

Research Article

Transportation and Transformation of Arsenic Species at the Intertidal Sediment-Water Interface of Bohai Bay, China

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Arsenic species including arsenite As(III), arsenate As(V), monomethylarsenate (MMA), dimethylarsenate (DMA), and some diagenetic constituents (Fe, Mn, and S²⁻) in porewaters along with the unstable arsenic species in sediments collected from a typical intertidal zone of Bohai Bay in China were measured. Their vertical distributions were subsequently obtained to reveal the transportation and transformation characteristics of arsenic at the intertidal sediment-water interface (SWI). Results show that the reduction of As(V) by microorganisms occurred in sediments, but the methylation of arsenic by microorganisms was weak in the intertidal zone. The distribution of As(V) was mainly controlled by Mn, whereas As(III) appeared to be more likely controlled by Fe. Arsenic in sediments mainly existed in a stable state, so that only little arsenic could be released from sediments when the environmental conditions at the SWI are changed. As(III) diffused from porewaters to the overlying water while the opposite was true for As(V) at that time when the samples were collected. The total diffusion direction for arsenic across the SWI was from porewaters to the overlying water with a total diffusive flux estimated at $1.23 \text{ mg}\cdot\text{m}^{-2}\cdot\text{a}^{-1}$.

1. Introduction

At present, our knowledge about the arsenic (As) cycles and species in marine sediments, especially in intertidal sediments, is limited. The ubiquitous use of anthropogenic arsenic associated with rapid economic development has significantly changed their original distribution patterns in natural environments, enabling their delivery to the intertidal zone from river catchments via fluvial transport, atmospheric deposition, and local wastewater discharge. The most common species of arsenic likely to be found in aquatic environments are arsenite As(III) and arsenate As(V) [1], but methylation probably occurs by activity of microorganisms, resulting in the formation of organic arsenic compounds such as monomethylarsenate (MMA) and dimethylarsenate (DMA) [2, 3]. The strong toxicity of inorganic arsenic leads to the important significance to carry out geochemical studies of arsenic in the environment.

Bohai Bay is a semienclosed shallow water basin located in the western region of the Bohai Sea in the northern China. Bohai Bay receives a vast amount of industrial and domestic sewage from Beijing and Tianjin through the Haihe River, Jiyunhe River, Yongdinxin River, and so on. It was reported that about $3.34 \times 10^5 \text{ kg/year}$ of arsenic was discharged into Bohai Bay via rivers [4]. Due to the large amount of contaminant inputs and poor physical self-cleaning capacity, Bohai Bay has become one of the most degraded marine systems in China.

The intertidal zone, strongly affected by both human activities and tide, is the transitional area of ocean and land, where sediment-water interface (SWI) is the boundary of the sediment and water phase. The generation, circulation, and migration of chemical substances are exceptionally active there [5]. The toxicity and mobility of arsenic in aquatic environment largely depend on its chemical form [6, 7]. For example, inorganic As(III) is generally more toxic than As(V), while organic arsenic compounds are generally

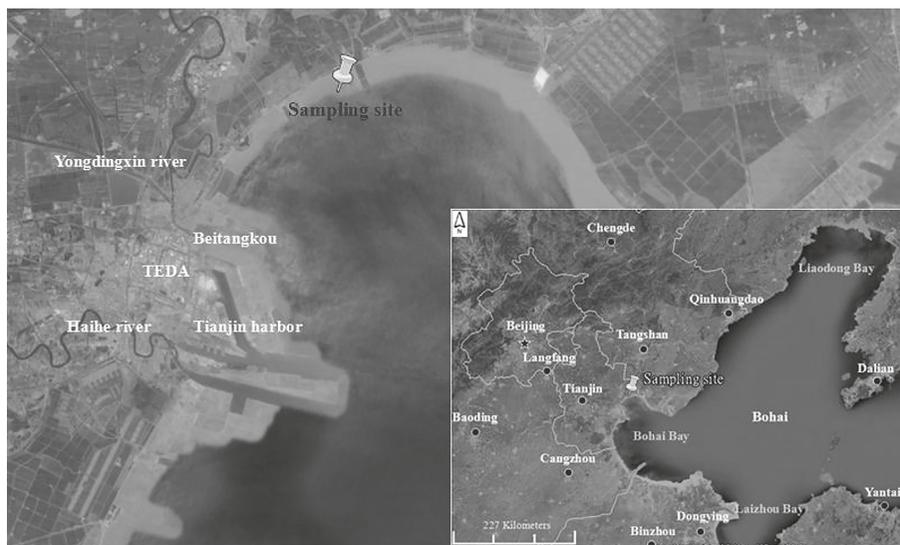


FIGURE 1: Geographic map of the sampling site.

nontoxic [6]. Therefore, studies of arsenic transportation and transformation at the intertidal SWI of Bohai Bay based on speciation analysis will be more significant. However, the geochemical cycle of arsenic is quite complicated here as many factors have strong effects on this process [8, 9]. At present, available data on arsenic in the intertidal SWI of Bohai Bay are insufficient for the evaluation of their physicochemical behaviors and total environmental impacts because the chemical states of arsenic in sediment and porewater need to be known to evaluate their mobility, bioavailability, and toxicity.

In this paper, we report the geochemical cycle of arsenic at the intertidal SWI of Bohai Bay based on the extraction of sediments using the method proposed by Ellwood and Maher [10] along with the measurement of dissolved arsenic species in porewaters and related physicochemical parameters at the SWI.

2. Materials and Methods

2.1. Site Description. Bohai Bay is one of the three bays of Bohai Sea in China covering an area of about $1.6 \times 10^4 \text{ km}^2$ with an average water depth of 12.5 m. There are several rivers flowing into Bohai Bay, by which it receives both industrial and domestic wastewater discharged from Beijing, Tianjin, and Hebei. All the wastewater through rivers and channels is directly drained into the near-shore water of Bohai Bay. Among 96 sewage outlets monitored in 2008, the water quality in the adjacent sea of 83% sewage outlets cannot meet the water quality requirement of marine function area, and the ecoenvironmental quality in the adjacent sea of 22% sewage outlets is in quite poor state. The exchange of water between Bohai Bay and the central Bohai Sea is relatively slow and weak for its semienclosed position and poor physical exchange capacity; thereby, most of the pollutants are gradually accumulated there.

The sampling site (Figure 1) is a typical intertidal zone of Bohai Bay in the Tianjin section ($N39^\circ12'23.6''$,

$E117^\circ54'54.2''$). The Tianjin coast belongs to an alluvial plain coast where the intertidal zone is quite broad and flat. With the rapid development of marine economy in Tianjin, the ecoenvironment of intertidal zone in the Tianjin section was seriously affected by the deposition of large amounts of different pollutants.

2.2. Sample Collection and Treatment. Overlying water at the surface was collected using a precleaned polyethylene container when the tide was at its highest in the daytime of that day. The temperature and pH of the overlying water were determined on-site using a thermometer and pH meter, respectively. An undisturbed surface sediment core about 30 cm depth was collected at the same place with a tailor-made Teflon sampler when the tide was at its lowest in the daytime. Subsequently, all samples were transported to the laboratory in an icebox. The sediment core was extruded in a glove-box bag filled with nitrogen gas to prevent oxidation. Sediment slices from each 1 cm depth were collected and placed in precleaned Teflon bottles and kept frozen at -80°C until analysis. Porewater sample in each sediment slice was obtained by centrifugation at 4°C .

2.3. Analytical Methods. The method for arsenic speciation analysis in water was established in our previous work [11]. Sulfides, dissolved iron (Fe), and manganese (Mn) were measured based on the National Standard Method in China (GB/T 17378.4-2007) and (GB/T 11911-89), respectively. The labile arsenic species including extracted arsenite [EAs(III)] and arsenate [EAs(V)] in sediments were determined according to the methods proposed by Ellwood and Maher [10]. Moisture, redox potential (Eh), total organic matter (TOM), and total arsenic (TA) in sediments were analyzed based on the National Standard Method in China (GB/T 17378.5-2007). In addition, sulfur species including elemental sulfur (ES), acid volatile sulfides (AVS), and mature pyrite sulfides (MPS) were determined by continuous

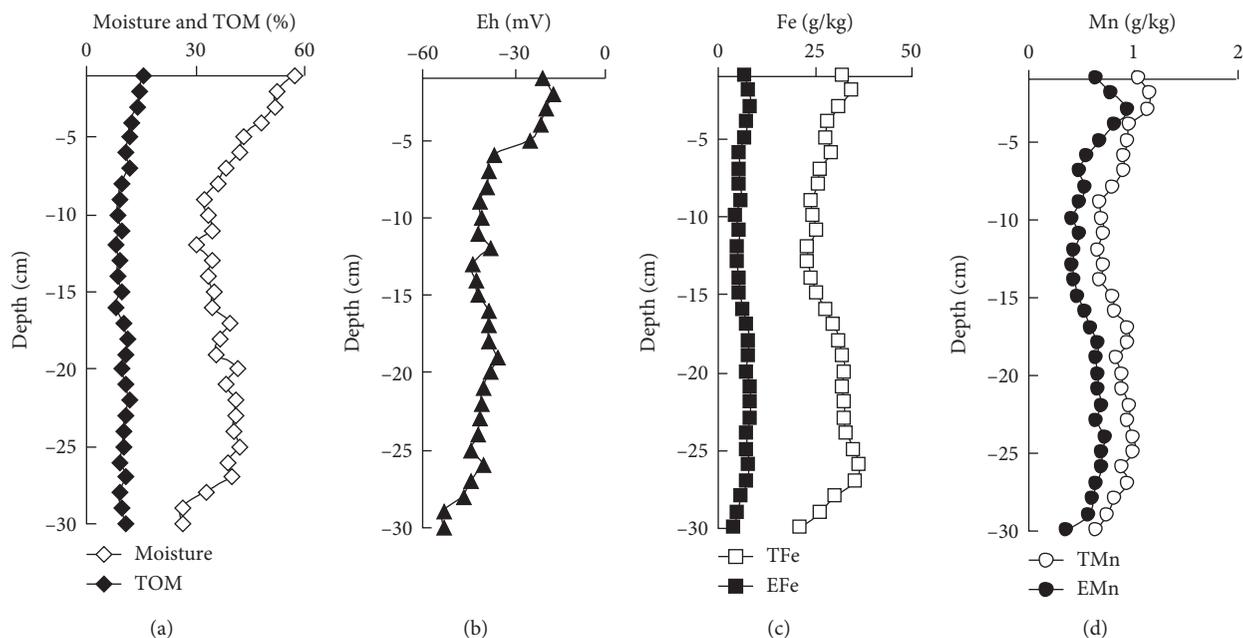


FIGURE 2: Profiles of moisture and TOM (a), Eh (b), Fe (c), and Mn (d) in sediments.

extraction iodometry [12]. Total iron (TFe) and total manganese (TMn) in sediments were analyzed by flame atomic absorption spectrometry (FAAS) according to the National Standard Method in China (GB/T 20260-2006).

3. Results and Discussion

3.1. Profiles of Labile Arsenic Species and Related Parameters in Sediments. The profiles of labile arsenic species and related physicochemical parameters in sediments are plotted in Figures 2–4.

Moisture reflects the porosity and the migration-diffusion rate of elements in sediments. The average moisture of sediments in this studied area was 38.65% and sharply decreased with depth at $-1\sim-12$ cm (Figure 2(a)), indicating a high diffusion rate in the top sediments. Organic matters in sediments provide the necessary survival conditions for bacteria that perform reduction of high valence elements such as Fe(III), Mn(IV), and S(VI). We found that TOM in this investigated site was especially high with an average content of 10.62% (Figure 2(a)). Redox potential (Eh) in intertidal sediments is strongly affected by oxygen in air, by which it presented gradual decrease with depth in the upper layer in Figure 2(b). The average Eh in this studied site was -38.1 mV indicating a weakly reducing environment in intertidal sediments. Fe and Mn hydrous oxides are the primary sources of soluble heavy metals in natural water. TFe in sediments was quite high with an average value of 29.36 g·kg $^{-1}$, while the labile EFe only accounted for about 23.13% (Figure 2(c)), indicating iron in intertidal sediments mainly existed in stable sulfides or in a residual state. On the contrary, TMn and EMn in sediments were quite low, and the labile EMn accounted for about 69.32% of the TMn content (Figure 2(d)).

Reduced sulfur, especially AVS (mainly composed of FeS), in sediments is an important control on the reactivity and toxicity of arsenic [13]. The average content of AVS in sediments was 0.15 g·kg $^{-1}$, and it was obviously higher in the bottom sediments than that in the upper layers as presented in Figure 3(b). It is noted that a sharp decrease of MPS in the top sediments was observed in Figure 3(c) with a maximum of 1.38 g·kg $^{-1}$ at the surface.

The average contents of EAs(III), EAs(V), and TAs in sediments were 0.51 , 2.18 , and 10.38 mg·kg $^{-1}$, respectively (Figures 4(a)–4(c)). In order to facilitate the comparison of trend and contents among EAs(III), EAs(V), and TAs, their profiles were plotted together in Figure 4(d). As expected, the average content of TAs was significantly higher than that of EAs(III) and EAs(V). It means that arsenic in intertidal sediments mainly existed in a stable state, or so called “inert” state. Labile arsenic [EAs(III) + EAs(V)] only accounted for about 25.9% of the TAs content, in which EAs(III) was only about quarter of EAs(V).

3.2. Profiles of Dissolved Arsenic Species and Related Parameters in Porewaters. The profiles of dissolved arsenic species and related physicochemical parameters in porewaters are plotted in Figures 5 and 6.

The pH of seawater is mainly controlled by the carbon dioxide-carbonate system, and its pH approximates 8.1 or fluctuates around this value. However, since the intertidal zone is affected by many factors, the measured pH was 8.24 (Figure 5(a)). Porewaters are often anoxic systems, in which the physicochemical reactions are remarkably different from the overlying water. We found that the pH of porewaters was significantly lower than that of overlying water in Figure 5(a). S $^{2-}$ is liable to react with dissolved iron to form FeS or FeS $_2$, and thereby the sulfides in overlying water and

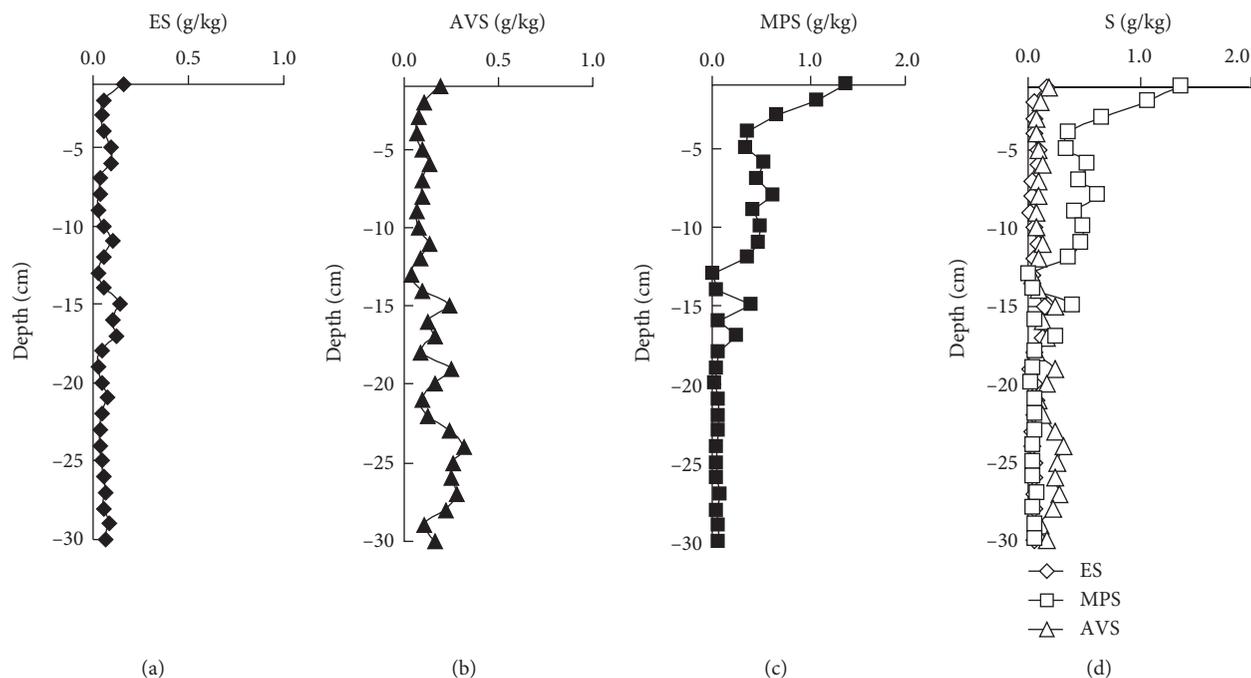


FIGURE 3: Profiles of ES (a), AVS (b), and MPS (c) in sediments and their comparison (d).

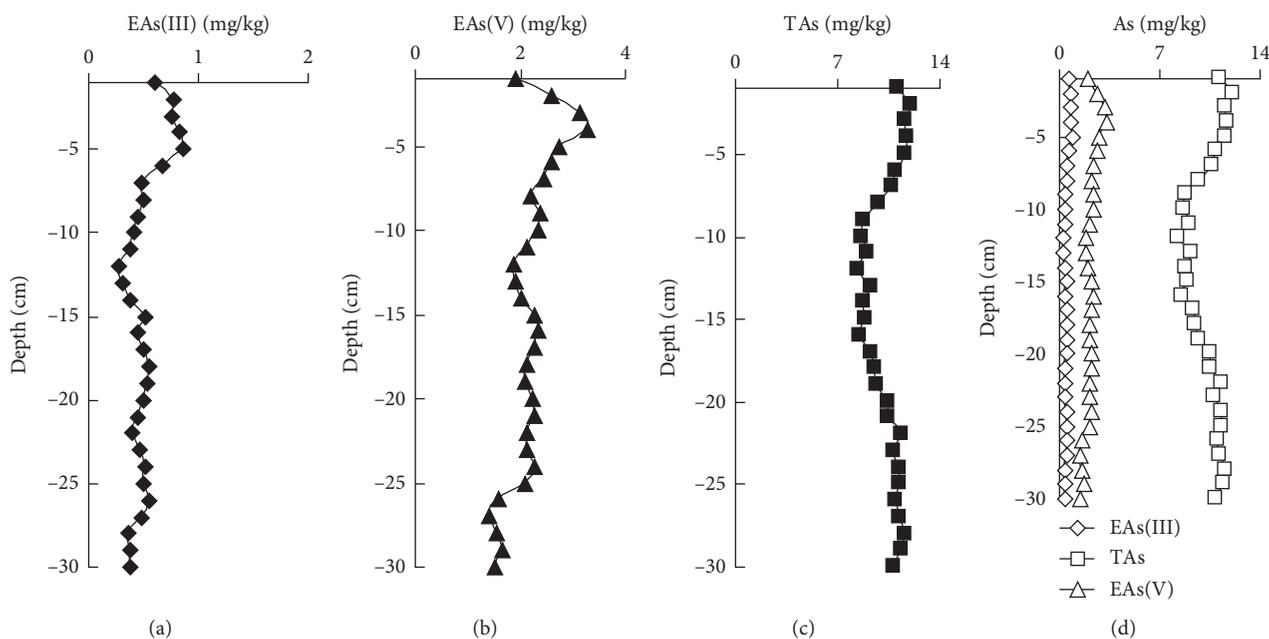


FIGURE 4: Profiles of EAs(III) (a), EAs(V) (b), and TAs (c) in sediments and their comparison (d).

porewaters were beyond the detection limit (Figure 5(b)). Dissolved Fe in overlying water was only $0.16 \text{ mg}\cdot\text{L}^{-1}$, but it reached an average of $0.60 \text{ mg}\cdot\text{L}^{-1}$ in porewaters (Figure 5(c)). Although dissolved Mn in bottom porewaters in Figure 5(d) was extremely low, an obvious concentration peak was presented in the profile of dissolved Mn in the upper sediment with a maximum of $11.13 \text{ mg}\cdot\text{L}^{-1}$ at the subsurface.

Dissolved oxygen (DO) in overlying water will cause a shift of redox equilibrium between As(III) and As(V)

toward As(V), by which the majority of As(III) is oxidized to As(V), by which the majority of As(III) is oxidized to As(V) because the tidal action increases the concentration of DO in the overlying water. Therefore, no As(III) could be detected in overlying water at the studied site, while As(V) reached $8.14 \mu\text{g}\cdot\text{L}^{-1}$. However, as it suffers less effects of DO in the porewaters, As(III) could be detected in porewaters at each depth with an average value of $2.11 \mu\text{g}\cdot\text{L}^{-1}$ (Figure 6(a)). As(V) was obviously higher than As(III) in porewaters in Figure 6(d), and the average concentration of As(V) was

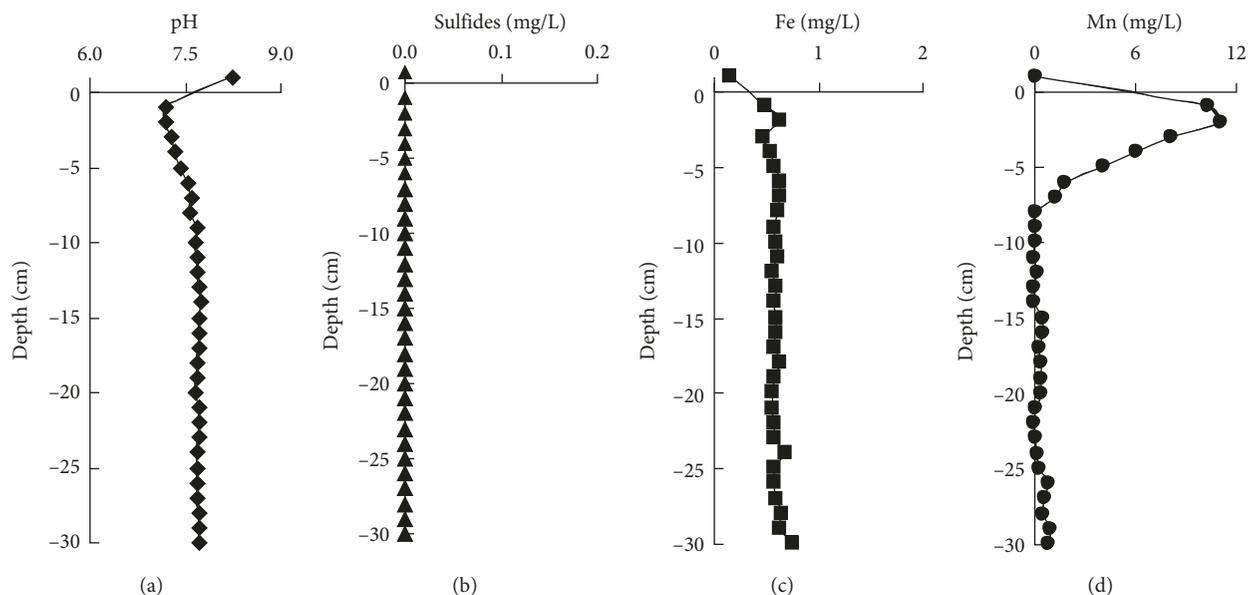


FIGURE 5: Profiles of pH (a), sulfides (b), dissolved Fe (c), and Mn (d) in porewaters.

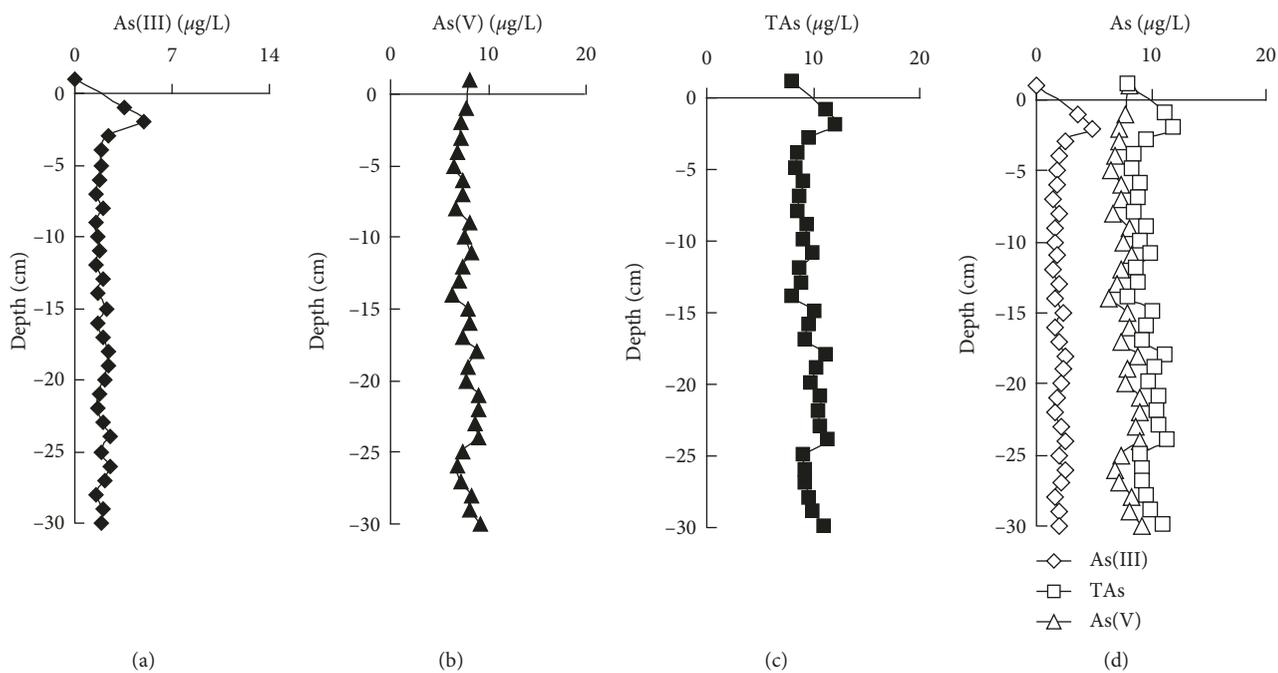


FIGURE 6: Profiles of dissolved As(III) (a), As(V) (b), and TAs (c) in porewaters and their comparison (d).

$7.75 \mu\text{g}\cdot\text{L}^{-1}$, which was about 3.7 times that of As(III). No organic arsenic compounds such as DMA and MMA were detected in porewaters, indicating the methylation of arsenic by microorganisms was weak in intertidal zone of the studied site.

3.3. Transportation and Transformation of Arsenic Species at the Intertidal SWI

3.3.1. Correlations of Related Parameters at the Intertidal SWI. In order to evaluate the correlations of different

parameters at the SWI, the coefficients of statistical correlation (r) are calculated and presented in Table 1.

Humus in sediments will seriously restrain arsenic adsorption onto the surface of iron oxides or hydroxide colloids by competitive adsorption [14]. Therefore, there should be a negative correlation between TOM and arsenic. However, both EAs(III) and EAs(V) presented positive correlations with TOM, suggesting that the distribution of arsenic in intertidal sediments was not dominated by TOM. Hydrous oxides of Fe and Mn are the main shelters of heavy metals in aqueous systems and thus have important effects

TABLE 1: Statistical correlations (r) between profiles of parameters at the intertidal SWI.

	TAs	EAs(III)	EAs(V)	As(III)	As(V)	TFe	EFe	Fe	TMn	EMn	Mn	ES	AVS	MPS	TOM	Eh	pH
TAs	1.00	0.53	0.11	0.40	0.03	0.58	0.44	0.04	0.68	0.70	0.52	-0.05	0.28	0.12	0.66	0.30	-0.49
EAs(III)		1.00	0.71	0.48	-0.37	0.37	0.46	-0.30	0.70	0.69	0.71	0.17	-0.06	0.45	0.68	0.81	-0.78
EAs(V)			1.00	0.11	-0.27	0.01	0.29	-0.41	0.44	0.46	0.45	0.00	-0.43	0.38	0.38	0.71	-0.58
As(III)				1.00	-0.07	0.46	0.39	-0.08	0.61	0.46	0.74	0.18	0.17	0.57	0.67	0.57	-0.66
As(V)					1.00	0.08	0.16	0.38	-0.07	-0.04	-0.29	-0.01	0.26	-0.30	-0.04	-0.40	0.36
TFe						1.00	0.86	-0.28	0.80	0.78	0.26	-0.02	0.58	-0.03	0.42	0.29	-0.21
EFe							1.00	-0.46	0.78	0.85	0.29	-0.09	0.33	-0.07	0.46	0.45	-0.25
Fe								1.00	-0.37	-0.43	-0.37	-0.14	0.19	-0.33	-0.32	-0.60	0.41
TMn									1.00	0.88	0.64	0.12	0.26	0.35	0.76	0.67	-0.64
EMn										1.00	0.55	-0.10	0.20	0.13	0.62	0.61	-0.55
Mn											1.00	0.29	-0.15	0.78	0.86	0.85	-0.96
ES												1.00	0.11	0.40	0.23	0.17	-0.22
AVS													1.00	-0.25	-0.01	-0.25	0.23
MPS														1.00	0.63	0.69	-0.82
TOM															1.00	0.73	-0.84
Eh																1.00	-0.90
pH																	1.00

on the distributions and speciation of arsenic at the SWI [15]. Arsenic is adsorbed on Fe and Mn oxides/hydroxides in sediments to form stable states, and positive correlations were obtained between TAs and TFe as well as between TAs and TMn, with correlation coefficients of 0.58 and 0.68, respectively. Usually, iron extracted by 25% hydroxylamine hydrochloride in acetic acid mainly exists as FeOOH in sediments, and a high correlation was found between extracted Fe and As (EFe and EAs) (Table 1).

A dynamic equilibrium exists between EAs(III) and EAs(V) in sediments. They will present negative correlation if the equilibrium is only controlled by Eh. However, the coefficient was 0.71, indicating that their distributions were not only controlled by Eh. EAs(III) will be oxidized at high Eh, and a negative correlation will thus be observed between Eh and EAs(III) if the distribution of EAs(III) is not influenced by other factors. However, a positive correlation ($r = 0.81$) was observed between Eh and EAs(III), demonstrating that the distribution of EAs(III) was not dominated by Eh. No significant negative correlation was found between As(III) and As(V) showing that the distribution of arsenic in porewaters was also not dominated by Eh. The correlation coefficients between As(III) and Fe and As(V) and Mn were -0.08 and -0.29 , respectively. Meanwhile, positive correlations between EAs(III) and EFe as well as between EAs(V) and EMn in sediments were presented. It is speculated that the distribution of As(V) was controlled by Mn, while As(III) was likely to be dominated by Fe.

3.3.2. The Flux of Dissolved Arsenic at the Intertidal SWI. Profiles of dissolved arsenic in porewaters and overlying water (Figures 6(a) and 6(b)) suggest probable diffusion of arsenic at the intertidal SWI. Based on the assumption that all dissolved arsenic diffusing upwards (or downwards) is transferred to the overlying water (or porewaters), the diffusive flux was estimated using the method applied by Ciceri et al. [16]:

$$J = \phi D_s \left. \frac{dAs}{dz} \right|_0, \quad (1)$$

where J is the flux of dissolved arsenic; ϕ is the porosity of sediment (0.50, calculated from the water content using the following formula: $\phi = V_{\text{water}} / (V_{\text{water}} + V_{\text{solid}})$ in which V is the volume obtained from mass and density of water and sediment); $(dAs/dz)_0$ is the concentration gradient calculated from the linear portion of dissolved arsenic concentration at the SWI; and D_s is the bulk sediment diffusion coefficient and is obtained from the formula [17]:

$$D_s = \phi D_0, \quad \phi < 0.7$$

$$D_s = \phi^2 D_0, \quad \phi > 0.7, \quad (2)$$

where D_0 is the free solution diffusion coefficient.

D_0 is affected by many factors such as pH, salinity, and so on. It is estimated based on the study by Tanaka et al. [18]. The average pH at the depths where the concentration gradient of arsenic species was calculated was used in calculating D_0 of As(III) and As(V) which are 10.7×10^{-6} and $7.6 \times 10^{-6} \text{ cm} \cdot \text{s}^{-1}$, respectively. The estimated diffusive fluxes for As(III) and As(V) across the SWI were 1.40 and $0.17 \text{ mg} \cdot \text{m}^{-2} \cdot \text{a}^{-1}$, respectively. However, it is noted that As(III) diffused from porewaters to the overlying water while the opposite was true for As(V), based on the variation direction of the concentration gradient. Therefore, the total diffusive flux for arsenic at the SWI was about $1.23 \text{ mg} \cdot \text{m}^{-2} \cdot \text{a}^{-1}$, and the direction was from porewaters to the overlying water.

3.3.3. Transportation and Transformation of Arsenic at the Intertidal SWI. Arsenic occurrence forms in the environment are affected by many factors. Thermodynamically, arsenic should mainly exist in As(V) species (H_2AsO_4^- , HAsO_4^{2-} , and AsO_4^{3-}) under oxidizing conditions and in As(III) species (H_3AsO_3 and H_2AsO_3^-) under reducing conditions. Commonly, there is an oxidized zone above the SWI and a reduced zone below the SWI. As shown in this

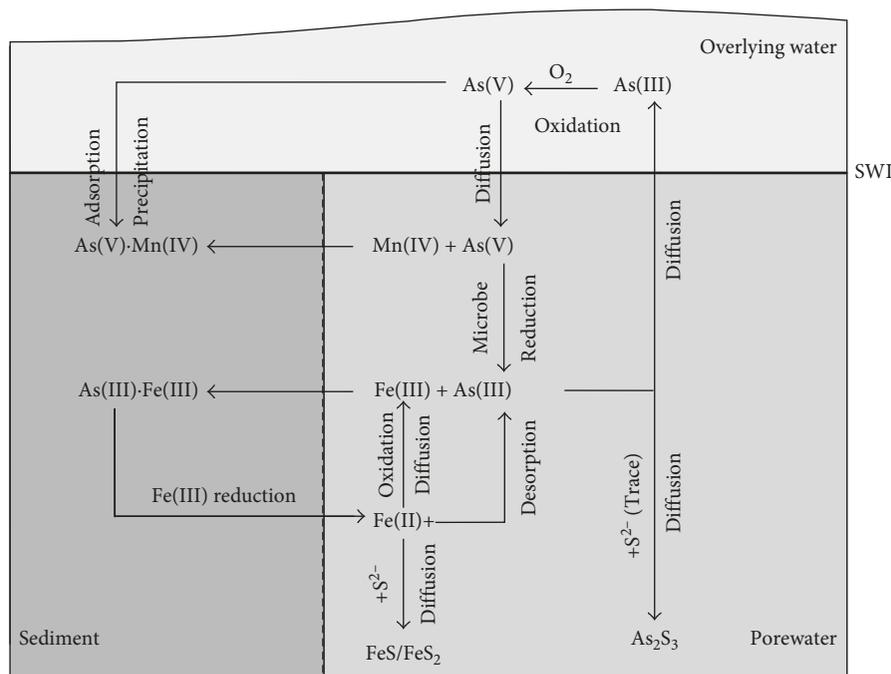


FIGURE 7: Transportation and transformation of arsenic at the intertidal SWI of Bohai Bay.

research, As(III) in the overlying water was below the detection limit, but it was detectable in the porewaters where reducing conditions prevailed. It is deduced that the transformation of arsenic species occurred in the sediments. Generally, arsenate cannot be reduced by other reagents in the environment, and thus, arsenite in porewaters mainly results from the reducing action of microorganisms [19]. Redox condition also has important effects on the release of arsenic in sediments. Arsenic can be adsorbed or coprecipitated into sediments in the oxidized layers during its migration toward the upper sediments. Sulfides of As(III) are thermodynamically stable in the presence of S^{2-} , which usually exists in a strongly reducing environment. Therefore, arsenic diffusing downward will be fixed by reacting with dissolved or particulate sulfides [20]. As shown in Figure 3(b), a high content of AVS in sediments was beneficial to the fixation of arsenic.

Arsenic is usually adsorbed on Fe oxides/hydroxides in an oxidizing environment, while Mn(IV) and Fe(III) are easily reduced to low valence and highly soluble Fe(II) and Mn(II) in a reducing environment, and then they migrate into the porewaters. Mn(II) is more stable than Fe(II) and more difficult to oxidize, so it can diffuse further than Fe(II), resulting in a peak of Mn(II) approaching the SWI (Figure 5(d)). Eh decreased with depth in sediments, but EFe did not decrease significantly (Figure 2(c)), implying that EFe was not visibly reduced to Fe(II) and subsequently entered into porewaters with the decrease of Eh. Therefore, no obvious increase of As(V) in porewaters appeared in Figure 6(b). It probably results from the slow dynamics of Fe(III) reduction after Fe(III) adsorbs arsenic and subsequently enters into sediments in the eodiagenesis. Based on the aforementioned analysis, the migration and transformation of arsenic at the intertidal SWI of Bohai Bay can be summarized and is shown

in Figure 7. It is obvious that the process is extremely complicated and further studies are required.

4. Conclusions

Different arsenic species and some diagenetic constituents (Fe, Mn, and S^{2-}) in porewaters along with the unstable arsenic species in sediments collected from a typical intertidal zone of Bohai Bay in China were measured to reveal the transportation and transformation characteristics of arsenic at the intertidal sediment-water interface (SWI). Results show that the reduction of As(V) by microorganisms occurred in sediments, by which 21% of the arsenic in porewaters was presented as As(III). No organic arsenic compounds were detected, indicating the methylation of arsenic by microorganisms was weak in the intertidal zone. Distribution of As(V) was mainly controlled by Mn, whereas As(III) appeared to be more likely dominated by Fe. Arsenic in sediments mainly existed in a stable state and was not easy to extract by the reagents used in this study. Significant diffusion of arsenic occurred at the SWI at that time when the samples were collected. As(III) diffused from porewaters to the overlying water, whereas the opposite was true for As(V). The net diffusion direction of arsenic across the SWI was from porewaters to the overlying water with a total diffusive flux estimated at $1.23 \text{ mg} \cdot \text{m}^{-2} \cdot \text{a}^{-1}$. Early diagenesis of arsenic at the intertidal SWI is quite complicated. Further studies need to be carried out to better understand and manage the overall system of the intertidal zone in Bohai Bay.

Conflicts of Interest

The authors declared that there are no conflicts of interest regarding the publication of this paper.

Acknowledgments

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