratory. Firstly, As contents in groundwater from 15-360 m in depth were tested by As Test (Hironaka, 1998) method in fieldwork, a total of approximately 550 samples. The results showed distribution of As contents in different well depths. Based on these results, around 100 groundwater samples from different depths and locations were collected to analyze in the laboratory. As contents were analyzed by High Resolution Coupled Plasma Mass Spectrometry with a detection limit for As of ~0.1 µg/l in Hoan Vu Analysis Service Center in Ho Chi Minh City, Vietnam (AOAC in laboratory - TCVN 6626-2000 Vilas). Moreover, other compositions were also analyzed with methods as bicarbonate (SMEWW 2320, 2012); sulfate, chloride, nitrate (SMEWW 4500, E-2012); calcium, magnesium (SMEWW 3125, 2012-ICP-MS) and total iron (SMEWW3500-Fe.B, 2012).

We collected 8 columnar sections of groundwater bore core drillings from 15-360 m in depth to identify characteristics of aquifers and aquitards. Moreover, three bore core sites, named DT2, DT3 and DT4 were drilled around 80-90 m in depth to clarify geological characters in detail (Figure 1, 2). These data are used to establish hydrogeological section (Figure 3).

4. Results and discussions

4.1. Geological characteristics of aquifers and aquitards

The identification of characteristic of aquifers and aquitards plays an important role in assessment of As concentration of groundwater (Ahmed K.M., 2004; Research Group for Applied Geology, 1999). Dividing the aquifers and aquitards is based on previous borehole data and results of sedimentary characteristics of 6 geological boreholes (Nguyen et al., 2009). Section AB is established along Tien River from Hong Ngu to Cao Lanh (Figure 1). Aquifers are often formed from the layers or lenses of fine to coarse sand and they are separated from each other by aquitards. The aquitards are composed of layers or lenses consisting of clayed silt or silty clay separated by upper and lower aquifers, but in some places there is the contact of two different aquifers. The results show that from top to bottom there are 5 aquitards and 4 aquifers named Clay 1, Sand 1, Clay 2, Sand 2,... and Clay 5. Particularly in the area of Hong Ngu district, Mesozoic basement is found at 190-260 m in depth. Geological sedimentary characteristics of 5 aquitards and 4 aquifers are described from top to bottom (Figure 3) respectively as follows:

4.1.1. Aquitard C1

Aquitard C1 is formed by layers or lenses of gray, gray-brown fedora, white gray yellow red patchy clayed silt or silty clay and has a thickness of approximately 20 to 60 m. This aquitard with bedding form is exposed on the surface extending from Hong Ngu to Cao Lanh with thickness from 15 to 60 m, or silty clay lenticular form with 40-60 m thickness is found at the depth of 30-65 m from Hong Ngu to Cao Lanh. Clay-silt-sand layers with a thickness of approximately 4-6 m often appear at the top of aquitard C1. They are natural levee deposits and probably quite favorable places for surface water recharges (river water and rain water). Grain size analysis results showed 18-32% sand, 28-50% silt and 24-32% clay; a high sand content of about 29-38% was found in some locations of Hong Ngu district. The clay, clayed silt lenses of

about 4-30 m thickness are usually found at depths of 15 to 60 m from north to south and divide aquifer S1 into upper and lower parts. This shows groundwater from upper and lower parts of aquifer S1 is often interconnected in the north and south areas.

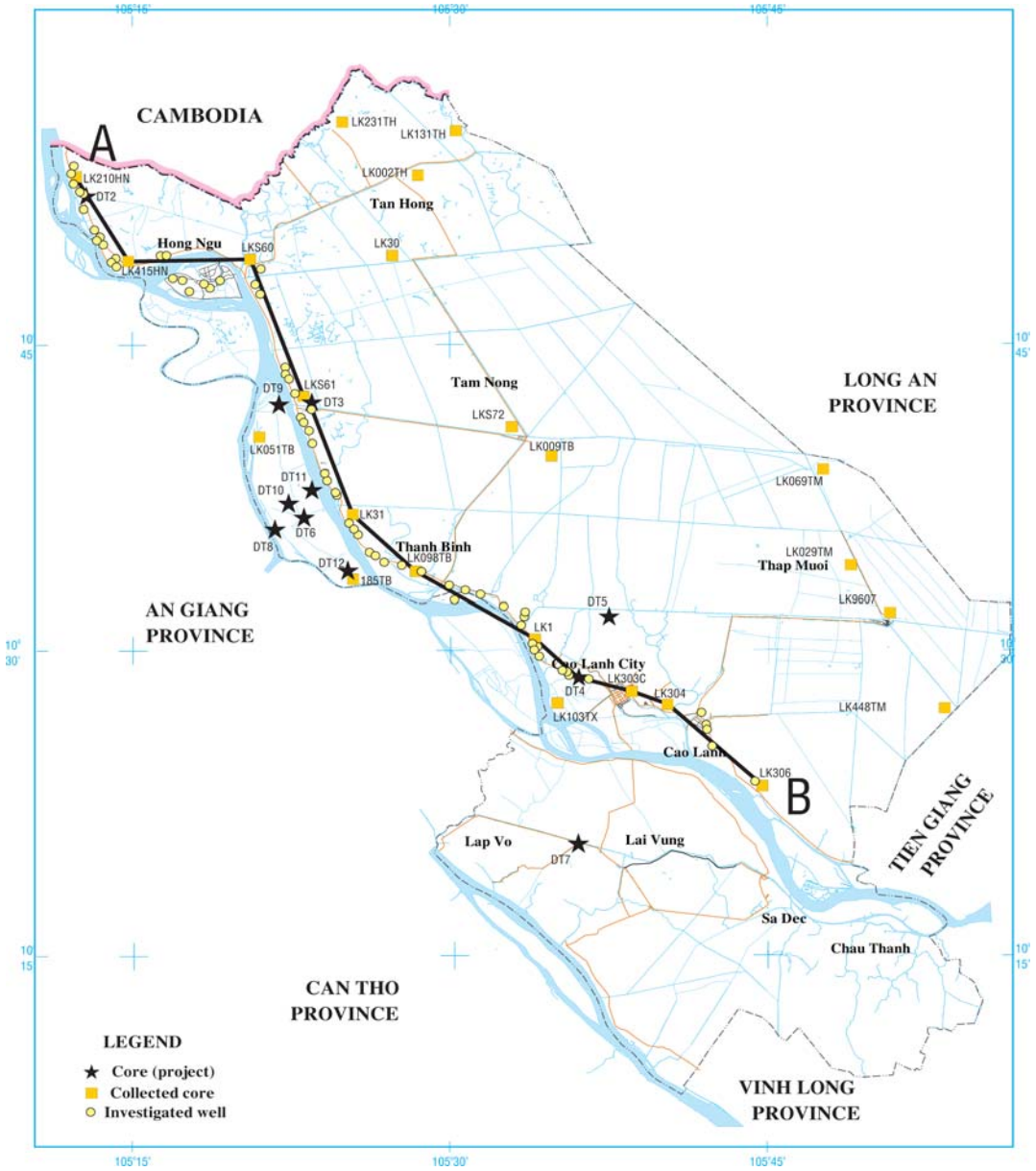

Figure 1. Location of cores and cross section AB in Dong Thap Province

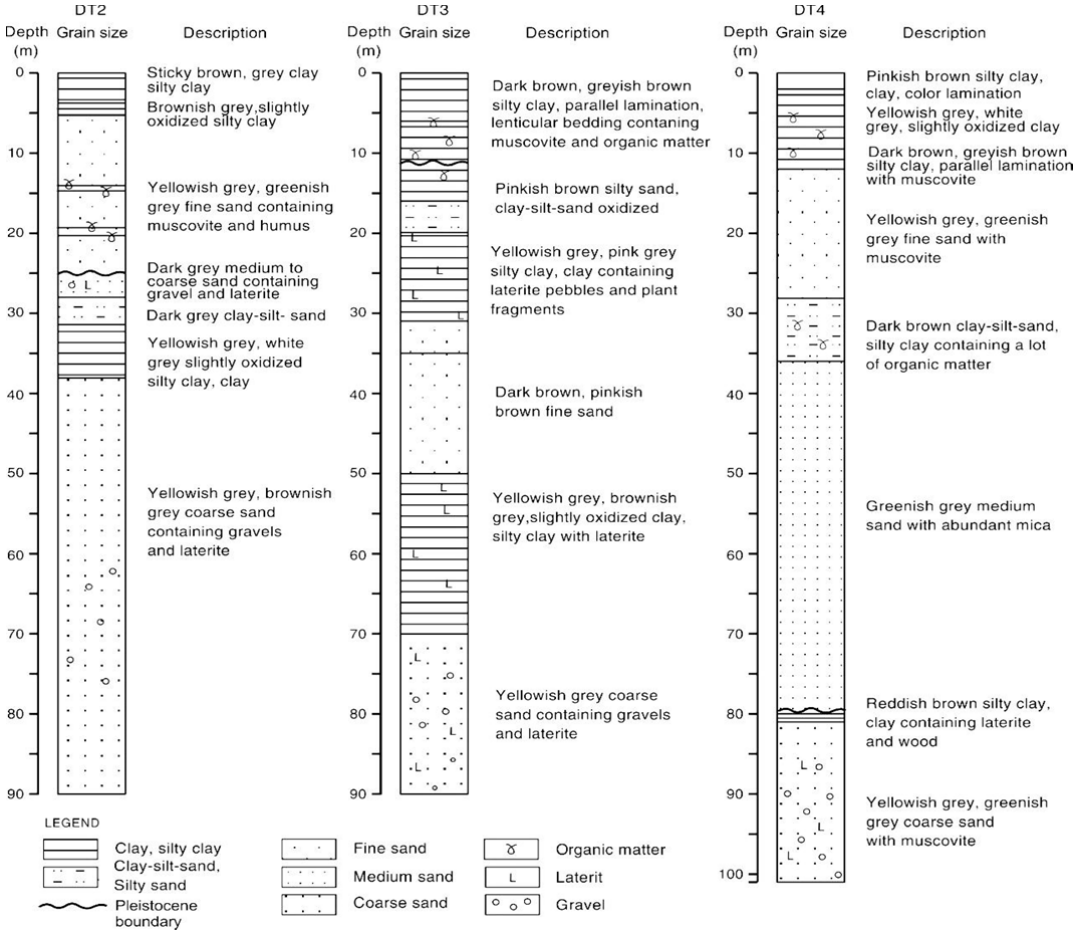

Figure 2. Lithological characteristics of DT2, DT3 and DT4 cores

4.1.2. Aquifer S1

Aquifer S1 with a thickness of about 40-80 m is divided into upper, middle and lower parts:

The upper part of aquifer S1 (S1a) has a thickness of about 10 to 15 m, is usually found at depths of 6 to 30 m. The S1a is composed of the alternate layers in the aquitard C1 with composition of fine, fine-medium and medium quartz sand and mica. Lens forms with a thickness of about 10 to 20 m are recovered in the northern area, layer forms with a thickness of about 8-15 m are found at depths from 10 to 30 m and extend in southern area.

The middle part of aquifer S1 (S1b) is found at depths of 30 to 60 m. It has a thickness of approximately 15 to 25 m and its composition is of mainly gray, light gray, gray-yellowish-brown fine, fine-medium sand. In the northern area such as Tan Hong and Hong Ngu, the S1b is found with alternate lenticular or layer forms of clayed silt, clayed silty sand.

The lower part of aquifer S1 (S1c) contains gray, yellowish gray-brown fine-medium, medium-coarse sand and some pebbles, gravels. The S1c is distributed fairly constantly from north to south area, its thickness is about 20 to 40 meters, specially

medium-coarse, coarse sand with a thickness of approximately 30 to 40 m was found at 40-115 m in depth from Hong Ngu to Thanh Binh. The top of S1c is commonly found at a depth of 60 m and the bottom of S1c is in depth of 80-115 m. This is an important aquifer and it has been exploited widespreadly. S1b and S1c parts are often connected with each other in the north and south areas such as Hong Ngu, Tan Hong, Tam Nong and Thanh Binh, however they are often separated by the silty clay lenses in Tam Nong, Thanh Binh, Cao Lanh, Lap Vo.

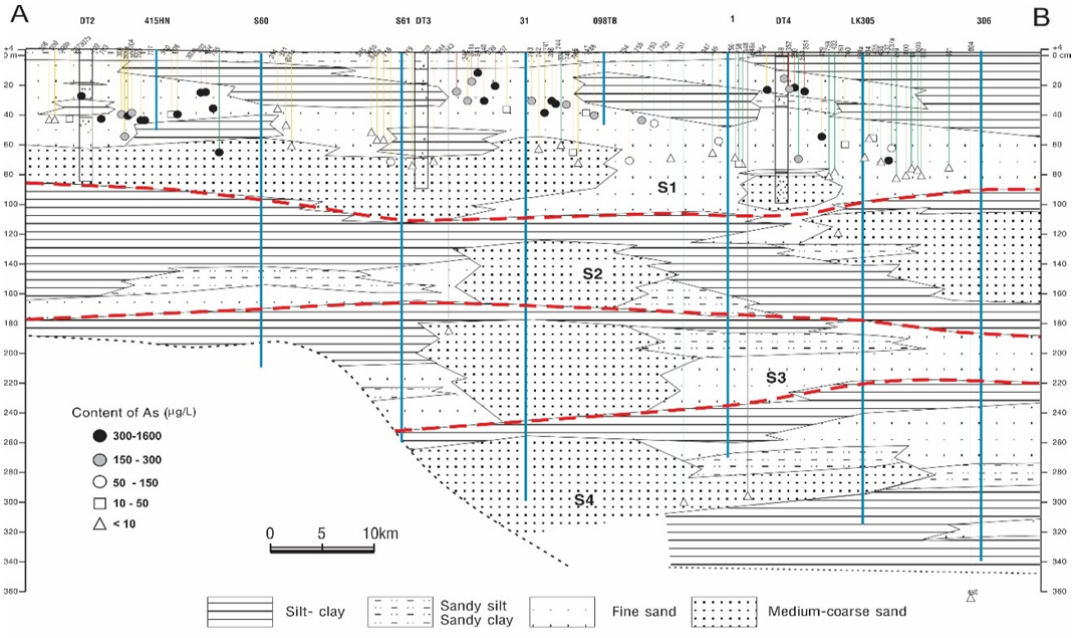

Figure 3. Distribution of arsenic contamination in the aquifers and hydrogeological section AB

4.1.3. Aquitard C2

Aquitard C2 with 20-80 m thickness is composed of gray, gray-green, gray-brown clay, silty clay. The top of aquitard C2 is found at depths of 80-90 m and the bottom is around 165-190 m in depth. This aquitard is rather typical by the largest thickness and distributes leaning from north to south area. In the north area from Hong Ngu to Tam Nong, the aquitard C2 has fairly stable thickness of about 40-80 m, so it plays an important role in separating aquifers S1 and S2. Particularly in the north and south areas such as Tan Hong and Cao Lanh, due to C2 formed by the lenses of about 15 to 45 m thickness, there is a transferability condition of groundwater between S1 and S2.

4.1.4. Aquifer S2

Aquifer S2 is composed of layers and lenses of light gray, yellowish gray-brown fine, fine-medium, and coarse sand with a thickness ranging from 15 to 60 m. The fine and fine-medium sand layers having thickness of about 15 to 20 m are usually found in Hong Ngu, Tan Hong and Tam Nong. They are protected fairly well by C2 and C3 aquitards in the upper and lower parts as shown in cross section AB. The medium-coarse and coarse sand lenses with a thickness of about 35-60 m appear at 105-170 m in depth from Thanh Binh to Cao Lanh. Besides, the medium-coarse and coarse sand lenses with a thickness of 30 to 60 m was found at depth of 105-165 meters in the south of the province from Cao Lanh City to Cao Lanh district. These lenses

are separated pretty well from the below aquifer S3 by the C2 aquitard, whereas they are often connected directly with the above S1 aquifer that can be through between S1 and S2 and contribute to formation of underground flow from northwest to southeast directions.

4.1.5. Aquitard C3

Aquitard C3 is composed of white-gray, gray-brown red clay, silty clay with a thickness of about 15 to 3 m. This aquitard appears in the form of 25-40 m thick layer covering Mesozoic basement at the depth of 170-210 m in the north of the province; in the thin layer form with a thickness of 15 to 17 m in the southern area. It is connected with C2 aquitard in Cao Lanh city.

4.1.6. Aquifer S3

Aquifer S3 is mainly composed of fine-medium, medium-coarse and coarse sand with a thickness of 25-80 m and alternated with clay, silty clay lenses. Aquifer S3 is found from the south of Tam Nong district to Cao Lanh. This is a pretty good aquifer separated from S2 and S4 aquifers by C2 and C4 aquitards respectively. Particularly in Thanh Binh area, aquifer S3 was found in coarse and medium-coarse sand lenses with a thickness of about 60m.

4.1.7. Aquitard C4

Aquitard C4 is composed of gray-brown, grey green clay, silty clay with a relatively constant thickness around 15 to 30 m. This aquitard is found at the depth of 200 to 270 m and inclined to the north. Besides, some 6-18 m thick sandy silty clay lenses are found in Tam Nong area and Cao Lanh City. Although the thickness of aquitard C4 is lower in comparison to others, due to lithological characteristics and spread properties of this aquitard, it is a pretty good aquitard between S3 and S4 aquifers.

4.1.8. Aquifer S4

Aquifer S4 is composed of fine-medium sand, medium-coarse and coarse sand, usually found at 235 m to over 340 m in depth. Aquifer S4 has a fairly stable thickness of 45-60 m and is found from north to south except basement area from Hong Ngu to Thanh Binh. Aquifer S4 is also of some sandy silty clay lenses and inclined toward the north. Due to the presence of Mesozoic basement, S3 and S4 aquifers are found only from Tam Nong to Cao Lanh.

4.1.9. Aquitard C5

Aquitard C5 is composed of gray-brown, white gray yellow silty clay, clay with 70 meter thickness and is found at depth of 275-340 m in the south of area. Particularly in Cao Lanh area, sandy silt, fine sand lenses with 3-8 m thickness are alternated in aquitard C5.

4.2. Distribution of As in aquifers and some chemical characteristics of groundwater

Result of As concentration in groundwater shows distinct regional patterns and depth trends. The high As content of 400-1650 µg/l was revealed along the Mekong and Bassac rivers. In general, higher levels of shallow well containing >50 µg/l As are concentrated in Mekong and Bassac riparian zone and channel bars in the upper delta plain in the depth of 15-60 m. Lower levels have been documented in the remainder of the upper delta plain and Late Pleistocene-Holocene sediments at the north part of the delta in the depth from 70 to over 300 m (Figure 4). This regional pattern at least in part appears to be related to the sedimentary facies. Holocene alluvial and deltaic deposits generally contain groundwater elevated in As; while concentrations are very rarely elevated in Pleistocene deposits (>50 ky.BP). This is indicated by the very high fraction of wells deeper than 120 m containing <30 µg/l As. Redox conditions play an important role because of the strong association of arsenic with Fe oxyhydroxides (Research Group for Applied Geology, 1999). Holocene deposits are generally gray in colour and contain high proportions of Fe^{2+}. In contrast, Pleistocene deposits that are typically orange in colour contain mainly Fe^{3+} and little Fe^{2+}. There is still not truly oxic aquifers, however, since

groundwater associated with Pleistocene deposits rarely contains detectable dissolved oxygen and dissolved Fe concentrations can reach ~1mg/l. Groundwater at 15-60 m is a type of Ca-Mg-HCO_3. pH is from neutral to rather alkali, pH of water in the upper layer aquifer is around 6.40-7.40. Total Fe is about 0.40-10.5 mg/l, particularly 14-15 mg/l. There is a close relationship between total Fe and total As, the total As is in direct proportion to the total Fe (Figure 5). Total Mn is 0.60-4.60 mg/l particularly 5-6 mg/l. High content of As is usually found in 15-70 m wells, almost located in the areas of the upper delta plain around the Mekong and Bassac rivers. Some areas with a high density of 70-80% tubewells have As levels of >500 µg/l located in Dong Thap and An Giang provinces. As level in most groundwater from 200 to 300 m in depth is rare or very low (Nguyen et al., 2008). Recently concentrations increased with high levels around 15-60 m in depth in the shallow aquifers, especially, at the present channel bars in the upper delta plain of MRD.

4.3. Relationship between high As content and sedimentary facies

A combination of As concentration in groundwater and sedimentary facies from the bore cores shows that there is probably a relationship between As concentration in groundwater and sedimentary facies in the MRD. Most high As content is found around the MR valleys in the Late Pleistocene and Holocene periods.

High As content in the Late Pleistocene aquifer is found in the Tan Hong district, Dong Thap province, in the north part of the MRD (Nguyen et al., 2006). The Late Pleistocene sediments are at 3-4.5 m above present sea-level, mainly composed of sandy silty and very fine sand. The sediments are elongated in northwest to southeast directions. Moreover, the medium-coarse sands with poor sorting of fluvial channel deposits are found at 20 to 45 m from that As content usually ranges from 15 to 150 µg/l, particularly over 300 µg/l in some places. This aquifer is composed of sand body of fluvial channel deposits containing fresh water, meanwhile, groundwater is not found or only saline groundwater exists in the same depths outside the fluvial channel.

High As content in both the Late Pleistocene and Holocene aquifers is found around the recent Mekong and Bassac rivers in Dong Thap and An Giang provinces. The Late Pleistocene aquifer with As content of 80-350 µg/l is found in the fluvial channel deposits at 35-50 m in depth. The Holocene aquifer with As content usually ranging from 80 to 1650 µg/l is also found in the fluvial channel and channel bar deposits at around 20-90 m. As level is about 5.5-24 mg/kg in brownish grey silty clay and sandy silt with organic matters ranging from 10 to 35 m in depth. Fe content is 2-4%. Most sandy layers do not contain As and their Fe content is also low about 0.92-2.3%. Moreover, As content of modern natural levee is 8-20 mg/kg, but modern channel sands do not have arsenic concentration. As concentration is 70-80 µg/kg in agricultural productions in some places where As contaminated groundwater was used for irrigation. In spite of forming in different periods, these fluvial channel and channel bar deposits are usually located in or below tidal and inner bay mud facies formed during Holocene transgression. In DT1 core, the tidal flat consisting of peaty layers and inner bay mud facies is 20-28 m thick and dated 10-8.3 ky.BP., and delta initiation is around 8.2-8 ky.BP. in the upper delta plain location now (Ta et al., 2005; Nguyen et al., 2010). Probably these deposits should be in high As content because As of 15-25mg/kg is found in the tidal flat muddy facies dated 8-6 ky.BP. in the DT3 core. This matter is also found in the Cambodia where high As content in groundwater is found in recent alluvial valley of Mekong river. Sedimentary facies shows that the 16.5 m thick-tidal deposits consisting of peaty layers and dated 9 to 7 ky.BP. were found from the KS bore core drilling at alluvial valley in Cambodia. It suggests that extensive early to middle Holocene tidal and shallow marine deposits exist under the upper Mekong River lowland

in Cambodia (Tamura et al., 2009). Moreover, high As content in groundwater related to wetlands was documented in the upper MRD (Polizotto et al., 2008). The above mentioned data indicate that the early to middle Holocene transgressive sediments would be deposited at the lower topography and/or incised valleys of the Mekong River. The tidal deposits overlay the fluvial deposits that could be aquifer with As concentration in 25-45 m in depth. It indicates that there is a relationship between high As content and muddy facies formed in the Holocene transgression.

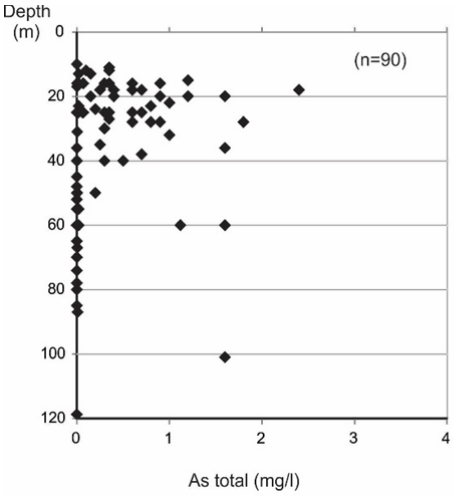

Figure 4. Relationship between of As total and depth

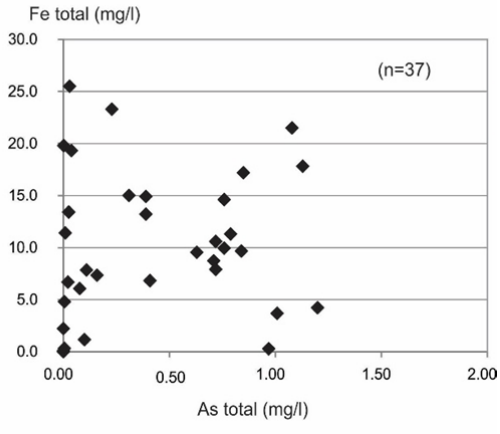

Figure 5. Relationship between of As total and Fe total

5. Conclusions

High content of As is usually found in 15-70m wells around the Mekong and Bassac rivers located in the upper delta plain of the MRD. Some areas with a high density of 70-80% tubewells have As levels of >500 µg/l located in Dong Thap and An Giang provinces. Most groundwater from 200 and 300 m in depth is safe.

A combination of As concentration in groundwater and sedimentary facies shows that there is a relationship between high As contents and tidal mud facies formed in the Holocene transgression. Most high As contents are found around the MR valleys in both Late Pleistocene and Holocene aquifers, but it is very high in the Holocene aquifer in comparison to the Late Pleistocene aquifer. This is the limited result, it is necessary to carry out further study on sedimentology and As concentration.

Acknowledgements

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