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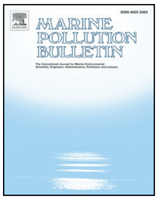

Heavy metals distribution and environmental quality assessment for sediments off the southern coast of the Shandong Peninsula, China

Shanshan Liu [a,b,c], Yong Zhang [a,b,*], Shipu Bi [a,b], Xiaobo Zhang [a,b], Xiaoyue Li [a,b,d], Manman Lin [a,b,e], Gang Hu [a,b]

[a] The Key Laboratory of Marine Hydrocarbon Resources and Environment Geology, Ministry of Land and Resources, China, 62 South Fuzhou Road, Qingdao 266071, PR China
[b] Qingdao Institute of Marine Geology, Qingdao 266071, China, 62 South Fuzhou Road, Qingdao 266071, PR China
[c] Geo-engineering investigation institute of Xiamen, China, 192 West Lianqian Road, Xiamen 361008, PR China
[d] Shijiazhuang University of Economics, China, 136 East Huaian Road, Shijiazhuang 050031, PR China
[e] Geophysical Exploration Academy of China Metallurgical Geology Bureau, China, 139 North Yangguang Street, Baoding 071051, PR China

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ABSTRACT

A systematic study was conducted on the distribution characteristics of heavy metals and on associated influencing factors in sediments off the southern coast of the Shandong Peninsula, China, based on the results of a heavy metals analysis on 157 surface sediment samples from coastal waters of the study area and on 46 samples from surrounding rivers flowing into the sea. An environmental quality assessment of heavy metals was performed using the Nemerow index. The results show that the distribution characteristics of sediment heavy metals in coastal waters outside of Qingdao can be divided into three classes: (1) Class I – Cr, Cu and Zn, with high-value areas extending from the northeast to the southwest in a banded or tongue-like pattern; (2) Class II – As, Cd and Pb, with high-value areas mainly distributed off the southeastern coast of Mt. Lao; and (3) Class III – Hg only, with high-value areas mainly in the northern area of Jiaozhou Bay. Integrated assessment based on the Nemerow index reveals that heavy metals pollution has occurred in surface sediments in a number of coastal water areas outside of Qingdao. This pollution is mainly found off the southeastern coast of Mt. Lao and in the northeastern section of Jiaozhou Bay. Results show that grain size of surface sediments, surrounding rivers and human activities are the main reasons for the element distribution pattern.

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Sediments act as a source and sink for heavy metals in water environment. Moreover, sediments serve as indicators of heavy metals pollution (Forstner, 1978; Livett, 1988). Therefore, heavy metal elements in sediments not only contain a high degree of valuable geological and environmental information but also reflect pollution conditions in the region. Sediment heavy metals serve as important tools for geochemical studies and ecological environment quality assessments (Ma and Wang, 2003; Liu et al., 2013). Environmental pollution problems associated with heavy metals have spurred worldwide concern since the middle of last century.

Research on the content of heavy metals that constantly accumulates in sediments helps increase understanding of the toxic effects of metal pollutants on benthic community structures. Meanwhile, heavy metals can be released from sediment into water, having long-term harmful effects on water environment. A typical example of water recontamination due to sediments is the case of Minamata disease in Japan (Sheng et al., 2008). It has been reported that sediment Pb content reaches up to 100×10^{-6}–150×10^{-6} in vast areas of Lake Erie and Lake Ontario in North America (Mudroch and Azcue, 1995). Pb, Zn and Cu contents in settling particles in seawater of the Jackson Port in Australia reaches as high as 365×10^{-6}–750×10^{-6}, 700×10^{-6}–1100×10^{-6} and 170×10^{-6}–280×10^{-6}, respectively (Taylor and Birch, 1996). Hg, Cd and Zn accumulate in sediments of the upper Elbe River of the North Sea, exceeding natural background values of 10–100-folds (Calmanl et al., 1995). The distribution of heavy metals in surface sediments is extremely variable in Jiaozhou Bay of Qingdao in Shandong Province, China. More specifically, high-value areas of heavy metals are mainly found in the estuary (Li et al., 2005; Zhang et al., 2011a,b); main heavy metal pollutants in intertidal sediments are derived from industrial discharge, domestic sewage, and precipitation and dust traffic pollution (Liu et al., 2010); there are relationships between heavy metals content, water depth, offshore distance and other factors (Yin et al., 2001). Regarding pollution assessments, methods commonly used to assess heavy metals pollution in water environment sediments include the pollution index, Hakanson potential ecological risk index, face graph, pollution load index, excess after regression analysis formula and geoaccumulation index (Sun et al., 1992; Gan et al., 2003; Wang et al., 2007; Wei et al., 2012; Liu et al., 2006, 2013; Chen, 1991; Wang and Yao, 2005; Li et al., 2011; Lin et al., 2013). Geoaccumulation and Hakanson index assessments have shown that overall heavy metals sediment pollution is minimal, while Hg pollution is relatively high in Jiaozhou Bay. With the exception of

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* Corresponding author at: Qingdao Institute of Marine Geology, Qingdao 266071, PR China.
E-mail address: qimgzy@163.com (Y. Zhang).

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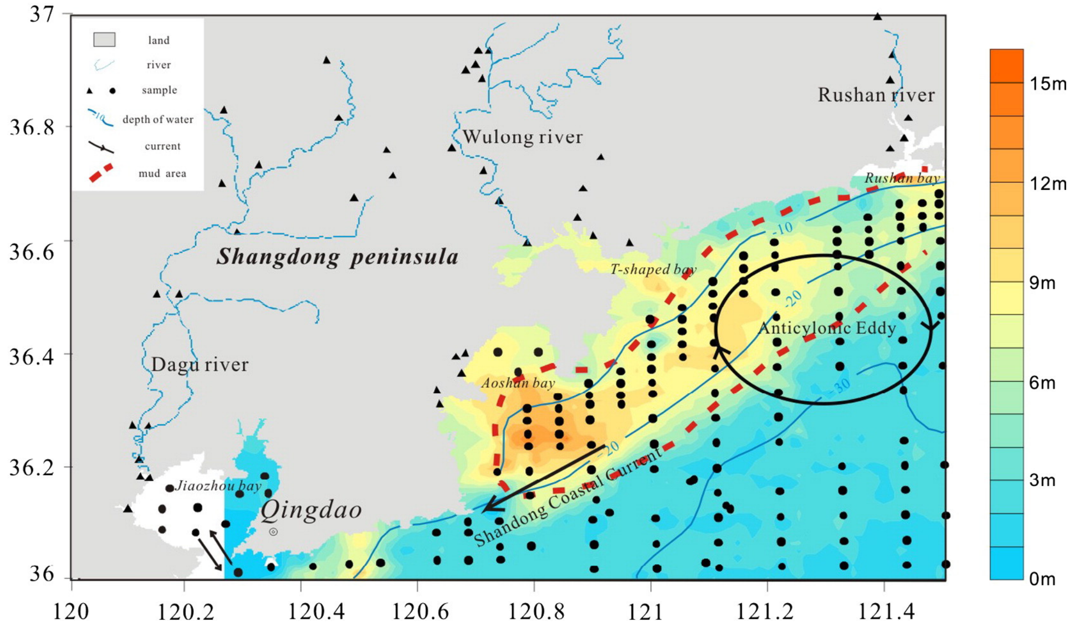

Fig. 1. Range and practical materials in mud area of the southern coastal waters off South Shandong Peninsula. (black dots show sample locations; black triangle dots are sample locations of rivers; blue lines characterize bathymetry counter lines; black line circles the Anticyclonic Eddy; black arrow means costal current; red dashed line circles the mud area; and counter map shows the mud thickness). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Cu and Cr, other heavy metals are found in higher quantities in July than in December in surface sediments of the eastern coast, with Cd pollution found in the highest quantity (Zhang et al., 2011a,b; Guo et al., 2012). In terms of overall ecological risk levels, heavy metal sediments present low potential ecological risks in the dumping ground outside of Jiaozhou Bay (Zhang et al., 2011a,b). The above findings show that research on heavy metal sediments in coastal waters off Qingdao have mainly focused on Jiaozhou Bay. Investigations of coastal waters in other areas of Qingdao are still scarce, and few geochemical reports on associated coastal waters are available. In this study, we collected and tested sediment samples from coastal waters off Qingdao and from surrounding rivers flowing into the sea. Using the obtained data, we examined the distribution characteristics of seven heavy metals (As, Cu, Cr, Cd, Hg, Pb and Zn) and their influencing factors in the study area and further assessed the environment qualitatively. The conclusions are the basis for appropriate policies to protect marine quality.

In July of 2011, the Qingdao Institute of Marine Geology launched the “Yezhizheng” research vessel for the collection of surface sediment samples from the seabed at 147 stations (Fig. 1). A sampling interval of 5 km was employed, and sample intensity levels were effectively increased in coastal areas. Each sample was at least 2 kg in weight. In October of 2011, surface sediment samples were collected from 46 stations in the flood plains of the Dagu, Wulong and Rushan Rivers around the study area. Sampling was performed using a wooden spatula, and the samples were stored in cloth bags. Each sample weighed at least 1 kg.

Sediment grain sizes were measured at the Experiment and Testing Center for Marine Geology of the Ministry of Land and Resources Department of the People's Republic of China. Portions (10–20 g) of the original sediment samples were soaked in H_2O_2 and HCl to remove organic matter and carbonates. Following salt leaching, the samples were dispersed in a $Na_6O_{18}P_6$ solution using ultrasound techniques. Grain sizes were measured using a Mastersizer-2000 laser particle size analyzer (Malvern, UK). Heavy metals testing was also conducted at the Experiment and Testing Center for Marine Geology of the Ministry of Land and Resources Department of the People's Republic of China. Sediment samples were dried at a constant temperature (<60 °C) and ground through a 250 mesh for elemental analysis. Cu, Pb and Zn were identified via X-ray fluorescence spectrometry; Cr and Cd were identified via inductively coupled plasma mass spectrometry; and As and Hg were identified via atomic fluorescence spectrometry. Blank and replicate samples were used for each of the testing items. The standard sample quality control results show high consistency with the reference values (relative deviation <5%), denoting that the test results are accurate and reliable.

Heavy metal elements include As, Cd, Cr, Cu, Pb, Zn and Hg, which are found in sediments at proportions of less than 0.10%. These heavy metals are critical components that affect the environmental quality of sediments. Statistical data on heavy metal sediment content are shown in Table 1 and Fig. 2. The content falls within the range of 4.3×10^{-6}–44.2×10^{-6}, with an average of 11.4×10^{-6}. High-As areas are concentrated off the southeastern coast of Mt. Lao. For Cd, high-value areas are mainly found off the southeastern coast of Mt. Lao and in the northern section of Jiaozhou Bay, with a sub-high-value area found in the northeastern section of Jiaozhou Bay. Cd content levels range from 32.0×10^{-9} to 210.0×10^{-9}, with an average value of 82.4×10^{-9}. High-Pb areas are mainly found in the northern Jiaozhou Bay and off the southeastern coast of Mt. Lao. Sediment Pb content is measured at 15.00×10^{-6}–41.2×10^{-6}, with an average level of

Table 1
Statistic values of heavy metals contents in surface sediments of Qingdao offshore.

	$Cu/\times 10^{-6}$	$Pb/\times 10^{-6}$	$Zn/\times 10^{-6}$	$Cr/\times 10^{-6}$	$As/\times 10^{-6}$	$Hg/\times 10^{-9}$	$Cd/\times 10^{-9}$
Mean	23.1	25.0	71.1	64.3	11.4	32.0	82.4
Standard deviation	5.0	4.4	17.5	13.0	5.6	9.8	28.7
Variation coefficient	0.22	0.18	0.25	0.20	0.49	0.31	0. 35
Minimum	8.2	15.0	21.3	20.0	4.3	12.0	32.0
Maximum	34.8	41.2	114.0	90.8	44.2	61.0	210.0

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