



# Spatio-temporal distribution and environmental risk of arsenic in sediments of the East China Sea

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## ABSTRACT

To understand the spatial and temporal distributions, environmental risks and sources of As, 38 surface sediments and three sediment cores were collected in the East China Sea (ECS). As concentrations in sediments ranged from 1.70 to 22.1  $\mu\text{g/g}$  with an average of 11.5  $\mu\text{g/g}$ . As concentration had positive correlations with clay + silt and total organic carbon (TOC) content with higher As values near the Changjiang Estuary and Hangzhou Bay mouth, indicating fine-grained sediments and riverine inputs as the main factors controlling As distribution. Chemical fraction analysis, sediment quality guidelines, together with geoaccumulation index, suggested that As in sediments of the ECS was at low risk, low adverse effect and moderately polluted level. Vertical profiles of As concentrations and burial fluxes, combined with  $^{210}\text{Pb}$  dating, reflected the As pollution history in the ECS within the last one hundred years. It showed a stable trend before one hundred years ago, a slight increase from the 1900s to 1990s, with peak values in the 1980s and 1990s, and a declining trend after the 1990s to 2009 with a relatively low value in 1998. These trends were closely associated with both natural and anthropogenic activities.

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## 1. Introduction

Arsenic as a widespread element exists naturally in the environment, and also comes from anthropogenic activities (Chapagain et al., 2009). As is of significant concern due to its acute and chronic toxicity and carcinogenic effects on marine organisms and humans (Cutter and Cutter, 1995; Oremland and Stolz, 2003; Routh and Hjelmquist, 2011). Thus, As was classified as a priority pollutant by the Environmental Protection Agency of the United States (USEPA, 1998) and China National Environmental Monitoring Centre (CNEMC, 1990).

The natural biogeochemical cycle of As has been changed by human activities, which gradually intensified the transfer of As from continents to rivers, seas, and even oceans. After entering into marine environments, As is adsorbed on suspended particulates and then deposited on sediments, leading to As enrichment in sediments (Szefer et al., 1995; Duan et al., 2011). On the contrary, when environmental conditions change, As associated with sediments is released to overlying water, threatening the aquatic biota. This suggests that sediments as the secondary source of As could affect water quality and ecology. The concentration and distribution of sedimentary As are closely related to the sedimentary environmental conditions, which mainly influences the chemical fractions of As. Besides, evaluation of potential toxicity and bioavailability of As is also dependent on chemical fractionation. Sequential extraction analysis has

been used to determine chemical fractions of As in sediments by many scientists (Tessier et al., 1979; BCR, 1993; Baig et al., 2009; Eiche et al., 2010; Wang et al., 2010; Routh and Hjelmquist, 2011).

On the other hand, in order to reflect the traces left by natural and anthropogenic activities in marine environment and to forecast their influence on environmental changes in the next few years, it is essential to reconstruct the environmental evolutions over the last several decades (Legesse et al., 2002). Due to its stability and non-biodegradation, As could accumulate and leave fingerprints in sediments over time. In core sediments, elevated As level can result from natural and anthropogenic occurrence. The anthropogenic sources of As mainly include insecticide, phytocide, paint and pigment production, leaded gasoline manufacture, fossil fuel combustion, smelting of arsenic-bearing ores and electronic industries (Flynn et al., 2002; Smedley and Kinniburgh, 2002; Paul et al., 2009). Therefore, the record of temporal variation of As in sediments could be used as an effective proxy to reflect its historical natural and anthropogenic (e.g., agricultural and industrial) inputs. The historical reconstruction of As pollution could be performed by determination of vertical distributions and burial fluxes of sedimentary As, combined with dating methods (e.g.,  $^{210}\text{Pb}$  and  $^{137}\text{Cs}$  dating) (Álvarez-Iglesias et al., 2007; Ruiz-Fernández et al., 2007; Duan et al., 2011).

Because of its potential toxicity and harm to marine organisms, As has been and should continue to be paid much more attention. To understand geochemical behaviors, evaluate ecological risks and reflect the historical evolutions of sedimentary As in the ECS, As concentrations and its chemical fractions in sediments were analyzed. The aims of the study were to (1) analyze spatio-temporal distributions

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of As and their controlling factors in the ECS; (2) evaluate the environmental risks of sedimentary As in the ECS; (3) discuss the origins of sedimentary As in the ECS; and (4) finally trace the time evolution of As in sediment cores.

## 2. Materials and methods

### 2.1. Study area

As one of the typical marginal seas in the western Pacific Ocean, the East China Sea (ECS) is surrounded by the Chinese mainland to the west, the Kuroshio Current to the east, Taiwan and the Taiwan Strait to the south, and the Yellow Sea to the north with the width and depth of 4500 km and 130 m, respectively (Fang et al., 2009; Song, 2010). Thus, a massive exchange occurs between the ECS and the Yellow Sea and Kuroshio Current (Chen et al., 1995). Besides, the Changjiang River as the world's fourth largest river based on suspended load has the average freshwater and sediment influxes of  $\sim 9.25 \times 10^{11}$  m<sup>3</sup>/yr and  $4.61 \times 10^8$  t/yr, respectively. It can discharge large amounts of terrigenous sediments to the ECS (Hori et al., 2001; Zhang and Liu, 2002).

With the rapid industrialization and urbanization development in the last few years, especially since 1978, when China's reform and opening up began, a high loading of industrial, agricultural and domestic sewage and solid wastes from coastal cities (e.g., Shanghai and Ningbo) was discharged into the Changjiang River and the ECS directly. Consequently, the ECS is suffering from environmental deterioration, and studies on the ECS are critical and necessary.

### 2.2. Sample collection

Thirty-eight surface sediments and three sediment cores from the ECS were collected during 10 to 29 May 2009 with the cruise of "Kexue 1", with the latitude of 29–32° N and longitude of 122–126° E (Fig. 1). Surface sediments (0–2 cm) were sampled by a box sampler. Immediately after collection, samples were kept in pre-cleaned polyethylene bags, sealed, and refrigerated until lab analysis.

Three sediment cores (G1, D2 and A5) were collected at the extension of the Changjiang River using a gravity corer deployed from the cruise of "Kexue 1" in May 2009 (Fig. 1). Core G1 (122°31.05'E, 29°30.25'N) is located on the shore of the northern Zhejiang and Fujian Provinces and the water depth is 32 m; core D2 (122°30.54'E, 31°01.37' N) is located near the Changjiang Estuary and the water depth is 16 m;

core A5 (124°59.66'E, 32°30.04'N) is located near the Cheju Island and the water depth is 59 m. Immediately after collection, cores were sectioned at 2 cm intervals and stored in pre-cleaned polyethylene bags, sealed, and refrigerated until lab analysis. Samples were dried in an oven at 60 °C for 72 h, and then were ground using an agate mortar and pestle for homogenization.

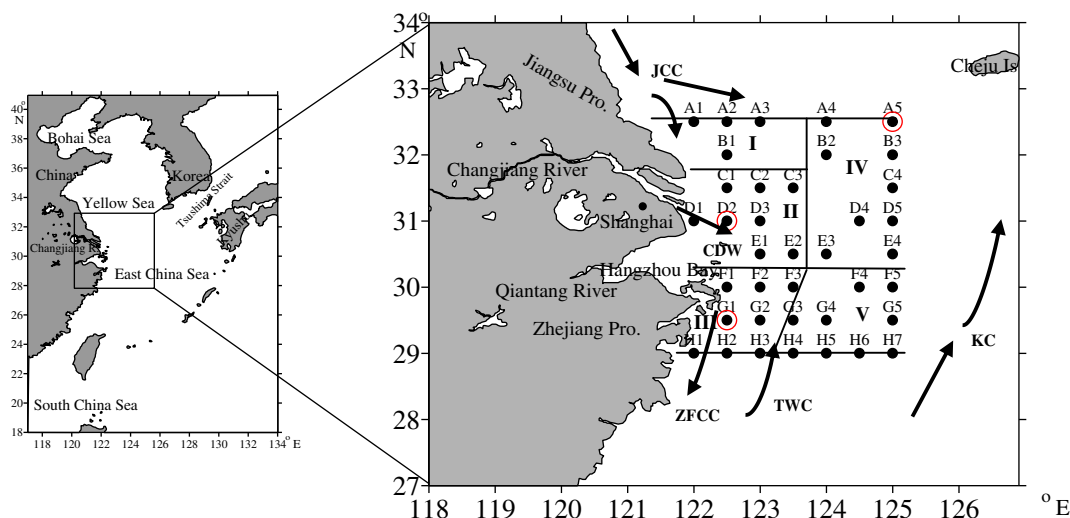
### 2.3. Sediment analyses

The analytical technique for As was conducted by modifying a method of "Technical Specification for Marine Chemistry Research" (NOB, 2006). About 0.2 g dry sample was digested with a mixture of 10 ml aqua regia in Teflon digestion vessel heated in a water bath for 1 h. Then, the solution was pre-reduced with 10% sulfocarbamide-ascorbic acid for 20 min. As concentration was determined by atomic fluorescence spectrometry coupled with a hydride generator (HG-AFS) using 0.7% KBH<sub>4</sub> with 0.05 M NaOH with a detection limit of 0.03 µg/g.

The sequential selective extraction method of Tessier et al. (1979) was performed to determine chemical fractions of As in sediments in this study. About 1.0 g dry sample was used for sequential extraction comprising the following steps.

The exchangeable fraction was extracted with 8 ml of 1 M MgCl<sub>2</sub> (pH 7.0) with sustained vibration at room temperature for 1 h. Residue from the first extraction step was further leached with 8 ml of 1 M NaOAc adjusted to pH 5.0 by HOAc with sustained vibration at room temperature for 5 h for the carbonates fraction. Residue from the second extraction step was leached with 20 ml of 0.04 M NH<sub>2</sub>OH · HCl in 25% HOAc with sustained vibration at 96 °C for 5 h for the Fe–Mn oxides fraction. Residue from the third extraction step was extracted with 3 ml of 0.02 M HNO<sub>3</sub> and 5 ml of 30% H<sub>2</sub>O<sub>2</sub> adjusted to pH 2 by HNO<sub>3</sub> with sustained vibration at 85 °C for 2 h, then 3 ml of 30% H<sub>2</sub>O<sub>2</sub> (pH 2 by HNO<sub>3</sub>) was added with sustained vibration at 85 °C for 3 h. After cooling down, 5 ml of 3.2 M NH<sub>4</sub>OAc in 20% (v/v) HNO<sub>3</sub> was added with vibration at room temperature for 0.5 h for the organic matter fraction. As in the residual fraction was obtained as the difference between total As concentration and the sum of these four extracted fractions.

After each extraction, the residue was centrifuged at 4000 rpm for 10 min, and supernatants were decanted into volumetric Nalgene flasks. Sediment residues were afterwards washed with Milli-Q water, centrifuged again, and the supernatants were combined. Extractants were treated with the same way as for total As, and then the As was determined by HG-AFS.



**Fig. 1.** Location of the stations in the East China Sea. ● indicates surface sediments, ○ indicates sediment cores. The currents are pointed out by arrows. CDW: the Changjiang Dilute Water; KC: the Kuroshio Current; JCC: the Jiangsu Coastal Current; ZFCC: the Zhejiang-Fujian Coastal Current; TW: Taiwan Warm Current.

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