

Distribution and chemical speciation of dissolved inorganic arsenic in the Yellow Sea and East China Sea

WU Bin^{1,2}, SONG Jinming^{1*}, LI Xuegang¹

¹ Key Laboratory of Marine Ecology and Environmental Sciences, Institute of Oceanology, Chinese Academy of Sciences, Qingdao 266071, China

² Key Laboratory of Marine Sedimentology and Environmental Geology, First Institute of Oceanography, State Oceanic Administration, Qingdao 266061, China

Received 21 February 2014; accepted 4 July 2014

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Abstract

Distribution and chemical speciation of dissolved inorganic arsenic were examined in Yellow Sea and East China Sea. Results demonstrated that: (1) both As(III) and As(V) were detected, with As(V) dominated at 40% stations of surface water and 51% stations of bottom water; (2) influenced by the exchange of fresh and sea water, the type of surface sediment and the transport of various water masses, large values in surface water were observed along the coastal region and in the same latitude of Changjiang River Estuary and Hangzhou Bay, and in bottom water found in the southern area where the Taiwan Warm Current and Kuroshio Current influenced; (3) As(III) behaved non-conservatively in Changjiang River Estuary. Man-derived inputs cause substantial positive deviation from the theoretical dilution. The negative correlation of As(V) to salinity in surface water suggested that it behaved conservatively during the transportation along Changjiang River Estuary. While, the occurrence of As(V) up to the linear fit in bottom water indicated the eventual transfer from dissolved phase to particulate phase, which was impossible to be determined without the knowledge of arsenic level and speciation in suspended particulate matter. Further study is needed on the arsenic source/sink relationships based on the distribution profiles.

Key words: dissolved inorganic arsenic, distribution, speciation, Yellow Sea, East China Sea

Citation: Wu Bin, Song Jinming, Li Xuegang. 2015. Distribution and chemical speciation of dissolved inorganic arsenic in the Yellow Sea and East China Sea. *Acta Oceanologica Sinica*, 34(6): 12–20, doi: 10.1007/s13131-015-0682-z

1 Introduction

Arsenic contamination in natural waters is a world-wide issue that has been increasingly highlighted. It is ubiquitously distributed as trace constituent and environmental contaminant in aquatic ecosystem as a result of rock weathering and human activities (Al-Abed et al., 2007; Choong et al., 2007). Arsenic from these natural and anthropogenic sources is transported into aquatic environment through atmospheric deposition and riverine input.

Arsenic can exist in a variety of oxidation states (-III, 0, III, V) as both inorganic and organometallic species, with their distinct biochemical and geochemical reactivity (Meharg and Hartley-Whitaker, 2002). In waters with relatively low level of organic carbon, the inorganic arsenic species predominate, with higher level of As(V) (H_3AsO_4 , H_2AsO_4^- , HAsO_4^{2-} , AsO_4^{3-}) found under oxidizing conditions and greater amount of As(III) (H_3AsO_3 , H_2AsO_3^- , HAsO_3^{2-}) under reducing conditions (Masuda et al., 2005). Under strong reducing conditions, biological methylation of inorganic arsenic can occupy up to 59% of the total amount in lake water (Anderson and Bruland, 1991). The toxicity, mobility and bioavailability of arsenic in the oceanic environment are largely controlled by its chemical form. The reduced inorganic arsenic species, arsenite (As(III)), is more toxic than the oxidized inorganic species, arsenate (As(V)), and the common organic forms,

monomethyl arsenic (MMA) and dimethyl arsenic (DMA), are less toxic than the inorganic forms (Faxneld et al., 2011). Furthermore, As(III) is of greater concern due to its enormous potential for mobility in environment as compared with As(V) (Ryu et al., 2011; Canales et al., 2013).

Distributions of dissolved inorganic arsenic have been investigated in Pacific (Andreae, 1979; Cutter and Cutter, 2006), Atlantic (Cutter and Cutter, 1995; Cutter et al., 2001), and North Sea (Millward et al., 1996). Horizontally, elevated arsenic concentration was commonly observed in coastal water strongly influenced by the riverine input, atmospheric deposition, selective uptake by algae, degradability of organisms (Andreae, 1979; Seyler and Martin, 1991; Windom et al., 1991), adsorption and suspended particles (Zhang, 1995), and human activities (Sanders, 1985), which poses increasing concern of potential ecological and health impact. Vertically, arsenic concentration usually shows minor surface-water depletion and deep-water enrichment because of the similarity between arsenic and phosphate who are both members of Group V in the Periodic Table. The nutrient-like depth profile in the ocean suggested the uptake by phytoplankton and regeneration from degradation of organic detritus (Andreae, 1979; Cutter and Cutter, 1998).

Arsenic is extensively cycled at the Earth's surface and has complex geochemical characteristics. More attention has been

paid to the coastal seas because of their huge potential adverse effects on human health. A better understanding and quantification of the physical and biogeochemical processes governing arsenic transport and mobility however still remain as challenging tasks. So far, detailed studies in less contaminated regions are scarce. In addition, few comprehensive researches of dissolved inorganic arsenic on geochemistry in the Yellow Sea and East China Sea have been ever conducted. Consequently, the goal of our study was to comprehensively examine the geochemical behavior of dissolved inorganic arsenic in Yellow Sea and East China Sea. To achieve this goal, we (1) determined the occurrence level and chemical speciation to evaluate the extent of contamination; (2) analyzed the distribution to assess the possible influencing factors such as Changjiang River discharge and Kuroshio Current incursion; and (3) specifically investigated the behavior of each speciation in Changjiang River Estuary to interpret the estuarine processes.

2 Materials and methods

2.1 Study area and sampling methods

Cruise of R/V *Kexue 3* was carried out in the Yellow Sea and East China Sea (26.5°–37.0°N, 120.5°–125.5°E) during 11 to 31 August 2011 (Fig.1). The water depth ranged 14–157 m (average of 57 m) and increased from the coast to the open sea, with the largest value observed in the southeast area. The investigated region is the key area for material exchange and energy flow, with various input rivers, major hydrodynamic flow patterns and many economically developed cities. Samplings were conducted in upstream directions deployed by conductivity temperature depth (CTD) rosette according to profiles of temperature and salinity. As displayed in Fig.1, water samples from 55 stations (symbol: ●) were collected at surface (0.5–1 m beneath the surface) and bottom (2 m above the sediment-water interface) using 2.5 L Niskin bottles. Meanwhile, stratified samples were conducted at 13 stations of L and PN transect (symbol: ⊙) in the same way. The bottle was previously acid cleaned by soaking in 10% HCl for 2 d, and rinsed 3 times by milli-Q water before the sample was collected. 1 000 mL water samples were filtered immediately on board through pre-cleaned 0.45 μm pore-size filters within clean bench. The filtrate at each site was stored at 4°C in acid-cleaned high density polyethylene bottle and preserved with ultrapure HCl to a pH<2. Plastic gloves were worn throughout and clean procedures were maintained to prevent sample contamination.

2.2 Analytical methods

Dissolved inorganic arsenic species were measured by hydride generation atomic fluorescence spectrometry (HG-AFS, XGY-1011A, Institute of Geophysical and Geochemical Exploration, Chinese Academy of Geological Sciences, China) (Ren et al., 2009, 2010; Yao et al., 2006) within 2 d in Laboratory after cruise. As(III) was directly reacted with KBH_4 to form H_3As by acidifying sample with 2 mol/L HCl. H_3As was subsequently stripped out of solution using Ar gas and measured by AFS. Total dissolved inorganic arsenic (DIAs, As(III)+As(V)) was measured in the same manner shown above after reducing As(V) into As(III) by reducing with 5% sulfocarbamide-ascorbic acid and 2 mol/L HCl for 20 min. Differences between the two determinations provided information for concentration of As(V). The detection limits for DIAs and As(III) by HG-AFS were 0.008 μg/L and 0.001 μg/L, respectively. To evaluate analytical precision, all samples were de-

termined in triplicate (precision as relative standard deviation better than 7% for all species). The accuracy of measurement was monitored using China national reference standard sample (GSB 05-1113-1999). Recoveries of the determination were 95%–105% assessed by standard additions method. The pH and temperature were measured by CTD (SeaBird-911) and the salinity was calculated from the CTD data. SPM was determined by drying at 60°C in laboratory and re-weighing the GF/F filters which filtered certain volume seawater samples on board and were stored at –20°C in plastic Petri dishes.

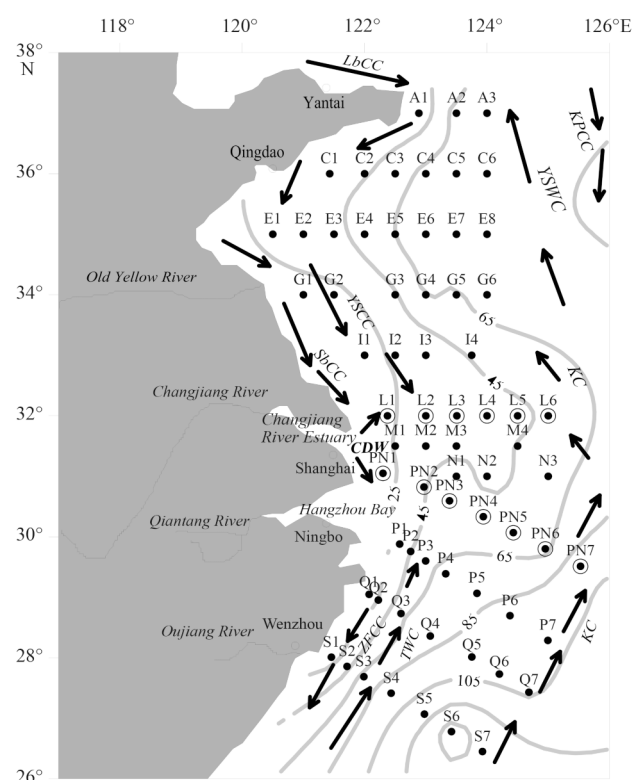


Fig. 1. Location of monitoring sites, various input rivers and major hydrodynamic flow patterns in the Yellow Sea and East China Sea. ● represents stations where surface and bottom waters were collected and ⊙ stations where stratified waters were collected. Dashed contours showed the water depth with unit of m. The arrow indicates the spreading direction of currents including LbCC: Lubei Coastal Current; SbCC: Subei Coastal Current; CDW: Changjiang Diluted Water; YSCC: Yellow Sea Coastal Current; YSCW: Yellow Sea Cold Water; ZFCC: Zhejiang Fujian Coastal Current; YSWC: Yellow Sea Warm Current; KPCC: Korean Peninsula Coastal Current; TWC: Taiwan Warm Current; and KC: Kuroshio Current.

2.3 Statistical analyses

Contour maps drawn by the software of Surfer 8.0 with the principle of interpolation method were used to exhibit the spatial distribution. Pearson correlation analysis by the software of SPSS 15.0 was used to explore the relationship of dissolved arsenic to environmental factors. In this work, we considered a value of $P < 0.05$ to indicate a significant difference and a value of $P < 0.01$ to indicate a highly significant correlation in all statistical analyses.

3 Results and discussion

3.1 Physicochemical parameters

Marginal sea is a mixing zone among marine, coastal and fluvial waters, and therefore is considered as the reactive zones for fluvial inputs. Temperature, salinity, SPM in our investigation was used to detect these features by precise and detailed oceanographic and geochemical observations (Fig. 2).

Temperature in surface water was within the range of 19.2–30.0°C, with a decrease from the south to the north. Low value observed in the northeast was contributed to the counter-clockwise circulation in the west of the southern Yellow Sea and high value found in the south was attributed to the incursion of Taiwan Warm Current (TWC) and Kuroshio Current (KC). By contrast, temperature in bottom water was within the range of 6.2–28.6°C, which was lower than that in surface water. It showed a decrease from the coast to off-shore sites, indicating that the c-

ontinental water input with low salinity diluted the seawater in the open sea. Influenced by the dilution of Changjiang Diluted Water (CDW), salinity of surface water varied 20.2–34.0 and exhibited the bidirectional extension feature as pointed by Wang (1998): one flowed toward north or northeast to enter the southwest of the southern Yellow Sea, and the other toward south or southeast to enter the East China Sea. Contributed by the incursion of KC, salinity of bottom water ranged 28.7–34.7, with high salt water extended to as far as the coastal region. In other areas, salinity in surface and bottom water vertically mixed similar to the demonstration in previous research (Wei et al., 2010). Concentrations of SPM measured in our research were highly variable (8.4–184.1 mg/L in surface and 12.3–706.8 in bottom). Higher levels were observed in the shore, since the coastal current intensified the sediment resuspension and continental weathering along the coast.

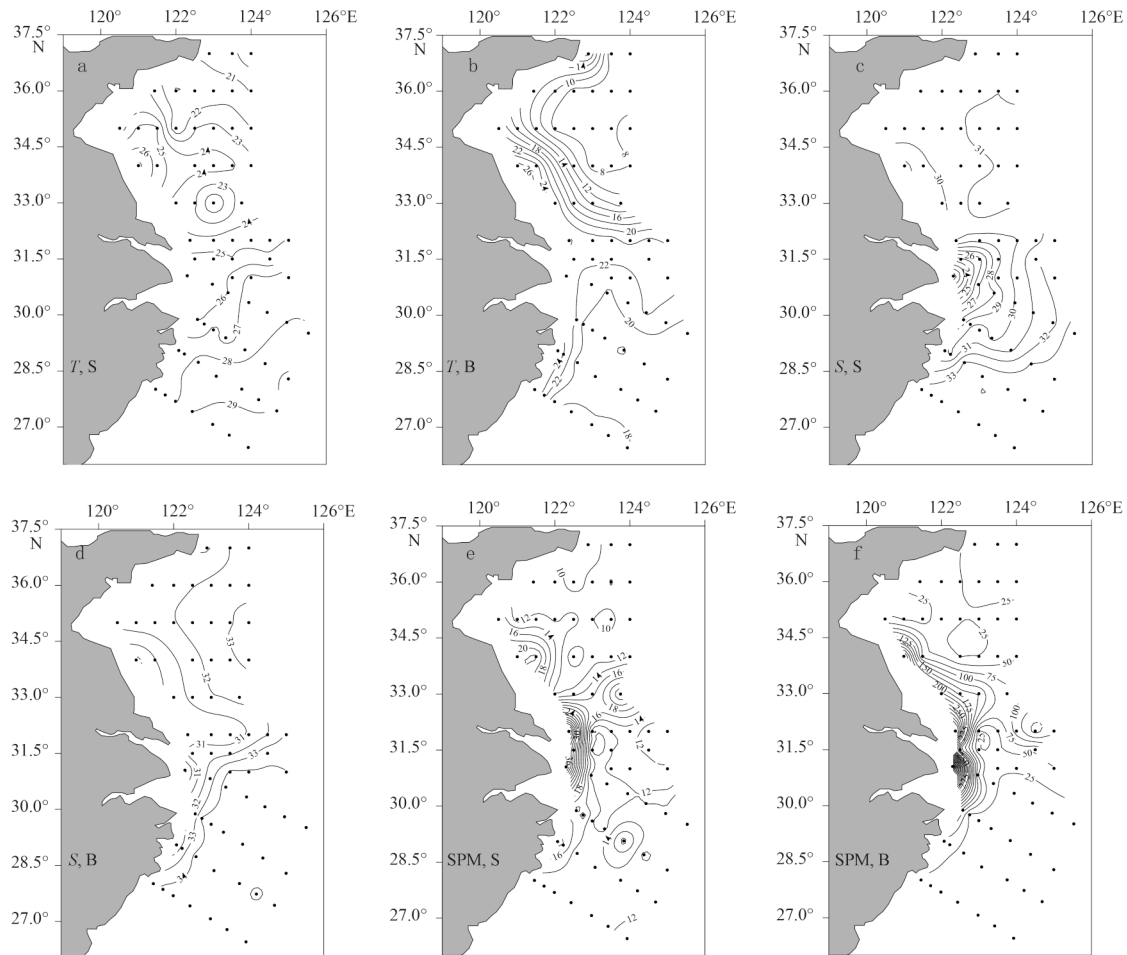


Fig. 2. Horizontal distribution of temperature (T , °C), salinity (S), SPM (mg/L) in the Yellow Sea and East China Sea. S represents surface and B bottom.

3.2 Occurrence level of arsenic in the Yellow Sea and East China Sea

Concentrations of dissolved inorganic arsenic (DIAs) in our study were within 0.50–4.18 $\mu\text{g/L}$ in surface water and 0.64–7.39 $\mu\text{g/L}$ in bottom water, which were lower than 10 $\mu\text{g/L}$, the World Health Organization (WHO) and US-EPA drinking water limit for dissolved arsenic (U.S. EPA, 2001; World Health Organization, 1993). The occurrence level of our investigation was similar to the

value determined in the Zhujiang River (Pearl River) Estuary (Yao et al., 2006) and Changjiang River (Yao et al., 2007), lower than that determined in the Tejo Estuary (Andreae et al., 1983), and higher than that determined in the Arabian Sea (Pettine et al., 1997), Mediterranean Sea (Cabon and Cabon, 2000), North Sea (Millward et al., 1996), Cape Basin (Statham et al., 1987) and Jiaozhou Bay (Ren et al., 2007) (Table 1). As listed in Table 1, interannual, seasonal and spatial variation of DIAs in seawater

worldwide indicated that occurrence level of dissolved arsenic depended on the sampling time and location, with high value generally observed in the wet season and in the estuary. Therefore, the relatively higher level of DIAs in our investigation are mainly due to the input from freshwater discharge (e.g., from the Changjiang River) in the west and the Kuroshio incursion in the east. The relatively higher content of DIAs in the Changjiang River (Yao et al., 2007) confirmed the great effect from the Changjiang discharge on the observed high value in our investigation.

3.3 Distribution of arsenic in the Yellow Sea and East China Sea

Occurrence level of arsenic is remarkably uneven and signi-

ficantly site-specific (Fig. 3), with the value higher than the world average baseline concentration in river water of 0.83 $\mu\text{g/L}$ and the level determined in the open sea of 1.42 $\mu\text{g/L}$ at most stations (Smedley et al., 2002). As(III) content varied from 0.08 to 3.21 $\mu\text{g/L}$ in surface and from 0.03 to 3.29 $\mu\text{g/L}$ in bottom. The highest value in surface and bottom was both found at Q1 which was adjacent to the Hangzhou Bay. The lowest one in surface was observed at N1 and in bottom at Q7. As(V) content ranged from 0.03 to 2.46 $\mu\text{g/L}$ in surface and from 0.03 to 7.28 $\mu\text{g/L}$ in bottom. The maximum in surface occurred at M4 and in bottom at Q7.

Dissolved arsenic concentration reflects the regional geochemical characteristics in the Yellow Sea and East China Sea

Table 1. Occurrence levels of dissolved inorganic arsenic in seawater worldwide

Research area	Sampling time	DIAs/ $\mu\text{g}\cdot\text{L}^{-1}$		Reference
		Surface water	Bottom water	
Yellow Sea and East China Sea	Aug. 2011	0.50–4.18	0.64–7.39	this study
Tejo Estuary	Apr. 1982	2.70–6.65	-	Andreae et al. (1983)
	Mar. 1992	1.03		
	Feb. 1993	0.97		
Arabian Sea	Jul. 1993	0.93	-	Pettine et al. (1997)
	Jul. 1994	1.17		
	Mar. 1995	1.42		
Mediterranean Sea	Dec. 1997	1.00	1.02	Cabon and Cabon (2000)
North Sea	Aug. 1988	0.15–1.11	-	Millward et al. (1996)
	Aug. 1989	0.62–1.08		
	Sep. 1989	0.84–1.57		
Cape Basin	Jan. 1979	1.32–1.67	1.48–1.87	Statham et al. (1987)
Jiaozhou Bay	Aug. 2001	1.38	1.29	Ren et al. (2007)
	Oct. 2001	1.17	1.20	
Zhujiang River Estuary	Jul. 1999	1.33–1.93	-	Yao et al. (2006)
	Jan. 2000	1.36–3.09		
Changjiang River	Apr.–May 1997	1.55–4.90	-	Yao et al. (2007)
	Apr.–May 2003	0.52–5.10		

(Fig. 3). Totally, elevated value in the Changjiang River Estuary was mainly from the riverine input. High value in the Yellow Sea was largely attributed to the KPCC and YSCC. The case in the East China Sea was due to the influence of the Changjiang River Estuary, Hangzhou Bay and inputs from water masses (ZFCC, TWC and KC). In surface water, As(III) and As(V) distributed similarly. Large values were observed in the estuary and near the shore, demonstrating the typical transport mechanism in marginal sea. Based on the distribution of salinity, large content of dissolved inorganic arsenic observed in the southern area was influenced by TWC, which could carry shelf water northward to as far as the Changjiang River Estuary (Beardsley et al., 1985; Shaw, 1992). In bottom water, high value of As(III) was occurred along the coast of Zhejiang Province where aquaculture prevailed, indicating the anaerobic environment induced accelerated the reduction from As(V) to As(III). As(V) concentration enriched in the southeast region where the KC reaches. Research on the interface of marine sediment-water is essential for the marine matter cycling. The highest value of As(III) in surface and bottom water was both found at Q1, where the surface sediment was dominated by clay (Qin et al., 1987). Finer grain size (<0.004 mm), with worse transmission and higher organic matter content, could effectively reduce As(V) to As(III). In comparison, low value of As(III) occurred in the region where the sediment was

dominated by sand with better transmission.

3.4 Speciation of arsenic in the Yellow Sea and East China Sea

Investigation to date has focused largely on total arsenic level in water, but it is now understood that toxicity varies with chemical form of arsenic (Jain and Ali, 2000). As reactivity and toxicity are altered by the transformation of chemical form, the observed variation in arsenic speciation has considerable geochemical and ecological significance. In order to assess the behavior and potential impact of dissolved inorganic arsenic on aquatic environment, a good understanding of speciation of arsenic is required. As(III)/DIAs ratio can reflect the predominant species and provide a signature for the source in the drainage area, which was used to examine the oxidation-reduction equilibrium and dynamic in water environment.

In surface water, the ratio varied between 0.05 and 0.95. In bottom water, the ratio was within the range 0.01–0.97. As(V) dominated at 40% stations of surface water and 51% stations of bottom water, with As(III)/DIAs ratio less than 0.5 at these stations. At the other stations with As(III)/DIAs ratio more than 0.5, As(III) was the main chemical form. Generally, As(III) mostly exists in reducing water and hydrothermal water, while As(V) is more often present in surface water and oxidizing groundwater

(Lizama et al., 2011). Yet, in present study, both chemical species were detected as previously reported (Andreae, 1979; Cutter and Cutter, 1995; Sanders, 1985). This discrepancy is often attributed to biological activity or kinetic effect (Cutter et al., 2001). Similar to dissolved antimony, a possible external source of As(III) in oxic waters is atmospheric deposition, and the internal source of As(III) includes biotic and abiotic reduction of As(V) (Choong et

al., 2007). In addition, algae bioaccumulation and detoxifying mechanism may be important because of their possible role in redox speciation (Mandal and Suzuki, 2002). In short, it is possible both biological uptake/release and geochemical transport affect the speciation of dissolved inorganic arsenic in water environment.

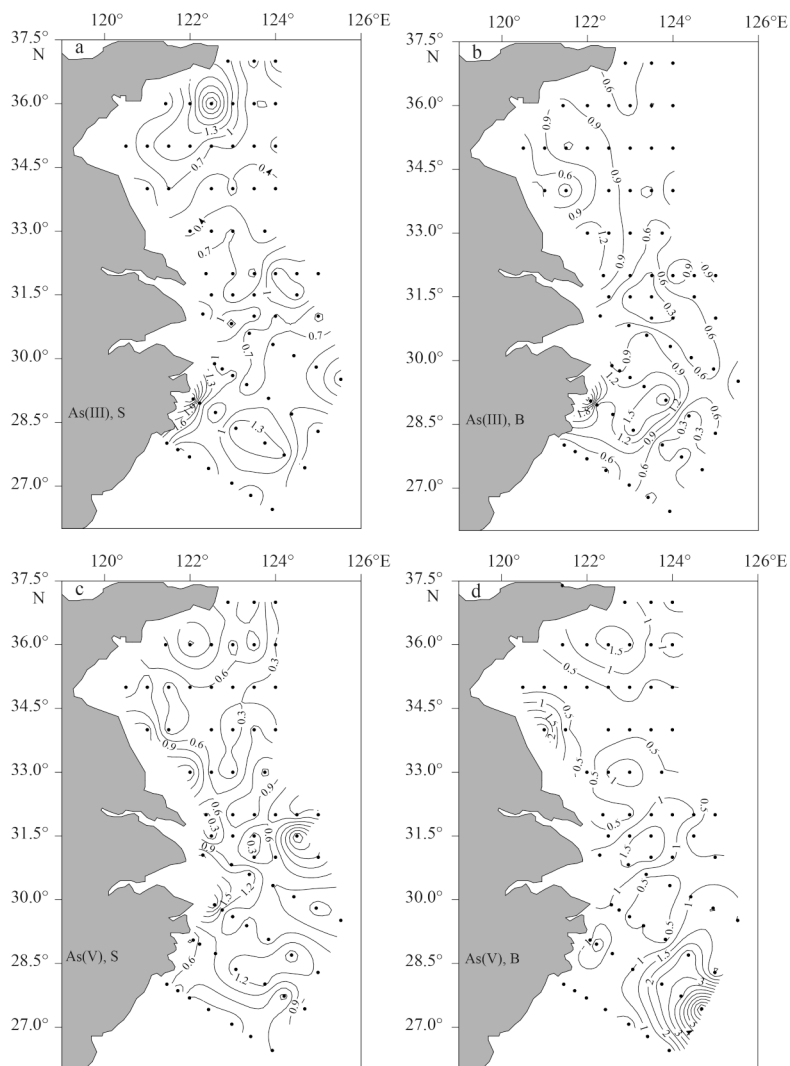


Fig. 3. Horizontal distribution of dissolved inorganic arsenic ($\mu\text{g/L}$) in the Yellow Sea and East China Sea. S represents surface and B bottom.

3.5 Conservative and non-conservative behavior of arsenic in the Changjiang River Estuary

Distribution of dissolved inorganic arsenic was dominated by physical process with a fairly stable freshwater end-member. Taking into account of the huge loads of water and sediment from the Changjiang River and relatively short flushing time of the East China Sea shelf, emphasis was given to two transects (L and PN). The L transect, with six stations sited along 32°N , was the boundary line between the Yellow Sea and the East China Sea. The PN transect was spreading from the Changjiang River Estuary (31°N , 122.5°E) to Ryukyu Islands (29.5°N , 125.5°E), which was influenced by a variety of water masses including CDW, TWC, Shelf Mixed Water (SMW), Kuroshio Surface Water (KSW), Kuroshio Subsurface Water (KSSW) and Kuroshio Inter-

mediate Water (KIW). The influence of various water masses on each transect was examined by T-S diagram (Fig. 4). Meanwhile, distribution of temperature, salinity and dissolved inorganic arsenic in each transect was displayed in Fig. 5.

For L transect, the CDW influenced the coastal stations and the SMW influenced stations far from the shore. Temperature in surface was higher than that in bottom, with notable stratification. Salinity in surface varied largely, with fresh water entering the sea from the west and meeting the high salinity water from the east. Concentration of As(III) ranged within $0.03\text{--}1.55\ \mu\text{g/L}$, with the lowest value occurred at 10 m depth of L2 and the highest at surface water of L4. Concentration of As(V) varied within $0.03\text{--}1.33\ \mu\text{g/L}$, with the lowest level appearing at the bottom of L6 and the highest value at the bottom of L4. For PN tran-

sect, temperature increased with the distance from the coast and the water depth. While, salinity was low in surface coastal water and high in the bottom open sea water. Content of As(III) was in the range of 0.11–1.91 $\mu\text{g/L}$, with the lowest value occurred at 30 m depth of PN4 and the highest one at 30 m depth of PN2. Carried by CDW, concentration of As(III) at PN1 and PN2 which

were near to the Changjiang River Estuary was higher. Content of As(V) was within the range of 0.01–1.63 $\mu\text{g/L}$, with the lowest occurred at the 10 m depth of PN2 and the largest one at the bottom of PN2. In PN transect, the mean value of As(III) (0.85 $\mu\text{g/L}$) was higher than that determined in September 2002 (0.03 $\mu\text{g/L}$) and 2003 (0.11 $\mu\text{g/L}$) and the average content of As(V) (0.86 $\mu\text{g/L}$)

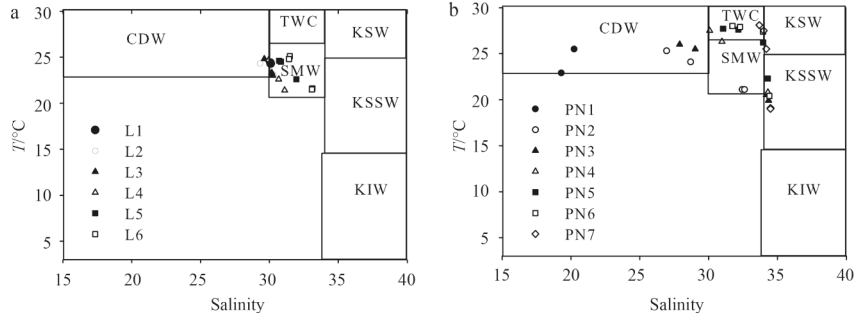


Fig. 4. T-S diagrams in L and PN transects. CDW represents Changjiang Diluted Water, TWC Taiwan Warm Current, SMW Shelf Mixed Water, KSW Kuroshio Surface Water, KSSW Kuroshio Subsurface Water, and KIW Kuroshio Intermediate Water.

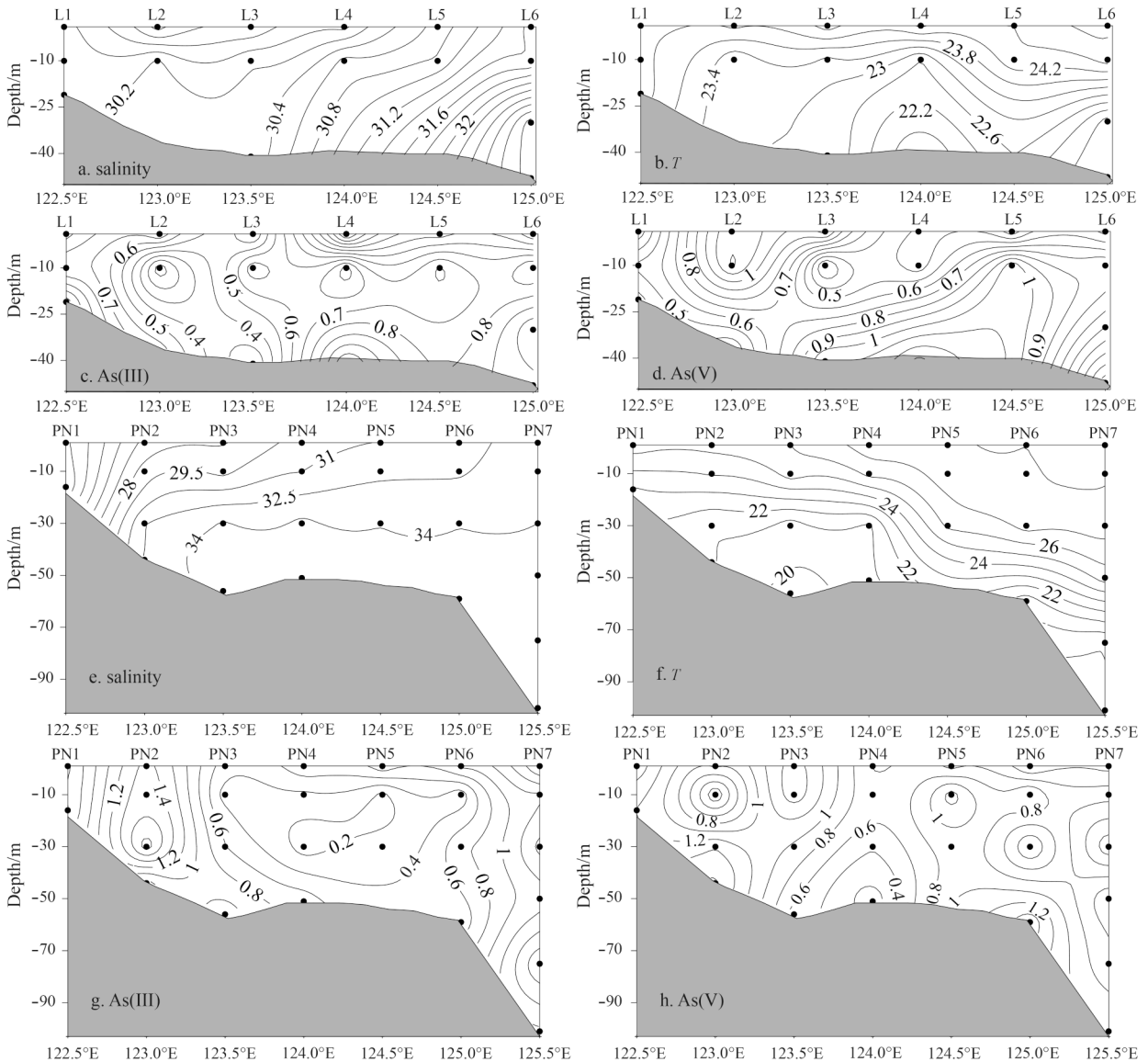


Fig. 5. Vertical distributions of temperature (T , $^{\circ}\text{C}$), salinity and dissolved inorganic arsenic ($\mu\text{g/L}$) in L (a-d) and PN (e-h) transects.

was slightly lower than that detected in September 2002 (1.17 $\mu\text{g/L}$) and 2003 (0.90 $\mu\text{g/L}$) (Ren et al., 2010). Result showed that KSW was rich in As(III) (1.83 $\mu\text{g/L}$, PN7), which was obviously higher than that in the Changjiang River Estuary (0.36 $\mu\text{g/L}$, PN1). Totally, As(III) concentration decreased from PN1 to PN4, since dissolved inorganic arsenic carried by the Changjiang River was diluted by the SMW. Whereas, with the distance from the coast, influenced by the KSSW, dissolved inorganic arsenic enriched in the bottom water.

To clearly examine the behavior of dissolved inorganic arsenic in the Changjiang River Estuary, relationship of dissolved inorganic arsenic to salinity in L and PN transect were displayed in Fig. 6. Results showed As(V) concentration negatively varied with the salinity in surface water ($R=-0.780$, $P<0.01$), suggesting that it had behaved conservative during transportation along the

Changjiang River Estuary. This behavior would be particularly enhanced during periods of high water discharge. While, As(V) concentration did not correlate to salinity in bottom water. The occurrence of As(V) up to the linear fit would suggest eventual transfer from dissolved to particulate phase in bottom water. Similarly, As(III) failed to relate to salinity in both surface and bottom water as well, indicating it behaved non-conservatively in the Changjiang River Estuary. These non-conservative behaviors were suggested to be related to some possible transfers from dissolved to particulate phases. Whereas, no significant relationship was found between dissolved inorganic arsenic and SPM in L and PN transects (Fig. 6). It becomes quite difficult to sort out which processes are of primary importance for dissolved inorganic arsenic behavior without the knowledge of arsenic levels and speciation in SPM.

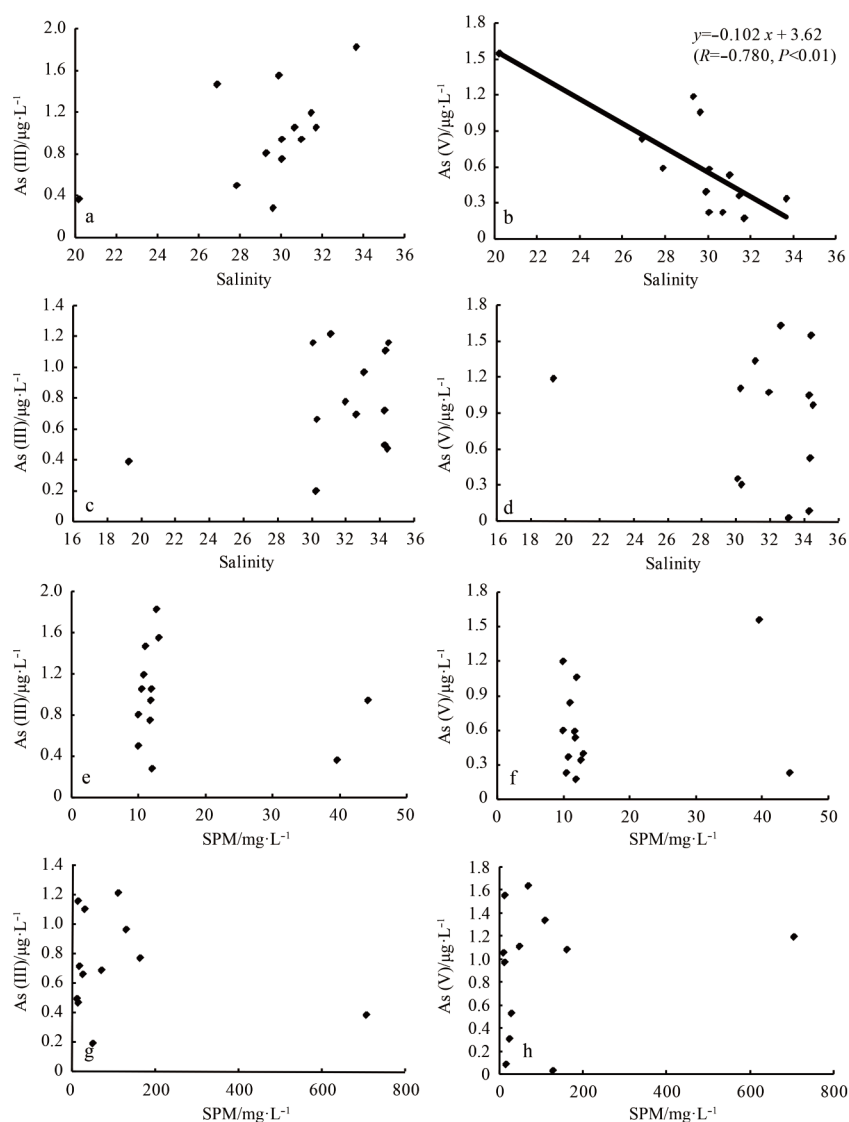


Fig. 6. Relationships of dissolved inorganic arsenic to salinity and SPM in L and PN transects. a, b, e and f. Surface water and c, d, g and h. bottom water.

The speciation and behavior of arsenic appeared to show considerable variation probably due to different biogeochemical process in various estuaries. Arsenic has been reported to be conservative in the southeastern U.S. estuaries (Froelich et al., 1985),

Rhone River Estuary (Garnier et al., 1996), Thames Estuary (Millward et al., 1997) and Humber Estuary (Kitts et al., 1994). In the Huanghe Estuary, arsenic behaves conservatively during the low water stage, while it presents a mid-estuarine maximum during

the high water stage (Huang et al., 1988). Non-conservative behavior, characterized by either removal or mobilization, has been found in other systems, such as the Seine Estuary in France (Michel et al., 1999), several UK estuaries (Langston, 1984), the Krka Estuary (Seyler and Martin, 1990), and the Schelde Estuary (De Gieter et al., 2005).

4 Conclusions

Overall, total dissolved inorganic arsenic concentration in the Yellow Sea and East China Sea are substantially heterogeneous: ranging from 0.50 to 4.18 $\mu\text{g/L}$ in surface water and from 0.64 to 7.39 $\mu\text{g/L}$ in bottom water. The average was significantly higher than the world average baseline concentrations in river water of 0.83 $\mu\text{g/L}$ and the level determined in the open sea of 1.42 $\mu\text{g/L}$ but all lower than the World Health Organization (WHO) and US-EPA drinking water limit of 10 $\mu\text{g/L}$. Combined with the distribution of salinity, higher values of dissolved inorganic arsenic found along the shore indicated the effect of land-source input from the adjacent rivers and those observed in southern stations imply the incursion of Taiwan Warm Current and Kuroshio Current. Both As(III) and As(V) were detected, with As(V) dominated at 40% stations of surface water and 51% stations of bottom water determined by the ratio of As(III)/DIAs. With regard to the behavior in the Changjiang River Estuary, As(V) behaved conservatively in surface water and non-conservatively in bottom. While, As(III) had non-conservative behavior in the whole estuary.

In future, more comprehensive field investigations are necessary to characterize the arsenic distribution pattern as well as possible explanatory factors. This will help better understand arsenic behavior in the coastal zones.

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