Modern (<100 years) sedimentation in the Taiwan Strait: Rates and source-to-sink pathways elucidated from radionuclides and particle size distribution

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A B S T R A C T

A large number of sediment cores collected during 2005–2010 from the Taiwan Strait were analyzed for radionuclides ($^{210}$Pb, $^{137}$Cs and $^7$Be) to elucidate sedimentation dynamics in this all-important gateway linking two largest marginal seas in the western Pacific (namely, the South China Sea and the East China Sea). Apparent sediment accumulation rates derived from $^{210}$Pb and $^{137}$Cs profiles vary from <0.1 to >2 cm/yr, averaging ~0.4 cm/yr and showing a spatial pattern closely related to hydrodynamics and sediment source-to-sink pathways. Spatial-temporal variation of $^7$Be activity in surface sediments off Taiwan’s west coast indicates episodic deposition of flood layers and their mobility from river estuaries toward the north. In conjunction with particle size distribution in surface sediments and the structure of sediment strata revealed by sub-bottom echo images; the radionuclide data can be used to outline three different sediment source-to-sink dispersal systems. Based on sediment loads of surrounding rivers and the distribution of sediment accumulation rates, lateral transport is required to account for the budget and size distribution of sediments in the strait.

1. Introduction

The Taiwan Strait (TS) is bounded by Taiwan, mainland China, the South China Sea (SCS) and the East China Sea (ECS) on its east, west, south and north, respectively (Fig. 1). About 350 km in length, 180 km in average width and 60 m in average depth, the strait connects two largest marginal seas in the western Pacific and receives sediments from the largest continent and a high-standing mountainous island known to have the highest erosion rate in the world (Li, 1976; Dadson et al., 2003; Huh et al., 2009a). Hydrodynamics and sedimentation processes in such a crucial setting are conceivably complex and play important roles in regulating the transport of water as well as sediments through it.

While circulation patterns in the TS have been fairly well studied and documented in the past 1–2 decades (see next section), sedimentation dynamics in the strait remains poorly understood by comparison. There is no doubt that $^{210}$Pb is the most commonly used and almost indispensable tool to date nearshore sediments. However, such data are virtually not available for the northern TS. Although a fairly comprehensive dataset has been established off southwestern Taiwan around the Gaoping submarine canyon recently (Huh et al., 2009a, b), there is as yet no report in the open literature of $^{210}$Pb-based sediment accumulation rates to the north of the Penghu Channel.

Published sediment accumulation rates in the TS were largely based on $^{14}$C dating of suitable carbon-bearing material found in sediment cores (Chen and Covey, 1983; Xu et al., 1989, 1990; Lan et al., 1993) plus estimates based on paleomagnetism (Chien and Leu, 1984) and a two-component (terrigenous vs. pelagic) $^{10}$Be mixing model (Lee et al., 1993). Rates from these methods are generally more than one order of magnitude lower than $^{210}$Pb- and $^{137}$Cs-based rates obtained in this work. As will be discussed later, such a discrepancy can be ascribed to the vastly different timescales involved in various methods and associated changes in environmental and sedimentation conditions with time.

The purpose of this work is to elucidate the sources, pathways and accumulation rates of sediments in the TS from seasonal to centennial timescales. Radionuclides ($^7$Be, $^{137}$Cs, $^{210}$Pb and $^{234}$Th) with different source functions and half-lives were used as tracers toward reaching this goal. Grain-size distribution in surface sediments was used to provide an additional constraint on sediment transport under the influence of hydrodynamic processes. Radionuclide and Grain-size data were integrated with sub-bottom echo images of relevant
sediment strata and their spatial distribution and variation in thickness to delineate sediment source-to-sink (S2S) pathways in the TS. Finally, from $^{137}$Cs- and $^{210}$Pb-derived sediment accumulation rates, a preliminary sediment budget in the TS was calculated and compared with sediment loads of adjoining rivers to assess sediment mass balance (or imbalance) on decadal to centennial timescales.

2. The oceanographic setting—bathymetry and circulation patterns

The bathymetry in the TS (Fig. 1) is characterized by several salient topographic features. The deepest part of the TS is the Penghu Channel (PHC) in the southeast. Surrounded by Taiwan (in the east), the Chan-Yuen Rise (in the north), the Penghu Archipelago (in the west) and the Taiwan Shoal (in the southwest); the funnel-shaped Penghu Channel is the most important gateway through which the Kuroshio Branch Current (KBC) derived from the Luzon Strait (also called Bashi Strait) flows into the TS (Jan et al., 2002). The Taiwan Shoal in the southwest is a shallow and barren sand bank (averaging 20 m in depth) lying between the rest of the TS and the South China Sea (in the south). The Chan-Yuen Rise (CYR) in the middle reaches of the TS is a topographic high impeding circulation between the Wu-Chiu Depression (WCD) in the southwest and the Kuan-Yin Depression (KYD) in the northeast. Constituted by a complex of sand bodies, the Chan-Yuen Rise can be divided by an intermediate swale into two parts: the alongshore-trending eastern CYR and the cross-shore trending western CYR. Except the Penghu Channel, the Wu-Chiu Depression and the Kuan-Yin Depression are the deepest basins in the TS; they collectively account for much of the strait’s area.

Fig. 1. Map showing the bathymetry of the Taiwan Strait and rivers on both sides of the strait. Western Taiwan rivers discharging directly into the strait are (from north to south): (1) Tanshui, (2) Touchien, (3) Chungkang, (4) Houlong, (5) Taan, (6) Tachia, (7) Wu, (8) Choshui, (9) Peikang, (10) Potzu, (11) Pachang, (12) Chishih, (13) Tsengwen, (14) Yenshui, (15) Erhjen and (16) Arkongtien Rivers. Sediment loads of these rivers are listed in Table 1. PHC: Penghu Channel, CYR: Chan-Yuen Rise, KYD: Kuan-Yin Depression, WCD: Wu-Chiu Depression, ECS: East China Sea, SCS: South China Sea. The inset in the upper left corner shows the mapped area (red rectangle) in the western Pacific.

As the year-round Kuroshio Branch Current (plus the South China Sea Current in the summer and fall seasons) is funneled through the Penghu Channel, the northbound current bifurcates in front of the Chan-Yuen Rise, with a deeper component turning northwestern around the western CYR and a shallow component crossing the swale between the eastern CYR and the western CYR (Fig. 2; also see Fig. 2 in Liao et al., 2008). The deep, western branch of KBC then flows along the eastern edge of the Wu-Chiu Depression, turning northeastward into the Kuan-Yin Depression at ~24.5°N to merge with the shallow, eastern branch of KBC behind the Chan-Yuen Rise. This combined current, commonly referred to as the Taiwan Warm Current (TWC), flows over the Kuan-Yin Depression and exits the TS in its north. The volume transport of the Taiwan Warm Current is the largest at the peak of the summer (southwest) monsoon and the lowest at the peak of the winter (northeast) monsoon (Jan et al., 2006).

On the western side of the strait is the southbound China Coast Current (CCC). Since the Chinese Coast Current and the Taiwan Warm Current flow in opposite directions, their waxing and waning are therefore opposite to each other in response to the annual cycle of summer and winter monsoons. When the southwest monsoon peaks in the summer, the Chinese Coast Current is virtually nonexistent in the TS (Fig. 2c). Conversely, when the northeast monsoon prevails in the wintertime (Fig. 2a), the Chinese Coast Current may extend southward to the middle reaches of the TS, with a part obstructed by the Cham-Yuen Rise and turning back northward to form a cyclonic eddy in the Kuan-Yin Depression (Jan et al., 2002; 2004; Lee and Chao, 2003; Wu et al., 2007).

Although the northeast monsoon is much stronger and lasts longer (September to May) than the southwest monsoon (June to August), wind forcing is generally less influential than remote
forcing via pressure gradient which drives northward flow through the TS constantly (Jan et al., 2002). Analyzing shipboard ADCP data collected during 1999–2001, Wang et al. (2003) obtained a net transport of 1.8 Sv \((Sv = 10^6 \text{ m}^3 \text{ s}^{-1})\) from the TS to the East China Sea. Based on a numerical modeling, however, Wu et al. (2007) reported a much smaller northward mean volume transport of \(~1\) Sv through the TS from 2000 to 2003, and it was argued that shipboard data would lead to an overestimate of northward transport due to a lack of wintertime observations.

Apart from circulation of water masses (i.e., mean current flows) at greater spatial and temporal scales, tidal currents also play an important role in the hydrodynamics as well as sedimentation dynamics in estuarine, coastal and shelf environments. Among tides of various frequencies, the semidiurnal \(M_2\) tide is by far the most predominant one in the TS (Wang et al., 2003; Jan et al., 2004). Fig. 3 shows \(M_2\) tidal current ellipses in the TS derived by Wang et al. (2003) based on shipboard ADCP observations during 1999–2001. In the eastern TS, the motions of \(M_2\) tides are generally
alongshore and of reversing-type, with the strongest flows found along the Penghu Channel and off the northwestern coast of Taiwan; from there the sizes of M₂ tidal current ellipses decrease while the tidal levels increase toward the middle of the TS (Hu et al., 2010). In the western side of the TS, especially the northwestern sector of the strait, the tidal ellipses are more rounded than those in the eastern TS, reflecting rotating-type motions which are more favorable for the accumulation of sediments. As will be discussed later, the shapes and sizes of tidal ellipses and their spatial variation have important bearings on the transport, deposition, size distribution, and hydraulic sorting of sediment particles.

3. Methods

3.1. Sampling

Sediment samples used for this work were collected from 16 cruises during 2005–2010 onboard R/V Ocean Researcher-I (OR1) and Ocean Researcher-II (OR2). The sampling sites are shown in Fig. 4. For radionuclide analyses, a total of 123 gravity cores (up to 2 m in length) and 57 box cores (up to 60 cm) were collected in the course of this study. Gravity cores were sectioned at 2-cm intervals in the upper 40 cm and 3-cm intervals thereafter. Selected box
cores were sampled at a high resolution for more detailed study. In addition to sediment coring, a Wildco Shiptek grabber was used to collect more surface sediments for \(^7\)Be and particle size analyses. Realizing that \(^7\)Be usually decreases rapidly with depth, the surface sediment samples were saved for \(^7\)Be analysis only when it was judged by naked eye that the uppermost part of the seabed sediment was not lost or disturbed to an unacceptable extent due to overturning, slumping, or serious mixing of sediments inside the sampling device.

The same procedures as described in our previous works (Huh et al., 2006, 2009a, b) were followed to determine water (and salt) content and process the samples for the subsequent analyses. Note that water contents were determined individually for every sampling interval throughout the cores, so that apparent depth (cm) can be converted to cumulative mass (g cm\(^{-2}\)) down-core. A mean sediment dry density of 2.6 g cm\(^{-3}\) was assumed in the calculation.

3.2. Analysis of radionuclides by \(\gamma\)-spectrometry

Six HPGe detectors were engaged in this study to count over four thousand sectioned sediment samples. These detectors include three different types: one GEM-type (ORTEC GEM-150240) with 150% efficiency (relative to 3 \(\times\) NaI), three GMX-type (ORTEC GMX-120265) with 100–120% efficiency and two LoAX-type (ORTEC LoAX-70450) with 70% efficiency. Each detector is interfaced to a digital gamma-ray spectrometer (DSpec Plus\(^{\circ}\)) and all of them are routed to a desktop PC. The GammaVision 32\(^{\circ}\) software was used for spectral analysis.

Covering a wider range of energies, the GMX detectors can be used to determine \(^{210}\)Pb, \(^{234}\)Th, \(^{214}\)Pb, \(^7\)Be and \(^{137}\)Cs simultaneously based on photon peaks centered at 46.52, 63.29, 351.99, 477.56 and 661.62 keV, respectively. However, the performance of GMX detectors is less suited than that of the GEM detector for counting high-energy (\(>\) 200 keV) gammas. Having the highest counting efficiency and resolution for high-energy gammas, the GEM detector is the best choice for measuring low activities of \(^7\)Be and \(^{137}\)Cs. On the other hand, the LoAX detectors have higher efficiency and better resolution for measuring low-energy (\(<\) 100 keV) gammas emitted by \(^{210}\)Pb and \(^{234}\)Th. For specific needs or selected cores, the GEM and LoAX detectors were used in combination with each other or with one of the GMX detectors to optimize the overall results.

Absolute counting efficiencies of the detectors for various photon energies were calibrated using IAEA reference materials 327 and 375 (Strachnov et al., 1996) for sample weight at 100 g as a reference, coupled with an in-house secondary standard for various masses (from 20 to 250 g) to calibrate the effect of sample mass on the attenuation of \(\gamma\)-ray of various energies. When a 120% efficiency GMX detector was used to count-2 g sediment samples, for example, absolute efficiencies are 14.9% for \(^{210}\)Pb, 28.3% for \(^{234}\)Th, 7.5% for \(^{214}\)Pb, 6.2% for \(^7\)Be and 5.0% for \(^{137}\)Cs. The counting time for each sample varied from a few hours to a few days.

\(^{204}\)Pb-214 was used as an index of supported \(^{210}\)Pb whose activity concentration was subtracted from that of the measured, total \(^{210}\)Pb to obtain excess \(^{210}\)Pb (\(^{210}\)Pb\(_{ex}\)). Th-234 (half-life = 24.1 days) could be measured only for selected cores due to its short half-life. To determine the levels of supported \(^{234}\)Th, the samples were re-counted 5 months later following the decay of excess \(^{234}\)Th (\(^{234}\)Th\(_{ex}\)). The \(^{210}\)Pb\(_{ex}\), \(^{234}\)Th\(_{ex}\) \(^7\)Be and \(^{137}\)Cs activities reported here were decay-corrected to the date of sample collection. All radionuclide data are calculated on salt-free dry weight basis. Error bars (as shown in Figs. 7, 8 and 11) represent \(\pm 1\sigma\) around the mean based on counting statistics and standard propagation of errors.

3.3. Particle size analysis

Both cored and grabbed surface sediments collected throughout the TS were analyzed for Grain-size distribution. The samples (\(<1\) g in wet weight) were washed with 15 ml of distilled water to remove sea-salts first and then treated sequentially with 10 ml of 15% \(\text{H}_2\text{O}_2\) and 7.5 ml of 10% \(\text{HCl}\) to remove organic materials and carbonates. Prior to the analysis, 10 ml of 1% \(\text{Na}_{2}\text{PO}_4\) was added to each sample followed by vigorous shaking. The samples were then ready for analysis using a Beckman Coulter LS-13 320 Laser diffraction particle size analyzer. More detailed procedures are described in Chang (2008).

3.4. Chirp sonar echo profiling

Three sub-bottom echo profiles were reviewed in this work to complement the results of this work and facilitate the data interpretation. Two profiles from the north of the strait were obtained on the OR1-794 cruise using a high resolution EdgeTech 0512i sonar sub-bottom profiler, which was set at the frequency range 0.5–12 kHz and towed near sea surface. One profile in the middle of the strait was collected on the OR1-524A cruise by a hull-mounted 3.5 kHz profiler. The profiles from the OR1-794 cruise were previously reported in Milliman et al. (2007) and Liu et al. (2008), while that from the OR1-524A cruise has not yet been published except in the Ph.D. thesis of one of the co-authors (Chiu, 2008).

4. Results and interpretations

With a total of 180 cores and more surface sediments collected for this study, the radionuclide and particle size data are too massive to be presented in their entirety here. All data used in this work are provided as supplementary material in the on-line version of this article. In what follows, figures are used to elucidate the major findings.

4.1. Apparent sediment accumulation rates determined from \(^{210}\)Pb and \(^{137}\)Cs profiles—the systematics and implications for sources and pathways of sediments

All sediment core samples were analyzed for \(^{210}\)Pb to estimate sediment accumulation rates. However, non-steady-state profiles are rather common, hampering the efficacy of excess \(^{210}\)Pb as a chronometer in this study. This is not unexpected considering frequent storms and floods around the TS; hence massive and erratic transport of sediments at some high-energy settings. To alleviate this problem, at least partially, many cores were also analyzed for \(^{137}\)Cs (by longer counting time) to trace sediments ever exposed to (and hence picked up) this anthropogenic nuclide following World War II. To this end, the penetration depth of \(^{137}\)Cs was estimated for the determination of mean accumulation rate in the past five to six decades, taking A.D. 1950 as the time horizon for the first appearance of this nuclide in marine sediments around Taiwan (Lee et al., 2004; Huh et al., 2006, 2009a). Fig. 5 is a schematic diagram showing how sediment accumulation rates are calculated from profiles of both nuclides. To derive \(^{210}\)Pb-based rates, semi-log plots were used. From the slope (m) of \(^{210}\)Pb\(_{ex}\) activity decrease (decay) downcore, sediment accumulation rates (S\(_{210}\)) were calculated by S\(_{210}\) = \(-\lambda/m\), where \(\lambda\) is the decay constant of \(^{210}\)Pb. As for \(^{137}\)Cs-based rates (S\(_{137}\)), they are calculated by: S\(_{137}\) = \(Z_2/T_0\), where Z\(_2\) represents the observed penetration depth of \(^{137}\)Cs and T\(_0\) denotes the time of sample collection (year, in A.D.). In theory, S\(_{210}\) and S\(_{137}\) should be the same if the high-energy setting mixture is negligible and downcore distribution of \(^{210}\)Pb...
and $^{137}\text{Cs}$ are governed simply by sediment accumulation and decay of these two radionuclides following burial.

Using the methods described above, we were able to obtain both $^{210}\text{Pb}$ and $^{137}\text{Cs}$ for 90 out of 118 cores which were analyzed for both nuclides (see Appendix II in the online publication). The correlation between the calculated $^{210}\text{Pb}$ and $^{137}\text{Cs}$ (Fig. 6) shows that the latter is systematically higher than the former, suggesting that $^{137}\text{Cs}$ as a sediment chronometer is preferentially transported downward. Indeed, downward transport of $^{137}\text{Cs}$ during early diagenesis associated with ammonia formation in pore water and ion exchange between $\text{Cs}^+$ and $\text{NH}_4^+$ was documented previously in both marine and lacustrine sediments (e.g., Sholkovitz and Mann, 1984; Comans et al., 1989). On average, $S_{^{137}\text{Cs}}$ is higher than $S_{^{210}\text{Pb}}$ by ~19%, with more than one-quarter of the data points on Fig. 6 deviating less than 10% from the 1:1 line. Shown in Figs. 7a and b are a dozen of those “well-behaved” profiles in cores raised from relatively stable benthic environments throughout the study area. These plots are reduced to smaller insets and tied to the coring sites shown in Fig. 8 to provide a geographic perspective of the characteristics of the profiles.

Since $S_{^{137}\text{Cs}} > S_{^{210}\text{Pb}}$ due to sediment mixing or diagenetic processes, $^{210}\text{Pb}$-based apparent sediment accumulation rates should be closer to true rates. Fig. 9 shows the spatial distribution of $S_{^{210}\text{Pb}}$ in terms of linear sedimentation (in cm/yr, Fig. 9a) and mass accumulation (in g cm$^{-2}$ yr$^{-1}$, Fig. 9b). Two depocenters are obvious in the TS (to the south of 26°N)—one near the estuary of the Choshui River, the largest sediment source from Taiwan (see Table 1; Water Resources Bureau, 1998), and the other in the northwest in the alongshore clinoform stretching from the Yangtze River’s estuary. The lowest rates are distributed over the Wu-Chiu Depression in the southwest and the Kuan-Yin Depression in the northeast, coinciding with the paths of the dominant current flows in the TS, namely the western branch of the Kuroshio Branch Current (along the eastern slope of the Wu-Chiu Depression) and the Taiwan Warm Current (over the Kuan-Yin Depression). Located between the above regions (of the highest and the lowest accumulation rates) is an area of intermediate water depth and sediment accumulation rates, which extends northward across the strait, roughly from the northern flank of the Chan-Yuen Rise to the nearshore area adjacent to the Min River’s estuary.

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**Fig. 5.** Schematic plots illustrating the calculation of sediment accumulation rates based on $^{210}\text{Pb}_{\text{ex}}$ and $^{137}\text{Cs}$ profiles. See text for more details.

**Fig. 6.** Correlation between $^{137}\text{Cs}$-based and $^{210}\text{Pb}$-based accumulation rates. The data can be found in Appendix II of the online publication.
4.2. Spatial and temporal variations of $^7$Be in surficial sediments—implications for flood dominated sedimentation and mobile mud dynamics

$^7$Be ($T_{1/2} = 53.3$ days), a cosmogenic nuclide concentrated in topsoils on land, has been shown to be a useful tool for tracing recent deposition of riverine fine sediments in nearshore settings (Sommerfield et al., 1999; Mullenbach et al., 2004; Moralles et al., 2006; Huh et al., 2009a). In ten of the cruises conducted since 2005 off the west coast of Taiwan, surface (~2 cm) sediments were collected by the Shiptek grabber for the determination of $^7$Be in a timely manner. The top of the grabbed sediment was judicially examined and only those judged to be reasonable well collected with minimal disturbance were used for this application. Although the same sampling locations were not reoccupied for all the cruises, the overlap of the covered area between cruises (Fig. 10) still allowed us to discern some spatial and temporal variations.
temporal variations. For instance, two obvious scenarios can be depicted as follows. At first, a mud belt highly enriched in $^7$Be (up to $>2$ dpm/g) was observed off the Tachia and Wu Rivers’ mouth during August 8–10, 2005 (Fig. 10a). This freshly deposited mud layer must be ascribed to Super Typhoon Haitang’s landing on and passage through Taiwan during July 18–19, 2005, which caused very intense rainfall ($>4800$ mm), and hence extensive sheet erosion in these rivers’ drainage basins and export of $^7$Be-enriched flood sediments to the rivers’ estuary. It is interesting to note that two months later (during October 17–23, 2005) the $^7$Be-laden mud patch vanished from the rivers’ mouth (Fig. 10b). Another pronounced change occurred between the end of June and July in 2006, about one month apart. During June 24–27, 2006 (Fig. 10d), a mud patch enriched in $^7$Be ($>1$ dpm/g) was found $\sim20$ km offshore (centered around $24^\circ20'$N and $120^\circ17'$E) in the west of the Wu and Tachia Rivers’ mouth. Meanwhile, a newly formed, $^7$Be-enriched mud patch emerged from the Chungkang River’s mouth to the north. One month later (during July 27–29, 2006; Fig. 10e), both patches mentioned above were gone, while another newly formed, $^7$Be-enriched mud patch emerged from Tachia River’s mouth, which was consistent with a SPOT satellite image taken on July 20 showing a plume out of the river’s mouth (Chang, 2008). We believe the plume was caused by Typhoon Billis during July 13–14, 2006. Grain-size analysis of estuarine and coastal sediments off western Taiwan following Typhoon Billis clearly showed that flood layers deposited in wake of the typhoon were transported toward the north (Chang, 2008).

The emergence and disappearance in space and with time of $^7$Be-labelled surface mud layers in the TS could reflect an episodic phenomenon induced primarily by floods. By integrating rainfall and river discharge data on land and the circulation pattern in the strait, the mobile mud dynamics can be explained by episodic riverine input followed by gradual migration of the ephemeral deposits, conceivably via resuspension and northward transport periodically by intensified tidal and current flows.

4.3. Mixing of surficial sediments revealed from $^7$Be and $^{234}$Th

Spatial distribution of $^7$Be-enriched mud deposits, as discussed above, was derived using the top 2 cm of the grabbed or cored
sediments without knowing the penetration depth of this short-lived nuclide. If the sediment is collected by the Shiptek grabber, it is impractical to use the collected sample for radionuclide profiling because grabbed sediment material is of limited thickness and is more susceptible to disturbance. For cored samples, however, it is possible to investigate the depth distribution of $^7\text{Be}$ if its activity is adequate and the samples were analyzed in a timely manner. This was specifically done on three box cores (BC8, 9 and 10) collected from the Tachia River’s estuary on the OR1-841 cruise (September 12–15, 2007) and one box core (BC3) taken from the mud wedge distributed along the southwestern flank of the Chan-Yuen Rise on the OR1-894 cruise (April 6–11, 2009). Profiles of $^7\text{Be}$ in these cores are shown in Fig. 11. In order to take sediment accumulation and mixing into consideration, profiles of $^{210}\text{Pb}_{\text{ex}}$ and $^{137}\text{Cs}$ in all 4 cores and $^{234}\text{Th}_{\text{ex}}$ in the last core mentioned above are also plotted. In the first three cores, $^{210}\text{Pb}_{\text{ex}}$ activity varied around 1 dpm/g without showing any trend with depth; $^{137}\text{Cs}$ penetrated through the entire lengths (440 cm) of the cores; while $^7\text{Be}$ was detected down to 14 cm depth in two of these three cores. Such a combination of downcore distribution of radionuclides suggests very rapid and dynamic deposition of sediment near the Tachia River’s mouth. In OR1-894 BC3, the penetration depth of $^{137}\text{Cs}$ is $20 \text{ cm}$, suggesting a mean accumulation rate of 0.34 cm/yr around the past 6 decades, in good agreement with $^{210}\text{Pb}$-derived apparent accumulation rate. In this latter core, however, both $^7\text{Be}$ and $^{234}\text{Th}_{\text{ex}}$ were detected down to 6 cm. If the decreases with depth of these two short-lived nuclides were interpreted solely by decay following burial, a mean deposition rate of $1.2 \text{ cm/month}$ in the past 5 months was calculated. Such a high rate is seemingly at odds with the much slower but fairly consistent accumulation rates derived from $^{137}\text{Cs}$ and $^{210}\text{Pb}$ on decadal to centennial timescales. It would be helpful to apply X-ray radiography to determine the effect of sediment mixing on down-core penetration of $^7\text{Be}$ and $^{234}\text{Th}_{\text{ex}}$. Unfortunately, we

Table 1

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<th>Name of river</th>
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<th>Sediment yield (ton km⁻² yr⁻¹)</th>
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| Rivers from mainland China | | | | |
| Yangtze | 1,808,500 | 337–513 | 186–284 | [2] |
| Min | 61,000 | 7.5 | 123 | [3,4] |
| Jiulong | 14,700 | 2.5 | 170 | [3,4] |
| Subtotal | 1,884,200 | 347–523 | 184–278 |

[2] Range of Yangtze River’s annual sediment load was measured at the Datong Station during 1951–2000. There was an obvious decrease during this period, with continued decrease to $\sim 150 \text{ Mt/yr}$ expected by the 2010s (Yang et al. 2002, 2003).
were not equipped to collect X-ray radiographs. At any rate, the
discrepancy between the short-term (seasonal) deposition rates
derived from $^7$Be- and $^{234}$Th$_{ex}$ and the longer-term accumulation
rates (based on $^{137}$Cs and $^{210}$Pb$_{ex}$) can be explained by short-term
deposition and subsequent erosion of the $^7$Be- and $^{234}$Th$_{ex}$-laden
particles. Such processes were observed off the mouth of the
Yangtze River (McKee et al., 1983).

It is almost certain that sedimentation near the Tachia River's
mouth is not at steady state. As for the mid-shelf site, it could be
more representative of the conditions at other sites where sedi-
ment accumulation rates were estimated. We take it as an example
to point out that, under more stable environments, sediment
mixing revealed by short-lived nuclides has limited impact to
sediment accumulation rates derived from much longer-lived
nuclides. Thus, insofar as the purpose of this study is concerned,
accumulation rates derived from $^{210}$Pb and $^{137}$Cs in much of the TS
are still useful and indispensable.

4.4. Grain-size distribution in surface sediments—implications for
sources and pathways of sediments

Fig. 12 shows the spatial variation of mean grain size in surface
($\leq 2$ cm) sediments, which bears some resemblance to that of
sediment accumulation rates (Fig. 9), with sand-sized sediments
covering areas of low accumulation rates (mainly the Kuan-Yin
Depression in the northeast and the Wu-Chiu Depression in the
southwest), while mud (silt+clay) overlaying areas of high and
intermediate accumulation rates. The correspondence suggests
that the distribution of particle size in surface sediments deposited
recently ($\sim 1$–10 years) has implications for sediment accumula-
tion over longer timescales (decades to 100 years), as dictated by
$^{210}$Pb and $^{137}$Cs chronology.

4.4.1. Sands over the western Chan-Yuen Rise

By analyzing the size distribution of surface sediments in
relation to seafloor morphology and hydrodynamics in the study
area, further information about sources, pathways and transport
dynamics of sediments can be gleaned. A case in point is the study
by Liao et al. (2008) of the Chan-Yuen Rise in the central-eastern
part of the TS. The sand bodies of the Chan-Yuen Rise are composed
of very fine-to-coarse sands (0–4 $\Phi$) in a progressive seaward
decrease in grain size. The configuration and spatial distribution
of the sand bodies in the Chan-Yuen Rise (Fig. 1) along with
corresponding decrease in tidal speed (Fig. 3) and in grain-size
offshore suggest that the sand bodies are deposited in a tide-
dominated regime and developed sequentially from the nearshore
to the offshore area, presumably in response to westward shifting
of the north-flowing tidal currents (Liao et al., 2008).

Well-sorted fine sands (Fig. 13a) and the alignment of well-
developed sand waves in the western Chan-Yuen Rise (Fig. 15; to
be discussed in Section 4.5) suggest that this sand body is actively
receiving sands from the south via the Penghu Channel (Liao et al.,
2008).

4.4.2. Size distribution of sediments transported northward from
the western Chan-Yuen rise and the west coast of Taiwan

To the north of the western Chan-Yuen Rise is an elongated sand
sheet (see Fig. 1, and Liao et al., 2008) where surface sediments are
composed largely of fine-to-coarse sands (2–4 $\Phi$; Chang, 2008),
suggesting that it is an extension of the western Chan-Yuen Rise
along the direction of the Kuroshio Branch Current. Surface
sediments in the south of the sand sheet (between 24 and 25° N)
are well sorted, with size frequency curves similar to those over the
western Chan-Yuen Rise, suggesting a common source from the
south. To the north of the sand sheet (at $\sim 25$° N), well-sorted sand
4.4.3. Size distribution of sediments along the Chinese coast

Discharged from western Taiwan rivers (Fig. 13f), broadband all the way from mud to coarse sand signifying sediments of fine to very fine sands from the western Chan-Yuen Rise and a sorted and their size frequency curves include a sharp peak indicative in Fig. 8) is in this zone of sediment convergence, where the $^{137}$Cs profile shows a mixed characteristics, where discussed in Section 4.1.

To the east of the above-mentioned sand sheet and north of the Chan-Yuen Rise is the Kuan-Yin Depression where surface sediments show a variety of size distribution resulting from mixing of fine sand from the west and mud from the southeast. Near the Tachia River’s mouth, surface sediments are composed entirely of newly delivered mud (4–10 ft; silt and clay; Fig. 13d). In the southeastern end of the Kuan-Yin Depression (around 24°30’N, 120°30’E) where $M_2$ tides are the weakest (Fig. 3), sediments are very poorly sorted and show a bimodal distribution (Fig. 13e). In the center of the Kuan-Yin Depression (25°–25.5°N, 120°–120.3°E), sediments are also poorly sorted and their size frequency curves include a sharp peak indicative of fine to very fine sands from the western Chan-Yuen Rise and a broadband all the way from mud to coarse sand signifying sediments discharged from western Taiwan rivers (Fig. 13f).

4.4.3. Size distribution of sediments along the Chinese coast

The distribution of mean grain size in surface sediments (Fig. 12) suggests a very sizable input of fine sediments from the Chinese coast toward the northern TS. Taking a close look of size frequency curves for samples from the middle of the northern TS, we reaffirm that there is an area in the middle of the strait receiving sediments from both shores across the strait.

4.5. Post-glacial sedimentation over transgressive surface revealed by sub-bottom sonar echo profiles

From Chirp sonar survey conducted on 12 cruises, Chiu (2008) compiled a fairly extensive set of echo profiles that can be used to map the distribution of sediment strata and their structure in the TS. Some of the results were reported in Milliman et al. (2007) and Liu et al. (2008) previously. An integration of the echo profiles and our data about modern sedimentation in the TS should be in order.

Illustrated in Fig. 14 are two contrasting Chirp sonar sections across the northern TS. The section crossing the northern Kuan-Yin Depression in the east is characterized by widespread exposure of scour furrows on the Holocene transgressive surface. Under strong tidal and current flows, the seafloor in that region is unfavorable for sedimentation. In contrast, the other section to the west, near the Chinese coast, is a low-energy setting with ample sediment supply, as reflected by thick layers of sediments derived from the northwest (i.e., the alongshore clinoform under the Chinese Coast Current) and the southeast (i.e., western Taiwan), with the former overlying unconformably over the latter and pinching out toward the east.

As with the Kuan-Yin Depression, the Penghu Channel is also a major pathway for sediment transport and, as expected, there is nothing but erosive features over the floor of the water channel due to strong tidal and current flows. In the north and at the back of the Penghu Channel, however, there is a mud wedge (or clinoform) deposited at the southwestern slope of the Chan-Yuen Rise (Fig. 15). It is important to note that core OR1-894 BC3 (discussed in Section 4.3) taken from this mud wedge is on the path of the Kuroshio Branch Current. According to Chiu (2008), the mud wedge can be traced over 100 km along the eastern slope of the Wu-Chiu Depression to form a mud belt. Fine-sized sediments accumulated in this belt are most likely sourced from Taiwan’s southwestern rivers (i.e., rivers 9 to 16 in Fig. 1) and are transported through the Penghu Channel by the Kuroshio Branch Current. The presence of $^{7}$Be and excess $^{234}$Th at the top of core OR1-894 BC3 (see Fig. 11) suggest very rapid source-to-sink transport along this pathway. Thus, in addition to the two well-recognized S2S systems which merge in the northern TS, there exists a third S2S dispersal system in the TS which is separated from the system sourced from Taiwan’s northwestern rivers (i.e., rivers 1–8 in Fig. 1) by the Chan-Yuen Rise in the middle of the strait.
5. Discussion

5.1. An appraisal of the validity of sediment accumulation rates derived from $^{210}$Pb and $^{137}$Cs

While synthesizing the rather large datasets obtained in this work, we found that steady-state (or quasi-steady-state) $^{210}$Pb$_{ex}$ profiles are more readily encountered in less energetic benthic conditions with weaker tidal and current flows. The southeastern part of the Kuan-Yin Depression and the mud belt along the Chinese coastline (esp. in the northwestern TS) are such environments. In these locations sediment accumulation rates derived from $^{210}$Pb and $^{137}$Cs usually agree better, suggesting more steady and stable supply of fine particles without, or with relatively less, post-depositional disturbances. On the other hand, sediments in high-energy benthic environments are usually coarser-grained and non-steady-state $^{210}$Pb profiles are more common. Under such conditions, we are obliged to resort to $^{137}$Cs for a ballpark estimate of mean accumulation rates in the past 5–6 decades since the advent of this man-made nuclide circa A.D. 1950.

In dealing with sediment budget from a S2S perspective, the timescales involved in the source and sink terms should be as compatible as possible. The source term can be reasonably estimated

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Fig. 12. Distribution of mean grain size in surface sediments. The data can be found in Appendix III of the online publication.
from sediment loads of Taiwan’s rivers using the database established by the Water Resources Agency since AD 1919 (WRA, 1998). To match this term in space and time, it behooves us to study sedimentation in the TS (and other neighboring seas) on decadal to centennial timescales. To this end, there are no better alternatives than using $^{210}$Pbex and $^{137}$Cs as sediment chronometers. However, without the effect of mixing quantified, sediment accumulation rates thus determined should be taken as upper limits.

5.2. Comparison of sediment accumulation rates derived from $^{210}$Pb and other methods

Sediment accumulation rates in the TS we derived from direct measurements of $^{210}$Pbex and $^{137}$Cs conflict with those based on $^{10}$Be concentration in surface sediments (Fig. 1 in Lee et al., 1993). The model-dependent $^{10}$Be sedimentation rates of Lee et al. (1993) have nothing to do with radioactive decay; they were derived using $^{14}$C-dated samples as a benchmark and assuming a constant $^{10}$Be flux throughout the TS. As such, $^{10}$Be concentrations in surface sediments were presumably inversely proportional to sediment accumulation rates at different sites. Sediment accumulation rates thus derived generally decrease offshore from the coastline of Taiwan, with highest values found in nearshore areas covered by reworked relict sand (e.g., the Chan-Yuen Rise) where there is actually no net accumulation of modern sediments. Here, we wish to point out that the assumption of constant $^{10}$Be flux for the entire Taiwan Strait is invalid. We believe that, as with $^{210}$Pb, most $^{10}$Be associated with Taiwan Strait sediments was derived from lateral transport of the Kuroshio Branch Current followed by boundary scavenging of this particle-reactive nuclide at ocean margins (Lee et al., 2004). Therefore, sedimentary fluxes and inventories of $^{10}$Be, like those of $^{210}$Pb (see Fig. 16), are bound to be dependent on particle fluxes and thus are highly variable from place to place in the Taiwan Strait.

Unlike the $^{10}$Be-based model results, the distribution of sediment accumulation rates reported here (Fig. 9) matches fairly well with the abundance of fine-sized particles in surface sediments (Fig. 12), indicating that modern sediments in the TS are fairly "muddy". Indeed, Kao et al. (2008) have provided unequivocal evidence that freshly delivered fluvial sediments are composed primarily of mud, with sand-sized particle accounting for only a minor fraction. Fig. 9 also matches well with the isopach map of the post-glacial fluvial sediment buried in the TS (Fig. 15 in Liu et al., 2008), again suggesting that sedimentation processes operating on short timescales may be relevant to the formation of sediment strata over much longer (from 100 to 10,000 years) timescales (Sommerfield and Nittrouer, 1999; Bentley and Nittrouer, 2003; Huh et al., 2009a).

As regards $^{14}$C-based sediment accumulation rates in the TS are generally lower than $^{210}$Pb-based rates by one order of magnitude, sometimes even more. However, similar to $^{210}$Pb results but unlike the model results of Lee et al. (1993); there is a general correspondence between higher $^{14}$C-based sediment accumulation rates and fine-grained sediments. In fact, the large disparity between accumulation rates derived from $^{14}$C and $^{210}$Pb is not an uncommon phenomenon;
Besides the “Sadler effect” (Sadler, 1981; Sommerfield et al., 2007), it may also be related to increased inputs of riverine mud during the transition from transgressive to high-stand sedimentation conditions in the late Holocene (Sommerfield and Wheatcroft, 2007).

5.3. Sediment provenances revealed from distribution of radionuclides

\(^{137}\text{Cs}\) and \(^{210}\text{Pb}_{\text{ex}}\) are useful not only as time tracers but also as process and provenance tracers in the TS. Fig. 16 shows very different distribution pattern of sediment inventories of \(^{137}\text{Cs}\) and \(^{210}\text{Pb}_{\text{ex}}\), indicating that sediments with substantially higher \(^{137}\text{Cs}\) activity and \(^{137}\text{Cs}/^{210}\text{Pb}_{\text{ex}}\) activity ratios are transported from the distal north along the Chinese coast. This is due to higher atmospheric fluxes of \(^{137}\text{Cs}\) at mid-latitudes coupled with lower sediment yield in Chinese rivers’ drainage basins. In contrast, with much higher sediment yield (Table 1) and relatively lower atmospheric fluxes of \(^{137}\text{Cs}\) in tropic to subtropic regions, sediment exported by Taiwanese rivers is \(^{137}\text{Cs}\)-depleted. Coupled with the intrusion of the Kuroshio Branch Current and thus enhanced boundary scavenging of \(^{210}\text{Pb}\) (Lee et al., 2004; Huh et al., 2009b) by sediments derived from Taiwanese rivers, lower inventory ratios of \(^{137}\text{Cs}/^{210}\text{Pb}_{\text{ex}}\) in the eastern part of the strait is quite conceivable.

Besides the targeted radionuclides, some other nuclides can also be measured coincidentally in this work by gamma spectrometry. These nuclides may also be used as plausible tracers of sedimentary provenance. For instance, the \(^{214}\text{Pb}/^{40}\text{K}\) activity ratios in sediments near the Min River’s mouth is lower than those off the Choshui River’s mouth by a factor of about 2 (data not shown here), indicative of a major lithological difference between the two rivers’ basins.

5.4. Varied characteristics of \(^{137}\text{Cs}\) profiles across the Taiwan Strait: Implications for source-to-sink transit time

It is informative to note from Fig. 8 that \(^{137}\text{Cs}\) profiles in the eastern side of the strait (sites 1–5 in Fig. 8) are characterized by an abrupt upward increase from nil to a subsurface maximum and a gentle decrease from the maximum toward the core top. This kind of \(^{137}\text{Cs}\) profiles are also commonly observed in other sedimentation environments receiving sediments from Taiwan’s drainage basins, such as the Southern Okinawa Trough off northeastern Taiwan (Huh et al., 2004, 2006; Lee et al., 2004) and the shelf/slope area off southwestern Taiwan around the Gaoping Submarine Canyon (Huh et al., 2009a). Based on \(^{210}\text{Pb}\) chronology established for these cores, the timings of both the penetration depth (circa A.D. 1950) and the subsurface maximum of \(^{137}\text{Cs}\) (circa A.D. 1963) are consistent with the history of nuclear fallout. In other words, the time span between these two
horizons (~13 years) is much shorter than that (~42–45 years) from the time of fallout maximum to the time of sample collection. In contrast to this type of 137Cs profiles, those in the western side of the strait (sites 7–12 in Fig. 8) show a different pattern, with the segment between the penetration depth and the subsurface maximum of 137Cs being longer than the upper part of the profiles. These two types of 137Cs profiles can be well explained by difference in drainage basin residence time of the source material. In the eastern strait receiving sediments mainly from Taiwan, 137Cs-bearing sediments are transported rapidly from their source areas via small mountainous rivers. As a result, the sedimentary record of 137Cs reflects its input history with little delay in time. In comparison, it conceivably takes much longer time for sediments to be delivered by rivers from large drainage basins in mainland China (see Table 1) and transported long distance to the western side of the strait.

A continuum of these two types of 137Cs profiles likely exists across the strait. Taking core OR2-1559 GC24 (i.e., site 6 in Fig. 8) for example, it was located in the middle of the strait at 25.42°N, 120.43°E where sediments from both sides merge (to be further discussed later). There, the subsurface maximum of 137Cs was found at a depth about one-half of the penetration depth of 137Cs. From this perspective, it may be possible to delineate the source

Fig. 15. Chirp sonar profile showing a mud clinoform in the southwestern flank of the Chan-Yuen Ridge and well-developed sand waves on the bank; the arrow on the profile indicates the location of core OR1-894 BC3 (nuclide profiles shown in Fig. 10). The mud wedge can be traced over 100 km in the middle of the TS (indicated in purple; Chiu, 2008).

Fig. 16. Spatial distribution of (a) 210Pbex inventory, (b) 137Cs inventory and (c) the 137Cs/210Pbex inventory ratio in Taiwan Strait sediments.
and pathways of sediments based on a systematic analysis of the shape and spatial variation of $^{137}$Cs profiles.

Another noteworthy feature is the bending or “concaving-up” trend in some of the $^{210}$Pb profiles in the eastern side of the strait, especially in the southern part of the Kuan-Yin Depression (i.e., sites 3–5) adjacent to Taiwan’s Choshui, Wu and Tachia Rivers. Since the time of deposition at the penetration depth of $^{137}$Cs calculated based on $^{210}$Pb decay is consistent with the time of first appearance (circa A.D. 1950) of $^{137}$Cs, the bending of the $^{210}$Pb profile cannot be attributed to sediment mixing. Rather, we believe it was caused by accelerated erosion due to inadequate soil and water conservation practices plus increased frequency of extreme precipitation events in Taiwan in the past several decades (Liu et al., 2009). This assertion is rigorously supported by the distribution of magnetic properties as provenance tracers in well dated sediment cores (Hung, C.-S., personal communication; manuscript in prep.).

### 5.5. A tentative evaluation of sediment budget in the Taiwan Strait

Sediment cores collected for this study were concentrated in the northern half of the TS. The southern part of the TS, consisting mainly of the Taiwan Shoal and the Penghu Channel, is essentially non-depositional and hence not amenable to sediment coring. From the data used in plotting Fig. 9, an area-weighted mean accumulation rate of $0.39$ g cm$^{-2}$ yr$^{-1}$ was derived for an area of about $4.1 \times 10^3$ km$^2$ between 24°N and 26°N in the northern TS. Multiplying these two values yields a sediment burial flux of about $160$ million tons per year (Mt/yr). This approximation is in all likelihood an overestimate of the sediment flux in the specified area not only because of the over-estimated values for sediment accumulation rates but also because it ignores the fact that there are areas without active accumulation where transgressive sand layer is still exposed over the seabed. Such environments exist in certain parts of the Kuan-Yin Depression and the area off northwestern coast of Taiwan under strong tidal and current forcing (see Fig. 14). Like the condition in most of the southern TS, the lack of muddy sediments in such environments renders it impossible to get box or gravity cores for this study.

Compared with the results obtained for the northern TS, the number of sediment cores collected from the southern strait is far less and the deduced sediment accumulation rates are much lower. Therefore, we contend that modern sediments in the TS primarily reside in the area specified above (i.e., between 24°N and 26°N).

As with a most recent study on the Poverty continental shelf by Miller and Kuehl (2010), we compared the burial flux of modern sediments with riverine sediment input to the TS. Listed in Table 1 are annual sediment loads of 16 Taiwanese rivers and 3 Chinese rivers which contribute to Taiwan Strait's sediment budget. Except the Yangtze River in the distant north, all other rivers empty directly into the TS. Of the 16 western Taiwan rivers, 8 in the north discharge a combined sediment load of $98$ Mt/yr (Table 1) directly into the Kuan-Yin Depression. The other 8 rivers in the south collectively discharge another $55$ Mt/yr toward the Penghu Channel, which is most likely entrained in the perennal flow of the Kuroshio Branch Current and transported to the northern TS. In the west side of the strait, the Min and the Jiulong Rivers from China’s Fujian Province supply $10$ Mt/yr. Thus, before taking Yangtze River’s contribution into account, sediment input to the TS from the nearby shores already amounts to $163$ Mt/yr, which is surprisingly comparable to the burial flux estimated above.

Judging from the spatial distribution of sediment accumulation rates (Fig. 9) and particle size in surface sediments (Fig. 12), the alongshore plume stretching from the Yangtze River’s mouth must be an important source of sediments to the TS, especially in the winter monsoon season. Thus, to account for the sediment budget in the TS on decadal to centennial timescales, it requires the export of a sizable fraction of Taiwan’s fluvial sediments out of the strait from its northeast. This export must at least equal to the amount imported by the Chinese Coast Current to the northern TS plus any amounts yet unaccounted for, such as the unknown quantity buried in the southern part of the TS or elsewhere. Were it not for this inevitable export, the overall mud content in the TS would have been much higher than what is actually observed.

### 6. Summary

By integrating all data obtained from this and previous studies, three major sediment dispersal systems can be defined in the TS. In terms of sediment load, the presumably largest system is sourced from a series of western Taiwan rivers including the Choshui River and seven smaller rivers to its north (Fig. 1). Sediments from these rivers’ drainage basins are discharged into the northeastern sector of the strait (mainly the Kuan-Yin Depression) and then transported by the Taiwan Warm Current toward the East China Sea. The second system is derived from other western Taiwan rivers to the south of the Choshui River, with their sediment loads delivered toward the Penghu Channel, entrained by the Kuroshio Branch Current and transported to the north. This system is separated from the first one by the Chan-Yuen Rise and is manifest by the mid-strait mud belt formed along its pathway. The third system is sourced from the Yangtze River and other Chinese rivers to its south, driven by the Chinese Coast Current and flowing in an opposite direction on the other side of the strait. Although the Yangtze River is the largest single sediment source in the region, most of its sediment load is deposited in the river’s subaqueous delta and alongshore clinoform in the north (Liu et al., 2007, 2008; Xu et al., 2009); the amount delivered to the TS remains to be further investigated. This southbound system encounters the northbound systems mentioned above in the middle of the northern TS, forming a distal depocenter there. To account for the sediment budget and the overall content of mud vs. sand in the TS on decadal to centennial timescales, it requires a considerable export of sediment, with mud-sized riverine sediments preferentially transported out of the strait in its northeast.

From the distribution of $^{210}$Pb-based sediment accumulation rates, $^{7}$Be activity and particle size in surface sediments, we conclude that sedimentation in the TS is controlled by flood, current and tidal flows. Higher sediment accumulation rates are associated with fine-sized sediment particles in low-energy areas where tidal and current flows are weaker but tidal levels are higher. The middle reaches of the strait and the coastal region in the northwest of the strait are such settings. Conversely, erosive or low deposition areas are dominated by stronger tidal and current flows, thus unfavorable for the accumulation of fine-sized sediments. The Penghu Channel, Chan-Yuen Rise, and the area off northwestern Taiwan are such settings. In general, information gathered from this work about modern sedimentation and sediment transport is consistent with the structure of sediment strata accumulated over much longer timescales as revealed by sub-bottom echo profiles.

Despite the findings summarized above, some of the notions developed from this work are still somewhat vague, suffering from internal problems, and require more work to tighten them. Questions remain to be resolved include the following:

1. How large are the lateral fluxes of sediments into and out of the TS? Can their difference, if any, be balanced by sediment burial in the TS on timescales dealt with in this study?
2. Of the three S2S systems described above, what is their net contribution to the sediment budget in the TS?
3. Where is the borderline (or mixing zone) between sediments sourced from western Taiwan and mainland China? Does it change with time (e.g., season) and how?
More sophisticated provenance studies are underway to address these issues.

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Appendix A. Supplementary materials

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.csr.2010.11.002.

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