# Arseniferous groundwater in relation with geochemical conditions, Mekong Delta, Vietnam

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*Abstract:* - The Mekong delta is occupied by Holocene sediments mainly composed of alluvial unconformably overlying the Late Pleistocene sediments. The burial of sediments, rich in organic matter that lead to strongly reduced conditions result in arsenic contamination. Moreover, the inherently abundance of acid sulphate soil and pyrite in the Mekong Delta, along with low pH are favorable conditions for the release of arsenic. Arsenic concentrations in sediments range from 4 to 45 mg/kg. Where concentration of arsenic and iron are high, the colour of sediments is yellowish brown to reddish brown as well as iron oxides/hydroxides are present. In contrast, arsenic concentration is low where the sediments contain pyrite. Arsenic concentrations in groundwater range from 1μg/l to 746 μg/l. High chloride (>1000mg/l) and sodium (>500mg/l) concentration result from transgression and marine processes. A good correlation is observed between As, Fe, Mn and $HCO_3$ concentration. This correlation attests reduction of FeOOH and released arsenic to groundwater with contribution of organic matter. Water samples with high arsenic concentrations (>100 μg/l) have high Fe concentration (>15mg/l) and neutral pH; while samples with low As (<10 μg/l) have variable Fe concentrations and low pH (<7). There is an annual cycle of near-surface oxidizing and reducing conditions, corresponding to fluctuations of water level due to effect of flood. The oxidizing conditions would be consistent with pyrite oxidation and partial dissolution. Severely high arsenic concentrations are obviously caused by complete reduction of Fe hydroxides whilst moderate to low arsenic concentrations are plausible due to incomplete reduction of FeOOH that depends upon distant distribution of organic matter.

*Key-words:* - Arseniferous groundwater, hydrogeology, groundwater chemistry, Mekong delta, Vietnam.

## 1 Introduction

Arsenic is widely distributed as a trace constituent in rock, soils, natural waters and organisms. Soil and water containing high levels of arsenic and other trace elements can easily contaminate plants, animals and human being in contact with them as they either produce toxic effects or accumulate in plants and thereby enter animal and human food chain [6]. Elevated concentration of arsenic in groundwater has been reported in several countries in the world, among them, natural contamination of arsenic in Bangladesh, India, Vietnam, Cambodia, and Nepal is well known as effects of fluvial or deltaic environment.

The area of Mekong Delta (Fig.1) is densely populated and with a convenient condition for agriculture. Groundwaters belong to aquifers formed in continental sedimentary deposits, developed in a wide Quaternary plain ambient. The physical and chemical characteristics of hydrogeological system are mainly determined by the form of accumulated materials and nature of alluvial deposits.

Arseniferous groundwater has been reported in both Red River Delta and Mekong Delta with concentration above 500μg/l [10, 11]. In Mekong delta, high concentrations of arsenic (>300μg/l) are often encountered in the shallow aquifer (around 50m depth) in the area located in Chau Doc and Cao Lanh (Fig.1). However, abnormally high arsenic concentrations were also found in deep aquifer (≈100m depth). Where arsenic presents in groundwater, iron and manganese also present at a consistently high level. Industrial sources of arsenic in Mekong Delta are trivial comparison to natural sources [17]. Arsenic content of alluvial sediments is in the range 4 - 45mg/kg, only slightly greater than typical values of 2 - 6 mg/kg [4; 20].

The aims of this paper is to investigate the hydrogeochemical characteristics of groundwater and geochemical conditions of the alluvial sediments in order to understand complex geochemical conditions that yield high arsenic concentrations in the Mekong Delta, South Vietnam.
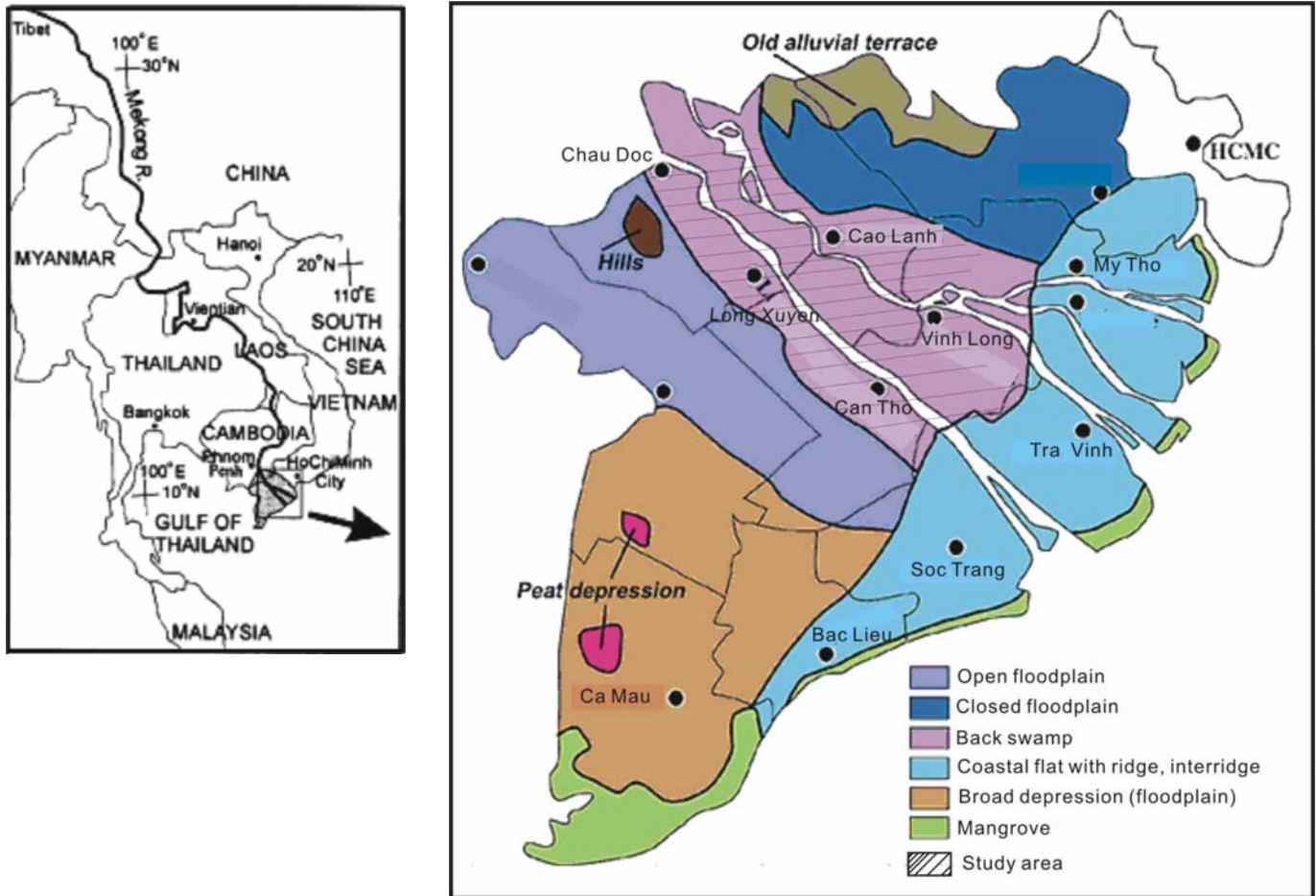


Fig.1. Location of study area and its topographical distribution.

## 2  Problem Formulation

### 2.1  Geological and Hydrogeological Setting

Most of the regions in Mekong delta have low elevation with the range of 0-4m above sea level and delta is very vulnerable to flood water from upper area and tidewater from lower sea (Fig.1). It is cited that delta plain show two parts: an upper delta plain dominated by fluvial processes, and the lower delta plain characterized by a well-developed beach-ridge system and mainly influenced by marine processes [13].

The study area is mainly conducted along Tien and Hau River (Fig.1, Fig.2). To northeast of Tien River, Dong Thap is a lowlying marshy plain, which contains potential aluminous soil. Along the two rivers there are high alluvial bank and natural dikes. Geologically, the area occupied by Holocene sediments mainly composed of alluvial unconformably overlying the Late Pleistocene sediments (Fig.2). The lowland flood-delta is entrenched over the Pleistocene terraces. Hills of older rocks occupied in the northwest of this area. The Late Pleistocene deposits or basement strata mainly composed of stiff, slightly oxidized, yellowish gray stiff silt, fine-medium sand bearing scattered quartz pebbles and laterite.

The Holocene sedimentary sequences are controlled by the changes in Late Pleistocene topography, sea level and sediment supply. These sequences have accumulated within the last 10 ky, with rapid transgression advancing the delta seaward. In addition to sea level changes, other factors influenced the thickness of Late Pleistocene strata and Holocene sediments are: (1) tectonic activities favoring fine-grained sediment deposition; and (2) high-energy fluvial events favoring coarse-grained deposits, led to variable thickness of the strata and sediments in Mekong Delta [14; 18]. During the Late Pleistocene low-

stand setting, medium sand and gravel beds were deposited. Parts of Pleistocene cover the delta in upland terraces in the west. These sediments were oxidized and well flushed because of low-stand settings. In the Early-Mid Holocene, sea level continued to rise and caused transgression, almost area in Mekong delta were inundated and occupied by tidal mangrove. In the interval of 6.5 to 4.5 ky, tidal predominant progradation occupied in the Dong Thap and An Giang [18]. The sediments comprise alternating layers of gravel, sand, silt, and clay forming several sub-aquifers.

Groundwater in the Holocene deposits is incised river silts and coastal sand dunes up to 20m thick. It is mainly saline water formed at the same time as sedimentation processes during the Flandiran transgression. However, it underwent changes by alluvial sedimentation covering the delta after the sea has retreated. Water level is from 0.5 to 3m. There is a hydraulic continuity between Holocene aquifers and surface waters.

Groundwater in the Upper to Mid Pleistocene is fresh to saline, 10-30 m thick widely used. This aquifer is mainly unconfined and underlain by Holocene aquifer
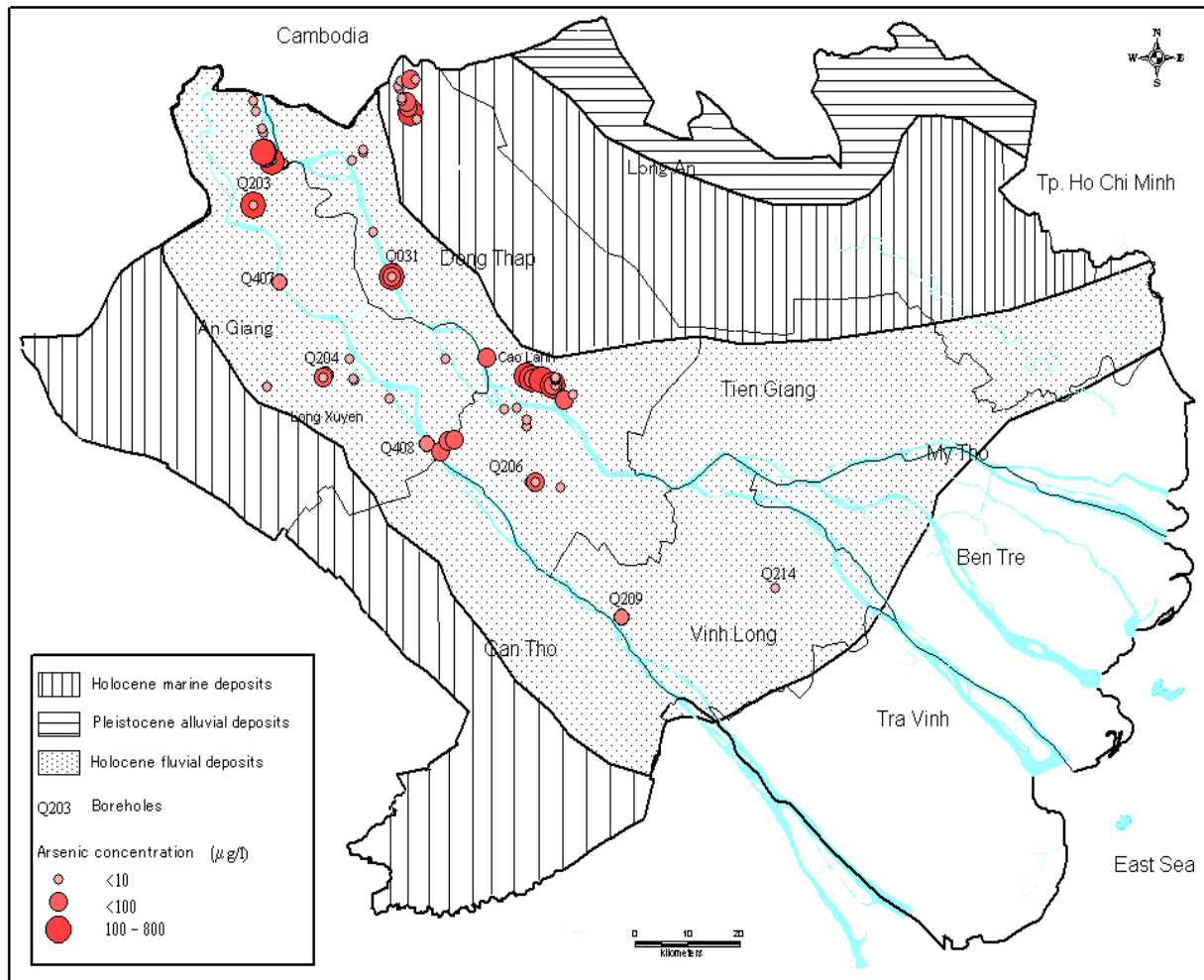


Fig. 2. Location of sampling sites and range of arsenic concentration in groundwater.

Generally, groundwater occurs in most of geological units, but not all form useful aquifers. It can be divided into two types of groundwater as (1) groundwater in the Holocene sediments is related to the leaching of pre-leaching salt and alum; (2) groundwater in the pre-Holocene sediments is believed to be fossil origin. There are silty and/or sandy aquifers sandwiched between thicker clay-rich aquicludes.

otherwise it distributes to surface in the east of the southern plain. Its recharge is predominant by rainwater and/or infiltration from river system in the area. Depth of the aquifer ranges approximately 44-73m. Aquifer consists of coarse sand intercalated gravels and grits. Water level reaches up to 21m.

## 2.2  Results

### 2.2.1 Groundwater Chemistry

Groundwater in this area has pH values in the range between 6.07 and 9.38 while field measure redox potential (Eh) values were variable from –303 to 625 mV. Electrical conductivity (EC) values were high, indicating an average value of 3043µS/cm.

Major compositions plotted on a Piper diagram indicate that the groundwater evolves from Na+K or mixed cations whereas rich bicarbonate and chloride for anion (Fig.3). In general, groundwater is dilute and mainly presented three types such as (1) Na-Mg- $HCO_3$ or Cl-Na-Mg- $HCO_3$; (2) Na-Cl-$HCO_3$; and (3) Na-Cl or Na-Mg-Cl. The occurrence of Na-Cl groundwater is predominant in Dong Thap. It is relatively high $SO_4$ (0-330 mg/l) but low $PO_4$ concentrations (0.013-5.25mg/l), probably due to anthropogenic influences. On the other hand, the large range of total dissolved ions primarily reflects the variations of $HCO_3^-$ (0-902mg/l) and $Cl^-$ (0-8570mg/l). Concentrations of dissolved $HCO_3^-$ reflect integrated microbial degradation of organic matter [17]. The origin of the chlorinity is apparently due to a transgression of East Sea up to Mekong Delta during Late Quaternary. DOC concentration in groundwater varied considerably below 1.5mg/l to as high as 58mg/l.

### 2.2.2 Total arsenic in groundwater and other redox species

Mean concentration of total arsenic in groundwater is about 90µg/l. The maximum value is 746µg/l, followed by 592µg/l and others around 300µg/l. In groundwater inorganic arsenic occurs in the 5+ and 3+ valency states [5]. The pH-Eh relationship indicated that groundwater in this area is more predominant of $As^{3+}$ than $As^{5+}$ (Fig.4). Arsenate and arsenite are mainly present as $HAsO_4^{2-}$ and $H_3AsO_3$, which are more stable species above pH 7 and up to pH 9, respectively. However, it can be explained that As(V) can be reduced to As(III) under moderately reducing conditions (Eh<100 mV), which can be induced by flood in Mekong delta (Fig.7a). High arsenic concentrations are obviously dominated by reducing conditions, only some samples present slightly oxidizing conditions (Fig.5).

In Dong Thap, concentration of arsenic exceed 600µg/l and most of high values are found (Fig.2). It is consistent with high concentrations of Mn and Fe distributed at depth (up to 100m) of aquifers (Fig.5). The peak of Mn concentration (20.8mg/l) shows at 36m depth while the highest concentrations of Fe in any depth even in deep aquifer (around 100m). Concentrations of $PO_4$ are range from 0.01 to 0.5mg/l; excluding samples have high values in deep aquifers, however, $SO_4$ decrease

downward. Values of DOC dominate in shallow aquifers around 20 to 30m depth (Fig.5). Arsenic concentrations are prevalent from 20 to 50m depth (Fig.6c). However, there are three samples present high arsenic content above 100m depth (Fig. 6c).

Samples were conducted in An Giang at depth above 30m. Most of groundwaters are free of arsenic (<50µg/l) (Fig.2). However, relative high arsenic concentrations (97-240 µg/l) were found around 50m depth of aquifers (Fig.6c). The maximum concentrations occur at depth of 57m (for As) (Fig.6c), 47m (for Fe) and 45m (for $PO_4$). Towards the base of deep aquifer, concentration of Fe, As and $PO_4$ decrease downwards.

It is obviously seen that concentrations of arsenic have good correlation with that of dissolved Fe and $HCO_3^-$ (Fig.7a, b). Both correlation of As and Fe show an axial intercept ≈ 200mg/l of $HCO_3^-$. This must represent the $O_2$ consumption and $NO_3^-$ reduction (Fig.6b, d) [12]. This relation will be used to elucidate FeOOH reduction in latter. Above 100m depth, arsenic concentration increase with depth (Fig.6c).

### 2.3.3 Arsenic concentration in sediment

Core sample of aquifer sediments was analyzed by XRF method for major elements and total arsenic. XRD method was used to determine mineral species in sediment. Arsenic concentrations in alluvial sediments in Mekong delta range from 4 to 45 mg/kg, but the great majority of measurements lie within the rang of 6-18 mg/kg. The variations in arsenic as well as Mn, Fe contents with depth are plotted graphically (Fig.7).

It is observed that clay and peat layer are richest in arsenic (37, and 45 mg/kg) while the silty and sandy layers contain significantly less. In generally, sand of aquifer is grey or slightly grey in colour over the entire depth of aquifer. At depth where sediments contain higher concentration of arsenic than other place, the aquifer sediments are brown, yellowish or reddish brown. In sediment samples, concentrations of arsenic and Fe are correlated relatively. High total arsenic and iron concentration occur where sediments contain iron oxide/hydroxide (goethite, hematite) mineralization at depth of 20-40m (37mg/kg), and 194m (45mg/kg) (Fig.7). The arsenic bearing hydrated Fe oxide coated clay (mainly illite, smectite) contain 101m (18mg/kg), 127.5m (15mg/kg), 155m (16mg/kg). The non-magnetic minerals consist of quartz, feldspar, and carbonate is low in arsenic content (around 4-5 mg/kg). Aside from these depth, arsenic concentration approach background levels of the rock such as 5-15 mg/kg for sandstones [12]. There is no appearance of arsenopyrite while pyrite is presented in sediments at any depth. All elements except manganese show a significant correlation with iron,

suggesting they were adsorbed by ferric oxyhydroxides. No sulfide minerals were found at any depth whereas small magnetite, randomly dispersed, companied by ferric oxide or hydroxide, illite and hematite minerals occur predominantly. It is mentioned that the arsenic is incorporated within the magnetite structure as an elemental substitution for perhaps $Fe^{3+}$, which is of similar atomic radius to that of As(III) (0.58Å) or As (V) (0.47Å) [5]. Relatively low arsenic and iron concentration below 230m depth due to probably insufficient iron oxyhydroxide sorbents present in the aquifer to be a significant sink for arsenic.

# 3  Problem solution

## 3.1  Mechanism of arsenic contamination

It is thought that arsenic mobilization occurs strongly under shifting red-ox conditions [4]. This may take place either during the oxidation of sediments present in reduced conditions resulting release of arsenic from sulfide bonding, or during the reduction of sediments present in oxidizing conditions then consequent release of arsenic during dissolution of the iron oxy-hydroxides coatings. Field redox potential values and redox sensitive species indicated relatively strong reducing conditions in groundwaters. This is consistent with the speciation of arsenic predominantly as arsenite (Fig.4). Groundwaters are anoxic because they contain high iron concentration. Moreover, concentration of $NO_3^-$ is very low and pH ranges from 6.36 to 7.79. Theoretically, well waters containing dissolved Fe are free of $NO_3^-$ that cannot be detected in most of water samples but intermediates of nitrate reduction, like nitrite, was found in groundwater in low concentration. The increases in sulfate and $Fe^{2+}$ below the depth where nitrate disappears indicates that nitrate reduction [2]. It is assumed that the onset of $NO_3^-$ and Mn(IV) reduction, as reflected in their respective decrease and increase Mn(II) in concentrations, is insufficient to mobilize As.

Arsenic is controlled primarily by sorption and precipitation: adsorption of As(V) onto Fe oxyhydroxides appears to control both arsenic mobility [3]. Under sufficiently reducing condition, however, precipitation of arsenic sulfides such as arsenopyrite control of arsenic levels. The source of sulphate in Mekong delta derived sufficiently reducing conditions, however, precipitation of As sulfides from acid sulphate soil as well as seawater intrusion, with water pH as low as 2.9. However, there is insufficient evidence to conclude that arsenic concentrations from acidic soils are significantly different from those from non-acidic areas of the delta.
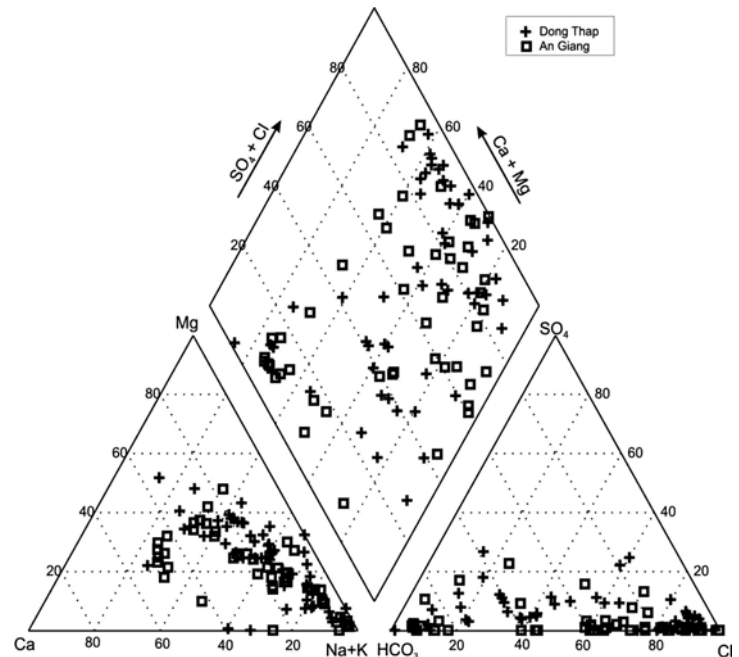


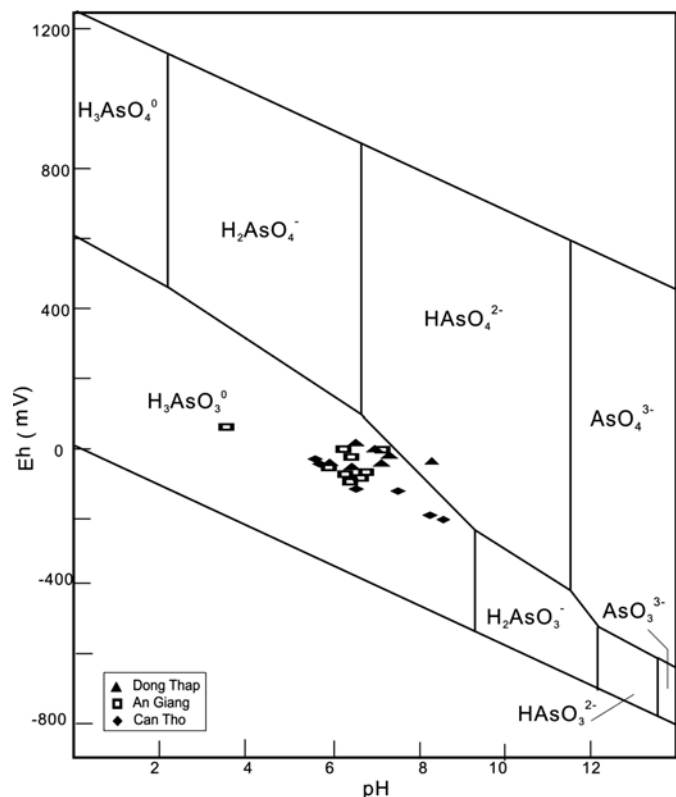Fig. 3. Plot of the major ion composition of the groundwater samples on a Piper diagram.



Fig. 4. Groundwater Eh-pH data plotted on an arsenic speciation diagram at $25^0$C, by Ferguson and Gavis (1972).
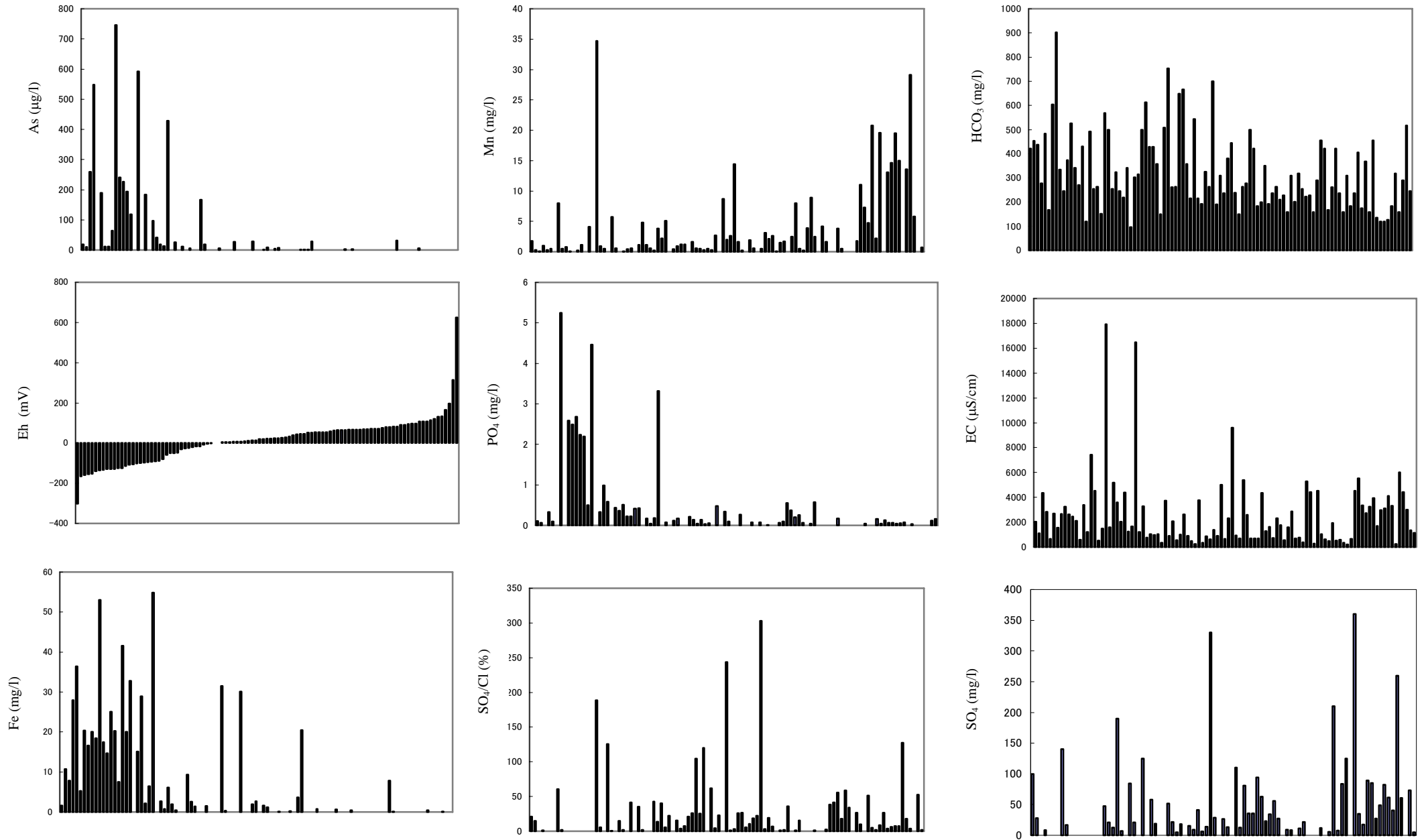
Fig. 5. Redox chemistry of groundwater samples. Samples were ordered in a sequence of increasing of Eh.
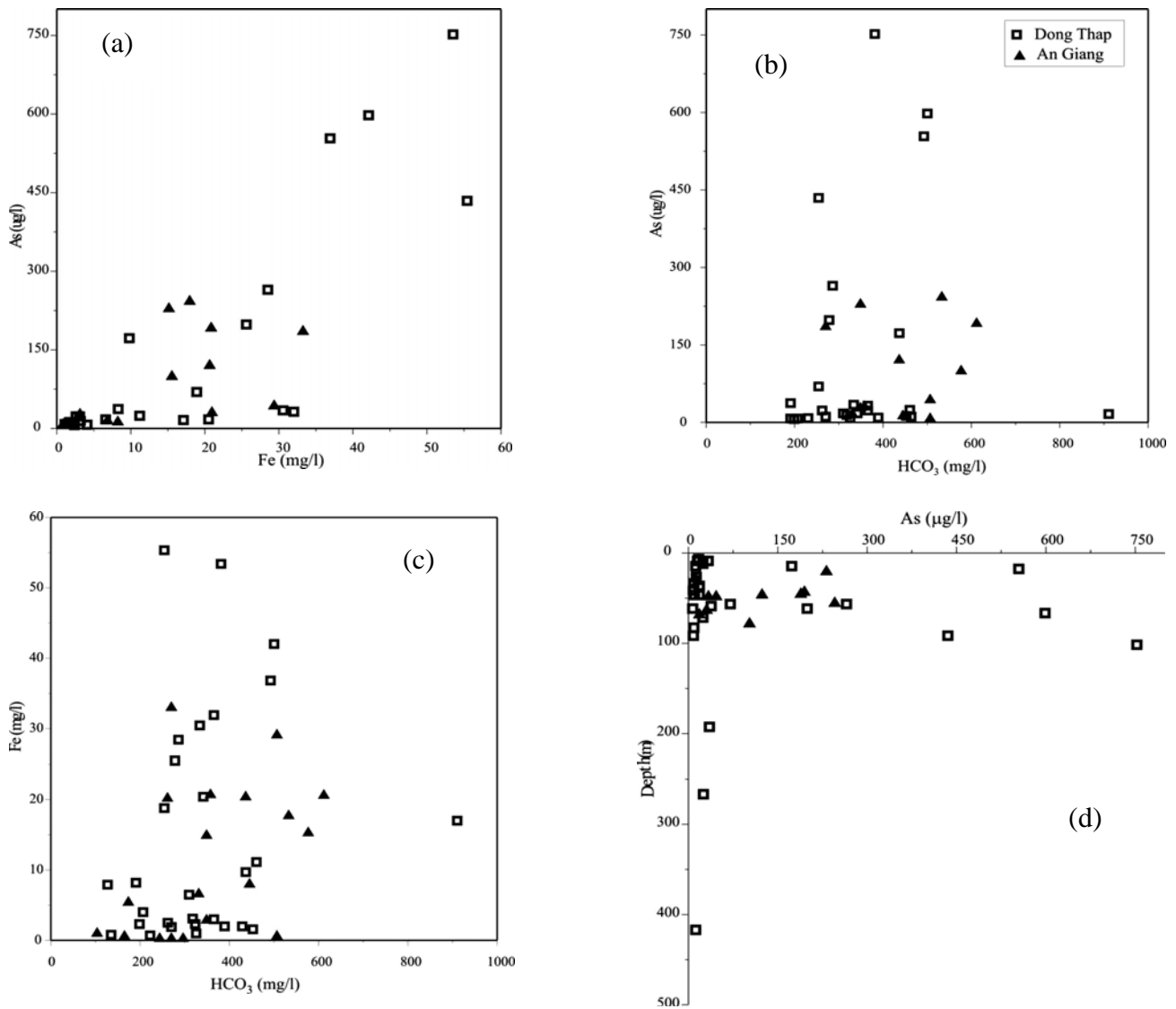
Fig. 6. Relation of (a) As to dissolved Fe; (b) As to HCO$_3$; (c) Fe to HCO$_3$; (d) As to depth.
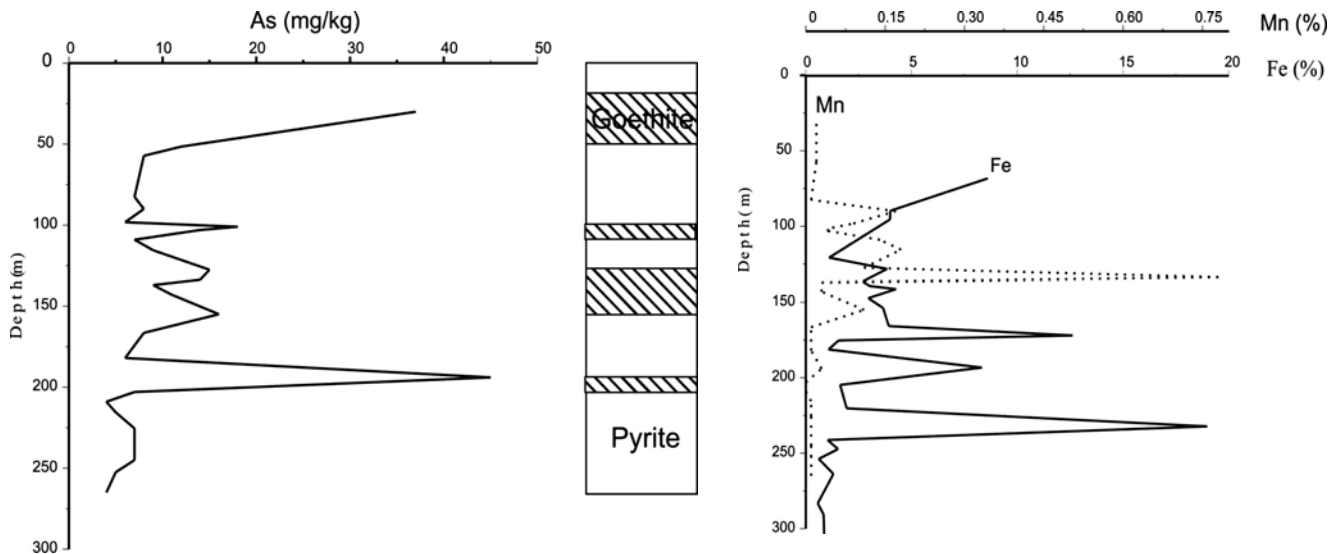


Fig. 7. Total arsenic, iron and manganese concentration in the sedimentary samples.

Aquifer sediment presents iron-rich clastic minerals such as iron oxide/hydroxide that carried arsenic with residual magnetite but predominant illite. Abundant organic matter in sediment contributed reducing ferric oxide and iron oxides (goethite, magnetite, and hematite) by iron reducing bacteria. Then adsorbed arsenic is released to groundwater and mobilized with ferrous ions. Pyrite mineral were also found in deep coarse sediment but sulphate reducing bacteria, organic carbon and under stronger reducing condition, precipitated pyrite instead of sorbed co-precipitated arsenic from groundwater [1]. In the other hand, molar concentrations of sulphate are many times greater that than of arsenic, suggesting that oxidation of arsenopyrite is not a significant source of sulphate [4]. Rapid oxidation of arsenopyrite is precluded by the low Eh (-80 mV) of the groundwater. According to Gordon et al. [10], there is influence of annual cycle of near surface oxidizing and reducing condition, corresponding to the dry and wet season to arsenic contamination in Mekong delta. During dry season, declination of groundwater levels facilitates exposure of sulfidic material to the atmosphere as well as oxidation process. The products of sulphide oxidation are rapidly dissolved by groundwater recharge when rainy season or annual flooding comes. Annually cycle in groundwater levels, which varies from at least 6m in some riparian environment, to only centimeters in more distant part of the flooding [10]. Therefore, in the rainy season, sediments rich in organic matter from both former reeds and mangroves are in anoxic environment. Then reducing process occurs in greater depth. Data suggest that arsenic is released to groundwater through reduction of arseniferous iron oxyhydroxides when anoxic conditions develop during sedimentary burial. The chemical evidences of Fe oxyhydroxide precipitation is supported by Fe staining of soil as well as bad taste of the associated high iron and aluminum concentration in water. Reduction of FeOOH is driven by the microbial oxidation of organic carbon [9]. The process dissolved FeOOH and releases to groundwater both $Fe^{2+}$ and sorbed load of Fe oxyhydroxide, which includes arsenic. The process generates $HCO_3^-$ ions and so produces relationship between $HCO_3^-$ and arsenic shown in Fig.6b. The reaction yields $HCO_3^-$ and $Fe^{2+}$ according to following reaction [11].

$$8FeOOH + CH_3COOH + 15H_2CO_3 \rightarrow 8Fe^{2+} + 16HCO_3^- + 12H_2O \qquad (1)$$
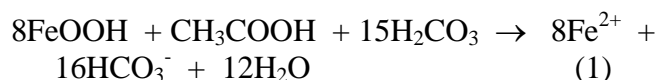
## 3.2    Resorption of arsenic to residual FeOOH.

It is suggested the possibility that in Late Pleistocene-Recent times, iron and arsenic-bearing sulfidic minerals in upper reaches of the Mekong Delta may have been exposed to atmosphere due to erosion, and oxidized. This resulted in mobilization of iron and arsenic, which were carried downstream by Mekong River. Mobilized iron precipitated as iron oxy-hydroxide and arsenic was adsorbed and/or co-precipitated with the iron oxy-hydroxide. At the present time, onset of reducing conditions in subsurface environment is causing dissolution of the iron oxy-hydroxide coating on soil particles and consequent mobilization of adsorbed/co-precipitated arsenic [18]. At depth where brown colour of the aquifer sand and concentration of arsenic in groundwater is low (10μg/l), whilst concentration of dissolved Mn and $SO_4$ are high. This was caused by incomplete reduction of FeOOH and Mn(IV) oxides. This incomplete reduction was attributed by distal from sources of organic matter. Concentrations of arsenic are kept low by re-sorption of arsenic to residual-FeOOH [12]. It also mentioned that arsenic released by reduction of $MnO_2$ must be re-sorbed to residual oxides of Mn and Fe, rather than remain in solution [8; 17]. Deep aquifers are free of arsenic (a very limited exceedance of 50μg/l of As in deep wells) can be explained by re-sorption assumption. Yellowish to pale grey Late Pleistocene sands are presented in deep aquifer sediment because they underwent a long period of oxidative weathering during the lowering of sea level at 18 ka. This weathering generated FeOOH by oxidation of Fe-bearing minerals in the sediment. Nevertheless, anoxic conditions in deep aquifer, as a consequence of deposition Holocene aquifer, has been sufficient to mobilize Fe into groundwater but has been insufficient to reduce all its FeOOH.

## 4    Conclusions

In this area, there are two possible conclusions of arseniferous groundwater drawn: firstly, arsenic concentration in excess of the 10μg/l acceptable limit occur at any depth; and secondly, relatively high concentrations are prevalent in two horizons such as Holocene aquifer (at 40m depth) and Pleistocene aquifer (at 100m depth). Iron oxide/hydroxide were found in the sediment where it contain high concentration of arsenic as well as yellowish brown and reddish brown in color. Arsenic carried in the water is adsorbed in iron rich clastic minerals, which have degenerated producing iron hydroxide coating on their surface. In addition, seasonally changing of groundwater level provokes oxidation and reduction cycle. It is assumed that oxidation of pyrite may contribute arsenic anomalies in shallow groundwaters. However, reduction of FeOOH that release to groundwater both $Fe^{2+}$ and arsenic is attributed by redox states of groundwater ($NO_3$, Mn, Fe concentration) as well as correlation between concentration of arsenic and $HCO_3$. That reduction of FeOOH is shown by high concentrations of dissolved iron concentration.

It is postulated that during the low-stand setting of the Late Pleistocene, the Mekong River system

entrenched their lowland basins and near surface sediments on these areas were exposed and oxidized. When transgressions occurred during the Early-Mid Holocene, the lower and adjacent parts were flooded and invaded by tidal mangrove, and development of fluvial marshes, swamps and peat. Arsenic sources were postulated that transport as phased of iron oxide/hydroxide with adsorbed arsenic and other trace metals, and deposited as sediment coating. Both deposit of arsenic-bearing iron mineral sediment, distribution of organic rich deltaic sediments (such as peat, swamp area) and development of severe reducing conditions facilitates complete reduction that causes more severe arsenic contamination in Dong Thap than in An Giang. Moderate to low arsenic but relatively high dissolved iron concentration is elucidated by incomplete reduction so that arsenic re-sorbed to residual FeOOH.

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