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REPORT

Ecological risk assessment of arsenic and metals in sediments of coastal areas of northern Bohai and Yellow Seas, China

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Abstract Distributions of arsenic and metals in surface sediments collected from the coastal and estuarine areas of the northern Bohai and Yellow Seas, China, were investigated. An ecological risk assessment of arsenic and metals in the sediments was evaluated by three approaches: the Sediment Quality Guidelines (SQGs) of the United States Environmental Protection Agency (USEPA), the degree of contamination, and two sets of SQGs indices. Sediments from the estuaries of the Wuli and Yalu Rivers contained some of the greatest concentrations of arsenic, cadmium, copper, mercury, lead, and zinc. Median concentrations of cadmium and mean concentrations of lead and zinc were greater than background concentrations determined for the areas. All sediments were considered to be heavily polluted by arsenic, but moderately polluted by chromium, lead, and cadmium. Current concentrations of arsenic and metals are unlikely to be acutely toxic, but chronic exposures would be expected to cause adverse effects on benthic invertebrates at 31.4% of the sites.

Keywords Arsenic and metals · Sediments · Contamination · Ecological risk assessment

INTRODUCTION

Contaminants that enter the marine environment and partition to particles are ultimately deposited in sediments that can be both a sink and a source of arsenic and metals (Förstner and Wittmann 1981). More than 90% of the arsenic and metals in aquatic systems can be accumulated in suspended particulate matter and sediment via several pathways: disposal of liquid effluents, terrestrial runoff, and leachate from numerous urban, industrial, and agricultural activities, as well as atmospheric deposition. Such accumulation can be greater in enclosed and semi-enclosed areas where exchange of water with the open sea is limited (Karageorgis et al. 2002).

Distributions of arsenic (As) and metals in sediments can provide evidence of human activities and their effects on ecosystems and aid in assessing the risks associated with discharged human waste. Accumulation of arsenic and metals in sediments has implications for local communities, as well as for qualities of river waters (Demirak et al. 2006; Zheng et al. 2008). Arsenic and metals are of ecological significance due to their toxicity, persistence, and bioaccumulation (Diagomanolin et al. 2004). The presence of arsenic and metals in sediments poses a potential threat to marine ecosystems, especially in coastal areas. Contamination of surface sediments with As and metals can affect the quality of overlying seawater, and affect the organisms that make up the base of the food chain and ultimately to human health (Christophoridis et al. 2009).

The northern portions of the Bohai and Yellow Seas, which are semi-enclosed, are bordered by northeast China and the Korean Peninsula. The region of the North Bohai and Yellow Seas are in an area of intense human activity and accounts for approximately 20% of the population and 25% of the gross domestic productivity (GDP) of China. Large amounts of industrial and municipal effluents containing As and metals have entered the offshore environment of the Bohai and Yellow Seas, and have caused deterioration of the coastal environment (Feng et al. 2003; Chen et al. 2004; Gao et al. 2008; Zheng et al. 2008; Xu et al. 2009). The ecosystem has experienced dramatic reductions in species diversity and abundance, fish farming, uncontrollable diseases in marine-cultured species and habitat loss. However, the distributions of As and metals have never been investigated comprehensively. Therefore, it has been difficult to determine the ecological risks, such that sources could be established and remedial action plans developed.

In this study, distributions of As and metals in the surface sediments of potential coastal and estuarine sources to the northern Bohai and Yellow Seas, China were investigated and visualized using Geographic Information Systems (GIS). In order to assess ecological risks for As and metals in sediments, three approaches were employed. The first was comparison of concentrations of As and metals to Sediment Quality Guidelines (SQGs) promulgated by United States Environmental Protection Agency (USEPA) (Giesy and Hoke 1990). This approach was used only to screen contaminants of concern in aquatic ecosystems. The second approach, developed by Håkanson (1980), introduces contamination factors and degree of contamination concepts as diagnostic tools for pollution control purposes. The third approach was to apply two sets of SQGs developed for marine and estuarine ecosystems (Long and MacDonald 1998) to assess the ecotoxicological sense of As and metal concentrations in sediments (a) the effect range low (ERL)/effect range median (ERM) and (b) the threshold effect level (TEL)/probable effect level (PEL) values.

The objectives of this study were to: (i) to examine the spatial distributions of concentrations of As and metals in surface sediments collected from coastal areas of the northern parts of the Bohai and Yellow Seas, China, using GIS, (ii) to evaluate anthropogenic contamination with As and metals, (iii) to assess the association between the concentrations of As and metals in sediments and potential for adverse effects on aquatic organisms based on the SQGs used in USA, Canada, Turkey, Portugal, Greece, and South Korea.

MATERIALS AND METHODS

Study Area

The northern Bohai and Yellow Seas are part of the Pacific Ocean and lie between the industrialized northeast coast of China, and the Korean Peninsula (Fig. 1). The coastal areas of the northern Bohai and Yellow Seas are composed of nine major cities (including Tangshan (TS) and Qinhuangdao (QH) cities in Hebei Province, and Huludao (HL), Jinzhou (JZ), Panjin (PJ), Yingkou (YK), Dalian (DL), and Dandong (DD) cities in Liaoning Province), where there are 7.1, 2.8, 2.96, 3.03, 1.27, 2.3, 6.03, and 2.4 million residents, respectively. The estuarine areas of the northern Bohai and Yellow Seas include ten main estuarine areas at the Dou, Qinglong, Luanhe, Liugu, Wuli, Daling, Liaohe, Daliaohe, Daqing, and Fuzhou Rivers flowing into the northern part of the Bohai Sea, and the six main estuaries at the Daqing, Fuzhou, Biliu, Yingna, and Yanghe, and Yalu Rivers flowing into the northern part of the Yellow Sea. As the main receiving coastal ecosystems, the

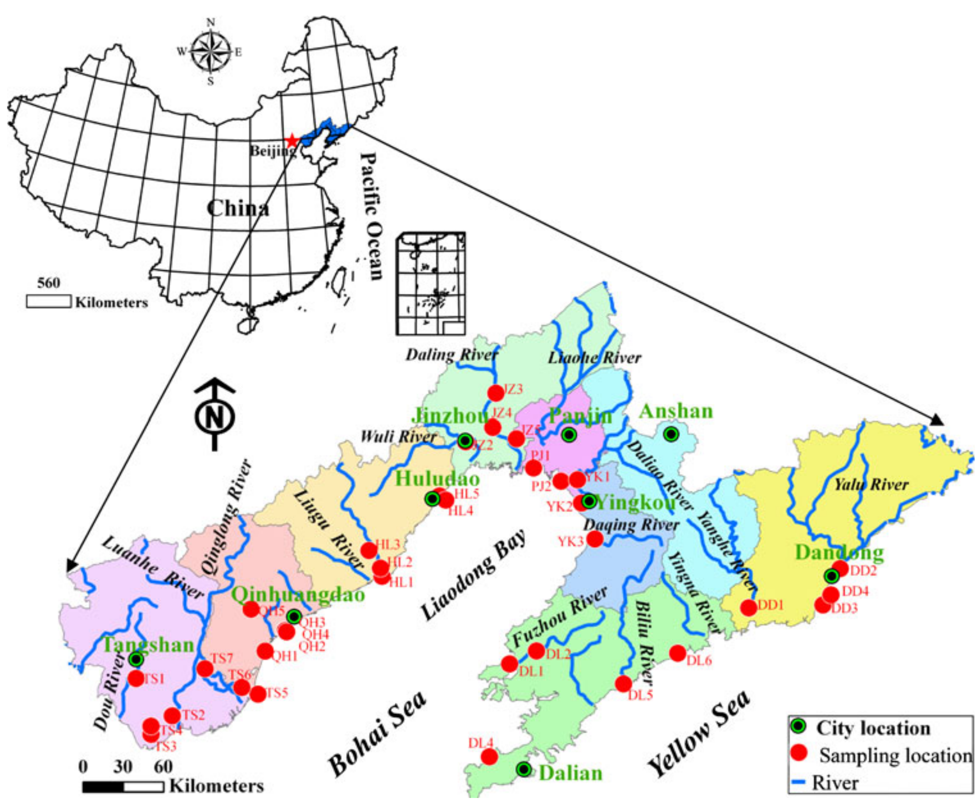

Fig. 1 Location map of surface sediments collected from the northern Bohai and Yellow Seas, China

water and sediment in the northern parts of the Bohai and Yellow Seas are influenced by industrial discharges and partially treated domestic effluents from coastal and estuarine areas, where many pollutant-generating enterprises (such as iron and steel, machine manufacture, shipbuilding, petrochemistry, metallurgy, spinning, medicine, and foodstuff) are located. For example, industries include the Jinxi Refinery and Huludao Zinc Plant (the largest zinc smelting plant in Asia), Bohai Shipyard, and Jinxi Chemical Complex (Chen et al. 2004; Zheng et al. 2008). The semi-enclosed nature of the northern parts of the Bohai and Yellow Seas restricts water exchange (the time of water renovation is more than 30 years), which leads to accumulation of pollutants in these seas.

Sampling of Sediments

Thirty-five surface sediment samples were collected from coastal and estuarine areas of Bohai and Yellow Seas in October, 2008. Sampling sites were selected to cover the coastal and estuarine areas of the northern Bohai and Yellow Seas, China. Sediment samples numbered as follows: TS1-7 in Tangshan, QH1-5 in Qinhuangdao, HL1-5 in Huludao, JZ1-5 in Jinzhou, PJ1-2 in Panjin, YK1-3 in Yingkou, and DL1-4 in Dalian were collected from the coastal area of northern Bohai Sea, while sediment samples numbered as follows: DL5-6 in Dalian and DD1-4 in Dandong were collected from the northern shore of the Yellow Sea (Fig. 1). Throughout the survey, a global positioning system (GPS) was used to locate and map all of the sampling sites. Surface sediments (top 10-cm layer) were collected using a trowel from the sedimentation basin of the bed close to the bank. Representative samples were prepared by mixing five subsamples from an area of about 5 m^2. Samples were placed in dark-colored polyethylene bags, refrigerated, and returned to the laboratory immediately. Samples were air-dried at 4°C, crushed, passed through a nylon sieve of 100 mesh, and then stored at 4°C in the dark before analysis of properties and concentrations of arsenic and metals.

Analysis of Arsenic and Metals

Sediments were digested with HNO_3 and H_2O_2 using USEPA Method 3051 (USEPA 1992) for quantification of As, cadmium (Cd), copper (Cu), chromium (Cr), lead (Pb), and zinc (Zn). The digestion for mercury (Hg) determination, which used a combination of H_2SO_4 and HNO_3, was based on USEPA Method 7471 A (USEPA 1992). Sample solutions and reagent blanks were analyzed for arsenic and metals using an inductively coupled plasma mass spectrometry (ICP-MS) (Agilent 7500). To monitor the quality of chemical analysis and examine the accuracy of the data, soil reference materials, GB ESS1 issued by State Oceanographic Administration of China, were analyzed at the same time as sediments. Average recoveries of As, Zn, Cr, Hg, Cu, Cd, and Pb in triplicate analysis were 98 ± 4.7, 93 ± 6.1, 97 ± 2.5, 101 ± 5.3, 95 ± 3.2, 94 ± 2 and 103 ± 4.0%, respectively. Analyses were conducted in duplicate. Analytical precision was <10% for all the elements.

Statistical analyses were conducted using SPSS 12.0 software (SPSS Inc., USA). ESRI ArcGIS 9.0 for Windows was used for the area digitization in order to display the spatial distribution of arsenic and metal concentrations in surface sediments and ecological risk indices.

RESULTS AND DISCUSSION

Arsenic and Metal Distribution in Surface Sediments

Concentrations of As and metals in surface sediments collected from coastal and estuarine areas on the northern parts of the Bohai and Yellow Seas are summarized (Table 1). These concentrations were compared to Marine SQGs (GB 18668-2002) issued by the China State Bureau of Quality and Technical Supervision (CSBTS) (Table 1). These are the primary sediment quality criteria that are applied to protect habitats for marine life including natural, rare, and endangered species as well as places for human recreation and sports (CSBTS 2002). However, the sediment criteria are limited in their utility since they do not take into account site-specific or ecosystem-specific information. By means of statistical methods and with the help of Pb^{210} age dating data, As and metal concentrations in the fine grains (<0.063 mm) from the dated sedimentary cores about 190 years ago were determined. These values were then used for background concentrations in the sediments of the Bohai Sea and adjacent estuaries (Li et al. 1994; Feng et al. 2003). Concentrations of As and all metals except Cd and Hg in the sediments followed normal distributions (K–S $p > 0.05$). Therefore, median concentrations of Cd and Hg were used as the indication of central tendency because there were a few extreme concentrations that could greatly influence the mean and distort what might be considered typical. Median concentration of Cd (0.1 mg kg^{-1}) and mean concentrations of Pb and Zn (26, 60 mg kg^{-1}, respectively) were greater than the background concentrations determined for the region (Table 1). Distributions of As and metals in the sediments collected from the northern Bohai and Yellow Seas, China are shown (Fig. 2). At only two sites (HL4 and TS5) were concentrations of Hg greater than its estimated background concentration (0.05 mg kg^{-1}). The greatest concentrations of

Table 1 Descriptive statistics for As and metals (mg kg^{-1}) in the sediments from the northern Bohai and Yellow Seas and SQG by USEPA

	As	Cd	Cr	Cu	Hg	Pb	Zn
Mean	8.5	0.15	47	13	0.028	25	60
Median	8.6	0.10	49	9.2	0.021	23	52
SD	1.9	0.14	22	10	0.028	9.2	40
Minimum	5.6	0.050	4.2	0.53	0.020	9.5	9.8
Maximum	13	0.83	94	35	0.18	49	1.7×10^2
Skewness	0.38	4.1	0.02	0.55	5.2	0.80	0.79
Kurtosis	−0.48	20	−0.73	−0.91	29	0.18	0.18
K–S *p*	0.82	0.030	0.85	0.33	0	0.27	0.78
Background concentrations[a]	10	0.069	60	19	0.050	11.5	57
Sediment primary standard criteria[b]	20	0.50	80	35	0.20	60	1.5×10^2
SQG[c]							
Non-polluted	<3	–	<25	<25	≥1.0	<40	<90
Moderately polluted	3–8	–	25–75	25–50	–	40–60	90–2.0×10^2
Heavily polluted	>8	>6	>75	>50	>1.0	>60	>2.0×10^2

[a] From Li et al. (1994) and Feng et al. (2003)

[b] CSBTS (2002)

[c] From Giesy and Hoke (1990)

Hg, Cd, Pb, and Zn were observed in sediments (HL4 and HL5) from the estuarine area of the Wuli River near Huludao city. Concentrations of As and metals in sediments from the estuary of this river exceeded background concentrations. The greatest As and Cu concentrations (13 and 35 mg kg^{-1}, respectively) occurred in the sediment (DD2) from the estuary of Yalu River. Concentrations of As and all the metals except Hg in the sediment of DD2 exceeded their respective background concentrations. The greatest concentration of Cr (94 mg kg^{-1}) existed in the sediment (TS2) from the estuarine area of the Qinglong River. Concentrations of arsenic and all the metals except Hg in sediments (DL1 and PJ2) from the estuaries of Fuzhou and Liaohe Rivers exceeded their estimated background concentrations. The other coastal and estuarine areas of the northern Bohai and Yellow Seas were also contaminated by arsenic and metals to some degree.

Assessment of Sediment Contamination According to SQGs by USEPA

Based on the SQGs proposed by USEPA (Giesy and Hoke 1990), sediments were categorized into three classes, non-polluted, moderately polluted and heavily polluted (Table 1) by comparing the mean or median concentrations of arsenic and metals in the sediments with the SQGs. Based on this comparison, sediments were classified as being heavily polluted by As and moderately polluted by Cr. However, there are significant spatial variations in the arsenic and metal concentrations (Fig. 2), of which some exceed the SQGs (Table 1). Nineteen sites (54.3% of the sites) distributed near the cities were heavily polluted by As and the remaining sites located at more upstream locations or far from the cities were moderately polluted. Concentrations of Cr in 74.3% of the sites were between 25 and 75 mg kg^{-1} (Fig. 2) which was classified as moderately polluted, while greater Cr concentrations, classified as heavily polluted (>75 mg kg^{-1}), were observed at three sites (DL1, DL6, and TS2). Concentrations of Zn at nine sites, primarily in the vicinity of Liaodong Bay and Dandong city, were classified as moderately polluted (90–200 mg kg^{-1}). Concentrations of Cu at five sites, primarily in downstream sections of the Qinglong, Liugu, Daliao, and Yalu Rivers, were classified as moderately polluted (25–50 mg kg^{-1}). Concentrations of Pb in three sites (HL4, HL5, and DD2), primarily in the area near Huludao and Dandong cities, were classified as moderately polluted (40–60 mg kg^{-1}). Neither Hg nor Cd occurred at concentrations that resulted in classifying sediments as polluted. The SQGs developed by the USEPA do not consider natural background or multiple metals. This is particularly important with As and metals, which may occur naturally at greater concentrations in some areas of the world. Therefore, the sediments were also classified relative to background concentrations.

Classification of Sediments Relative to Degree of Contamination

In order to overcome the limitations of the USEPA SQGs, concentrations in sediments were compared to background or reference values (Burton 2002) by calculating

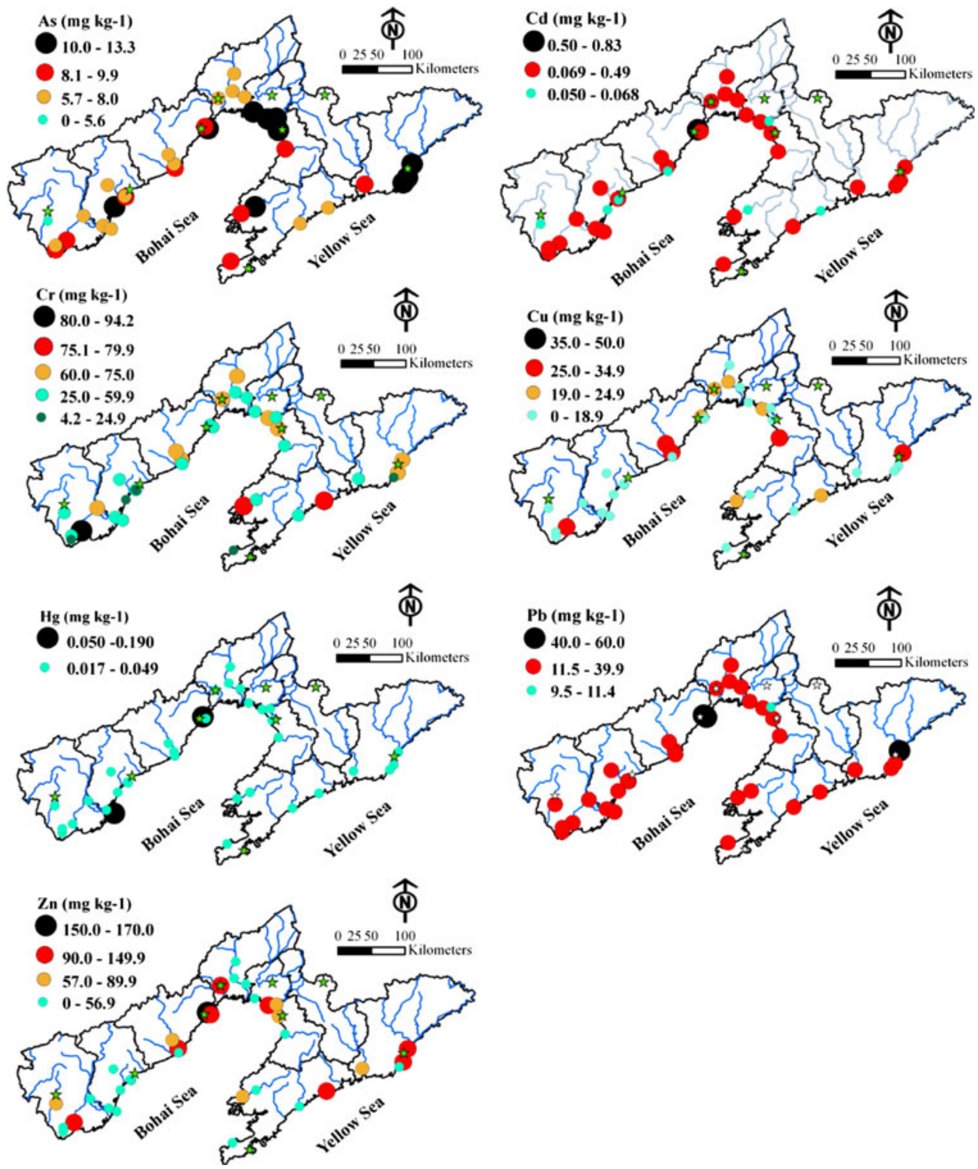

Fig. 2 Distributions of arsenic and metals in the sediments collected from the northern Bohai and Yellow Seas, China

contamination factors $\left(C_f^i\right)$ and degree of contamination (C_d) (Håkanson 1980). The contamination factor $\left(C_f^i\right)$ i.e., the sum of all contamination factors was calculated by use of Eq. (1).

$$C_f^i = \frac{\overline{C}_{0-1}^i}{C_n^i} \qquad (1)$$

where $\overline{C}_{0-1}^i$ is the mean content of the substance; C_n^i is the reference value for the arsenic and metal. The following terminologies are used to describe the contamination factor: $C_f^i < 1$ low contamination factor; $1 \le C_f^i < 3$ moderate contamination factors; $3 \le C_f^i < 6$ considerable contamination factors; $C_f^i \ge 6$ very high contamination factor. The degree of contamination (C_d), defined as the sum of all contamination factors for a given basin was calculated by use of Eq. (2).

$$C_d = \sum_{i=1}^{7} C_f^i = \sum_{i=1}^{7} \frac{\overline{C}_{0-1}^i}{C_n^i} \qquad (2)$$

Sediments were classified based on their degree of contamination, into the following groups: $C_d < 7$ low degree of contamination; $7 \le C_d < 14$ moderate degree of contamination; $14 \le C_d < 28$ considerable degree of contamination; $C_d \ge 28$ very high degree of contamination.

Contamination factors for sediments, which compared individual As and metals with their background concentrations, are given (Fig. 3). Moderate contamination of sediments with Pb was found at 82.9% of sites, while five sites (14.3% of the total sites), were classified as considerably contaminated. For Cd concentrations, approximately 6% of total sites were classified as being moderately

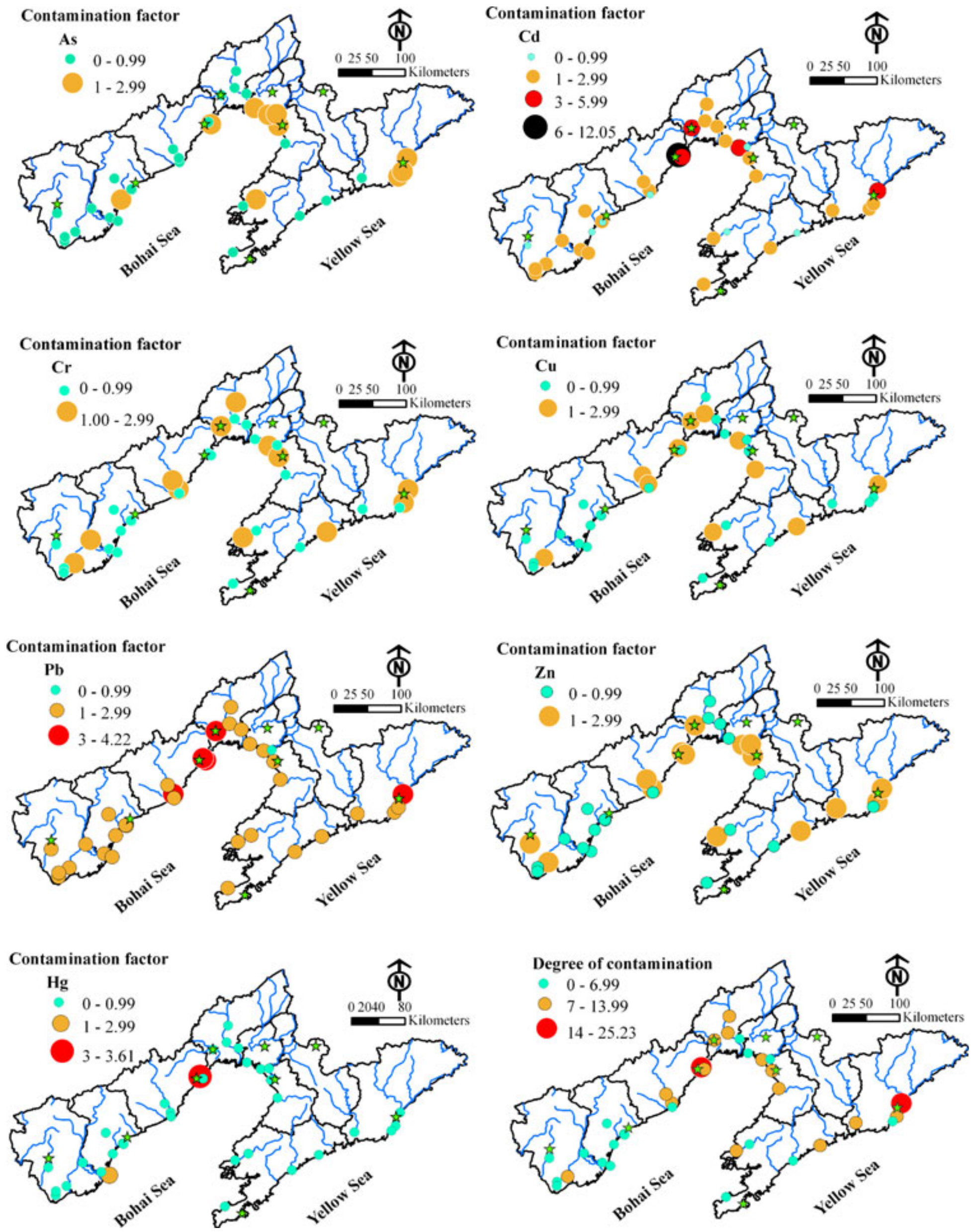

Fig. 3 Assessment of sediment contamination by the contamination factor and the degree of contamination

contaminated, three sites were classified as being considerably contaminated and one site was classified as being highly contaminated. Values for C_f^i were between low and moderate for Zn, Cu, Cr, and As at all sites. Sediments classified as moderately contaminated based on concentrations of Zn, Cr, Cu, and As accounted for 42.9, 34.3, 31.4, and 28.6% of the total sediments, respectively. The contamination factors for arsenic and individual metals were of the following order: Pb > Cd > Zn > Cr > Cu > As > Hg.

Contamination degree (C_d) values were calculated (Fig. 3). According to the Håkanson (1980) classification, C_d values at two sites, HL4 and DD2, indicate a considerable degree of contamination, which suggests pollution from human activities at these sites. Site HL4 is located on the Wuli River where sediment was reported to be heavily polluted by As and metals from a number of industries, especially the Huludao Zinc Smelting Plant (Zheng et al. 2008). Site DD2 is in the downstream section of the Yalu River near Dandong city which has light industries such as automobile manufacturing, paper making, chemical engineering, printing and dyeing, and spinning. Upstream areas of the Yalu River have some of the richest copper ore deposits in Asia, which have been mined since the beginning of the twentieth century. It has been reported that the annual discharges of Zn, Cu, and Pb from the Yalu River to the Yellow Sea in 1980s were 255.1, 119.5, and 45.2 t, respectively. According to the Bulletin of China Oceanic

Table 2 Classification of sediment samples based on the proposed SQGs

	SQGs (mg kg^{-1})[a]				Percentage of the samples exceeding SQGs (%)		
	TEL	PEL	ERL	ERM	ERL	ERL-ERM	>ERM
As	7.2	41.6	8.2	70	45.7	54.3	0.0
Cd	0.68	4.2	1.2	9.6	100	0.0	0.0
Cr	52.3	160.4	81	370	97.1	2.9	0.0
Cu	18.7	108.2	34	270	97.1	2.9	0.0
Hg	0.174	0.486	0.15	0.71	97.1	2.9	0.0
Pb	30.2	112.2	46.7	218	97.1	2.9	0.0
Zn	124	271	150	410	97.1	2.9	0.0

[a] Long et al. (1995)

Environmental Quality, the estuary of Yalu River is one of most heavily arsenic and metal contaminated coastal areas in China (Gao et al. 2008). Fifteen sediment samples (42.9% of total sediments), mostly located near the cities of Huludao, Jinzhou, Panjin, Yingkou and Dandong, were classified as moderately contaminated.

Ecotoxicological Significance of As and Metal Concentrations in Sediments

Concentrations of As and metals were evaluated in a screening-level ecological risk assessment, by comparing to numerical SQGs such as TELs and PELs, ERL and ERM (Long et al. 1995; Long and MacDonald 1998). Low-range values (i.e., ERLs or TELs) are concentrations less than which adverse effects upon sediment dwelling fauna would not be expected. In contrast, the ERMs and PELs represent chemical concentrations above which adverse effects are likely to occur (Long and MacDonald 1998). The incidence of toxicity was determined among samples in which none of the substances equaled or exceeded the ERL concentrations, in which one or increasing numbers of substances exceeded ERL concentrations, but none exceeded any ERM; and in which one or increasing numbers of substances exceeded ERM concentrations (Pekey et al. 2004; Zheng et al. 2008). The same approach was used to evaluate the predictive ability of the TELs/PELs (Long et al. 1998). The U.S. National Oceanic and Atmospheric Administration (NOAA) guidelines provide two values for each chemical, classifying the sediment either rarely (<ERL), occasionally (≥ERL and <ERM) or frequently (≥ERM) associated with adverse biological effects (Birch and Taylor 2006; Christophoridis et al. 2009).

The SQG values for As and metals and a classification of the samples based on these guidelines are shown (Table 2). The results of classifying sediments based on the SQG suggest that Cd in sediments would rarely be expected to cause adverse effects on biota. Only a small percentage of sediments would be classified as possibly presenting an occasional threat to organisms due to concentrations of Cr, Cu, Hg, Pb, or Zn. Fifty-four percent of sediments would be expected to occasionally be associated with the toxic effects on aquatic organisms due to As. Classifications of sediments based on SQGs are given (Fig. 4). Most sites in the lower reaches of the rivers or near the cities would be expected to occasionally be associated with adverse biological effects. No concentrations of As or metals exceeded either ERM or PEL values. However, concentrations of at least one metal in 11 sediment samples (31.4% of sediments) exceeded the TEL value. Thus, at concentrations of As or metals in sediments greater than the TEL, toxic effects from long-term exposure to As and metals would be predicted to occur.

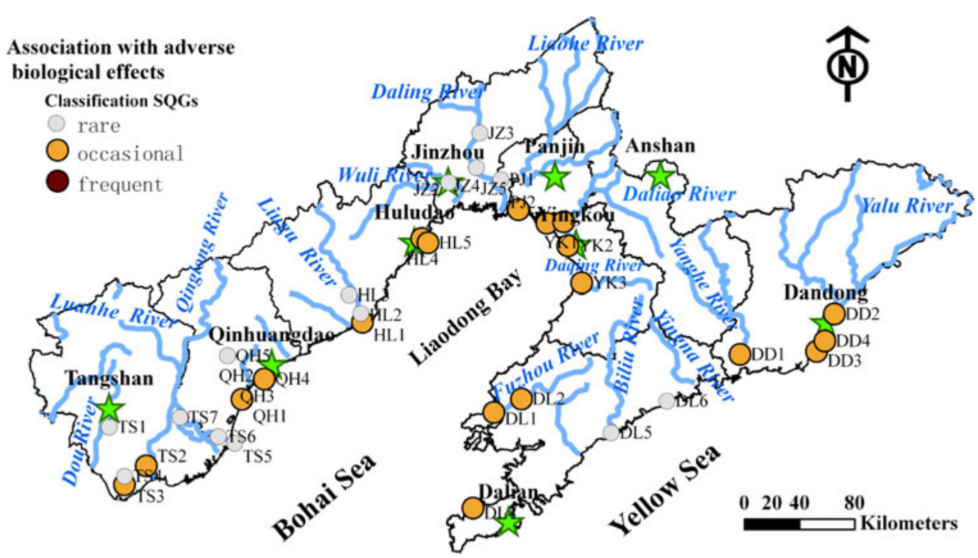

Fig. 4 Classification of the surface sediments based on SQGs (*Star* indicates city location)

Although SQGs have been found to be relatively good predictors of significant site contamination, they have several limitations (Burton 2002). False-positive and false-negative predictions are frequent for at least some. The SQGs are chemical-specific and do not establish causality where chemical mixtures occur. SQGs should be used only in a "screening" manner or in a "weight-of-evidence" approach. Aquatic sediments should be assessed in a "holistic" manner (i.e., concerned with, or dealing with wholes or integrated systems rather than with their parts) in which multiple components are assessed (e.g., habitat, hydrodynamics, resident biota, toxicity and physic-chemistry) by using integrated approaches.

CONCLUSIONS

The greatest concentrations of Hg, Pb, Cd, and Zn in the sediments were found at the estuarine area of the Wuli River and along the coast of the North Bohai Sea. The greatest concentrations of As and Cu occurred in sediments at the estuary of the Yalu River and along the North coast of the Yellow Sea, and the greatest concentration of Cr were observed in sediment from estuarine area of the Qinglong River. Based on the USEPA SQGs, As and Cr were most likely to cause adverse effects. However, based on the contamination factor and degree suggested by Håkanson, the contamination factor for arsenic and metals in the sediments follows in the order of Pb > Cd > Zn > Cr > Cu > As > Hg and the sediments in the Wuli and Yalu Rivers were classified as having a considerable degree of contamination. Based on comparisons to the effect-range classification for arsenic and metals, arsenic would be a potentially toxic to benthic organisms in sediments of the northern parts of the Bohai and Yellow Seas. Meanwhile, due to differential bioavailabilities, the potential effects of arsenic and metals can not be determined by only measuring their concentrations. A complementary study should be considered in order to provide a more accurate appraisal of arsenic and metal contamination and risks in the sediments.

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