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The enrichment, release, distribution and geochemical transport of arsenic in the south side formation of the Chou-Shui River alluvial fan (I)

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共同主持人：

計畫參與人員：　王聖瑋、鄭世尉

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Abstract

High arsenic, As, concentrations in deep well water have been verified to be associated with blackfoot disease, which was once common on the Chianan plain in Taiwan. This is the first-year report of a three-year project. An investigation of shallow groundwater quality revealed that high arsenic, As, concentrations were found in both aquifers and aquitards in the southern Choshui river alluvial fan of Taiwan. 655 geological core samples from 13 drilling wells were collected and analyzed. High As contents were found primarily in aquitards, to a maximum of 590 mg/kg. The contents were correlated with the locations of the marine sequences. Additionally, strong correlations among the As concentrations of core samples, the clay and the geological age of the Holocene transgression were identified. Most of the As in groundwater originated from the aquitard of the marine sequence. The high As content in marine formations with high clay contents may be attributable to the bioaccumulation of As in the sea organisms, which accumulated and were deposited in the formation. A preliminary environmental model of the origin of the high As concentration in the shallow sedimentary basin of the Choshui river alluvial fan of Taiwan is proposed. The enrichment and release mechanism will be investigated in the second and third years study to establish a model of the ecological cycle of As in Choshui river alluvial fan in Taiwan.

Keywords: arsenic; occurrence; marine sequence; sedimentary basin

摘要

砷為一具有毒性且致癌之重金屬，於嘉南平原曾造成大規模之烏腳病疫情，相關研究報告說明此與當地居民飲用含砷之深層井水有關。而根據歷年研究報告指出鄰近嘉南平原之濁水溪沖積扇南端之淺層地下水中亦含有高砷含量，唯兩地區地下水中發現高砷含量之深度有所差異，故本研究為三年期計畫之第一年，挑選濁水溪沖積扇南端之 13 口地質鑽探井，針對 655 個岩心樣本之砷含量進行分析，並蒐集地層岩心質地資料與地年分析結果，探討此區地層岩心之砷含量、土壤質地與沉積年代三者之關係，最後歸納此區地下水中砷含量可能之主要來源。此區沉積地層中相互交錯之三個海相層與三個陸相層為冰河時期之海進海退現象所造成，而海相層與陸相層即分別相當於現今之阻水層與含水層，由本研究結果發現濁水溪沖積扇南端地層中，高砷含量主要發生於多屬黏土質之淺層阻水層，其顆粒較小，易吸附高砷含量，反之，土壤顆粒較大之含水層岩心，其砷含量則較低，且岩心定年結果約為 3,000 至 9,000 年前之全新世沖積土壤，此與嘉南平原深層地層岩心之定年結果相近，即濁水溪沖積扇南端與嘉南平原兩者之高砷含量地層雖具不同深度，卻屬同一時期形成，此結果說明地下水中高砷含量之發生與岩心質地及其所屬年代呈現相關。另外，利用阻水層岩心與含水層岩心之平均砷濃度分別與地下水中之砷濃度進行迴歸分析，發現阻水層岩心砷含量與地下水砷含量為顯著正相關 (R^2=0.51)，而含水層岩心砷含量與地下水砷含量之 R^2 則為 0.21 且非顯著，此結果說明地下水中砷含量不僅由地層中傳輸而得，阻水層岩心更可能為主要來源。本研究同時提出濁水溪沖積扇地下水砷來源之概念模式，然而，地層中砷含量之富集與釋放機制為未來所必須深入探討，以期建構完整之砷循環模式。

關鍵詞：砷、來源、海相序列、沈積盆地

1. Introduction

High arsenic, As, concentrations in deep well water have been verified to be associated with blackfoot disease, which was once common on the Chianan plain in Taiwan (Tseng et al., 1997; Chiou, 1996; Ch'I et al., 1968; Shen and Chin, 1964). Total As concentrations in deep well water range from 470 to 897 μg/L in this hyperendemic blackfoot disease area (Chen et al., 1994). The high contamination of groundwater by As has also been observed in shallow tubewells in the Bangladesh to depths of 15-30m (Kinniburgh, 2001). The highest As concentration of groundwater has been generally monitored in the southeastern Bangladesh, close to the Bay of Bengal (Gaus et al., 2001). Extensive As contamination in groundwater in Bangladesh represents a significant health risk to millions of people (Chowdhury et al., 2000; Karim, 2000). Smedley and Kinniburgh (2002) indicated that extreme As concentrations in natural water are rare, but are most frequently observed in groundwater. Released from natural sources is the dominant cause of elevated As concentrations in groundwater (Nordstrom, 2002). From 1999 to 2003, the Taiwan Sugar Company (2003) undertook a groundwater quality survey in the Choushui river alluvial fan in the north of the Chianan plain. The As concentrations of 34 samples in groundwater exceeded the current drinking water supply standard of 50 μg/L in Taiwan in 1999. The results in 2000 and 2003 were similar. The concentrations of As in groundwater measured in these three years followed a log-normal distribution, as determined by a chi-square test and a K-S test. They have a geometric mean of 0.262 mg/L with a maximum of 0.60 mg/L and a geometric standard deviation of 1.492 mg/L. These monitoring wells were distributed over the southern Choushui river alluvial fan, which is close to the blackfoot disease hyperendemic area. However, the As concentration in groundwater was high in shallow monitoring wells with depths of about 20-70m underground in Yun-Lin county, the south part of the Choushui river alluvial fan, and differs from that in deep wells with depths of 70 to 300m, in the blackfoot disease hyperendemic area (Tainan Hydraulic Laboratory, 2002; Taiwan Sugar Company, 2000).

The As contents of core samples at the Ghaigang of the Chianan plain were analyzed (Bi, 1994). The average concentration of the core samples was 9.8 mg/kg. The maximum concentration of 26 mg/kg was found 105m below the ground. Several works have addressed the high As content in the deep strata of the Chianan plain, indicating that the high As contents of the core samples are related to the formations of the marine sequences (Chen, 2003; Lin, 1995; Yen et al., 1980). Groundwater is utilized abundantly as an alternative to surface water, especially in the southwestern coastal region, including in the Yun-Lin county, where surface water resources are seriously deficient, because of the high domestic, irrigational, aquacultural and industrial demand. The quantity of groundwater pumped annually in Yun-Lin is 660 million tons, which is three times that in Tainan county (Agricultural Engineering Research Center, 1999; Jang, 1996). Residents may be directly exposed to As through drinking water or indirectly exposed to As through various paths, including ingesting aquacultural and agricultural products, which pose risks to human health. Accordingly, the purposes of this study are to analyze As concentration distributions in formations; determine the correlations among As concentration distribution, material in the core sample and geologic ages of formation, and to postulate the As sources of groundwater in the southern Choushui river alluvial fan of Taiwan. These results can be applied to evaluate As contamination potential and safety concerning the domestic use of groundwater in the future.

2. Materials and methods

2.1. Study area

The Choushui river alluvial fan of Taiwan includes Chang-Hwa county and Yun-Lin county. The latter is located in the southern part of the Choushui river alluvial fan, and is surrounded by the Taiwan Strait to the west, the Central Mountain to the east, the Chou-Shui river to the north and the Pei-Kang river to the south (Fig. 1). A hydrogeological study revealed that the Choushui river alluvial fan was form in the late Quaternary and is partitioned mainly into the proximal-fan, the mid-fan and the distal-fan areas (Central Geological Survey, 1999) (Fig. 2). According to the subsurface hydrogeological analysis to a depth of around 300 m, the formation is divided into six interlayered sequences, including three marine sequences and three non-marine sequences, in the distal-fan and the mid-fan areas, respectively. Generally, the non-marine sequences of formation, with coarse sediment, ranging from medium sand to gravel of high permeability, can be regarded as aquifers, whereas the marine sequences of formation, with fine sediment can be regarded as aquitards. The hydrogeological formation of the proximal-fan, which consists entirely of gravel and sand, is considered to be an unconfined aquifer and an important groundwater recharge area. Huang (1996) employed accelerator mass spectrometry (AMS) C-14 dating to determine the ages of mollusk shells within sedimentary basin of the Choushui river alluvial fan. He determined that the marine sequence 1 was deposited 3,000-9,000 years ago during the Holocene transgression; marine sequence 2 was dated 35,000-50,000 years ago, and marine sequence 3 was probably deposited 80,000-120,000 years ago during the more recent interglacial transgression (Fig. 3). These data were collected and analyzed to determine the correlations among the As concentration distribution, material in the core samples and geologic ages of the formations.

2.2. Sampling and analysis

Geological core samples were collected from 13 drilling stations in southern Yun-Lin county, which are close to coastal and vicinal blackfoot disease hyperendemic areas (Fig. 1). The As concentration of groundwater was high in the shallow monitoring wells of the Choushui river alluvial fan (Tainan Hydraulic Laboratory, 2002), so core samples were collected at 3m intervals to a depth of 100m below sea-level, and at 10m intervals from that depth to the bottom of the well. The As contents of a total of 655 geological core samples were analyzed.

30% H_2O_2 and 9.6M HCl were added to remove organic matter and digested geological core samples, to enable the total As concentration to be determined. After filtering, all samples were examined using an electro-thermal atomic absorption spectrometer (AAS, Perkin-Elmer AA100) and a hydride generation (HG) system (Perkin-Elmer FIAS100); 0.5% $NaBH_4$ in 0.25% NaOH and 1M HCl were added to reduce arsenic to arsine. Finally, the total As concentration was determined.

3. Results and discussion

3.1. Distribution of arsenic contents in sedimentary basin

The analyzed As concentrations of total 655 core samples have geometric mean, geometric standard deviation, minimum and maximum, of 2.27 mg/ kg, 1.43 mg/kg, 0.45 mg/kg and 590 mg/kg, respectively. These results are consistent with a log-normal distribution as determined by performing a chi-square test and a K-S test. The numbers of samples with As concentrations of 0-10 mg/kg, 10-50 mg/kg and ＞50 mg/kg are 486, 155 and 14, respectively. These values exceed the average concentrations in the earth’s crust, 1.8 mg/kg and in sandstone or limestone, 1 mg/kg (Valette-Silver et al., 1999). Pearce et al. (2001) found As concentrations of 0.4-10.3 mg/kg (with a median of 2.1 mg/kg) in 21 sediment samples from Chapai Nawabganj, Faridpur and Lakshmipur in Bangladesh. Foster et al. (2000) found As concentrations of 1-16 mg/kg in a sediment profile from Brahmanbaria, north-east Bangladesh. Although, in their studies, the maximum As concentration of 264 mg/kg was found in an iron-rich clay layer, the As content in this study exceeds the values found in sedimentary formations.

The horizontal and vertical cross sections of the As concentration distributions demonstrate that the As content is high primarily in aquitards (Fig. 4), and the As concentrations decrease from the west coastal area, which has six interlayered sequences of formation, to the east mountainous area where the formation becomes a single gravel aquifer. Furthermore, the As concentrations in the shallow strata to depth of 50m, which were formed during the Holocene transgression, are higher than those in the other strata. At drilling stations 5, 6, 8 and 10, As concentrations are high at depths of below 200m, because these drilling stations are near the Chianan plain where deep groundwater had high As concentrations. The As concentration is highest at a depth of 54m at drilling station 1 and the As content of groundwater in this region is also highest in the Choushui river alluvial fan. Drilling stations 3 and 4, which are located near the coast, have high As contents in both shallow and deep strata. The locations of drilling well stations 3 and 4, in the transition zone between shallow to deep have high As content formations, which may be responsible for the high As concentrations found in both shallow and deep strata.

3.2. Correlations among As contents, material and geological ages of core samples

As content (>50mg/kg) is high mainly at drilling stations 1, 3 and 10 (Table 1), but the depths at which the high As content vary among the stations. Drilling station 1 exhibited a high As content in the shallow stratum; drilling station 3 had a high As content in the deep stratum and drilling station 10 had a high As content in both shallow and deep strata. The high As contents in the cores may be strongly associated with the material of the core samples and the geological ages of their formation (Fig. 3). The material of the core samples in the drilling station 1 associated with marine sequence 1 is clay, but the clay layer in drilling station 3 was during the formation of marine sequence 2. Additionally, clay layers were found in both marine sequences 1 and 2 at drilling station 10. Not only dose the high As content of the core samples correlate closely with the presence of clay, but also the medium to low As content correlates with the presence silt and sand materials. Figure 5 reveals that the logarithm of the As concentrations of the three materials of the core samples - clay, silt and sand - are determined by the goodness-of-fit to a normal distribution, as determined by a chi-square test with a 5% level of significance. The logarithm of the As concentrations in clay, silt and sand are 0.94±0.38 (log (mg/kg)), 0.79±0.35 (log (mg/kg)) and 0.69±0.29 (log (mg/kg)), respectively. The small grains of the core materials have a large surface area, increasing the adsorption of As onto the surface of the solid. This result explains why the As concentration decreases from the distal-fan area, which consists of fine sediments, to the proximal-fan area, which consists of primarily coarse sediments.

The data from the AMS C-14 dating of mollusk shells in the core samples of the Choushui river alluvial fan (Central Geological Survey, 1999) suggest that the geologic ages of the core samples can be grouped (Table 2) as follow; 2,931-5,364 years ago, 7,090-9,230 years ago, and >36,400 years ago. The first two intervals are associated with the formation of marine sequence 1, and the third is associated with the formation of marine sequence 2. The As concentrations of the core samples in the second interval exceed those in the other two intervals, and the highest As concentration is 51.35 mg/kg. Based on the classification of the sedimentation sequences (Fig. 3), the second interval corresponds to the bottom of marine sequence 1. The distribution of clay in marine sequence 1 is more extensive than that in marine sequences 2, causing considerable As accumulation.

Notably, the sedimentary basin of Bangladesh is mostly alluvium and deltaic deposits (Alam et al., 1990) and its geological age, especially in the Ganges, Brahmaputra and Megna river floodplain (BGS & DPHE, 2001), ranges from 3,000 to 10,000 years ago, during the Holocene transgression (Umitsu, 1993). The correlations among the As contents, material and geological ages of the core samples observed in Bangladesh are similar to those found in the Choushui river alluvial fan of Taiwan.

3.3. Arsenic sources in groundwater

The correlations between the As concentrations in the groundwater and in the core samples of the aquitard or aquifer are statistically analyzed to determine the primary source of As in groundwater. The source of the As concentration in groundwater is assumed to be the solid phase of the formation. The formation is further classified as an aquifer or an aquitard based, on the material of the core samples and the hydrogeological setting. Accordingly, the As in the groundwater may be released from the aquifer and the aquitard. Before the correlative analysis is performed, the vertical influence range of the concentration of As released from the upper and lower aquifer and the aquitard to the groundwater must be defined. The influence range of the As concentration in groundwater released from aquifers and aquitards is assumed to be half of the interval measured vertically from the center of the well screen to the upper and lower well screens (Fig. 6). The measured As concentrations in the core samples of the aquifers and aquitards within the influence range were each averaged; the averages were used to evaluate the correlation between the As concentrations in groundwater and those in each of the core samples from the aquifers and the aquitards. Figure 7 shows that the As concentrations of core samples in the aquitards were positively correlated with the As concentration in groundwater, using $p<0.01$ as the threshold for significance ($R^2=0.51$), but the As concentrations of the core samples in the aquifers were poorly correlated with those in the groundwater, $R^2=0.21$ and $p>0.05$. This result indicated that the As concentration of the core samples in the aquitards substantially influenced the As contents in vicinal groundwater and may be the primary source of As in groundwater.

Liu (2003) applied factor analysis to assess the quality of groundwater in Yun-Lin county in Taiwan, and stated that the arsenic pollutant factors included As, total organic carbon (TOC) and alkalinity (Alk), which were all significantly positively correlated to each other. Anawar (2003) demonstrated that the As of groundwater in the Ganges delta were strongly correlated with the bicarbonate content and dissolved organic carbon (DOC), and that the combined effects of $NaHCO_3$ and pH mobilized As from sediments. Harvey (2002) indicated that the DOC and dissolved inorganic carbon (DIC) of groundwater in Bangladesh was also positively correlated to As content, to the peak depth. The analysis of the carbon isotopes of groundwater revealed the presence of young DIC and old DOC in the same water sample and demonstrated As was released from sediments to groundwater. The results in our study provide that the As in the groundwater of the Choushui river alluvial fan originated from the sediment and was mostly from marine sequence.

This study proposes a preliminary environmental model of the origin of the high As concentration in the shallow sedimentary basin of Choushui river alluvial fan of Taiwan. Changes in sea level significantly influence the composition and structure of the geological environment. The sedimentary basin of the Choushui river alluvial fan is formed by the alternating invasion and retreat of seawater with interlayered formations of marine and non-marine sequences. The marine sequences comprise mainly fine sedimentary clay, ranging from clay through silt to fine sand. The high As content in the marine formation with the high clay content may be attributed to the bioaccumulation and biotransformation of As in the sea organisms, which As is then accumulated and deposited in the formation (Francesconi et al., 1998; Francesconi and Edmonds, 1997).

4. Conclusions and suggestions

High As concentrations in shallow strata, and especially in aquitards, are responsible for high As concentrations in groundwater in the Choushui river alluvial fan. The distribution in the sedimentary basin is caused by the alternating invasion and retreat of sea water with interlayered formations of marine and non-marine sequences. High correlations were found among the As contents of core samples, the type of clay material and the geological ages of the Holocene transgression (marine sequence 1). The marine sequence were typically formed with a high clay content, increasing the As content. High As contents are thus present in the marine sequences, particularly in marine sequence 1. Although the depth at which the As contents were high in samples from the Choushui river alluvial fan differed from that in samples from the Chianan plain, the geologic ages are all in the range of 3,000-9,000 years ago or the Holocene transgression. Additionally, a preliminary environmental model is developed to describe the origin of high As concentration in the shallow sedimentary basin of the Choushui river alluvial fan in Taiwan. The source of As in groundwater is postulated to originate primarily from aquitard formations of marine sequences.

The release of As from the formation to the groundwater has been examined globally. Related studies have investigated the oxidation of As-bearing sulfides, the desorption of As from oxides and hydroxides, the reductive dissolution of As-bearing oxides and hydroxides, the release of As from geothermal waters, and evaporative concentration, and the leaching of As from sulfides by carbonates (Smedley, 2003; Kim et al., 2000; Welch et al., 2000). Some researchers have proposed that the oxidation of sulfides, such as arsenopyrite or As-rich pyrite, is the dominant mechanism of the release of As. Others have posited that the bacteria-mediated mineralization of organic matter and the reductive dissolution of iron and manganese oxyhydroxides are responsibly for the high concentrations of As in groundwater of a sedimentary basin, such as that in Bangladesh (Anawar et al., 2003; Nickson et al., 2000; Bhattacharya et al., 1997; Dipankar et al., 1994). The process of formation of the high As content in the Choshui river alluvial fan is similar to that of As enrichment in the alluvial aquifers in Bangladesh, so the aforementioned release processes may be valuable in the future for determining the As release mechanism of the Choshui river alluvial fan.

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Table 1
Drill stations and depths of core samples with As concentrations greater than 50 mg/kg

Drill station	Depth (m)	As (mg/kg)
1	54	590.00
3	162	117.00
3	180	107.50
8	130	102.60
1	63	76.80
1	6	75.35
10	39	75.08
10	272	74.64
10	152	67.48
11	30	61.88
10	21	56.55
8	12	54.90
1	60	52.35
3	51	51.35

Table 2
Geological dating and As contents of core samples

Drill station	Depth (m)	Dating (year)	As (mg/kg)
1	108.4	>52000	5.32
3	32.7	2931±81	29.34
3	48.9	8819±99	51.35
3	117.9	43900±1500	13.16
5	157.8	>45000	5.43
6	39.4	3936±64	11.31
7	23.0	5364±89	4.83
7	42.7	8440±90	8.27
7	111.4	39900±1100	110
7	129.6	>50000	1.71
8	91.6	>50000	34.04
9	25.1	7370±100	10.89
9	25.1	7620±80	10.89
9	48.8	9230±60	2.61
9	68.6	36400±500	6.63
9	105.1	>50000	17.22
13	38.2	8330±60	11.51
13	38.8	7920±70	11.51
13	55.0	7090±60	3.37
13	60.0	7850±60	3.01
13	99.0	43300±130	4.61

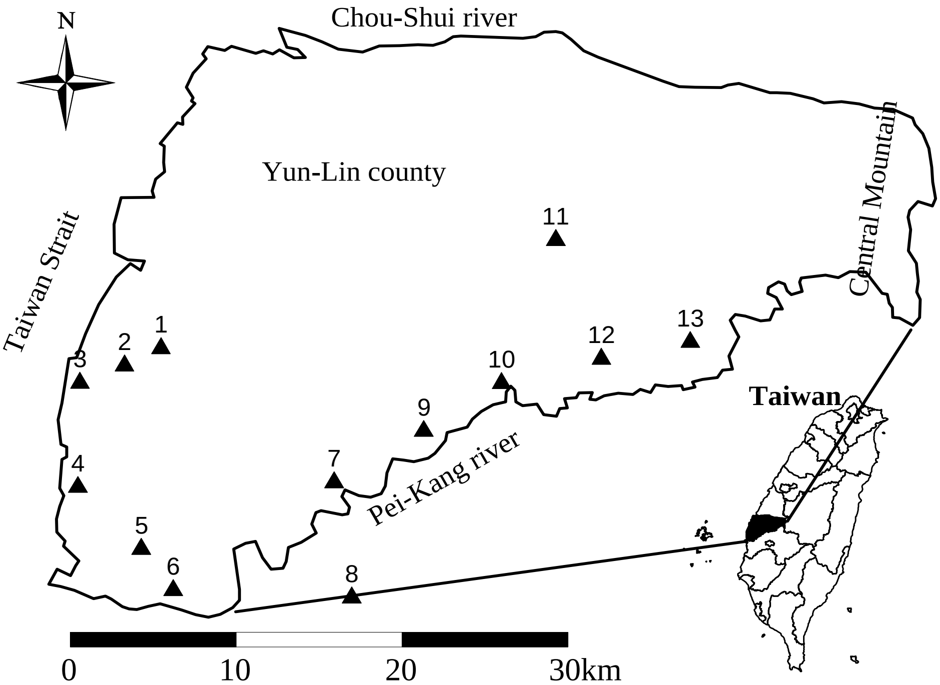


Fig. 1. Study area. Geological core samples were collected from 13 drilling stations in Yun-Lin county, at 3m or 10m intervals to a depth of 300m.

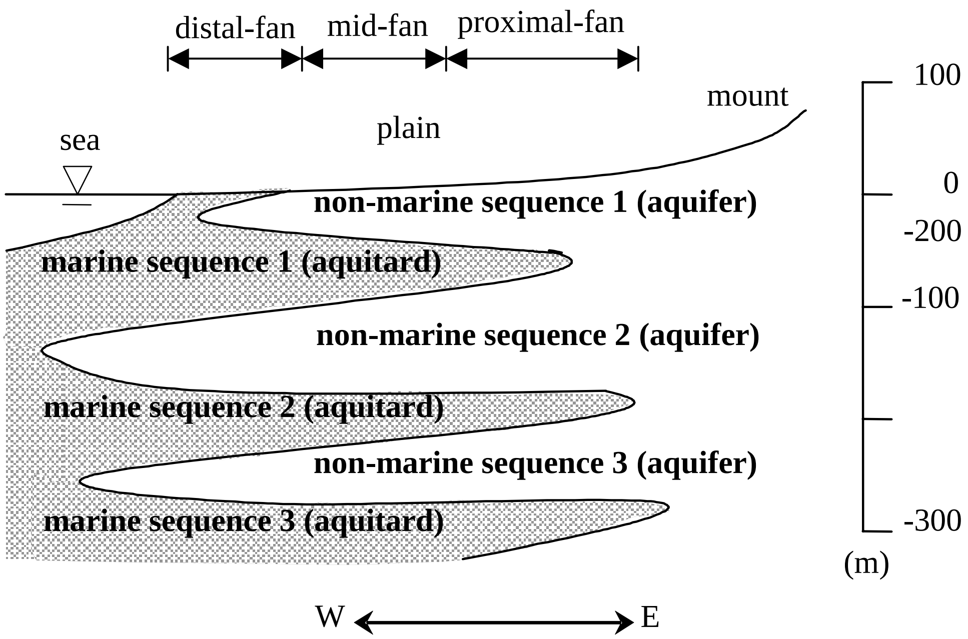


Fig. 2. Conceptual hydrogeological profile for Yun-Lin county in the south of the Choshui river alluvial fan. Six interlayering sequences of formation, including three marine sequences and three non-marine sequences, are considered.

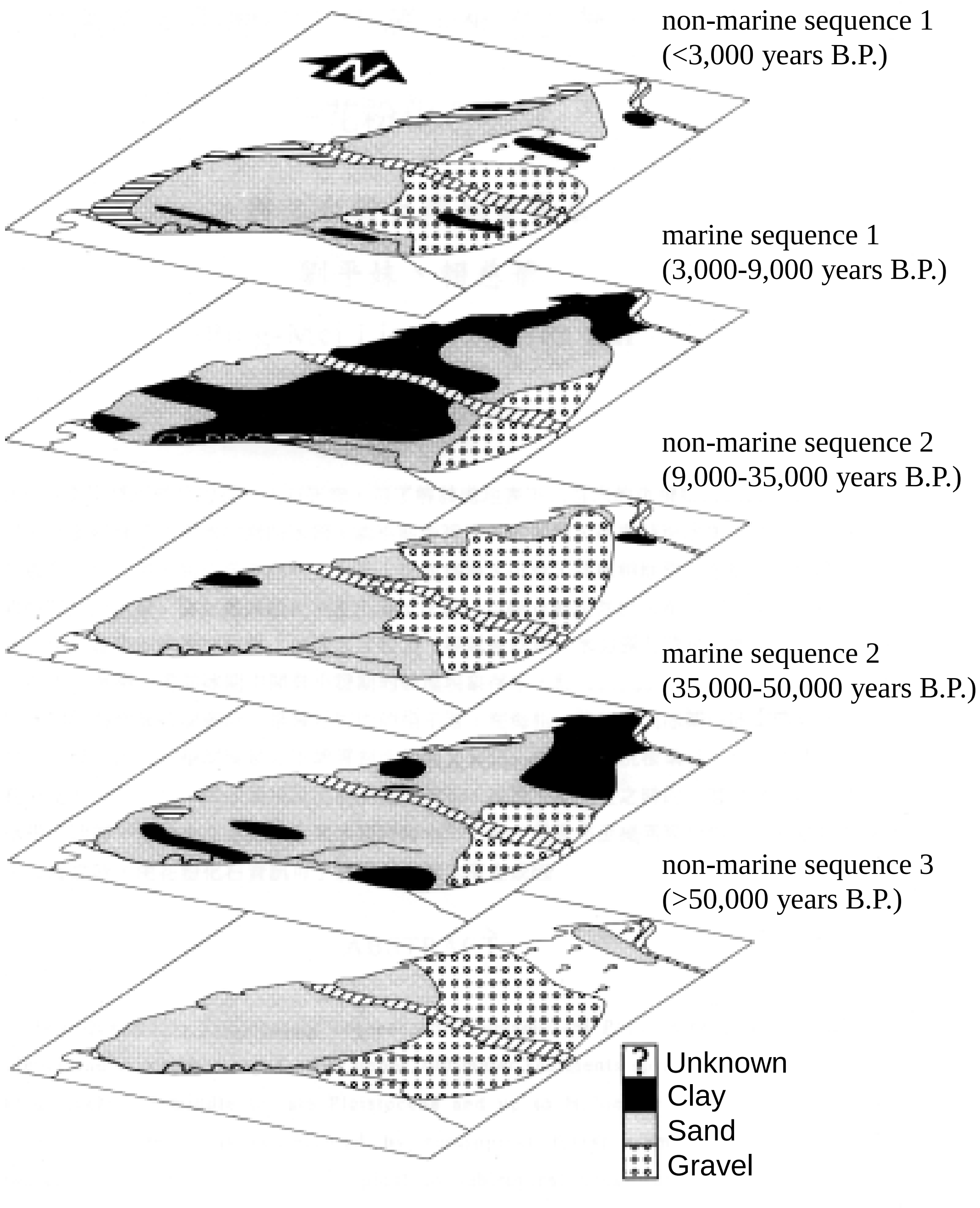


Fig. 3. Distributions of the marine and non-marine sequences in the sedimentary basin of the Choushui river alluvial fan (Huang, 1996).

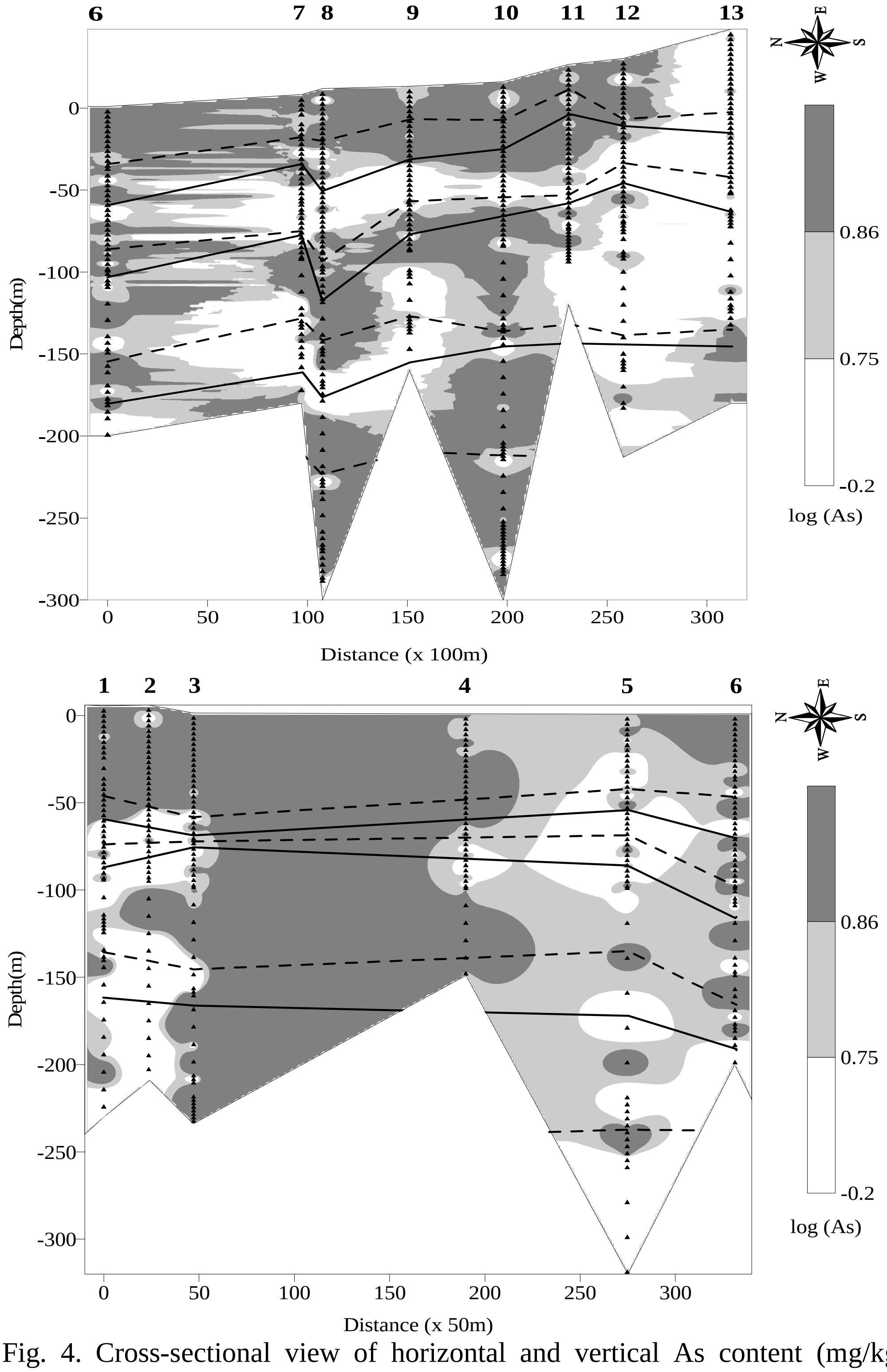


Fig. 4. Cross-sectional view of horizontal and vertical As content (mg/kg) distributions. (a) Drilling stations in the west-east direction. (b) Drilling stations in the north-south direction. The dotted and solid lines represent the tops of the aquitards and the aquifers, respectively. The solid triangles represent the locations at which the cores were sampled. The number on the top of each figure specifies the drilling station.

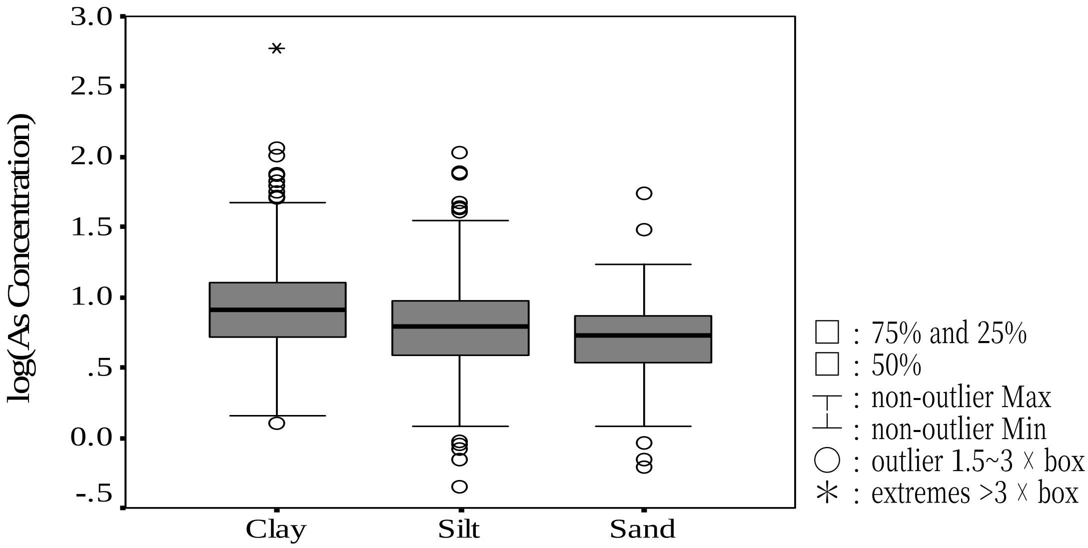


Fig. 5. Box-plot of the logarithm of As concentration in clay, silt and sand of core samples.

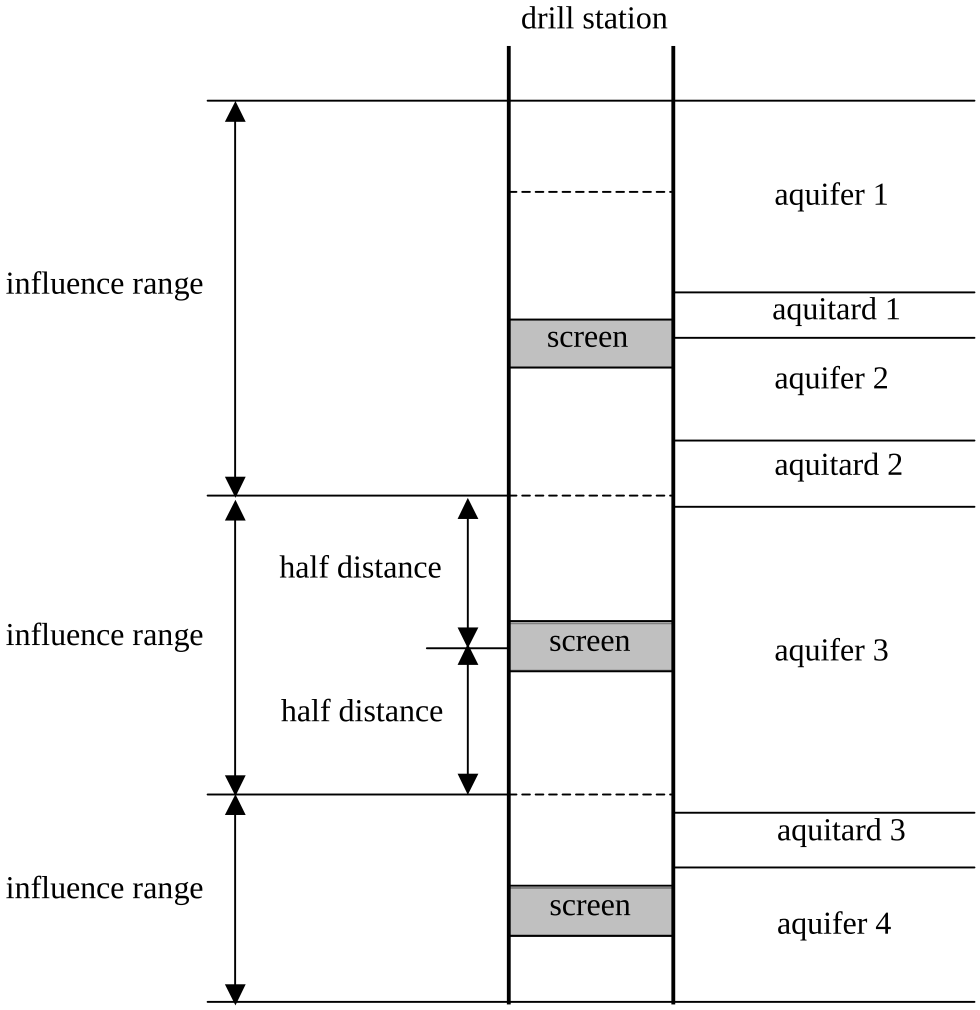
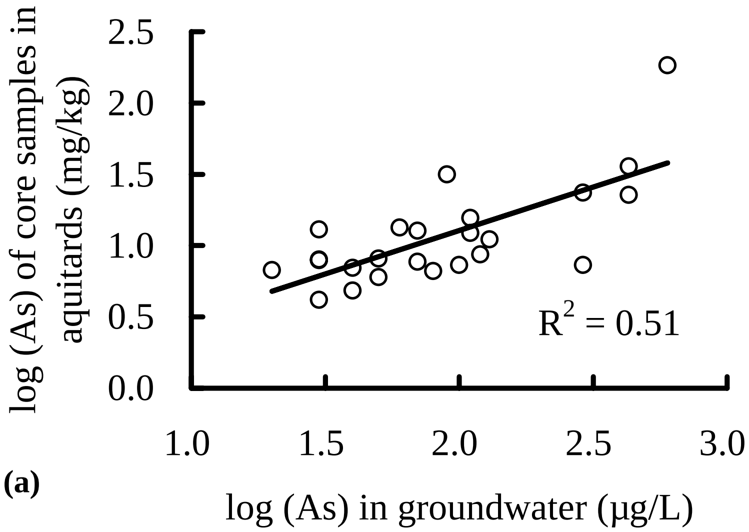


Fig. 6. Vertical influence range for the upper and lower aquifers and aquitards of As concentration in groundwater monitoring well (not to scale).



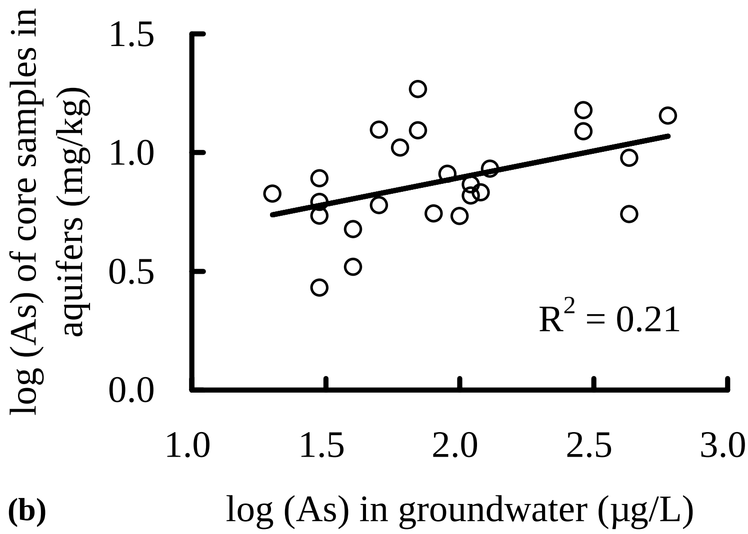


Fig. 7. (a) Correlation between As contents of core samples in aquitards and As concentration in groundwater. (b) Correlation between As contents of core samples in aquifers and As concentration in groundwater.