Detrital fine-grained sediment contribution from Taiwan to the northern South China Sea and its relation to regional ocean circulation

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\textbf{A R T I C L E  I N F O}

\textbf{Article history:}
Received 6 November 2007
Received in revised form 3 July 2008
Accepted 5 August 2008

\textbf{Keywords:}
clay minerals
surface sediments
provenance
South China Sea
Taiwan

\textbf{A B S T R A C T}

Results of clay mineralogy in 140 samples collected in major rivers and lakes in southwestern Taiwan and on the seafloor off Taiwan in the northeastern South China Sea (SCS), combined with clay mineral records of the Pearl River drainage basin, rivers in Luzon, and the South China shelf and slope, are used to semi-quantitatively evaluate the detrital fine-grained sediment contribution of Taiwan to the northern SCS. The clay mineral assemblage of the Taiwan-sourced sediments consists dominantly of illite (average 56%) and chlorite (41%), with very scarce kaolinite and smectite. Their respective distribution from the rivers and lakes to the seafloor off Taiwan does not show obvious basin-wide differences. Linear correlations of illite chemistry index with illite crystallinity and of illite crystallinity with kaolinite (%) present two end-members of provenances, the Pearl River and Taiwan, for the South China shelf and slope. Assuming that kaolinite in the northern SCS is provided completely from the Pearl River, the contribution of Taiwan in clay minerals is evaluated as 29% to the South China shelf and 23% to the South China slope, respectively. Accordingly, the contribution of the Pearl River to the South China shelf and slope is 52% and 31%, respectively. The Luzon Arc accounts for the rest of clay mineral components for the northern SCS mainly by providing smectite. The Bashi Strait-crossed branches of the southward deep North Pacific Deep Water and the northward surface Kuroshio Current in the western Pacific may transport Taiwan-sourced suspended sediments westwards to the northern SCS.

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\section{1. Introduction}

Determining the provenance of detrital sediments in the ocean is prerequisite for understanding environmental and climatic conditions that occurred in land-source areas. Sources of terrigenous matters in marginal seas with bounded lands are particularly complicated because they may involve diverse geological settings. One of such cases is the South China Sea (SCS), the largest marginal sea in the world oceans (Milliman and Syvitski, 1992). The river-borne terrigenous sediments compose ~80% of total SCS surface sediments (Huang, 2004) and have formed high sedimentation-rate deposition in the geological past (Wang et al., 2000), especially for sediment drifts on the northern slope, where the Holocene sedimentation rate can reach as high as 70 cm/kyr (Bühring et al., 2004). The terrigenous detrital sedimentary archives in the SCS, therefore, have become ideal to study the high-resolution history of the East Asian monsoon evolution (e.g., Wang et al., 1999; Wehausen and Brumsack, 2002; Clift et al., 2002; Liu et al., 2003, 2005; Wang et al., 2005; Boulay et al., 2005; Wan et al., 2007).

The northern SCS has offered a special attraction for marine geologists because of the very high sedimentation rate of its detrital sediments (e.g., Wang et al., 2000). However, its sedimentary source is not well established because the geology of two major potential source areas, South China and southwest Taiwan, is similar with same cratonic Mesozoic–Cenozoic sedimentary rocks (Commission for the Geological Map of the World, 1975) and Nd isotopic values (Li et al., 2003; Boulay et al., 2005). Most of previous studies mainly at Ocean
Drilling Program (ODP) Leg 184 sites (partly shown in Fig. 1A) on the northern slope of the SCS considered the Pearl River as the main sedimentary source (Wehausen and Brumsack, 2002; Clift et al., 2002; Tamburini et al., 2003; Li et al., 2003; Boulay et al., 2003) with an additional volcanic source (Boulay et al., 2005). But Taiwan presents one of the highest erosion rates in the world and exports 384 Mt/yr of suspended sediments to the ocean, with about half of the total sediments directly to the SCS (Dadson et al., 2003). Such a fine-grained sediment discharge is even larger than any other provenance surrounding the SCS including the Pearl, Red, and Mekong rivers (Table 1) and must have played a significant role in sediment components in the SCS, especially on the northern slope.

One way to distinguish the fine-grained sediments of South China from Taiwan is to use clay mineralogy of the sediments because clay minerals in river and marine sediments reflect the intensity of chemical weathering of their source areas (Chamley, 1989). In South China, the more or less flat hinterland of the Pearl River drainage basin has undergone deep and long-term lateritic weathering during Neogene. The river sediments with kaolinite and Al-rich illite dominance indicate a strong chemical weathering (Liu et al., 2007a,b). In Taiwan, steep erosional gradients, short transport distances, and short storage times have significantly increased the intensity of physical weathering. The sediments present only a moderate chemical weathering on the basis of major and trace element geochemistry (Selvaraj and Chen, 2007), indicating a strong physical erosion; ratios above 0.5 are found in Al-rich illite (biotite, mica), which are characteristic of physical erosion; ratios above 0.5 are found in Al-rich illite (muscovite), which are released following strong hydrolysis (Esquevin, 1969; Gingele et al., 1998). Illite crystallinity was obtained from the glycolated curve. Illite chemistry index refers to a ratio of the 5 Å and 10 Å peaks at around 5 Å. Lower values represent the higher crystallinity, characteristic of weak hydrolysis in continental sources and arid and cold climate conditions (Chamley, 1989; Krumm and Buggisch, 1991; Ehrmann, 1998).

In order to determine the provenance of detrital fine-grained sediments in the northern SCS, literature data of clay minerals in 38 surface samples from the Pearl River drainage basin (Liu et al., 2007a,b), 16 surface samples from the South China shelf (Boulay et al., 2004), and 9 surface samples from the South China slope (Boulay et al., 2004) evaluate the detrital fine-grained sediment contribution of Taiwan to the northern SCS.

2. Materials and methods

A set of 140 samples from southwestern Taiwan and off Taiwan in the northeastern SCS (19 from rivers, 5 from lakes, and 116 from the seafloor) are examined for clay mineralogy (Fig. 1B; C; Supplementary data Table S1). River and lake samples, covering with all major mountainous rivers and lakes in southwestern Taiwan, were collected from surface muddy channel/bed deposits to avoid contamination from bank sediments. The seafloor samples with different tectonic settings, including 11 from the SW Taiwan shelf (tectonically stable), 87 from the accretionary prism east of the Manila trench (tectonically active), and 18 from the passive continental margin west of the Manila trench (tectonically stable), were selected from surface sediments or top parts of box and piston cores, taken by the R/V Ocean Researcher I & III during 2003–2006. In order to compare with clay mineral compositions derived from Taiwan, 7 samples from three largest rivers in Luzon, northern Philippines (3 from the Cagayan River, 2 from the Pamrnga River, and 2 from the Agno River) were collected during the summer season of 2007 (Fig. 1A, Supplementary data Table S1). All samples were analyzed for clay minerals.

Clay minerals were identified by X-ray diffraction (XRD) using a PANalytical diffractometer at the Laboratoire IDES, Université de Paris XI on oriented mounts of non-calcareous clay-sized (~2 µm) particles (Holtzapffel, 1985). The oriented mounts were obtained following the methods described in detail by Liu et al. (2004). Three XRD runs were performed, following air-drying, ethylene-glycol solvation for 24 h, and heating at 490 °C for 2 h. Identification of clay minerals was made mainly according to the position of the (001) series of basal reflections on the three XRD diagrams. Semi-quantitative estimates of peak areas of the basal reflections for the main clay mineral groups of smectite (including mixed-layers) (15–17 Å), illite (10 Å), and kaolinite/chlorite (7 Å) were carried out on the glycolated curve (Holtzapffel, 1985) using the MacDiff software (Petschick, 2000). Relative proportions of kaolinite and chlorite were determined based on the ratio from the 3.57/3.54 Å peak areas. Following the laboratory routine at both Laboratoire IDES of Université de Paris XI and Laboratoire PBDS of Université de Lille I (Liu et al., 2003, 2007b), the weighting factors introduced by Biscaye (1965) are not used when generating relative weight percentages of each clay mineral. Replicate analyses of a few selected samples gave a precision of ±2% (2σ). Based upon the XRD method, the semi-quantitative evaluation of each clay mineral has an accuracy of ~5%. Additionally, some mineralogical characters of illite were determined on the glycolated curve. Illite chemistry index refers to a ratio of the 5 Å and 10 Å peak areas. Ratios below 0.5 represent Fe–Mg-rich illite (biotite, mica), which are characteristic of physical erosion; ratios above 0.5 are found in Al-rich illite (muscovite), which are released following strong hydrolysis (Esquevin, 1969; Gingele et al., 1998). Illite crystallinity was obtained from half height width of the 10 Å peak. Lower values represent the higher crystallinity, characteristic of weak hydrolysis in continental sources and arid and cold climate conditions (Chamley, 1989; Krumm and Buggisch, 1991; Ehrmann, 1998).

In order to determine the provenance of detrital fine-grained sediments in the northern SCS, literature data of clay minerals in 38 surface samples from the Pearl River drainage basin (Liu et al., 2007a,b), 16 surface samples from the South China shelf (Boulay et al., 2004), and 9 surface samples from the South China slope (Boulay et al., 2004)
3. Results

The clay mineral assemblages in rivers and lakes of southwestern Taiwan are similar, predominantly consisting of illite (44–66%), and chlorite (33–48%), with very scarce kaolinite (0–4%) and smectite (0–8%) (Supplementary data Table S1). Their average percentage is 55% for illite, 43% for chlorite, 1% for kaolinite, and 1% for smectite, respectively (Table 2, Fig. 2). Clay minerals on the southwestern Taiwan shelf contain only illite (57–61%, average 59%) and chlorite (39–43%, average 41%), without measurable kaolinite and smectite (Fig. 2). The accretionary prism east of the Manila trench is characteristic of dominating illite (49–64%, average 56%) and chlorite (35–42%, average 39%), with scarce kaolinite (0–5%, average 2%) and smectite (0–15%, average 3%) (Fig. 2). The passive continental margin west of the Manila trench has a similar clay mineral distribution with the accretionary prism, with slight increase in smectite (2%) and decrease in illite (2%), respectively (Table 2, Fig. 2). To sum up, the results of all samples from the rivers and lakes in southwestern Taiwan to the shelf, accretionary prism and passive continental margin on the seafloor off Taiwan present dominating illite (average 56%) and chlorite (41%), with very scarce kaolinite and smectite (Fig. 2). In addition, the illite chemistry index (0.26–0.40) and illite crystallinity (0.12–0.22°Δ2θ) also indicate concentrative values at 0.36 and 0.18°Δ2θ, respectively (Supplementary data Table S1, Table 2).

For three Luzon rivers, all samples present predominant smectite (83–92%, average 88%), with minor kaolinite (average 9%) and scarce chlorite and illite (Supplementary data Table S1). There is no obvious difference in clay mineral distribution among various river drainage basins in Luzon, suggesting the major contribution of smectite from Luzon to the SCS.

4. Discussion

The similar clay mineral distribution from southwest Taiwan to the northeastern SCS (Fig. 2) suggests a provenance-controlled consistency in the clay mineral assemblage. Both the active accretionary prism east of the Manila trench and the passive continental margin west of the Manila trench receive illite and chlorite all from southwestern Taiwan, not depending on their tectonic settings. The dominating illite and chlorite results are generally in accordance with a few literature data not depending on their tectonic settings. The dominating illite and chlorite results from the ones in the Pearl River drainage basin, in the Luzon rivers, and on the shelf and slope of South China (Fig. 3). The Taiwan-sourced clays are chiefly illite (average 56%) and chlorite (41%) with scarce kaolinite and smectite, the Pearl River sediments consists mainly of kaolinite (46%), illite (26%), and chlorite (25%) with scarce smectite (3%) (Liu et al., 2007a,b), whereas clays from the Luzon rivers are principally smectite (88%) with minor kaolinite (9%) and scarce chlorite and illite (Table 2). The South China shelf and slope sediments contain abundant smectite (19–46%), illite (26–37%), and chlorite (19–

![Image](https://example.com/image.png)

**Fig. 2.** Distribution of average clay mineral assemblages in lakes and rivers in southwestern Taiwan and on the shelf, accretionary prism east of the Manila trench, and passive continental margin west of the Manila trench off Taiwan. See **Fig. 1** for their geographic locations and Supplementary data Table S1 for detailed GPS positions and clay mineral proportions. N, number of surface samples.

**Table 2**

<table>
<thead>
<tr>
<th>Region</th>
<th>Sample number</th>
<th>Chlorite (%)</th>
<th>Illite (%)</th>
<th>Smectite (%)</th>
<th>Kaolinite (%)</th>
<th>Illite chemistry</th>
<th>Illite crystallinity (°Δ2θ)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SW Taiwan rivers</td>
<td>19</td>
<td>43</td>
<td>55</td>
<td>1</td>
<td>1</td>
<td>0.33</td>
<td>0.16</td>
</tr>
<tr>
<td>SW Taiwan lakes</td>
<td>5</td>
<td>42</td>
<td>55</td>
<td>1</td>
<td>2</td>
<td>0.38</td>
<td>0.18</td>
</tr>
<tr>
<td>SW Taiwan shelf</td>
<td>11</td>
<td>41</td>
<td>59</td>
<td>0</td>
<td>0</td>
<td>0.36</td>
<td>0.16</td>
</tr>
<tr>
<td>E Manila trench</td>
<td>87</td>
<td>39</td>
<td>56</td>
<td>3</td>
<td>2</td>
<td>0.35</td>
<td>0.16</td>
</tr>
<tr>
<td>W Manila trench</td>
<td>18</td>
<td>39</td>
<td>54</td>
<td>5</td>
<td>2</td>
<td>0.35</td>
<td>0.16</td>
</tr>
<tr>
<td>Pearl River</td>
<td>38</td>
<td>25</td>
<td>26</td>
<td>3</td>
<td>46</td>
<td>0.62</td>
<td>0.30</td>
</tr>
<tr>
<td>Luzon rivers</td>
<td>7</td>
<td>2</td>
<td>1</td>
<td>88</td>
<td>9</td>
<td>/</td>
<td>/</td>
</tr>
<tr>
<td>S China shelf</td>
<td>16</td>
<td>30</td>
<td>37</td>
<td>19</td>
<td>13</td>
<td>0.54</td>
<td>0.24</td>
</tr>
<tr>
<td>S China slope</td>
<td>7</td>
<td>19</td>
<td>26</td>
<td>46</td>
<td>9</td>
<td>0.51</td>
<td>0.23</td>
</tr>
</tbody>
</table>

Illite chemistry index and illite crystallinity of the Luzon river samples were not measurable because their illite is very scarce. Data of the Pearl River from Liu et al. (2007a,b); data of the shelf and slope of South China from Liu et al. (2003) and Boulay et al. (2004, 2005).
30%) with minor kaolinite (9–13%), with increasing smectite and decreasing illite and chlorite seawards (Liu et al., 2003; Boulay et al., 2004, 2005). The physical segregation during sedimentation of clay minerals is not remarkable and can’t play a major impact on their decrease illite and chlorite seawards (Liu et al., 2003; Boulay et al., 2004, 2005). Therefore, these variations indicate that Taiwan discharges sediments characterized by abundant illite and chlorite and no kaolinite and smectite to the northern SCS. Instead, the Pearl River provides abundant kaolinite and no smectite, and Luzon provides mainly smectite to the sea.

Illite and chlorite on the shelf and slope of South China may also be derived from the Pearl River, where average illite and chlorite contents are 26% and 25%, respectively (Table 2). But the illite contents on the shelf (37%) and slope (26%) are higher than or equal to the Pearl River values (Table 2), precluding the Pearl River as a unique major source for illite. Because illite and chlorite in the SCS are considered to have same provenance, the Pearl River and Taiwan are estimated to be the two major potential source areas, the Pearl River and Taiwan sources (Table 2), we here further determine the source of illite in the northern SCS to account for the provenance of combined illite and chlorite. Illite chemistry index and illite crystallinity of all mentioned samples are employed, because the two indicators present a large volume of basalt is present in the volcanic Luzon Arc (Commission for the Geological Map of the World, 1975) and could represent, by weathering, a huge amount of smectite with average 88% in rivers of Luzon. The Luzon provenance, therefore, is suggested to account for all smectite contribution in sediments on the South China shelf and slope (19% for the South China shelf and 46% for the South China slope, Table 2). Assuming that kaolinite in the northern SCS is provided completely from the Pearl River, a linear correlation of illite crystallinity with kaolinite (%) is observed for all surface sediments, with two end-members of the Pearl River and Taiwan sources and transitional South China shelf and slope sediments (Fig. 5). Taking into account the kaolinite contents (Table 2), the combined illite and chlorite contribution ratios, and the unique smectite source of Luzon, we estimate the contributions of clay minerals as 52% from the Pearl River and 29% from Taiwan to the South China shelf, and as 31% from the Pearl River and 23% from Taiwan to the South China slope (Figs. 5 and 6).

River and an increase contribution from southwestern Taiwan. In consideration of average illite crystallinity values of 0.16°Δθ for the Pearl River samples, 0.23°Δθ for the South China slope samples, 0.24°Δθ for the South China shelf samples, and 0.30°Δθ for the Pearl River samples (Table 2), we approximately calculate the combined illite and chlorite contribution ratio of southwestern Taiwan to the Pearl River as 3:4 for the South China shelf and as 1:1 for the South China slope (Fig. 4).

A large volume of basalt is present in the volcanic Luzon Arc (Commission for the Geological Map of the World, 1975) and could represent, by weathering, a huge amount of smectite with average 88% in rivers of Luzon. The Luzon provenance, therefore, is suggested to account for all smectite contribution in sediments on the South China shelf and slope (19% for the South China shelf and 46% for the South China slope, Table 2). Assuming that kaolinite in the northern SCS is provided completely from the Pearl River, a linear correlation of illite crystallinity with kaolinite (%) is observed for all surface sediments, with two end-members of the Pearl River and Taiwan sources and transitional South China shelf and slope sediments (Fig. 5). Taking into account the kaolinite contents (Table 2), the combined illite and chlorite contribution ratios, and the unique smectite source of Luzon, we estimate the contributions of clay minerals as 52% from the Pearl River and 29% from Taiwan to the South China shelf, and as 31% from the Pearl River and 23% from Taiwan to the South China slope (Figs. 5 and 6).

Fig. 3. Comparison of clay mineral assemblages among southwestern Taiwan, the Pearl River, Luzon, and the northern SCS. Data of the Pearl River from Liu et al. (2007a,b), data of the shelf and slope of South China from Liu et al. (2003) and Boulay et al. (2004, 2005).

Fig. 4. Correlations of illite chemistry index with illite crystallinity of surface sediments in southwestern Taiwan, the Pearl River, and the northern SCS. The coarse dash line shows a linear correlation between illite chemistry index and illite crystallinity.

Fig. 5. Correlations of illite crystallinity with kaolinite (%) of surface sediments in southwestern Taiwan, the Pearl River, and the northern SCS. The coarse dash line shows a linear correlation between illite crystallinity and kaolinite content.
Thus, the Pearl River contributes approximately half of its clay minerals to the South China shelf and only one third of clay minerals to the slope, whereas southwestern Taiwan contributes about one fourth of its clay minerals to both the shelf and slope (Fig. 6). The results have raised a question: how are the Taiwan-sourced clays delivered to the South China shelf and slope with a much higher percentage than previously thought? As the westward branches of the North Pacific Deep Water (NPDW) and the Kuroshio Current (KC) in the western Pacific cross the Bashi Strait, they may transport the Taiwan-sourced sediments to the northern SCS. Lüdmann et al. (2005) suggested a branch of the southward deep NPDW crossing the Bashi Strait and carrying eastern and southern Taiwan-sourced re-suspended fine sediments into the northern SCS (Fig. 6). The westward deep currents are then pushed upslope at the southeast slope of the Dongsha Islands and release sediments to form high sedimentation-rate drifts on the South China slope (Lüdmann et al., 2005). Instead, Shao et al. (2007) considered the bottom currents originated also from the branch of NPDW but moving southwestwards along the slope to form high sedimentation-rate drifts on sides of NE–SW bottom channels. Recently, high-resolution seismic data revealed that the high sedimentation-rate drifts are actually composed of a series of sediment waves that have migrated upslope (Zhong et al., 2007), confirming the existence of westward deep currents in front of the South China slope. Despite the absence of observed deep-sea current data, we accept the assumption of the deep-sea currents (Lüdmann et al., 2005) that carry the Taiwan-sourced sediments to the South China slope. In addition, a branch of the northward surface Kuroshio Current (KC) may also transport suspended Taiwan-sourced sediments westwards when it enters into the SCS through the Bashi Strait (Caruso et al., 2006). The current is also one of the most important carriers accounting for smectite contribution to the northern SCS from the Luzon Arc and the western Philippine Sea (Wan et al., 2007).

5. Conclusions

Our results indicate that the clay mineral assemblage of surface sediments in major rivers and lakes in southwestern Taiwan and on the seafloor off Taiwan in the northeastern SCS consists dominantly of illite (average 56%) and chlorite (41%), with very scarce kaolinite and smectite. Their respective distribution from the rivers and lakes to the shelf, accretionary prism, and passive continental margin on the seafloor off Taiwan does not show obvious basin-wide differences. By combining with clay mineral records of the Pearl River drainage basin, the Luzon rivers, and the South China shelf and slope and assuming that kaolinite in the northern SCS is provided completely from the Pearl River, the linear correlations of illite chemistry index with illite crystallinity and of illite crystallinity with kaolinite (%) are observed for all surface sediments, with two end-members of the Pearl River and Taiwan sources and transitional South China shelf and slope sediments. The contribution of Taiwan in clay minerals is semi-quantitatively evaluated as 29% to the South China shelf and 23% to the South China slope, respectively. Accordingly, the contribution of the Pearl River to the South China shelf and slope is 52% and 31%, respectively. The Luzon Arc accounts for the rest of clay mineral components for the northern SCS mainly by providing smectite. The Bashi Strait-crossed branches of the southward deep NPDW and the northward surface KC may transport Taiwan-sourced suspended sediments westwards to the northern SCS.

Acknowledgments

We thank the Central Geological Survey of Taiwan to provide samples off SW Taiwan for this study, and Hailing Ren and Hui-Ling Lin for assistance in collecting the samples. We specially thank Gert J. De Lange and two anonymous reviewers for their constructive reviews on the early version of this paper. This study was supported by the National Basic Research Program of China (2007CB815906), the National Natural Science Foundation of China (40776027 and 40621063), the Shanghai Rising-Star Program (07QH14014), the Shanghai Shuguang Program (07SG23), the Fok Ying Tung Education Foundation (101018), and the Doctoral Program of Higher Education of the Ministry of Education of China (20060247032) to Z. Liu. Partial funding for this study was