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Clay mineral distribution and provenance in the Heuksan mud belt, Yellow Sea

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Abstract The Heuksan mud belt (HMB), located in the southeastern Yellow Sea, runs parallel to the southwest coast of Korea. In this study, the distribution and relative contribution of four major clay minerals are investigated using 101 surface sediment samples collected in the course of KIOST (2001, 2010, 2011) and KIGAM (2012) cruises, as well as 33 river sediment samples (four from the Huanghe River, three from the Changjiang River, and 26 from Korean rivers) in order to clarify the provenance of fine-grained sediments in the HMB. Based on this currently largest and most robust dataset available for interpretation, the clay mineral assemblages of the fine-grained sediments in the HMB are found to be on average composed of 64.7% illite, 17.9% chlorite, 11.4% kaolinite, and 5.9% smectite. Overall, the clay mineral assemblages are similar in both the northern and the southern parts of the HMB, although smectite seems to be relatively enriched in the southern part, whereas kaolinite is slightly more dominant in the northern part. This clearly indicates that the clays are mostly derived from Korean rivers and, in the southern part of the HMB, partly also from the Huanghe River in China. The new data thus confirm and strengthen the tentative interpretation of some earlier work based on a more limited dataset.

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Introduction

Large amounts of fine-grained sediments have been transported from numerous sources to be deposited in the world oceans. Based on the estimation of global fluxes, more than 23 billion tons of sediments have been transported annually to the world oceans by rivers, ice, atmosphere, and the sea itself (Milliman and Meade 1983). Among these, rivers are the most important agent of sediment transport, supplying about 20 billion tons per year of suspended or dissolved loads into the oceans (Milliman and Farnsworth 2011). Most river-suspended and ice- and atmosphere-transported sediments are of terrigenous origin. About 60% of all these materials are assumed to be clay minerals (Weaver 1989), and thus about 10 billion tons of clay minerals are delivered to the ocean every year.

A variety of mineralogical, geochemical, sedimentological, and geophysical methods as well as remote sensing technologies have been used for sediment provenance studies. Specific minerals, especially quartz (e.g., Nascimento et al. 2015), heavy minerals (e.g., Yang et al. 2009), and clay minerals (e.g., Li et al. 2014), have been shown to be suitable provenance indicators. Among these, clay minerals are the most commonly used minerals, not only because they form an important component of fine-grained marine deposits but also because they reflect the geology of drainage basin particularly well. Nevertheless, clay mineral assemblages in marine environments are quite complex and influenced by many factors such as source geology, hydrodynamic sorting, depositional alteration, and submarine weathering (e.g., Petschick et al. 1996; Pache et al. 2008; Liu et al. 2010). With respect to source geology, the production of clay minerals strongly depends on the nature of the parent rocks and the weathering conditions on land. For these reasons, clay mineral assemblages are particularly useful in reconstructing sediment sources and climatic conditions of adjacent continents (Hiller 1995).

The Yellow and the East China seas have for many decades been studied in terms of late Quaternary land–ocean interactions and paleo-environmental changes. Of the two seas, the Yellow Sea occupies the epicontinental shelf between the Chinese mainland and the Korean Peninsula, whereas the East China Sea is characterized by a broad shallow shelf dominated by a prominent submarine delta (Chough et al. 2000). Large and well-defined mud deposits occur in many places of the Yellow and adjacent seas (Ren and Shi 1986; Lee and Chough 1989; Alexander et al. 1991; Yang et al. 2003; Shi et al. 2012; Fig. 1a). Recently, Dong et al. (2011) suggested that the formation mechanisms of mud deposition in shallow and deep waters of the Yellow Sea differ because of their different affinity to river inputs and different, though complex hydrodynamic conditions. The mud deposits in the Yellow Sea are mostly composed of fine-grained sediments of terrigeneous origin, being mainly associated with the discharge of the Huanghe and Changjiang rivers in China and the Han, Geum, and Yeongsan rivers in Korea (e.g., Yang et al. 2003).

To determine the source and transport pathways of fine-grained sediments in these mud deposits, mineralogical, geochemical, geophysical, and sedimentological methods as well as circulation pattern studies have been applied (Chough and Kim 1981; Xu 1983; Milliman et al. 1985; Park and Khim 1992; Choi and Kim 1998; Wei et al. 2000; Yang et al. 2003; Lim et al. 2006, 2007; Yang and Youn 2007; Moon et al. 2009; Choi et al. 2010; Cho et al. 2011; Shi et al. 2012; Ha et al. 2013; Li et al. 2014). Despite intensive studies over the last three decades, controversy still persists with respect to the provenance of the sediment. For example, based on clay mineralogy, the central Yellow Sea mud (CYSM) has been suggested to have been supplied by the Huanghe River (e.g., Park and Khim 1992), whereas geochemical studies suggest multiple sources (e.g., Lim et al. 2006; Yang and Youn 2007). Recently, Li et al. (2014) suggested that the CYSM is most likely a mixing product of fine-grained sediments supplied by the Changjiang River and small rivers of North Korea. This interpretation has recently been supported by the findings of Lim et al. (2015) based on clay minerals, rare earth element fractionation parameters, and conservative trace elements, which suggest that the dominant sediment source in the CYSM was the Changjiang River together with some clay-sized particulate input from the Huanghe River and smaller Korean rivers.

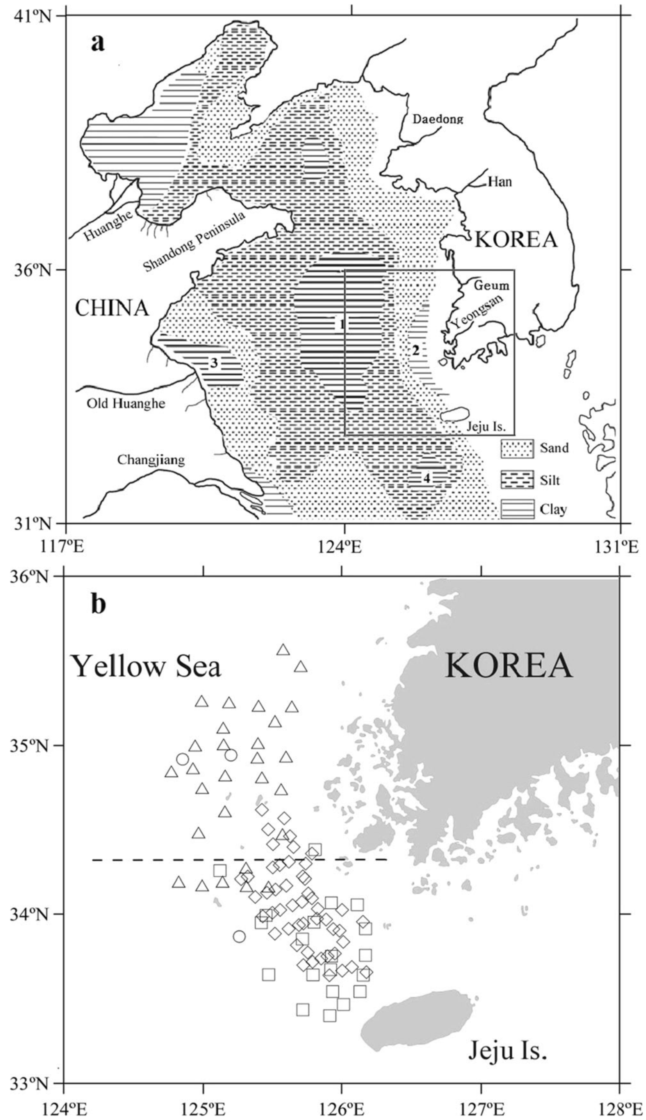


Fig. 1 **a** Map showing the fine-grained sediment deposits in the Yellow Sea and the East China Sea (modified after Yang et al. 2003). *1* Central Yellow Sea mud, *2* southeastern Yellow Sea mud (i.e., Heuksan mud belt, HMB), *3* Old Huanghe River delta mud, *4* southwestern Cheju Island mud. *Box* Study area. **b** Map showing sampling locations of surface sediments in the HMB collected during the KIOST cruise of 2001 (*circles*), 2010 (*rectangles*), and 2011 (*triangles*), and during the KIGAM cruise of 2012 (*diamonds*)

The so-called southeastern Yellow Sea mud (SEYSM, cf. Park and Khim 1992) forms a 20–50 km wide and over 200 km long belt extending parallel to the southwest coast of the Korean Peninsula. This mud deposit has been renamed the Heuksan mud belt (HMB, after the nearby Heuksan Island) by Lee and Chu (2001). Although numerous studies have attempted to clarify the formation of the HMB deposit, the governing processes are still controversially discussed (Jin and Chough 1998; Park et al. 2000; Chough et al. 2002; Shinn et al. 2007; Lee et al. 2014). For example, the sequence stratigraphy of the transgressive deposits and sediment budgets of the HMB differ substantially between the studies. Also inconsistent are opinions on sedimentary processes and dispersal mechanisms, along with their spatial variability. Nonetheless, Lee et al. (2014) suggest that the uppermost unit of the HMB is composed of recent mud derived from the Geum River in Korea.

Similar to the disagreement concerning the formation mechanism of the HMB, the provenance of fine-grained sediments in the HMB is also still being debated (Chough and Kim 1981; Schubel et al. 1984; Park and Khim 1992; Lee and Chu 2001; Lim et al. 2007; Moon et al. 2009; Cho et al. 2011; Ha et al. 2013). The two main opposing views are (1) that the muds largely originated from Korean rivers and (2) that the northern and southern parts of the HMB differ in origin. For example, hydrographic measurements together with satellite imagery clearly indicate that the suspended plume toward the HMB emanates from the west coast of Korea (i.e., from the Geum River; cf. Lee and Chu 2001). In contrast, Ha et al. (2013) reported that rare earth elements in sediments from the southern part of the HMB closely resemble those of Chinese rivers, although the major elements are characteristic of Korean river-derived sediments. Compared with the numerous clay mineral studies in the Yellow and East China seas, clay mineral data from the HMB are still rather scarce. In addition, most previous clay mineral studies in this region are based on less than 20 samples from limited areas. A comprehensive picture of clay mineral distribution and provenance in the HMB is thus still lacking.

Within this context, the present study concentrates on the documentation of clay mineral distribution and composition (i.e., of illite, chlorite, kaolinite, and smectite) in the HMB and the identification of their potential sources. For this purpose, 101 surficial sediment samples were collected from the HMB, complemented by 33 comparative samples from Korean and Chinese rivers discharging into the Yellow Sea.

Materials and methods

Surface sediments in the HMB were collected with different sampling tools in the course of several research cruises (Fig. 1b). A total of 51 samples were obtained by the Korean Institute of Ocean Science and Technology (KIOST) in the course of cruises in 2001, 2010 and 2011 using a grab sampler. Another 50 samples were subsampled from box or piston cores collected by the Korean Institute of Geoscience and Mineral Resources (KIGAM) in 2012. The sample positions together with sand, silt and clay contents are listed in ESM Table 1 of the electronic supplementary material available online for this article. In addition, 33 river sediment samples (four samples from the Huanghe River, three from the Changjiang River, and 26 from Korean rivers) were collected for comparative purposes aimed at tracing potential sediment sources of surficial sediments in the HMB (for more information, see Song and Choi 2009).

Semi-quantitative clay mineral analyses were carried out by X-ray diffraction (XRD) on preferred-oriented specimens of clay-sized particles (<2 μm). Clay-sized fractions were prepared according to Stoke's law after removal of organic matter using 6% H_2O_2, wet sieving through a 63 μm (230 mesh) sieve, and centrifuging at 7,400 rpm for 5 minutes (Kim et al. 2011). The concentrated clay paste was spread thinly onto glass slides following the "smear-on" method to minimize grain-size effects (Stokke and Carson 1973).

The XRD analysis was conducted using a Siemens/Brucker D5005 analytical X-ray system with CuKα radiation and monochrometer at 40 kV/40 mA and 3~30° (2-theta) conditions. Two XRD runs were performed on each sample, which were air-dried and ethylene-glycol (EG) treated for more than 6 h at 65 °C prior to analysis. Semi-quantitative calculations of each peak were carried out using the Bruker EVA 3.0 program. The relative contents of the four major clay minerals smectite (S), illite (I), kaolinite (K), and chlorite (C) were estimated through the particular peak (001) area ratio of each clay mineral on the EG diffractogram. In each case, the weighting factor was multiplied with the estimated area to revise the intensity (001) difference of each mineral (Biscaye 1965). The relative contents of kaolinite and chlorite were obtained using the peak area ratios of the chlorite (004) and kaolinite (002) peaks (Biscaye 1964).

Results

The clay mineral compositions of Chinese and Korean river sediments are compiled in Table 1. Without exception, and in descending order, the clay mineral assemblages of all river sediments are composed of illite, chlorite, kaolinite, and smectite. It is worthy to note, however, that the Huanghe River sediments can be distinguished from the Korean and Changjiang river sediments by their relatively high smectite content, and low kaolinite and chlorite contents (Table 1).

According to Choi et al. (2010), Korean river sediments are enriched in kaolinite and chlorite (>44%), but depleted in smectite (3.6%), compared with Chinese river sediments. Changjiang River sediments are illite-rich (62%) and smectite-poor (3.9%), whereas Huanghe River sediments are enriched in smectite (12.0%). The clay mineral compositions of river sediments analyzed for this study are in good agreement with those of Choi et al. (2010), except for a few differences such as the chlorite and kaolinite contents of Chinese river sediments, and the illite and chlorite contents of Korean river sediments. Overall, the clay mineral compositions of the river sediments from around the Yellow Sea can be distinguished from each other in that (1) the Huanghe River sediments are characterized by high smectite contents; (2) the

Table 1 Clay mineral composition of the river sediments around the Yellow Sea (*n* sample size, present study)

River	*n*	Illite (%)	Chlorite (%)	Kaolinite (%)	Smectite (%)	References
Old Huanghe		59.0	8.1	8.9	24.0	Xu (1983)
		63.7	8.3	7.7	20.5	Qin and Li (1983)
Modern Huanghe		59.0	9.3	8.5	23.2	Xu (1983)
		65.0	12.0	9.0	14.0	Milliman et al. (1985)
		62.0	16.0	10.0	12.0	Yang et al. (2003)
		56.3	22.4	9.3	12.0	Choi et al. (2010)
	4	56.7	17.7	14.1	11.5	This study
Changjiang		68.0	13.9	12.7	5.5	Xu (1983)
		53.0	10.0	18.0	19.0	Milliman et al. (1985)
		66.0	12.0	16.0	6.0	Yang et al. (2003)
		61.7	24.5	9.8	3.9	Choi et al. (2010)
	3	61.0	19.6	16.3	3.2	This study
Han		70.0	16.8	12.5	0.7	Park and Khim (1992)
	14	59.9	21.4	16.5	2.1	This study
Geum		63.7	19.3	17.0	0.1	Choi (1981)
		59.3	17.4	18.9	4.4	Yang et al. (2003)
	9	54.5	23.5	20.9	1.1	This study
Yeongsan		63.9	16.8	19.2	0.1	Park and Khim (1992)
		59.5	13.8	13.3	13.4	Yang et al. (2003)
	3	63.5	19.5	15.2	1.7	This study

Changjiang River sediments are relatively enriched in illite; and (3) the Korean river sediments show relatively high kaolinite and chlorite contents. These features and differences are highlighted in a ternary diagram illustrating the I–S–(K+C) ratios (Fig. 2), and in a crossplot between the S/I and (K+C)/I ratios (Fig. 3).

The clay mineral compositions of surficial HMB sediments found in the present study are compared with those of previous studies in Table 2. Although the clay mineral contents differ by small amounts in the various studies, the results of the present study are in good agreement with those of the previous investigations. For example, the illite content of the marine sediments shows great similarity to those of the river sediments. As in the case of the river sediments, clay mineral assemblages of the HMB sediments also consist predominantly of illite, chlorite, kaolinite, and smectite, in decreasing order. The distribution patterns of the four clay minerals in the HMB are shown in Fig. 4.

From Table 2 it can be seen that Wei et al. (2000) reported different clay mineral compositions between the northern and the southern part of the HMB. To cross-check this, the clay mineral data of the present study were subdivided into a northern and a southern part separated at 34°20′N where the shape of the HMB changes from a NS to a NW–SE direction. In general, the northern and southern parts of the HMB sediments of the present study show similar clay mineral

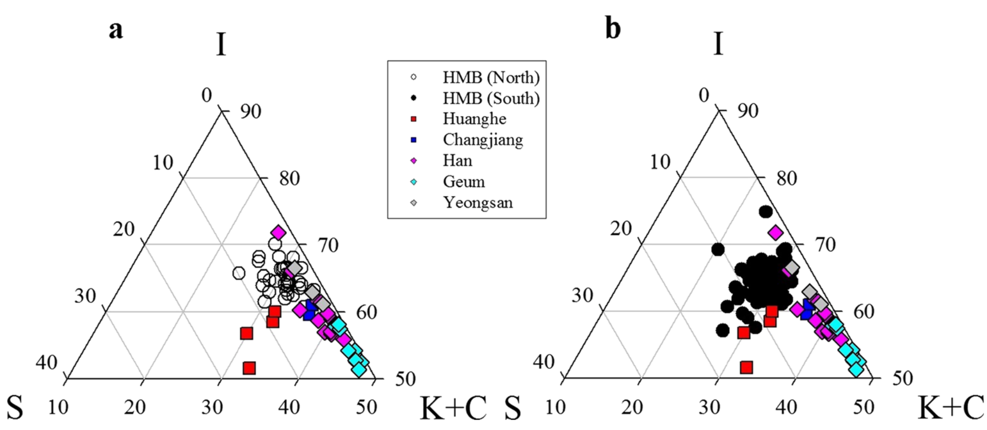


Fig. 2 Ternary diagram of I–S–(K+C) ratios. *I* Illite, *S* smectite, *C* chlorite, *K* kaolinite. **a** Sediments in the northern part of the HMB in comparison with river sediments around the Yellow Sea. **b** Sediments in the southern part of the HMB in comparison with river sediments around the Yellow Sea. For locations of HMB sediment samples, see Fig. 1b

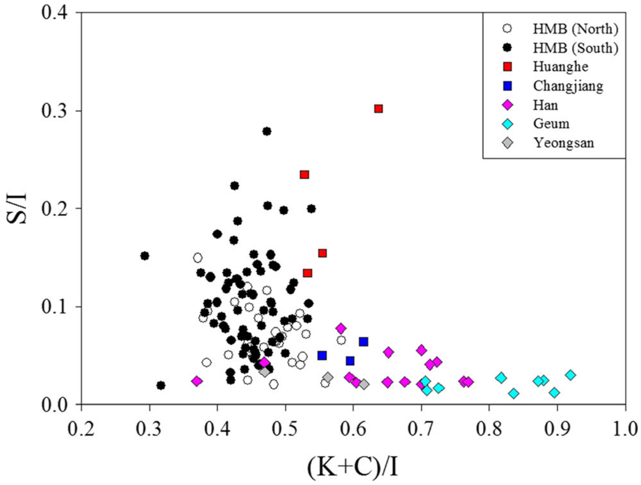


Fig. 3 Crossplot between S/I and (K+C)/I ratios of samples from the northern and southern parts of the HMB in comparison with river sediments from around the Yellow Sea. For locations of HMB sediment samples, see Fig. 1b

assemblages, the order of contribution of the dominant minerals being illite, chlorite, kaolinite, and smectite (Table 3). Whereas the overall distribution patterns of illite and chlorite are similar, those of smectite and kaolinite are reversed (Fig. 4), the former being relatively enriched in the southern part of the HMB, the latter being somewhat more dominant in the northern part.

Discussion

Overall, the results of this investigation are in agreement with those of previous studies (Choi 1981; Qin and Li 1983; Xu 1983; Milliman et al. 1985; Park and Khim 1992; Yang et al. 2003; Choi et al. 2010). The discrimination of the river sediments on the basis of clay minerals is supported by other geochemical properties of the same rivers (Lim et al. 2006; Yang and Youn 2007; Song and Choi 2009). Although various sedimentological, mineralogical and geochemical approaches have been conducted to clarify the provenance of sediments in the HMB (e.g., Park and Khim 1992; Park et al. 2000; Lim et al. 2007), the issue has remained controversial. Many Korean scientists contend that the HMB sediments are mostly supplied by Korean rivers, based on their distribution patterns, as well as sedimentological, geophysical, mineralogical, and geochemical evidence (Chough and Kim 1981; Lee and Chough 1989; Park and Khim 1992; Jin and Chough 1998; Park et al. 2000; Lee and Chu 2001; Chough et al. 2002; Shinn et al. 2007; Moon et al. 2009; Cho et al. 2011; Lee et al. 2014). In contrast, most Chinese scientists, supported by some scientists from Korea, propose that, on the basis of the large sediment budgets and high accumulation rates, as well as other sedimentological, mineralogical, and geochemical evidence, the HMB deposit represents mixtures of sediment from sources including Korean rivers, the Huanghe River and/or the Changjiang River (Schubel et al. 1984; Ren and Shi 1986; Alexander et al. 1991; Lim et al. 2007; Ha et al. 2013). Wei et al. (2000) proposed such a multiple origin, suggesting that the sediments in the northern HMB came from Korean rivers, whereas those in the southern HMB were mixture of materials supplied from different sources.

Clay mineral studies have been widely carried out to identify the origin of the sediment in the Yellow Sea (Chough and Kim 1981; Xu 1983; Milliman et al. 1985; Park and Khim 1992; Choi and Kim 1998; Yang et al. 2003; Yang and Youn 2007; Moon et al. 2009; Choi et al. 2010; Wei et al. 2000; Cho et al. 2011; Shi et al. 2012; Li et al. 2014). All these studies agree that clay minerals in Yellow Sea sediments are terrigenous in origin, being primarily composed of illite (generally more than 60%) with subordinate contributions of chlorite, kaolinite, and smectite. Among the major clay minerals, smectite was regarded as possible indicator on the basis of which sediments from Chinese and Korean rivers could be distinguished, primarily because smectite contents are much lower in Korean river sediments (<5%) as compared to those of the Changjiang (>5%) and Huanghe (>10%) rivers (Table 1), although clay mineral contents also depend on sampling processes, analytical conditions, and calculation methods (Choi et al. 2010). In addition, the distribution patterns of clay minerals show that illite contents increase and smectite contents decrease eastward within the Yellow Sea. On the other hand, Yang et al. (2003) and Shi et al. (2012) report very irregular distribution patterns of clay mineral contents, and therefore expressed doubts about the applicability of smectite for the discrimination between Chinese and Korean river sediments.

The ternary diagram of I–S–(K+C) ratios shows that only Huanghe River sediments plot within the smectite-rich region

Table 2 Clay mineral composition of surficial sediments in the HMB

Illite (%)	Chlorite (%)	Kaolinite (%)	Smectite (%)	References	Remarks
71.0	16.0	14.0	<2.0	Park and Khim (1992)	Core top
69.4–71.0	12.4–13.5	15.5–17.0	0.01–1.1	Cheng (2000)	
<58	>17	>16	7~10	Wei et al. (2000)	South
>58	<17	<16	<7	Wei et al. (2000)	North
55~70	12~18	12~17	<10	Lee and Chu (2001)	Core top
64.7	17.9	11.4	5.9	This study	

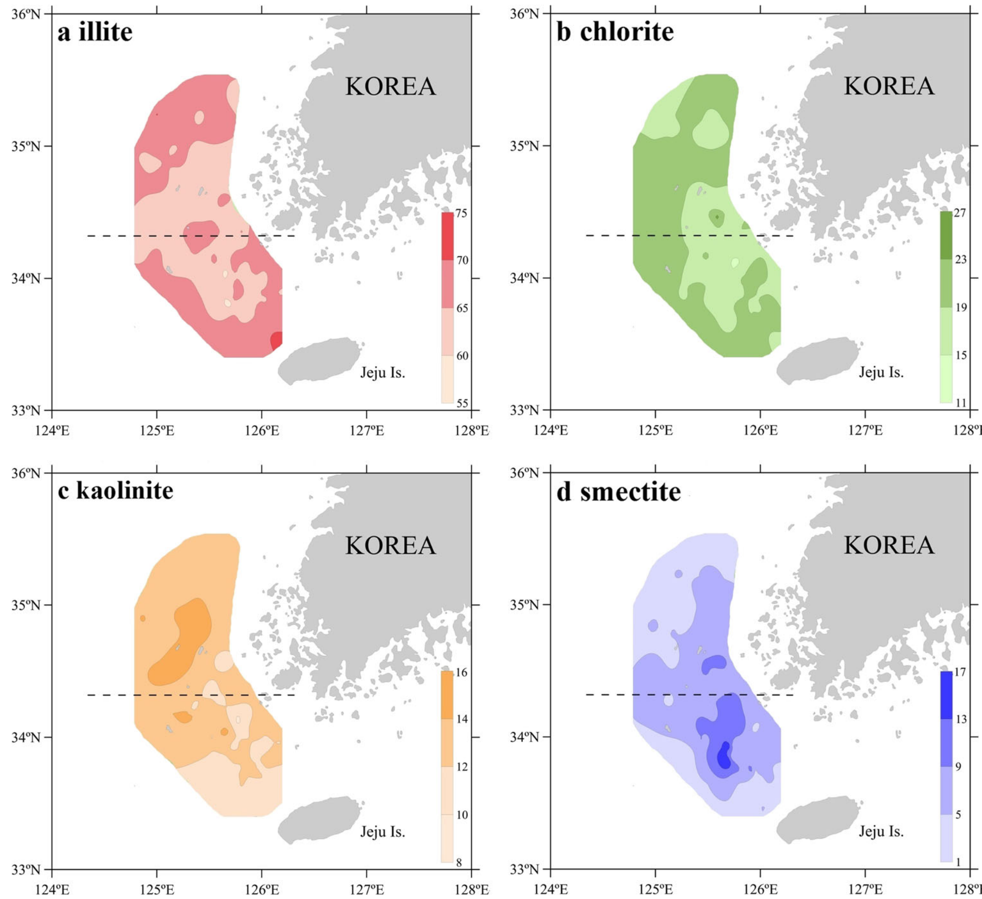


Fig. 4 Distribution patterns of four major clay minerals (illite, chlorite, kaolinite, smectite) in surficial sediments of the HMB

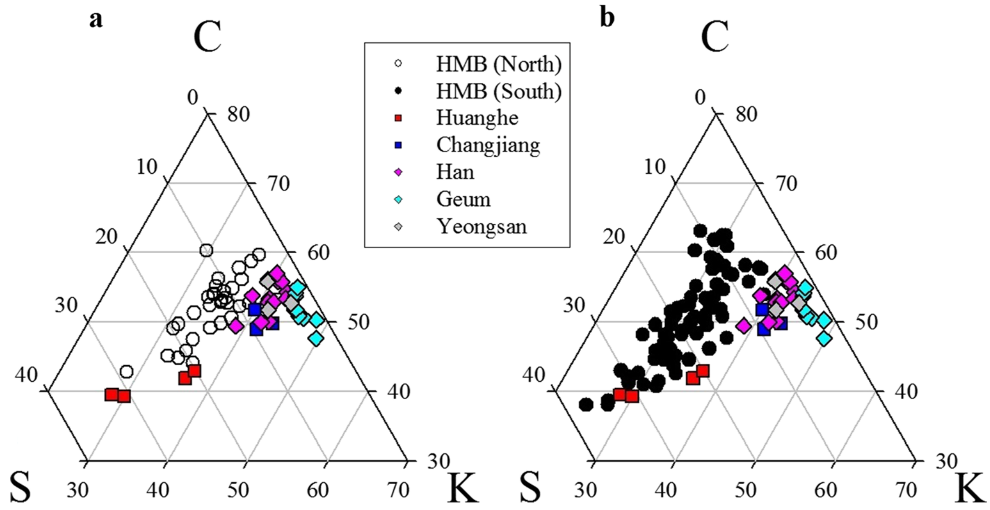
(Fig. 2), whereas most of the samples from the northern part of the HMB plot close to the region representative of Korean rivers and the Changjiang River (Fig. 2). This demonstrates that the northern part of the HMB has a close affinity to Korean rivers and the Changjiang River. This relationship is supported by the crossplot between S/I and (K+C)/I ratios (Fig. 3). In contrast, the ternary diagram illustrating I–S–(K+C) ratios shows that the clay mineral compositions in the southern part of the HMB plot mostly in the area between the Huanghe River and the Korean rivers or Changjiang River (Fig. 2). This can also be observed in the crossplot between S/I and (K+C)/I ratios (Fig. 3). It can be inferred that the southern part of the HMB is composed of sediment mixtures originating from the Huanghe River and Korean rivers or Changjiang River.

The low smectite and high kaolinite contents of the samples from the northern part of the HMB argue strongly in favor of their supply from Korean rivers. By the same token, the high smectite and low kaolinite contents of the samples from the southern part of the HMB favor Korean rivers as the primary source with a subordinate supply from the Huanghe River. Although the Jeju Island consisting of widespread basaltic rocks is a possible source of smectite, the clay mineral distribution of surficial sediments near the island lack smectite, because there is no potential delivery (e.g., rivers) of smectite from Jeju Island to the East China Sea or the Yellow Sea (Yang et al. 2004; Youn 2009). This interpretation is thus in agreement with the recent findings of Wei et al. (2000) based on clay mineralogical analyses. Although the major elements suggest domination by Korean river-derived sediments, rare earth elements in sediments from the southern part of the HMB are closely related to the sediments of Chinese rivers (Ha et al. 2013).

Table 3 Clay mineral compositions of surficial sediments in the northern and southern parts of the HMB (*n* sample size, present study)

	Illite (%)	Chlorite (%)	Kaolinite (%)	Smectite (%)
Northern part of HMB (*n*=32)				
Average	64.9	18.3	12.2	4.6
Minimum	60.7	14.3	9.8	1.3
Maximum	70.1	23.7	14.2	9.8
Southern part of HMB (*n*=69)				
Average	64.6	17.7	11.1	6.5
Minimum	55.9	11.9	8.4	1.4
Maximum	74.8	21.3	13.7	15.9

Fig. 5 Ternary diagram of S–C–K ratios. *S* Smectite, *C* chlorite, *K* kaolinite. **a** Sediments in the northern part of the HMB in comparison with river sediments around the Yellow Sea. **b** Sediments in the southern part of the HMB in comparison with river sediments around the Yellow Sea. For locations of HMB sediment samples, see Fig. 1b



Unfortunately, there is currently no reliable indicator to differentiate between sediments from the Changjiang River and those of Korean rivers. The ternary diagram of S–K–C ratios (Fig. 5), however, may provide a possible clue to distinguish between the two sources. Thus, most Korean river sediments plot above the 50% chlorite line, whereas the Changjiang River sediments plot below this. In general, the samples from the northern part of the HMB plot above the 50% chlorite line, suggesting that the sediments of this part of the HMB originate from Korea. In addition, the surface current regime in the Yellow and East China seas supports this interpretation. Thus, the surface currents in the Yellow Sea are characterized by a basin-wide cyclonic gyre consisting of a northward-flowing warm oceanic current (Yellow Sea Warm Current) and a southward-flowing coastal current (Jiangsu Coastal Current or Yellow Sea Coastal Current; Beardsley et al. 1985; Hu and Li 1993). This cyclonic gyre probably plays an important role in transporting sediments into the HMB.

During the summer, the northward-flowing Taiwan Warm Current is influenced by the discharge of the Changjiang River, resulting in the formation of the Changjiang diluted water (CDW) off the river mouth. However, most of the Changjiang sediments are transported southward along the coast into the East China Sea (Alexander et al. 1991; Dong et al. 2011). During peak floods, however, the CDW extends southeastward up to Jeju Island (Guan 1994). Although some studies suggest that the turbid water from the Changjiang estuary may be dispersed far enough to mix with the waters of the South Sea of Korea around Jeju Island (Lee et al. 1998; Ahn et al. 1999), it is more commonly assumed that Changjiang-derived sediments can generally not escape the estuarine trapping mechanism to extend eastward beyond 123°E (Hu and Li 1993; Zhang 1999; Gao et al. 2000). Thus, because Changjiang River sediments are unlikely to be transported into the HMB, the slightly higher smectite content of the southern part of the HMB probably reflects the influence of Huanghe River sediments.

In addition, the Yellow Sea is characterized by a semi-diurnal tidal regime with tidal ranges exceeding 4 m near the coast (Choi 1980). The long axes of the tidal ellipses are oriented NE–SW in the mid-eastern Yellow Sea, and N–S to NW–SE in the southeastern Yellow Sea (Larsen et al. 1985; Lee and Jung 1999). With current velocities of 40–100 cm/s, the tidal flow is sufficiently strong to cause local resuspension and/or bedload transport in dependence of grain size, and should hence also play an important role in the sediment dynamics of the HMB (Sternberg et al. 1985).

Conclusions

Based on clay mineral studies of river sediments from around the Yellow Sea and surficial sediments in the HMB, the following conclusions can be drawn:

1. Korean and Chinese river sediments are composed of illite, chlorite, kaolinite, and smectite in descending order. Smectite is relatively high in Huanghe River sediments, whereas kaolinite and chlorite are relatively high in Korean river sediments.
2. Fine-grained sediments in the HMB are also dominated by illite, whereas chlorite and kaolinite are subordinate components and smectite is the smallest contributor, the latter being slightly higher in the southern part of the HMB compared to the northern part.
3. Sediments of the northern part of the HMB essentially originate from Korean rivers, whereas those of the southern part are predominantly derived from Korean rivers with minor contributions from the Huanghe River in China, thus confirming the tentative interpretation of Wei et al. (2000) based on a more limited dataset.

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Conflict of interest The authors declare that there is no conflict of interest with third parties.

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