

Selenium and tellurium fractionation, enrichment, sources and chronological reconstruction in the East China Sea



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ABSTRACT

Surface and core sediments of the East China Sea (ECS) were collected to study spatio-temporal distributions and chemical fractions of Se and Te and to evaluate their enrichment and sources. Higher Se and Te concentrations in surface sediments appeared in the inner shelf and near the Cheju Island. It seemed to be controlled by sources and sediment characteristics. Enrichment factors (EFs) showed that Se and Te were at minor and moderate enrichment, respectively. Sequential extraction suggested that non-residual fractions of Se and Te accounted for high percentages ($29.5 \pm 16.2\%$ and $50.9 \pm 13.2\%$) in total, combined with risk assessment code (RCA), indicating that Se and Te were at medium and high risks, respectively. All temporal profiles of abundances, EFs and burial fluxes (BFs) of Se and Te displayed higher values before 1900, in 1989 and 2009, and in the period of 1960–1980 with a peak in 1970. These higher values were closely associated with biological and anthropogenic activities.

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1. Introduction

Coastal waters and estuaries are often surrounded by urban and industrial areas and are frequently loaded to a significant level with various pollutants. The ECS adjacent to the metropolis Shanghai and Ningbo Cities, receives massive agricultural, municipal, residential and industrial waste products, containing lots of nutrients, heavy metals and trace elements (e.g., Se and Te). A good deal of research on nutrients and heavy metals in the ECS has been studied (Lin et al., 2002; Wang et al., 2003; Yuan et al., 2004; Fang et al., 2009), whereas rather less attention has been focused on trace elements, such as Se and Te, which are particularly sensitive to the surrounding environment because of their very low abundances.

Selenium is an essential element to organisms, but it can cause anomalies in organisms at low concentration and it is toxic at high concentration. Of the two oxidized forms, selenium (Se(IV)) is generally the more toxic form and more easily bioaccumulated (Maier and Knight, 1993). Tellurium in the same group as Se in the periodic table, is a relatively rare element without significant biological role. However, Te and its compounds are considered to be toxic and need to be handled with care (Řezanka and Sigler, 2008). Organotellurium compounds can

damage cells, e.g., by oxidizing sulfhydryl groups and depleting endogenous reduced glutathione in a variety of tissues. Thus, due to their bioavailability, Se and Te can induce environmental problems.

Natural biogeochemical cycles of Se and Te have been changed by anthropogenic activities, which transferred Se and Te from continents to seas via rivers and atmosphere. After entering into marine environments, Se and Te were adsorbed on suspended particulates or utilized by organisms and then deposited eventually on sediments, leading to Se and Te enrichment in sediments and making sediments being an important deposit of Se and Te in aquatic environment (Szefer et al., 1995; Duan et al., 2010). Sediments could be a secondary source of pollutants, once environmental condition was changed. When sedimentary environmental condition changed, Se and Te associated with sediments would be released to overlying water, threatening the aquatic biota. Therefore, it was necessary to assess enrichments and potential environmental risk levels of sedimentary Se and Te. Different methods, such as enrichment factor and geoaccumulation index have been widely applied to evaluate enrichment of trace elements and apportion their natural vs. anthropogenic contributions (Silva et al., 2009; Chandía and Salamanca, 2012; Hasan et al., 2013; Zahra et al., 2014). Moreover, depending on sediment conditions, Se and Te were mainly associated with carbonates, Fe–Mn oxides, sulfur and various organic compounds, either complexed or built into molecular and high-molecular. All these species could be

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presented in sediments concomitantly, which made the geochemical behaviors of Se and Te in environmental compartments quite complex (Wright et al., 2003). Hence, estimating the potential bioavailability and risks associated with Se and Te in sediments required a detailed understanding of their biogeochemical behaviors and chemical speciation. Sequential extraction has been generally used to determine chemical speciation of Se and Te (Tessier et al., 1979; Ponce de Leon et al., 2003; Wright et al., 2003; Tolu et al., 2011; Savonina et al., 2012). Chemical speciation normally included exchangeable, carbonate, reducible, oxidizable and residual fractions. In the absence of anthropogenic influence, trace metals in sediments were mainly associated with silicates and primary minerals, and therefore had limited mobility. Trace elements introduced by human activities were mainly associated with carbonates, Fe–Mn oxides and organic matters, showing high mobility (Passos et al., 2010). When environmental conditions (e.g., salinity, pH, temperature and redox state) changed, Se and Te existing as labile fractions would be migrated, released and used by organisms (Atkinson et al., 2007; Duan et al., 2010).

In recent years, eutrophication become heavier and red tides occur frequently in the ECS due to a large amount of nutrients is discharged into the ECS (Liu et al., 2013), which might introduce more biological Se and Te to sediments. Thus, in order to reflect the traces left by anthropogenic and biological activities in marine environment, it is essential to reconstruct environmental evolution over the last several decades (Legesse et al., 2002). Due to their stability and non-biodegradation, Se and Te could accumulate and left fingerprints in sediments over time. Therefore, temporal variations of Se and Te in sediments could be used as effective indicators to essentially reflect their historical inputs by natural, anthropogenic and biological activities.

Due to their species having diverse properties, Se and Te are of environmental and ecotoxicological concern. To understand and evaluate biogeochemical behaviors and environmental impacts of Se and Te, the distributions, fractionations, geochemical enrichments and historical evolutions of Se and Te in sediments of the ECS were studied in this paper. The objectives of this study were to (1) determine Se and Te distributions and their influencing factors in sediments of the ECS; (2) analyze chemical fractions of Se and Te in surface sediments of the ECS; (3) evaluate geochemical enrichments and risks of Se and Te in the ECS; and (4) trace the historical evolutions of Se and Te in the ECS.

2. Materials and methods

2.1. Study area and sampling

The ECS is one of the larger marginal seas in the western Pacific Ocean of the northern hemisphere and surrounded by 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). The ECS receives a large amount of terrigenous sediment from the Changjiang River, which is the world's fourth largest river based on suspended load. The average freshwater and sediment fluxes of the Changjiang River are approximately $9.25 \times 10^{11} \text{ m}^3/\text{yr}$ and $4.61 \times 10^8 \text{ t/yr}$, respectively (Hori et al., 2001; Zhang and Liu, 2002).

According to the latitude of 29–32° N and longitude of 122–126° E, 38 surface sediments and a sediment core from the ECS were sampled in May 2009 with the cruise of “Kexue 1” (Fig. 1). Surface sediments (0–2 cm) were collected using an Ekman–Birge box sampler. Immediately after collection, samples were placed in pre-cleaned polyethylene bags, sealed and refrigerated until lab analysis.

Sediment core G1 (122°31.05'E, 29°30.25'N) was collected at the extension of the Changjiang River using a gravity corer (Fig. 1). Core G1 is located in the inner shelf along the northern Zhejiang and Fujian Provinces with water depth of 32 m. Immediately after collection, core G1 was sectioned at 2 cm intervals and stored in pre-cleaned polyethylene bags, sealed and refrigerated until lab analysis.

Both of surface and core sediment samples were dried in an oven at 60 °C for 72 h. Dried aliquots were ground using an agate mortar and pestle for homogenization, and prepared for analysis. Data reported in this study were calculated as dry weight.

2.2. Sediment analyses

The analytical technique for Se and Te was performed by modifying a method of Mercone et al. (1999). Briefly, about 0.2 g dry sample was digested with a mixture of 10 ml aqua regia in Teflon digestion vessel heated in water bath for 1 h. After cooling, solution was pre-reduced by boiling at an acidity of 5 M HCl for 30 min. And then the reduced solution was determined by atomic fluorescence spectrometry coupled with a hydride generator (HG-AFS) using 0.7% KBH_4 with 0.05 M NaOH under Se and Te hollow cathode lamps for Se and Te determination, respectively.

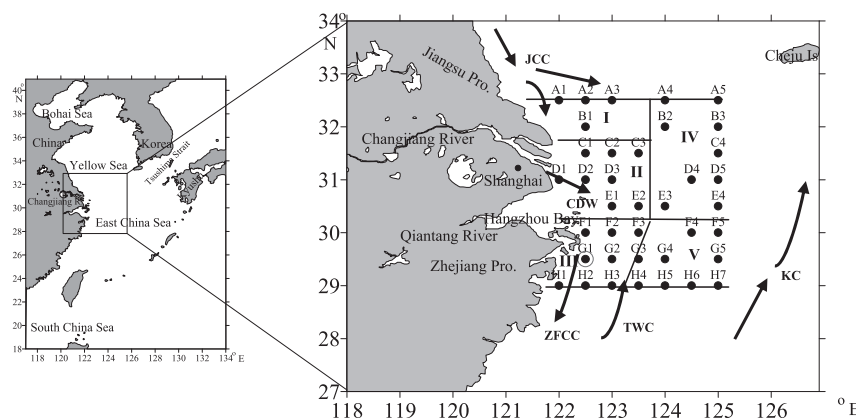


Fig. 1. Location of the stations in the East China Sea. ● indicates the surface sediments, ○ indicates core G1. The currents were pointed out by arrows. CDW: the Changjiang Dilute Water; KC: the Kuroshio Current; JCC: the Jiangsu Coastal Current; ZFCC: the Zhejiang-Fujian Coastal Current; TW: Taiwan Warm Current.

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