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Distribution and accumulation of heavy metals in carbonate and reducible fractions of marine sediment from offshore mid-western Taiwan

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ABSTRACT

Two marine sediment cores from offshore mid-western Taiwan were subsampled and pre-treated using a sequential extraction procedure to separate carbonate and reducible fractions. Aliquots of these extracts were analyzed to determine their chemical composition to evaluate the geochemical processes responsible for heavy metal distribution and accumulation in the coastal environment. Our data demonstrate that sedimentation rates derived from excess ^{210}Pb associated with metal fluxes show large increases circa A.D. 1990. A well-synchronized increase in metal flux in both geochemical fractions was found and validated by Pearson's correlation. Principal component analysis revealed the heavy metal fluxes to be highly correlated with the sediment deposition rate, with metal contamination potentially driven by a sole contributor. This study emphasizes the changes in sedimentation rate is potentially caused by activities associated with the inland economic development during this time, rather than by raising heavy metal pollution dominated the accumulation offshore mid-western Taiwan.

1. Introduction

Sediments are often considered as both a major sink for anthropogenically-produced heavy metals in aquatic systems, and as a potential source of trace metals for the overlying water column and benthic biota (Chapman et al., 1998). A systematic investigation of heavy metals in Asian riverine sediments revealed the major source of such pollution to be anthropogenic activities, including the release of domestic and industrial wastewater (Fang and Yang, 2010). A risk assessment of the bioavailability of metals in contaminated sediments has therefore been taken into serious consideration (Fan et al., 2002).

Sedimentary particulate matter typically consists of various geochemical fractions, including (1) ion-exchangeable, (2) carbonate, (3) reducible (Fe-Mn oxide), (4) organic matter, (5) sulfides, and (6) residues (Abollino et al., 2011; Leleyter and Probst, 1999; Maher, 1984; Rao et al., 2008). A number of previous studies have assessed heavy metal mobilization and redistribution via the analysis of the total metal concentration in bulk sediment (Hsu et al., 2006; Hung and Hsu, 2004). However, sequential extraction procedures, isolating different metal species among sorbent phases, can provide further information regarding metal geochemical behavior in advance (Caplat et al., 2005; Li et al., 2007; Wang et al., 2010; Yuan et al., 2004). Tessier's popular sequential extraction procedure, as well as BCR schemes together with their modified protocols, have been extensively adopted for various matrices, including river sediments (Yu et al., 2001a), contaminated soils (Meers et al., 2006; Munoz-Melendez et al., 2000), agricultural soils (Bäckström et al., 2004), estuarine sediments (Riba et al., 2002), and marine sediments (Wang et al., 2010).

Over the past few decades, a considerable number of studies have investigated spatial/temporal variations in heavy metals bounded to different sequentially-extracted marine sediment fractions (Abollino et al., 2011). There is fairly general agreement that metal binding with different geochemical fractions fluctuates considerably among different heavy metals, sedimentation environments and even seasons (Bostick et al., 2001; O'Day et al., 2000; Souch et al., 2001). Heavy metal mobility and availability depend on the binding behavior of the elements in question, and their reactivity with the components of the geochemical matrices. As a result, the evaluation of heavy metal accumulation in sediments cannot be carried out based simply on metal concentration values in individual geochemical phases or bulk sediments. In the present study, heavy metal flux variations recorded in two major metal carriers, carbonate and reducible, were evaluated in two sediment

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cores obtained near the coastal region off a major river mouth in mid-western Taiwan. The main objectives are (1) to study the relationship between the heavy metal fluxes and sediment accumulation rates, and (2) to evaluate any correlation in heavy-metal binding patterns among different geochemical fractions in the sedimentary cores. This information is critical, as significantly enhanced levels of heavy metals associated with the transportation of riverine sediment has affected coastal regions of Taiwan following the country's industrialization over recent decades.

2. Study area and materials

Taiwan is situated at the collision boundary between the Philippine oceanic plate and the Eurasian continental plate. This boundary is also an active orogenic belt and has the highest uplift rate in the world, with an average of 2–10 mm $year^{-1}$ (Liu and Yu, 1990). Similarly, the rates of physical denudation and chemical weathering in the region are among the highest in Asia (You et al., 1988), if not in the world (Horng et al., 2012). The high relief Central Mountain Range (CMR) located in central Taiwan has a well-developed radial drainage system which transports riverine materials to the Taiwan Strait, with an estimated sediment load of up to 60–150 Mt y^{-1} (Liu et al., 2008).

The bathymetry of the Taiwan Strait is characterized by several salient topographic features (Fig. 1); the deepest of these is the Penghu Channel (PHC) in the southeast, while the Chan-Yuen Rise (CYR), and the Kuan-Yin Depression (KYD) located north of PHC surround the Taiwanese coast in the eastern Taiwan Strait. Two sediment cores, BC7 (24°30.02′N, 120°30.00′E) and GC24 (24°20.23′N, 120°10.19′E), were taken from the north of CYR near the estuary of the Choshui River (CR) on May 28, 2007 during the OR2-1442 cruise. CR has a long-term average sediment load of 60 Mt y^{-1} and is the largest river in Taiwan. The Wu River (WR) has a sediment load of 10 Mt y^{-1} and is one of the large rivers in the central Taiwan. Together, these two rivers discharge more than half the sediment transported from Taiwan into the strait. Therefore we believe that accumulated in these two cores are sediments that are representative of the weathering products removed from major drainage systems in western Taiwan. The configuration and spatial distribution of sediments are indicative of depositional environments, a tide-dominated regime affected by the northeast-flowing alongshore current (Liao et al., 2008; Huh et al., 2011). The particle size of these surface sediments is dominated by very fine-to-coarse sands (0–4Φ), with grain size progressively decreasing seaward. Clay accounts 13.26% and 6.93% only in BC7 and GC24, respectively. TOC is preferentially transported northeastward by the strong alongshore current nearby CR river mouth. Our preliminary assessment indicates that only insignificant organic matters and sulfide fractions can be deposited in the study area. Investigations of heavy metal accumulation were focused on the two major metal carriers of carbonate and reducible fractions. The sampled cores were sectioned at 2 cm intervals in the upper 40 cm, and at 3 cm intervals thereafter. More details about the bathymetry and hydrodynamics of the Taiwan Strait and the procedures for sub-sampling of the cores can be found in Huh et al. (2011).

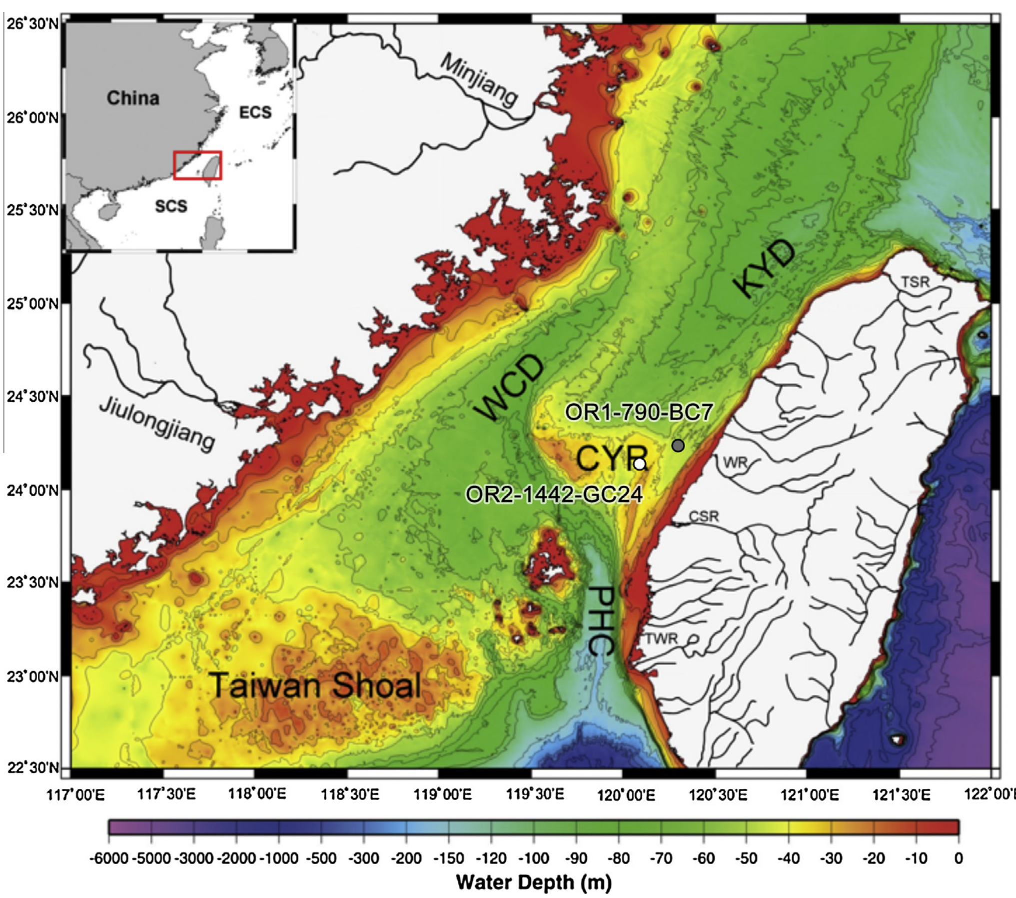

Fig. 1. Locations of sediment cores and bathymetry of the Taiwan Strait.