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Assessment of heavy metal contamination in the surface sediments: A reexamination into the offshore environment in China

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ABSTRACT

The contents of Cu, Pb, Zn, Cd, Cr, Hg and As in the surface sediments of over 668 sites were monitored in a comprehensive program for assessing the degree of heavy-metal pollution and adverse biological effects of the offshore sediments in China. The contamination factor and geoaccumulation index indicated that As and Pb might be two of the most influential pollution loading in these metals. Cluster analysis separated 19 areas in China's near seas into 7 groups with different pollution characteristic, where the sediments along Zhejiang coast were the most highly-contaminated. Based on biological adverse effects index and sediment quality guideline for As, nearly half of sites in China's near seas induced slight adverse biological effects. With correlation analysis, self-organizing map and factor analysis, different sources as well as various adsorption mechanisms/anthropogenic factors were suggested to be important roles in altering the concentration of heavy metals in the sediments.

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1. Introduction

Since the year 2000, China has experienced serious environmental problems along with its industrial expansion (Fig. S1), and brought with it a serious environmental dilemma, such as pollution by persistent organic compounds and heavy metals. The contamination of heavy metals throughout China has caused several eco-related and health-related concerns (Khan et al., 2008; Pan et al., 2015; Rodríguez-Lado et al., 2013; Wei and Yang, 2010; Wong et al., 2002). One of these problems is heavy metal pollution in marine environments. Some heavy metals may have a pronounced adverse effect on marine organisms. For instance, Cd has been included on the endocrine disruptor list and has a toxic effect on hepatic tissue as well as an apoptotic effect on the testis of marine teleost *Gobius niger* (Migliarini et al., 2005). Hg is capable of causing significant hemocyte mortality in the Pacific oyster *C. gigas*, and even a slight excess in Cu concentrations can affect phagocytic activity and bacterial clearance in the blue mussel *M. edulis* (Renault, 2015).

In addition to industrial wastewater and industrial coal combustion, other anthropogenic sources also release vast amounts of heavy metals into the marine environment each year, such as nonpoint sources and waste incineration. Along with these processes, a considerable proportion of heavy metals is deposited into the offshore sediments. The contamination level of sediments is cumulative over time. Therefore, it is suitable to judge the effects of long-term pollution by investigating pollution in sediments. Meanwhile, metals in the marine environment are associated with different sediment components in various adsorption mechanisms including precipitation, ion exchange, complexation and partition. Changes in environmental conditions (pH, ionic strength, redox potential or biological activities) may cause the mobilization of adsorbed metal/metalloid into the liquid phase and the subsequent contamination of seawater. Therefore, the quality of seawater may be directly affected by trace element diffusion from surface sediments.

Most existing reports investigating heavy metal pollution in marine sediments have focused on the survey of highly polluted and semi-closed bays or estuaries either in China (Gao and Chen, 2012; Li et al., 2015b; Yu et al., 2010) or other regions (Alyazichi et al., 2015; Batayneh et al., 2015; Pejman et al., 2015). However, these studies were carried out based on different estuarine environment and various local sources, and little attention was paid to common features among heavy metal pollution sources and mechanisms. Meanwhile, studies based on the data collected from the open sea were limited. As for China's near seas, Meng et al. has investigated the concentrations of Hg in the surface sediments in the Bohai Gulf, Yellow Sea, East China Sea and Pearl River Estuary (Meng et al., 2014). Studies concerning the metals Cu, Pb and Zn in the sediments in the East China Sea (Fang et al., 2009) and Cu, Hg, Pb, Zn, Cd and As in the Yellow Sea (He et al., 2009) were also performed to uncover any common patterns of heavy metal distribution in the open sea. Based on the Chinese national dataset, the present study addresses sedimentary quality assessment using both background levels derived from time-scale/geochemical background concentration and screened sediment quality guidelines (SQGs) developed by several institutions (including the National Oceanographic and Atmospheric Administration in the United States) to

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estimate the degree of pollution and possible biological adverse effects of sedimentary heavy metal contaminations in China's near seas.

2. Materials and methods

2.1. Studying areas and monitoring sites

China's eastern coastal region, which consists of nine provinces and two municipalities directly under the central government, only occupies approximately 10% of the land area in China, but contained 42% of the population, and generated 58% of the gross domestic product and 62% of the industrial value added in 2007 (Li et al., 2008). Among all of these provinces/municipalities, the industrial value added accounted for over half of the local gross domestic product in the north, including Shandong, Tianjin and Jiangsu, whereas in southern provinces, industrial manufacturing is not as dominant. The industrial value only accounts for approximately 20% of the gross domestic product in Hainan (Table S1). Coastal sewage outlets may be important sources of heavy metals.

Traditionally, China's near seas were divided into the Bohai Gulf and three marginal seas, namely, the Yellow Sea (including the West Korean Sea), East China Sea (ECS) and South China Sea (SCS). The Yangtze River, Pearl River and Yellow River are the three major rivers flowing into the ECS, SCS and Bohai Gulf, respectively, and the discharges of the three rivers were up to 850, 268 and 11 km^3/year (Chu et al., 2015; Wua et al., 2012), respectively. The annual streamflows of several main rivers in China were almost unchanged between 1956 and 2005, except for that of the Yellow River (Zhang et al., 2011). The plume of the Yangtze River is a natural boundary between the ECS and the Yellow Sea; it inputs hundreds of millions tons of sediment into China's near seas, carrying heavy metals from sources along the river. Sediments from these major rivers can be dispersed up to 300 km across the shelf (Ridgway and Shimmield, 2002).

Aside from sewage outlets and rivers, nonpoint sources, such as rain-wash, were also responsible for the input of metals and were difficult to measure. Precipitation played a dominant role in in transferring originating from nonpoint sources into the sea. China has a predominantly subtropical monsoon climate in the south and a monsoon climate typical of the mid-latitudes in the north. Therefore, the precipitation along coastal areas between the southern and northern regions of China experienced a nearly stepwise attenuation. Based on the dataset provided by the National Climate Center of China (SURF_CLI_CHN_PRE_MON_GRID_0.5), the absolute maximum annual precipitation in 2007 occurred in the Guangdong coastal region (up to 2422.8 mm), whereas the minimum value occurred at the Liaoning coast (474.5 mm). The 1500 and 1000 mm contour lines of precipitation were in Shanghai and Shandong, respectively.

The surface sediments in China's near seas were studied in October 2007 during 19 expeditions carried out for a comprehensive marine pollution survey and assessment program, organized by the State Oceanic Administration of China (SOA). The scope of investigation for this program covered the waters between latitudes 17.41°N and 40.75°N and longitudes between 107.63°E and 127.00°E. Thousands of available data were obtained from 668 sites (Fig. S3). The farthest site was approximately 470 km from the coastline. This program conducted the only comprehensive observation of marine pollution to cover nearly the entire extent of China's near seas over the past decade. Based on this program, several average values and ranges for metals in the Bohai Gulf/Yellow Sea/ECS/SCS have been calculated and published in our previous report without statistical analysis, assessment or thorough discussion (Ji, 2011).

2.2. Sampling and chemical analysis

The sampling and chemical analysis procedures were conducted following the same standard methods. Sediment samples were collected at each station by mixing three to four random samples from the top 5 cm of the surface sediment using a grab sampler. The 500 g–600 g sediment samples were sealed in clean plastic bags for the determination of heavy metals, and samples of roughly equal weights were sealed in clean wide-mouthed glass jars for the determination of total sulfides (TS), total organic carbon (TOC), total nitrogen (TN) and total phosphorus (TP). The sediment samples collected were preserved at 277 K before analysis. Oxidation-reduction potentials (E_h) were measured in-situ using an oxidation reduction potentiometer. For heavy metals analysis, samples were digested with concentrated HNO_3 and $HClO_4$ in a Teflon beaker following the standard procedures (Li et al., 2015a; SOA, 2007). Cu/Zn was analyzed by flame atomic absorption spectrometry equipped with deuterium background correction, with As/Hg by atomic fluorescence spectrometry and Cd/Cr/Pb by graphite furnace atomic absorption spectrometry. TN was determined by titration for total Kjeldahl nitrogen in sediments (SOA, 2007), similar to EPA method 351.3 (USEPA, 1978). TP was determined by a method developed from the vanadomolybdophosphoric acid colorimetric method (Pacey, 1999; SOA, 2007). TS were determined by the iodometric method (SOA, 2007), modified from EPA methods 9030b and 9034 (USEPA, 1996a, 1996b). TOC was estimated using a modified Walkley-Black method (SOA, 2007; Walkley A, 1934). E_h was measured using a Pt electrode; therefore, the measured values can only be considered as relative redox characteristics (Vershinin and Rozanov, 1983). Total petroleum hydrocarbon (oil) was extracted using n-hexane and detected by UV fluorescence spectroscopy (UNEP, 1992).

The accuracy of the analytical procedure was <10%, which was checked by analyzing the following Standard Reference Materials (SRMs): the offshore marine sediment standards of China (GBW07314), marine sediment standards of the Yellow Sea (GBW07333) and marine sediment standards of the South China Sea (GBW07334). Blanks were included in each batch of samples to prevent artificial contamination of the samples.

2.3. Statistical analysis

The correlations among the chemical parameters were analyzed by Spearman's rank correlation coefficients, as the data for some of the parameters did not follow a normal distribution.

Hierarchical agglomerative cluster analysis (CA) was performed on the normalized dataset of heavy metals, TS, TN, TP, E_h and TOC using Ward's method with squared Euclidean distances as a measure of similarity. Since CA analysis based on the average values was apparently affected by the sites with high loads of pollution, analysis based upon median values was also carried out.

The source apportionment process was performed using R-mode factor analysis (FA) after the Kaiser-Meyer-Olkin test (KMO = 0.639) and Bartlett's test of sphericity (significance = 0.000). Varimax rotation with the Kaiser Normalization scheme was used to extract factors to explore the potential relationship between the variables (concentrations of heavy metals, TOC, TN, TP and TS) in offshore marine surface sediments. An Eigen value above 1 represented a significant contribution by the corresponding factor.

As a neural network model projecting a high-dimensional vector onto a low-dimensional array of neurons in an orderly and nonlinearly fashion (Kohonen et al., 1996), the self-organizing map (SOM) algorithm can detect nonlinear relationships intuitively (Dollhopf et al., 2001). This gives the SOM an important advantage over other methods, such as CA, principle component analysis and FA, which are based upon the assumption of a linear relationship among variables (Dollhopf et al., 2001). Here, a two-layer SOM model could form: an input layer that has the same number of neurons as input variables (e.g., trace metals) and an output layer formed by neurons organized on a regular low-dimensional grid. Input neurons were assigned to every output neuron. The training used competitive learning. This competition occurred repeatedly over a number of iterations, resulting in clusters of winning

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