



## Full length article

## Clay mineralogy indicates the muddy sediment provenance in the estuarine-inner shelf of the East China Sea



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## ARTICLE INFO

## Keywords:

Sediment provenance  
Clay minerals  
Source-to-sink  
Yangtze River  
Estuarine-inner shelf  
East China Sea (ECS)

## ABSTRACT

The estuarine-inner shelf mud regions of the East China Sea (ECS) are valuable for studying the source-to-sink processes of fluvial sediments deposited since the Holocene. In this study, we present evidence of the provenance and environmental evolution of two cores (S5-2 and JC07) from the estuarine-inner shelf regions of the ECS over the past 100 years based on <sup>210</sup>Pb dating, high-resolution grain size measurements and clay mineral analyses. The results indicate that the clay mineral assemblages of cores S5-2 and JC07 are dominated by illite, followed by kaolinite and chlorite, and present scarce amounts of smectite. A comparison of these clay mineral assemblages with several major sources reveals that the fine sediments on the estuarine-inner shelf of the ECS represent a mixture of provenances associated with the Yangtze and Yellow Rivers, as well as smaller rivers. However, the contribution of each provenance has varied greatly over the past hundred years, as indicated by the down-core variability due to strong sediment reworking and transport on the inner shelf and the reduction of the sediment load from the Yangtze River basin. In the mud region of the Yangtze River estuary, the sediment from 1930 to 1956 was primarily derived from the Yangtze River, although the Yellow River was also an important influence. From 1956 to 2013, the Yellow River contribution decreased, whereas the Yangtze River contribution correspondingly increased. In the Zhe-Min mud region, the Yangtze River contributed more sediment than did other rivers from 1910 to 1950; however, the Yangtze River contribution gradually decreased from 1950 to 2013. Moreover, the other small rivers accounted for minor contributions, and the East Asian winter monsoon (EAWM) played an important role in the sediment transport process in the ECS. Our results indicate that the weakening/strengthening of the EAWM and a decrease in the sediment load of the Yangtze River influenced the transport and fate of sediment on the estuarine-inner shelf of the ECS.

## 1. Introduction

Rivers represent the primary link between land and sea and transport a large quantity of sediment, equivalent to approximately 15–20 billion tons/yr, to the global ocean (Milliman and Meade, 1983). Most of this sediment is deposited on continental shelves and the slopes of marginal seas. As one of the largest river-dominated marginal seas in the world, the East China Sea (ECS) is primarily influenced by fine-grained sediment supplied by major rivers (and small mountain rivers) in eastern mainland China, as well as by ocean circulation and monsoon

patterns (Lee et al., 2004; Diekmann et al., 2008; Yuan et al., 2008; Dou et al., 2010). The mud deposits in the Yangtze River estuary and the inner-shelf mud region of the ECS are formed by a large quantity of river-deposited terrigenous sediment and are affected by tidal currents, estuarine processes and shelf circulation (Guo et al., 2003; Liu et al., 2007a,b,c). The rate of sediment deposition is high (Satio et al., 2001; Hori et al., 2002; Xiao et al., 2006; Lim et al., 2007; Liu et al., 2007a,b,c, 2010a,b), especially in the Yangtze River estuary, where the sedimentation rate over the last hundred years has been 3–5 cm/yr (DeMaster et al., 1985; Hu et al., 2001; Chen et al., 2004; Liu et al.,

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<https://doi.org/10.1016/j.jseaes.2017.11.036>

Received 13 February 2017; Received in revised form 23 November 2017; Accepted 24 November 2017

Available online 24 November 2017

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2006; Qiao et al., 2017). Therefore, the ECS is the ideal area to study sediment source-to-sink transport processes, environmental changes, storm records and East Asian monsoon evolution (Li et al., 2016, 2015; Zhao et al., 2016a,b; Feng et al., 2016; Wang et al., 2014).

In recent decades, determining the provenance of river-dominated sediments in this region has become a major issue because of the difficulties presented by the geology, climate conditions and weathering of the two major material source areas, the Yangtze River and Yellow River basins, as well as those of minor river basins, including the Qiantang, Ou, and Min River basins and small mountainous river basins in Taiwan (Yang et al., 2015). Most approaches to studying the sediment sources of the estuarine-inner shelf of the ECS have focused on the magnetic properties (Liu et al., 2010a,b; Kim et al., 2013), elemental composition (Yang et al., 2006a,b), rare earth elements (Xu et al., 2011) and Sr-Nd isotopic compositions (Yang et al., 2007). However, the transport of fine-grained sediment from terrigenous sources to mud deposits in the estuarine-inner shelf region is poorly understood.

The clay minerals in recent marine sediments are mainly detrital in origin (Griffin et al., 1968; Velde, 1995) and can be transported over long distances and settle far away from their sources, especially if they are re-transported within the nepheloid layer (Jones, 1984; Gingele et al., 2001; Sionneau et al., 2008). Therefore, clay minerals have been widely used to trace the provenance of terrigenous particles and to determine the intensity of continental weathering in the source region (Gingele et al., 2001; Chamley, 1989). Hydrodynamic sorting can greatly affect clay mineral contents in estuarine and coastal sea environments. Most previous studies of clay mineral assemblages in the ECS have focused on the surface sediments of estuarine areas and the Okinawa Trough over long periods (e.g., the Holocene) to ensure the inclusivity of different sediment sources and partitioning processes during transport (Fan et al., 2001, 2007; Diekmann et al., 2008; Dou et al., 2010; Liu et al., 2014). Chen (1978) found that the clay mineral assemblage of the surface sediments in the ECS is dominated by illite and chlorite. Guo et al. (1995) and You et al. (1993) showed that the illite content increases southward in the ECS from 65% in the Yangtze River estuary to 73% in the northeastern Taiwan Strait. Liu et al. (2006) showed that the Yellow River is not a major supplier of mud to the estuarine-inner shelf of the ECS, although it transports suspended sediments into the ECS. Furthermore, Liu et al. (2014) investigated the clay mineral composition of Core MZ02 from the inner shelf of the ECS and suggested that the clay is associated with a mixed sourced from the Changjiang, Minjiang, and Taiwan Rivers. However, few studies have investigated the provenance of clay minerals in the mud deposits on the estuarine-inner shelf of the ECS over short periods, such as during the past hundred years. Thus, in this study, we used clay mineral data to provide a continuous record of continuous terrigenous “source-to-sink” processes based on two cores retrieved from the estuarine-inner shelf region of the ECS. The objectives of this paper are to (1) estimate the sediment sources of the cores and their temporal changes and (2) further discuss the links between the clay mineral assemblages and climate changes in the mud region of the ECS over the past hundred years.

## 2. Study area

The study area is located in the coastal mud area of the ECS (Fig. 1). The continental shelf of the ECS is one of the largest continental shelves in the world, with an average width of 500 km and a shallow depth of 130 m. The shelf is situated on the margin of the northwestern Pacific Ocean. Numerous rivers drain into the western Pacific and provide the major routes for the transport of terrigenous sediments to the ocean (Xu et al., 2009a,b). The Yangtze River is the largest river on the Asian continent and is one of the world's great rivers. Geologically, the Yangtze River catchment comprises complex rock types in the mainstream and its major tributaries. The upper basin is characterized by Paleozoic carbonate rocks, Jurassic red sandstone and Mesozoic igneous rocks, and the middle-lower basin mainly consists of Paleozoic

marine and Quaternary fluvial and lacustrine sedimentary rocks (Yang et al., 2004). The Yangtze River has transported approximately 470 Mt/yr of terrigenous suspended sediment into the ECS over the past century (Milliman and Farnsworth, 2011), and a huge quantity of sediments has accumulated in the estuary and on the adjacent ECS shelf, especially on its southern margin along the Zhejiang-Fujian coast (Milliman and Meade, 1983; Liu et al., 2006, 2007a,b,c; Xu et al., 2009a,b; Yang et al., 2011a,b; Zhao et al., 2016a,b). These deposits have formed coastal mud areas, such as the Yangtze mud area and Zhe-Min coastal mud area. The sediment load transported by the Yellow River into the ECS has been approximately  $< 0.2\text{--}1.6 \times 10^8$  t/yr over the past hundred years (Demaster et al., 1985; Su and Huh, 2002). In addition to these large rivers, several small rivers, such as the Min, Ou, and Qiantang Rivers (less than  $2.0 \times 10^7$  t/yr of sediment) also play important roles in sedimentation in the ECS (Deng et al., 2006). Moreover, rivers the flow from Taiwan annually deliver  $2.6 \times 10^8$  t/yr of sediment to the ocean (Ren, 2003), and a portion of this sediment is carried northward by the effects of the Taiwan Warm Current (TWC).

The continental shelf circulation current and the East Asian winter monsoon are the dominant physical drivers of the sedimentary processes in the study area (Xu et al., 2015). The main currents governing the circulation in the study area are the Zhejiang-Fujian Coastal Current (ZFCC) and the TWC. In the summer, the TWC intensifies and the coastal currents weaken because of prevailing southeast monsoons, leading to the deposition of a large suspended sediment load from the Yangtze River at the river mouth (Shen, 2001; Liu et al., 2006). In contrast, in the winter, the ZFCC is stronger and carries the freshwater and sediment load from the Yangtze River southward along the inner shelf (Milliman et al., 1985; Su, 2001).

## 3. Materials and methods

Two sediment cores (S5-2 and JC07) were collected on December 20, 2013, from the estuarine-inner shelf regions of the ECS using a stainless-steel gravity core sampler (Fig. 1). Core S5-2 (30.37°N, 122.76°E) was collected from the mud area of the Yangtze River estuary at a water depth of 22.6 m, and core JC07 (28.23°N, 122.15°E) was collected from the Zhe-Min mud area at a water depth of 48.3 m. Moreover, twenty-one suspended sediment samples were collected from the main branch and major tributaries of the Yangtze River basin in June–July 2015. These samples were filtered in situ using membranes with a pore size of 0.45 μm to acquire water samples. The sampling sites were distributed throughout the entire river system (Fig. S1). The grain size measurements were performed using a Mastersizer 2000 instrument (Malvern Instruments Ltd., UK), which measures grains in the range of 0.02–2000 μm and has a relative error within 3% for replicated measurements. Before the grain size analyses, the air-dried sediment samples were successively pretreated with H<sub>2</sub>O<sub>2</sub> (30%) to remove organic matter and with HCl (10%) to remove carbonates. Aggregates were then dispersed by the addition of (NaPO<sub>3</sub>)<sub>6</sub> and subsequent ultrasonic treatment. Detailed particle-size parameters were calculated using the moments method provided by McManus (1988). The radioactive nuclide <sup>210</sup>Pb was used to calculate the deposition rates and to date the sediments. In this study, <sup>137</sup>Cs was not used because the total amount of <sup>137</sup>Cs in the sedimentary environment has decreased by 65% and because <sup>137</sup>Cs is potentially mobile in the sedimentary profile (Pan et al., 2011; Gao et al., 2017). The <sup>210</sup>Pb activity was measured based on α-rays (HPGe alpha spectrometer) at the Ministry of Education Key Laboratory for Coast and Island Development, Nanjing University. The sedimentation rates were calculated following the method of Goldberg and Koide (1963), and the pretreatment process for the determination of Pb isotopes was the same as that described by Zhao et al. (2016a,b).

The clay minerals were identified via standard X-ray diffraction (XRD) using a D8 ADVANCE diffractometer at the Key Lab of Marine Sedimentology and Environmental Geology (MASEG), State Oceanic Administration (SOA), China, on oriented mounts of non-calcareous

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