On the Arsenic Distribution along the Coastline Area of Southeastern Taiwan

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The arsenic anomaly along the coastline near to the solar-salt-making area in the southeastern Taiwan is investigated. Relevant chemical, mineralogical, and sedimentological analysis revealed that anomaly is restricted to the underground water and sediments and it can possibly be excluded from the seawater in the area. The anomaly is correlated with the slate ingredient content in the sediments. Slate sand can be an arsenic scavenger that has eliminated the arsenic pollution possibly occurring in the adjacent sea, and instituted the arsenic enrichment in the sediments. The conspicuousness of the enrichment may be attributed to the source of the slate sands and the sites where the sands deposited. Since the slate sand has been the depositional loci of arsenic as well as a good material for making up an aquifer due to its larger particle size, the enriched arsenic may be re-liberated into the environment, resulting in the arsenic anomaly in groundwater.

1. Introduction

Arsenic anomaly in the coastline area of southwestern Taiwan occurs in the underground water. It once reflected in the black-foot disease prevailing in the area [1,2], although no clear direct proof of relationships exists. However, seawater and by which the solar salt produced in the area, there reveals no sign of even slightly higher in arsenic content. A study examining the arsenic anomaly and its geo- chemical cycle occurring in the area has fulfilled using ninety-one sediment samples collected in the field through viewpoints of geochemistry, mineralogy, and sedimentology [3]. This study is part of the examination. It attempts to relate the origination of the limited anomaly with the mechanism of eliminating the arsenic pollution prevailed in this area.

2. Basic data

The average amount of arsenic (ppm) in natural environment is listed in Table 1 [4]. Shales and slates may content higher arsenic (5~ 70ppm) than any other rocks. Because the gneiss and the schist which were derived from the metamorphism of slates show less arsenic content than that of the parent rocks, it is believed that the element may be released during the metamorphism [4]. Previous studies show that arsenic anomaly may exist along the coastline area of southwestern Taiwan. Water produced from artesian wells in this area generally shows higher arsenic content, especially those from deeper wells (Table 2).

Table 1.
The average amount of arsenic content in natural environment (ppm)

Materials	ppm or mg/l
Igneous rocks	1.5
Shale	13
Sandstone	1
Limestone	1
Soil	5 ~ 10
Seawater	0.003
River	0.002
Under ground water	0.x ~22
Continental crust	1.8

After Onishi [4].

The earliest study reporting the arsenic

Table 2.
Arsenic contents of aquifers varies with the depth

Depth, m	As, ppm
45	0.05
130 ~ 135	0.38
130 ~ 150	1.24
130 ~ 170	1.45
195 ~ 200	1.13

After S. Yeh [2]

anomaly that occurs in this area was found in underground water analysis collected from deep wells in 1951 [1]. Chemical analysis for some aquatic plants, animals, and soil/ sediments sampled in the vicinity of the solar salt ponds also showed higher arsenic contents (Table 3). It is also reported that higher arsenic content of sediment is strongly related to the presence of slate particles in the sediment [3].

Table 3.
Data of arsenic contents (ppm) of some samples collected in the study area.

Aquatic plants	5 ~10 (ppm)
Soil	7.08
River mud	8, 11
Shoreline sand	3 ~30
Well cores	2.7, 3.3
River	0.0x
Underground water	0.0x~30
Seawater	0.00x

The slate ingredients are originated from the Central Mountain Range of Taiwan. They are transported by two rivers via north (Hsilou Chi) and south (Kauping Chi) into the coastline near the area then delivered to the study area by long-shore current. A series of studies dealing with arsenic contents of the sediments deposited in the area were fulfilled in 1963 [3] Slate particles transported into this area from south and north show different arsenic contents. Generally those from north shows higher values. However, both of them deposited into the area and once formed a lagoon environment in which abundant aquatic plants existing. This possibly resulted in a favorable environment for arsenic accumulation. Sediments collected from sand barriers, including that inside and outside of the present lagoon, both contain more than 15 % of slate ingredient. Particle size analysis finds that the slate particles are coarser than the mean size of the sediments and contain higher amount of arsenic than the parent rock. Subsequently the origination of the arsenic anomaly is considered to be resulting from the presence of the slate particles in the sediments

3. Arsenic enrichments on slate particles

Figure 1 demonstrates the arsenic content of sediment samples increases as the slate ingredient (weight percentage) increases. Each line represents

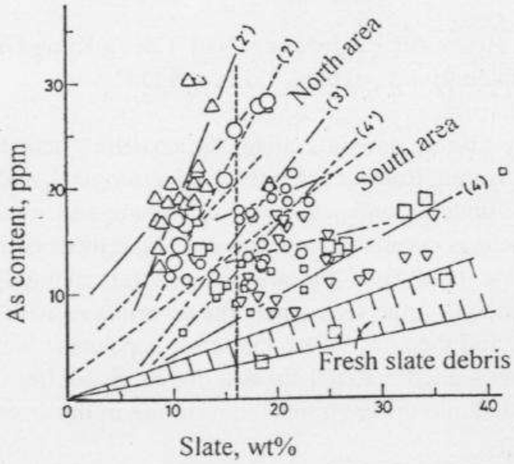

Figure 1. Arsenic contents of sediments are related to the presence of slate ingredients in the sediments. Lines represent different environments.

one sedimentary environment of discrepancy. It is obvious, the arsenic content is highly correlated with the presence of slate particles. And the average arsenic contents of the slate particle in the samples are higher than that of the original slate.

Since the arsenic accumulation on the slate particles may result from the physical-chemical reactions on the slate surface, investigation for the phenomena was carried out based on the following assumptions. Assume that arsenic contents of the samples are mainly resulted from the presence of the slate particles. And there can be two different types of arsenic presentation: on the particle surfaces and inside the particle. Concerning the former part and assuming the particle being a sphere with a radius r, then the weight of one slate particle is $f_1(b,r^3)$. Here b is the density of the slate particle. If the sample concerned contains wt % of slate, then the number of the slate particle is wt / $f_1(b,r^3)$. And because each particle has surface area $f_2(r^2)$, the total surface area of slate particles in the samples is $f_3(\mathrm{wt}\ /\ r)\ /\ \mathrm{b}$. Subsequently, the arsenic content of the sample contributed by the surface area of the slate particles is

$$\begin{aligned} As_s &= k' f_3(\mathrm{wt} / r) / b \\ &= k_s f_3(\mathrm{wt} / r) \end{aligned} \quad (1)$$

If the slate contains C% of arsenic inside the particle, then the total arsenic content of the sample is

$$\begin{aligned} As &= C\mathrm{wt} + k_s f_3(\mathrm{wt} / r) \\ &= \mathrm{wt}\,(C + k / r) \end{aligned} \quad (2)$$

Transforming (2) into

$$As/\mathrm{wt} = C + k / r \quad (3)$$

and drawing a graph of As/wt versus $1/r$, it is possible to examine whether the arsenic accumulation occurred onto the surface of the slate particles. Moreover, from the intercept C, it is possible to evaluate the amount of the arsenic content that is contributed by the solid portion of the slate particles. Figure 2 discloses the results obtained in the study. It is obvious that the accumulation

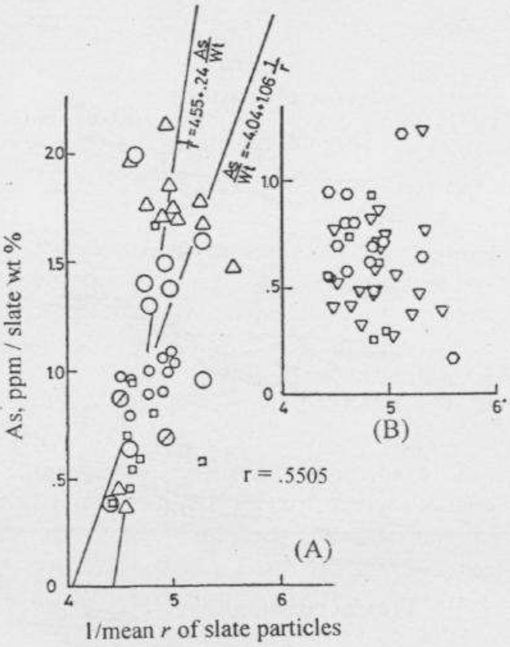

Figure 2. Arsenic accumulation on the surface of slate particles is manifested in the northern part (A) but obscure in the southern part of the study area.

phenomena are evident in the northern part of the area. In southern part, it is relatively obscure. Additionally, it is found that, comparing the two parts of arsenic content on the surface and inside the body of the slate particles, the surface accumulation can be more important.

4. Weathering test

A weathering test [5] on the fresh slate debris (powdered rock samples) and the weathered slate particles (separated from sediments) were compared (Table 4). It is interesting to find that the properties of the arsenic reacted with the fresh and the weathered slate debris are different. At the lithosphere – atmosphere contact, nearly one thirds of the element is released from the fresh rock. But under the same conditions, can only 1/40 ~1/50 of the quantity be released from the weathered one.

5. Arsenic anomaly in artesian water

During the years 1970 ~1980, a series of analysis on the arsenic content of artesian water was conducted by some laboratories in the area [6]. Significant conclusions were drawn to correlate the anomaly with the depth of the wells. In the analysis, samples from a total number of 43,566 wells were taken. Among them, 17.8 % of the well contained more than 0.1 ppm As, and 4.4 % over 0.35 ppm. The highest value was 3.0 ppm. It was noted that over 90% of the deeper wells showed high As content (Table 2). It was also noted that the depth that anomaly occurred was related to the location of the wells; the far the well located from the shoreline, the deeper was the anomaly occurred in the well. This involves the development of the shoreline sedimentary environments.

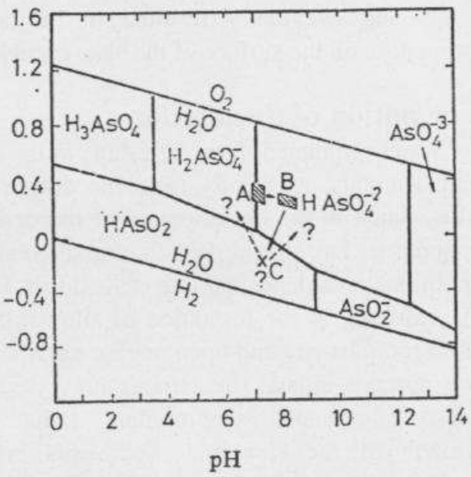

Figure 3. Arsenic cycle in geochemistry. Area A: contact zone between stream and seawater, B: lagoon, and C: underground water.

Table 4.
Comparison on arsenic releases between fresh slate debris and sand samples collected in the coastline

Sample		As release, ppm					
Environment	As cont.	4 days		7 days		14 days	
	ppm	Pure	Acidic	Pure	Acidic	Pure	Acidic
Lagoon	13.18	.14	nd	.20	.20	.19	.22
"	9.71	.12	"	.13	.13	.15	.17
"	16.73	.14	"	.18	.18	.19	.18
Sand bar	9.95	.14	nd	.18	.18	.18	.22
"	9.18	.14	"	.18	.17	.19	.18
"	13.04	.14	"	.18	.17	.18	.18
Fresh	14.22	2.78	4.32	4.05	9.00	4.26	9.76
"	25.82	6.43	8.33	8.80	17.64	9.09	16.90

6. Arsenic cycling in geochemistry

Figure 3 displays the oxidation –reduc-tion relations among some naturally occurring arsenic compounds [7]. Areas *A* and *B* represent the environments of the contact zones between streams and seawater (*A*) and seawater and lagoon (*B*), respectively. The area C is the environment of underground water based on Garrel *et. al.*[8]. It is possible that the arsenic released from its parent rocks would experience the former two environments before it re-released in the third that we find in the under-ground water. The arsenic, when dissolved in surface water was $H_2AsO_4^-$ [*A*]. It transformed into $HAsO_4^{-2}$ when it encountered seawater, especially the lagoon environment. The transformation can be a factor that improved the attractive force between the element and the slate particles and eventually resulted in the arsenic accumulation on the surface of the slate particles.

7. Formation of the aquifer

Water produced from artesian wells shows arsenic anomaly, especially from the deeper ones, can be related to the development of the delta and the shoreline. During the last thousands years, the rivers in this area deposited lots of load into Taiwan Strait, resulting in the formation of alluvial coastal plain, lagoon, barrier and open marine environments as we observe today. The terrigenous – sedimentation – dominated environment induces the outgrowth of the shoreline. Sediments bringing slate-debris transported into the area was buried at the base, with the later coming sediments covered (Fig. 4). As a result, the advance of the delta front or the shoreline and the thickening of the deposited Sediment, the As gathering slate debris were buried deeper and deeper land-ward and turned into the important aquifer, due to the coarser particle sizes. It is believed that the As is then re-liberated and the higher As –containing underground water is formed.

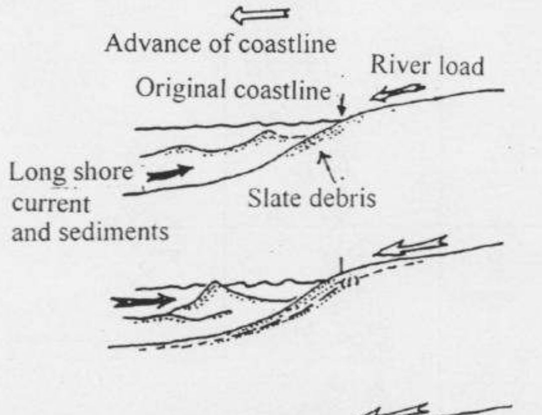

Figure 4. Schematic depiction of the outgrowth of coastline and the burial of slate – containing sediments.

8. Conclusions

Evidences show there is an arsenic anomaly existing in the underground water along the coastline area of southwestern Taiwan. However, the seawater and the solar salt produced using the seawater as the raw material show no sign of even slightly higher of As content. Arsenic found in this area is positively correlated with the slate ingredient content in the sediments. Examinations revealed that slate would be the source rock for delivery the element to the area. However, the slate sand that derived from the slate rock can be also an arsenic scavenger that has gathered the element and eventually eliminated the arsenic pollution possibly occurring in the adjacent sea. During the outgrowth of the coastline, the slate particles then buried at the base of the sediments, forming the aquifers in this area. In such a case, the arsenic may re-liberate into the environment, resulting in the arsenic anomaly in the artesian water.

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