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Heavy metal enrichments in the Changjiang (Yangtze River) catchment and on the inner shelf of the East China Sea over the last 150 years

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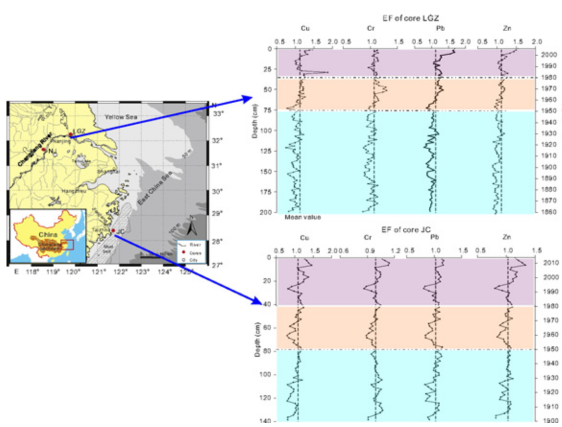
HIGHLIGHTS

- Three cores were collected from East China to evaluate the heavy metal enrichment.
- The major sources of heavy metals come from natural weathering detritus.
- Enrichment of Cu, Cr, Pb and Zn has increased over the last five decades.
- The heavy metal enrichment synchronizes with enhancing human activities.

GRAPHICAL ABSTRACT



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ABSTRACT

Compositions of heavy metals including Cu, Zn, Cr and Pb in three sediment cores recovered from the lower basin of the Changjiang (Yangtze River) and the inner shelf mud of the East China Sea were analyzed by traditional X-ray florescence (XRF) and XRF Core Scanner. This study aims to investigate the accumulation of heavy metals in the fluvial sediments and to decipher the influence of anthropogenic activities within the large catchment over the last 150 years. The data suggest that the heavy metals, especially Pb and Zn, show obvious enrichments in concentrations since 1950s, and the small and consistent variations of heavy metal concentrations before 1950s can represent geochemical background values. After removing the grain size effect on elemental concentrations, we infer that the sources of heavy metals predominantly come from natural weathering detritus, while human contamination has increased over the last half century. The calculations of both enrichment factor and geoaccumulation index, however, indicate that the pollution of these heavy metals in the fluvial and shelf environments is not significant. The rapid increase in human activities and fast socioeconomic development in the Changjiang catchment and East China over the last five decades accounts for the enrichments of heavy metals in the river and marine sediments. The inner shelf of the East China Sea, as the major sink of the Changjiang-derived fine sediments, provides a high-resolution sediment archive for tracing the anthropogenic impacts on the catchment.

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1. Introduction

Heavy metals of Cu, Cr, Pb and Zn accumulated in terrestrial and marine environments can cause serious problems to ecosystem due to their toxicity, persistence and bioaccumulation (Kelderman and Osman, 2007; Hu et al., 2013). There are two major sources of heavy metals in detrital sediments, i.e. natural inputs and anthropogenic contaminations, anthropogenic sources include biomass and fossil fuel combustion (coal, petroleum and natural gas), waste incineration, as well as mining and smelting industries (Nriagu and Pacyna, 1988; Pan and Wang, 2012). The relative contributions of anthropogenic sources to global emissions of heavy metals have considerably varied with time. Terrigenous sediment in fluvial and marine environments often serves as a major sink for heavy metals, and has become an environmental archive for the investigation of anthropogenic contamination in the past (Qiu et al., 2007; Krom et al., 2009). Geochemical composition of dated sediment cores has shown to be an excellent tool for evaluating the effects of anthropogenic and natural processes on depositional environments. A number of recent researches have used sediment cores from marine and lake environments to investigate and reconstruct historical records of contaminant inputs in these environments (Audry et al., 2004; Chen et al., 2004; Kelderman and Osman, 2007; Morelli et al., 2012; Townsend and Seen, 2012; Azoury et al., 2013; Duan et al., 2014; Vallius, 2014; Xu et al., 2014; Choi et al., 2015).

The Changjiang (Yangtze River) is the largest river in China and one of the largest in the word in terms of its huge water and sediment discharges, which plays a critical role on terrestrial material cycle and ecosystem health in the western Pacific marginal seas (Milliman et al., 1985; Saito et al., 2001; Yang et al., 2003; Wang et al., 2008). As the largest river in Asia, the Changjiang has the water and sediment discharges of about 896.4×10^9 m^3/year and 390×10^6 ton/year respectively, based on the multi-year observations at Datong gauge station (1950–2010) (Changjiang Water Resources Commission, http://www.cjw.com.cn/). Since the middle Holocene, a major portion of the Changjiang-derived sediments have been trapped in its estuary to build a large delta, and the remainder being mostly transported southeastward by the coastal currents, and formed a unique muddy depositional system on the inner shelf of the East China Sea (ECS), offshore the coastal Zhejiang and Fujiang Provinces (Qin, 1979; Liu et al., 2006; Liu et al., 2007; Xu et al., 2009; Yang et al., 2014). Thus, the sediment of the mud belt on the ECS shelf is predominantly from the Changjiang River.

In recent years, the Changjiang River is severely disturbed by anthropogenic activities, with the construction of the world's largest hydroelectric project (Three Gorges Dam, TGD) as a typical example. As a result, the sediment discharge to its estuary has been decreasing rapidly since the impoundment of TGD in 2003, averaging at only about 155×106 ton/year over the last ten years (2003–2013) (Changjiang Water Resources Commission, http://www.cjw.com.cn/). The Changjiang catchment has a population of about 500 million and has become one of the most important areas in the socioeconomic development of China. Thus, it is of great significance to evaluate the impact of anthropogenic activities on the fluvial and coastal sea environments.

Several studies have investigated the temporal and spatial distributions of heavy metal concentrations in the Changjiang catchment, which suggest that heavy metal enrichment has become serious over the last decade compared to the 1990s (Liu and Fan, 2011; Dong et al., 2012); the enrichments of organic pollutants and Pb in the Changjiang estuarine sediments are closely related to economic development (Shen et al., 2006; Guo et al., 2006). Generally, the estuary and coastal area have complicated hydrodynamic regimes subject to dynamic marine processes, and the sediments therein may be derived from both terrestrial and marine sources. Thus, it is difficult to reliably reveal the anthropogenic activity on the accumulation of heavy metals in the estuary and coastal ocean. However, the inner shelf mud of the ECS, as a major sink of the Changjiang-derived sediments during the mid-late Holocene (Liu et al., 2006; Liu et al., 2007; Xu et al., 2009), is characterized by continuous deposition with higher sediment accumulation rates (Gao and Wang, 2008; Yang et al., 2014), which thus provides a good archive for the high-resolution environmental study. Similarly, the lower valley of a river acts as the sink of sediment from the whole catchment, which makes it a desired area to evaluate the impact of human activity on the fluvial environment.

In this study, three short gravity cores with sediments ultimately from the Changjiang River were taken from the lower Changjiang mainstream (cores LGZ and NJ) and from the mud belt on the ECS inner shelf (core JC). The major purpose of this research is to investigate the anthropogenic impacts on the Changjiang catchment over the last 150 years. Based on the variations of geochemical elements and grain size of the core sediments, sources of heavy metals will be examined and the degree of heavy metal enrichment is assessed by using the proxies of enrichment factor and geoaccumulation index.

2. Materials and methods

The cores LGZ and NJ were taken from the lower mainstream of the Changjiang River in March 2008 and March 2010, respectively, and core JC was taken offshore the Zhejiang coast in April 2014 (Fig. 1). Core LGZ is located in a newly emerging bar in Yangzhong County, Jiangsu Province. The geographic coordinate is 32°18.393′ N and 119°45.218′ E. Core NJ was taken from the north bank of the Changjiang River near Nanjing City, with the geographic coordinate of 31°59.149′ N and 118°39.817′ E. Core JC was taken from the mud belt on the ECS inner shelf with the geographic coordinate of 28°39.017′ N and 121°41.300′ E, and the water depth of 10 m. No obvious agricultural and industrial activities are observed near the cores of LGZ and NJ, which makes them natural depositional environments in the Changjiang catchment and suitable for the study of environmental reconstruction.

Sediment cores LGZ and NJ were obtained by pushing about 200 cm long of PVC pipes into the ground, and subsequently pulled out manually. All the cores were taken to minimize disturbance and kept in a vertical position during transport from the field to the laboratory. This sampling technique has been used successfully in a range of sediment types and it yields undisturbed samples of sediment (Moura et al., 2004; Morelli et al., 2012). In laboratory, the cores were cut and split into two halves with a nylon string for minimizing metal contamination, and were stored at 4 °C until analysis.

The absolute and relative concentrations of elements of the core sediments were analyzed using X-ray fluorescence (XRF, PANalyticalAxiosMAX) and XRF Core Scanner (Avaatech Company). For the XRF Core Scanner analysis, the split-core surface was first flattened and covered with a thin Ultralene film to avoid contamination of the measurement prism of the core scanner, which allows continuous and non-destructive analysis of elements range from aluminum through to uranium (Richter et al., 2006). The relative concentrations of elements in the core sediments were acquired by scanning the core with a 0.5 cm resolution by the XRF Core Scanner. China Steam Sediment Reference Material (national geostandard GSD-15) was analyzed before and after the core scanning in order to confirm the stability of the equipment. The basic unit of the scanning elements is total counts or counts per second (cps). This unit implies the elemental intensity that is proportional to the chemical concentration (Tjallingii et al., 2007). In this study, the XRF-scan data will be presented as unprocessed intensities, i.e. cps.

After the XRF scanning, the cores LGZ, NJ and JC were sliced at 1–2 cm sampling intervals for the analyses of traditional XRF and grain size. A total of 101, 182 and 138 subsamples were respectively collected from cores LGZ, NJ and JC. For the XRF analysis, the subsamples were oven dried at 60 °C and ground to a fine powder with a size of about 200 mesh. The powder samples were heated again in the oven for 2 h at 120 °C and kept overnight at 60 °C before the XRF analysis. About 4 g samples were put into the tablet mold with boric acid prepared, and then the elemental concentrations were measured on the compressed disks.