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Distribution and pollution assessment of heavy metals in surface sediments in the Yellow Sea

Xin Jiang [a,b,c], Ankang Teng [d], Wenzhe Xu [d], Xiaoshou Liu [a,*]

[a] College of Marine Life Sciences, Ocean University of China, Qingdao 266003, PR China
[b] Key Laboratory of Tropical Marine Bio-resources and Ecology, South China Sea Institute of Oceanology, Chinese Academy of Sciences, Guangzhou 510301, PR China
[c] University of Chinese Academy of Sciences, Beijing 100049, PR China
[d] Department of Biology and Chemistry, City University of Hong Kong, Hong Kong, China

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ABSTRACT

Heavy metal concentrations in surface sediments at 56 stations during two cruises in the Yellow Sea in summer and winter, 2011 were analyzed by inductively coupled plasma-mass spectrometry. The pollution status was assessed via the Geoaccumulation index and Hankanson potential ecological risk index. Higher concentrations of heavy metals (except for Mn) were found in the central Southern Yellow Sea and the western Northern Yellow Sea. The higher contents of Mn were much closer to Shandong Peninsula. Correlation analyses indicated that Pb, Cu, Fe, Ni, Zn and Co probably had the same origin and were controlled by grain size and total organic carbon. Pollution assessment showed that most areas of the Yellow Sea were not or lowly contaminated with the exception of the northwest and south parts of the Southern Yellow Sea showing Cd-contamination. The pollution status of the Yellow Sea in summer was worse than that in winter.

Heavy metal pollution in the aquatic environment is one of the critical issues due to the toxic and persistent characters (Zhan et al., 2010; Varol and Şen, 2012; Gao and Chen, 2012). Heavy metal pollutants pose potential threats to ecosystems because they could be concentrated or accumulated in organisms and biomagnified at higher trophic levels (Zhan et al., 2010; Ghrefat et al., 2011; Gao and Chen, 2012), and partly converted to more toxic organic compounds (Liu et al., 2009). Heavy metals in seas originate from both natural processes and anthropogenic activities. Natural processes like atmospheric inputs and aeolian processes set the background values for heavy metals (Zhang et al., 2003). With the rapid industrialization and urbanization in coastal regions, anthropogenic inputs are the main sources of pollution in the marine environment, and heavy metals are increasingly introduced to the estuarine and coastal environments through riverine discharge and oceanic dumping (Choi et al., 2007; Zhang et al., 2012; Yuan et al., 2012). After being introduced into the aquatic environment, heavy metals from the aqueous phase eventually become deposited to sediment through physical, chemical or biological mechanisms (Zhang et al., 2007; Zhan et al., 2010; Yuan et al., 2012).

The Yellow Sea is a semi-enclosed epicontinental sea of the northwestern Pacific, separated from the Bohai Sea in the east. It is located between the China Mainland and the Korea Peninsula, between 31°40′N–39°50′N and 119°10′E–126°50′E. The Yellow Sea covers 380,000 km^2, with an average depth of 44 m and a maximum depth of 140 m in the northern area of Cheju Island. It is divided into the Northern Yellow Sea (NYS) and the Southern Yellow Sea (SYS) by a line linking Chengshanjiao on the Shandong Peninsula and Changsan-got on the Korea Peninsula (Yang et al., 2003). The Yellow Sea receives billions of tons of particulate materials annually from both Chinese and Korean Rivers, such as the Changjiang River, Yellow River, Yalujiang River of China and the Han, Kum and Yeongsan Rivers of Korea (Qin et al., 1989). The statistics from the State Oceanic Administration, PR China (SOA, 2010, 2011, 2012, 2013) indicate that 6530, 9540, and 16,530 km^2 of offshore areas of the Yellow Sea are heavily polluted (i.e. the water quality is worse than class 4) in the years of 2010, 2011, and 2012, respectively. The polluted areas of those years are significantly larger than in 2009 (2150 km^2).

The distribution of metals in sediments might provide evidence for human activities and their effects on ecosystems. For this reason are sediments commonly chosen as environmental indicators of the quality and potential risks within aquatic systems (Unlu et al., 2008; Luo et al., 2010; Zhan et al., 2010). The Geoaccumulation

* Corresponding author. Tel./fax: +86 532 82031735.
E-mail address: liuxs@ouc.edu.cn (X. Liu).

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index and Hankanson potential ecological risk index are the most popular methods used to evaluate the ecological risk posed by heavy metals in sediments (Varol and Şen, 2012; Li et al., 2012). Most of the existing studies, on the distribution of heavy metals in surface sediments, divided the Yellow Sea into the NYS and the SYS; however, few studies considered the Yellow Sea as a whole. Concentrations of Ni and Co and their seasonal variability are rarely studied neither. The main objectives of the present study are to determine the distribution of eight heavy metals (Pb, Cd, Cu, Fe, Ni, Mn, Zn and Co) concentrations in surface sediments of the Yellow Sea, and to assess the pollution status of this area with the Geoaccumulation index and Hankanson potential ecological risk index.

The study area is located between 31°58′N–38°45′N and 121°00′E–124°40′E, near the coast of China. Sediments were collected onboard the vessel "*Dongfanghong II*" from 34 sampling stations (23 in the SYS and 11 in the NYS, Fig. 1a) during 13 to 30 June, 2011, and 22 sampling stations (16 in the SYS and 6 in the NYS, Fig. 1b) from the 20th November to 7th December, 2011. Surface sediments were taken using a stainless steel box-corer. The top 3 cm of samples were then placed in polyethylene bags, and refrigerated at −20 °C. After transport to the laboratory, samples were air-dried at room temperature (20 °C), crushed, passed through a nylon sieve of 160 meshes (96 μm), homogenized, and then stored in polyethylene bags for further analysis.

About 0.5 g dried sediment samples and 10 mL concentrated HNO_3 were placed together in a plastic digestion tube and a microwave digestion system was used to remove any organic matters present in the sediment. The digested samples were then diluted with water to 50 mL. Sample solutions and blanks were analyzed for Pb, Cd, Cu, Fe, Ni, Mn, Zn and Co with inductively coupled plasma-mass spectrometry (ICP-MS, Optima 2100 DV ICP System, Perkin Elmer). All samples were analyzed in duplicate, and the data shown herein represent average values of the duplicates. In order to evaluate the precision, the GBW-07314 reference material (The Second Institute of Oceanography, the State Oceanic Administration of China) was measured. All plastic and glassware were pre-cleaned by soaking in HNO_3 (v/v = 1:3) for at least 24 h, followed by soaking and rinsing with de-ionized water.

Surfer 8.0 (Golden Software Inc., USA) was used for drawing the distribution maps of metal concentrations in surface sediments. Statistical analyses (i.e. two-tailed t test, bivariate correlation, one-way ANOVA) were conducted with SPSS 19.0 software (SPSS Inc., USA).

In this study, two methods of pollution assessment of heavy metals are conducted, the geoaccumulation index (Müller, 1969) and Hankanson potential ecological risk index (Hakanson, 1980). The geochemical background values of Cu, Pb, Zn and Cd were 15.92, 14.54, 60.00 and 0.103 μg/g, respectively (Lu and Zhu, 1987). However, the background values of other metals studied here were not available.

The geoaccumulation index (I_{geo}) is defined by the following equation (Eq. (1)):

$$I_{geo} = \log_2(C_n/kB_n) \quad (1)$$

where C_n is the concentration of metals examined in sediment samples and B_n is the geochemical background value of metals. Factor k is the background matrix correction factor due to lithospheric effects, which is usually defined as 1.5 (Müller, 1969; Rubio et al., 2000; Ghrefat et al., 2011; Shafie et al., 2013). The geoaccumulation index consists of seven classes:

Class 0 (practically uncontaminated): $I_{geo} \leqslant 0$.
Class 1 (uncontaminated to moderately contaminated): $0 < I_{geo} \leqslant 1$.
Class 2 (moderately contaminated): $1 < I_{geo} \leqslant 2$.
Class 3 (moderately to heavily contaminated): $2 < I_{geo} \leqslant 3$.
Class 4 (heavily contaminated): $3 < I_{geo} \leqslant 4$.
Class 5 (heavily to extremely contaminated): $4 < I_{geo} \leqslant 5$.
Class 6 (extremely contaminated): $I_{geo} > 5$.

The Hankanson potential ecological risk index (RI) is defined as follows (Eqs. (2)–(5)):

$$C_f^i = \frac{C_i}{C_n^i} \quad (2)$$

$$C_d = \sum_i^m C_f^i \quad (3)$$

$$E_r^i = T_r^i \cdot C_f^i \quad (4)$$

$$RI = \sum_i^m E_r^i \quad (5)$$

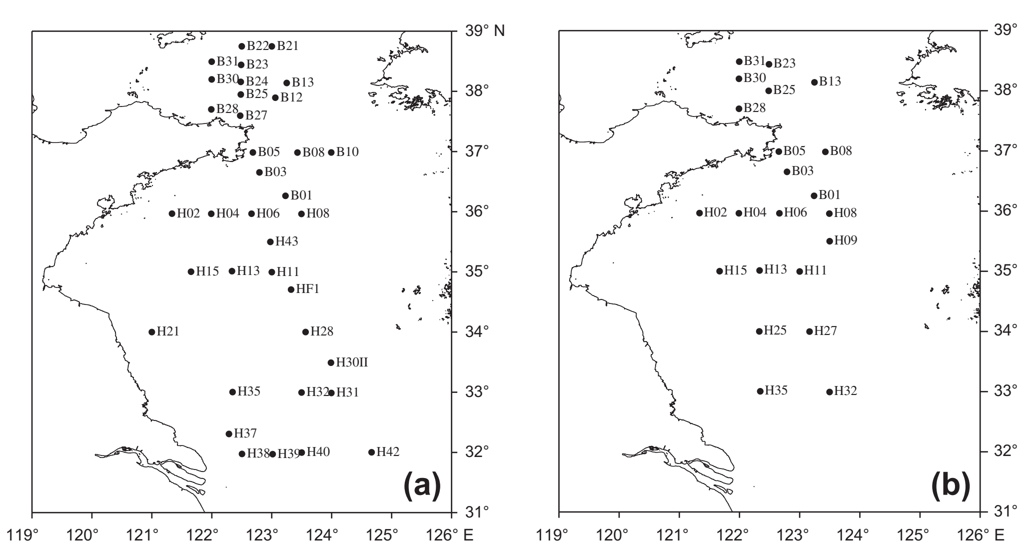

Fig. 1. Map of the Yellow Sea, showing the sampling stations during summer (a) and winter (b) in 2011.

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