Assessing effective provenance methods for fluvial sediment in the South China Sea

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Abstract: Sediment is delivered by the rivers of SE Asia to the South China Sea where it provides an archive of continental environmental conditions since the Eocene. Interpreting this archive is complicated because sediment may be derived from a number of unique sources and the rivers themselves have experienced headwater capture that also affects their composition. A number of methods exist to constrain provenance, but not all work well in this area. Anthropogenic impacts, most notably agriculture, mean that the modern rivers contain more weathered materials than they did up until about 3000 years ago. The rivers have also changed their bulk chemistry and clay mineralogy in response to climate change, so that these proxies, as well as Sr isotopes, are generally unreliable provenance indicators. Nd isotopes resolve influx from Luzon, but many other sources in SE Asia have similar values and clear resolution of end members can be difficult. Instead, thermochronology methods are best suited, especially apatite fission track, which shows more diversity in the sources than either U–Pb zircon or Ar/Ar muscovite dating. Nonetheless, even fission track is best used as part of a multiproxy approach if a robust quantitative budget is desired.

The South China Sea is one of the largest marginal basins in the Western Pacific and has been the repository of large volumes of clastic sediment delivered to the continental margins by the large rivers that drain the East Asian continent. In theory, these sedimentary records could be used to decipher the history of tectonism, surface uplift and environmental evolution in this region. Such data are essential if we are to understand the relationships between solid Earth evolution and the development of climate in the aftermath of the India–Asia collision, most notably the intensification of the East Asian monsoon whose history of activity is still not well understood (Sun & Wang 2005; Clift et al. 2014). However, if we are to read and interpret these sedimentary records then it is necessary to first understand where the sediment in each sub-basin was derived from because changes in sediment source may also result in changes in composition and mineralogy, which could be mistaken for changes in environmental conditions when in reality they merely reflects derivation from source bedrocks with different bulk compositions. Unless the sediment source can be properly assigned then other data sets cannot be used to their full potential in understanding how processes such as the intensification of the East Asian monsoon have impacted the continental environment over the last 65 million years. In this paper I review a number of the more commonly applied methods for constraining the source of sediment into this basin and explore the effectiveness of each in allowing us to understand how sediment is dispersed after its delivery to the ocean.

Sediment provenance is particularly important in the South China Sea because such methods have been used to track sediment transport within the basin and subsequently to infer the influence of bottom currents, which are in turn controlled by the opening and closure of gateways (Lei et al. 2007), especially those into the Western Pacific, such as the Bashi Straits between Taiwan and Luzon (Fig. 1). Furthermore, the three large rivers that drain into the basin, the Mekong, the Red (Song Hong) and the Pearl Rivers have all been proposed to have experienced significant amounts of headwater drainage capture, as a result of the eastwards tilting of the Asian continent during the uplift of the Tibetan Plateau (Brookfield 1998; Clark et al. 2004). The timing of this reorganization is controversial (Clift et al. 2006a; Robinson et al. 2013; Zheng et al. 2013), but is also important for testing models of surface uplift in Tibet and surrounding regions. If we are not able to pinpoint the source of the sediment within the delta and submarine fan systems in the marginal seas then it is impossible to fingerprint the influence of one river compared to another and thus to isolate the potential impact of drainage capture. Developing robust provenance tools is the first stage in addressing this process.
Most of the methods that I discuss are not particularly novel, but represent standard methodologies that are applied to many sedimentary basins around the world. The South China Sea represents a special challenge because of the diversity of possible sources, but also provides an example of how, even in such a relatively complicated setting, effective sediment provenance reconstructions can be achieved.

**Geological setting**

The tectonic origins of the South China Sea have been strongly debated, but it is reasonably clear that the region began to experience significant continental extension during the Eocene (Ru & Pigott 1986; Franke 2013), culminating in the onset of seafloor spreading around 28 Ma (Briais et al. 1993; Barckhausen et al. 2014). Seafloor spreading ceased...
by 17 Ma, after which time the region has largely been affected by thermal subsidence, although disrupted by localized neotectonic activity, such as the volcanism and uplift in Hainan island (Shi et al. 2011). The most important tectonic event to have influenced sediment supply to the basin after the end of extension has been the collision between the oceanic island arc of Luzon with the passive margin of China, as manifest in the island of Taiwan (Suppe 1984; Huang et al. 2006). Taiwan is one of the world’s great sediment sources and exceeds the Ganges–Brahmaputra delta in supplying sediments to the ocean despite its small size (Milliman & Syvitski 1992).

As well as receiving sediment from eastern and southern Taiwan, the South China Sea is supplied by sediment from smaller rivers on the Philippine Islands to the east, from Borneo to the south, but, more importantly, from three major rivers along the western and northern sides of the basin, and namely the Mekong, Red and Pearl rivers (Fig. 1). These, respectively, carry loads of 160, 170 and 69 Mt a\(^{-1}\) (Milliman & Syvitski 1992; Le et al. 2007). These numbers can only be considered as a rough guide, as the budgets are typically only for the suspended load, are not always taken near the river mouth and may be affected by anthropogenic disruption of the basin. Despite the large size of some of these drainage systems, the two largest rivers in SW Taiwan – the Tsengwen and the Kaoping rivers – have measured pre-modern sediment loads of 31 and 49 Mt a\(^{-1}\), respectively, and the long-term recent discharge into the Taiwan Strait exceeds 380 Mt a\(^{-1}\) (Kao & Milliman 2008). It is noteworthy that Taiwan is also struck by many large typhoons originating in the central Pacific. These typhoons result in significant erosion and sediment discharge, with Typhoon Herb in 1996 alone being responsible for the discharge of 142 Mt into the South China Sea in a single week (Milliman & Kao 2005). Although Taiwan cannot have been a significant sediment source before the uplift of the present ranges after around 6 Ma (Huang et al. 2006), it is certainly worth considering the sediment-producing potential of each possible source when making volumetric assessments of the contribution from different end members to the deeper part of the South China Sea.

**Basis of provenance**

The ability to distinguish and estimate the amount of sediment derived from a given source is mostly based on the concept that the bedrock sources providing the sediment are distinguished from one another in different parts of the potential source area on the basis of their chemistry, geochronology or tectonic evolution and that these differences are transferred from the bedrock to the sediment in the rivers. Southeast Asia is remarkably suitable for such provenance work because of the assemblage of a number of contrasting tectonic blocks or terranes in this region (Fig. 2). These were largely brought into juxtaposition during the Triassic Indosinian Orogeny (Carter et al. 2001; Lepvrier et al. 2004; Carter & Clift 2008), with later additions, especially in the Tibetan headwaters of the large rivers during the final collision between India and Eurasia. Because of their contrasting geological histories, tectonic blocks shown in Figure 2 produce sediment of different composition or geochronological age, which can then be detected in the sediments deposited in the South China Sea. The geological evolution of each of these blocks is relatively complicated, but it is possible to say that essentially southern China, Cathaysia, represents a tectonic block that collided with the Yangtze Craton at approximately 800 Ma and that subsequently this was the host to a Mesozoic volcanic arc complex (Hutchison 1994; Fletcher et al. 2004). In contrast, the central part of China is dominated by the ancient crust of the Yangtze Craton, which itself collided with the North China Block during the Triassic (Hu et al. 2006). The Songpan Garze Terrane, which no longer provides sediment directly into the South China Sea, represents an accretionary complex formed during this collision between north and southern China (Zhou & Graham 1996; Huang et al. 2003; Enkelmann et al. 2007).

On the western side of the basin, the Indochina Peninsula is dominated by a separate tectonic block, but one that was also involved in the Indosinian Orogeny (Carter et al. 2001). Indochina has undergone more recent deformation as a result of the emplacement of flood basalt sequences in the Central Highlands of Vietnam during the Late Miocene (Carter et al. 2000). This is one of a number of rather enigmatic volcanic provinces around the basin, which provide isotopically unique sediment from newly emplaced primitive volcanic sequences. Likewise, on the eastern side of the basin, the island arc of Luzon has provided some sediment into the basin in the recent geological past, although it is worth noting that its location to the east of the main deep-water basin is a relatively recent development following the northwards drift of the arc and the collision of the arc with the southern margin of mainland Eurasia (Fig. 3). During the latest Miocene, plate tectonic reconstructions show that Luzon was positioned somewhat to the south of the basin and has only been able to supply sediment to the basin in the last few million years (Hall 2002).

The rivers draining the island of Borneo are probably the least well defined of any potential source around the sea, but they too have had a
limited impact on sediment flux into the ocean, partly because they were separated from the present basin by a palaeo-South China Sea until around 16 Ma when the southern margin of the basin, dominated by the Dangerous Grounds, began to collide with the northern margin of Borneo (Hutchison 2005; Clift et al. 2008a). Even since that time, direct sediment supply from Borneo into the deeper parts of the South China Sea has been restricted by the tectonic topography of the Dangerous Grounds (Hutchison & Vijayan 2010), and particularly by the long and deep North Borneo Trough that separates Borneo from the main part of the sea and which acts as a very effective sediment trap (Hutchison 2010), so that only plumes of suspended relatively fine-grained sediment can be transported deep into the basin.

Consideration of such plate tectonic reconstructions is very important when making provenance assessments because it is clearly impossible to derive sediment from a tectonic block that was not in the vicinity of the South China Sea at the time of sedimentation. Conversely, convincing provenance data can help us to better define the tectonic development of SE Asia by showing which blocks were present at any particular time.

**Sediment mixing on the continental shelf**

Provenance analysis of sediment in the deep basin does not necessarily reflect the contribution of a single river system to the overall basin budget because of filtering of the signal through the continental shelf. Changes in sea level and, therefore, in the position of the river mouth relative to the continental shelf edge affects how important that river will be in supplying sediment into the deep basin. For example, the Mekong River appears to have been important in supplying sediment into the deep SW part of the basin during sea-level lowstands, but has been relatively cut-off following post-glacial sea-level rise (Szczuciński et al. 2013). The same is true of the Red and Pearl rivers, with their wide continental shelves. In contrast, areas where the continental shelf is very narrow, such as offshore central Vietnam, may be important sources of sediment to the deep basin even during periods of lowstand.

**Fig. 2.** Simplified tectonic terrane map of East and SE Asia showing the major blocks discussed in this paper and the courses of the rivers. After Metcalfe (1996).
relatively high sea level (Schimanski & Stattegger 2005; Lahajnar et al. 2007; Szczuciński et al. 2009).

Reworking on the continental shelf is important in influencing the net contribution to the deep basin. Sediment originating from the Mekong River catchment to the deep basin is now delivered, not from modern sediment discharge at the river mouth, but from erosion of the lowstand Mekong delta at the shelf edge (Dung et al. 2013). Newly delivered sediment is, instead, sequestered close to the coast. Geochemical data from South Asia has reinforced the idea that the continental shelf is a location in which sediment from different sources may be mixed via longshore transport and that reworking of older deposits, due to storms or bottom currents, may influence the composition of sediment delivered to the deep sea, and, in particular, this does not necessarily reflect the large river mouth in close proximity (Limmer et al. 2012).

As a result, fingerprinting of sediments in modern rivers can help to resolve the influence of onshore basins in supplying sediment layers to the continental shelf and to the deep ocean. Sediment deposited in the offshore, even close to the river mouth, cannot be considered a reliable fingerprint of provenance.

Clay minerals

Clay minerals can provide an important source of sediment provenance data, especially in distal fine-grained sediment sequences where single grain methods may not be applicable, most sand being sequestered on the shelf in many systems. The method is based on the contrasting mineralogy of sediments in the different river systems that currently supply the basin and the documented changes in clay mineralogy of the modern seafloor across the basin (Chen 1978). The method suffers from being only ‘semi-quantitative’, in that relative proportions of different key minerals are determined
by a variety of methods such as X-ray diffraction data, with different methods producing slightly different estimates that may not all be completely in accordance with one another (Holzappfel 1985; Moore & Reynolds 1989; Hillier 2003). The accuracy of the methods is not entirely clear and is best applied to resolving relatively larger differences in assemblages (>5%). Smaller differences in clay mineral assemblage cannot be considered robust and are not considered as effective provenance tools.

A number of studies have now highlighted the fact that the Pearl, Red and Mekong rivers, as well as smaller rivers draining Borneo and the Malay Peninsula, are characterized by different clay mineral assemblages that roughly correlate with the intensity of chemical weathering in the fluvial basin (Liu et al. 2007b, 2012). Although some of the differences in clay mineral content are related to source rock compositions, much of the contrast is the result of a variable intensity of chemical weathering, which in turn is linked to the topography. The intensity of the summer monsoon, which provides much of the moisture and related warmth to the area, also modulates the rate of chemical weathering and, thus, the clay mineralogy (West et al. 2005), accounting for the abundance of smectite and kaolinite in the more tropical regions of the southern South China Sea, especially the Sunda Shelf (Aoki 1976; Chen 1978).

Composition is important in the case of the rivers draining Luzon because these rivers are rich in smectite, which is a product of the chemical breakdown of volcanic rocks (Liu et al. 2009) that are abundant in this volcanic arc, as well as in the ranges of central Vietnam (Jagodziński 2005). In contrast, although the rocks of Taiwan originated as sediments on the passive margin of China, these have experienced significant metamorphism and are now dominated by illite and chlorite, which largely represent the products of physical erosion of the high mountains in Taiwan rather than the products of chemical weathering of pre-existing bedrock. Meanwhile, sediment delivered by the Pearl River is dominated by kaolinite with some illite and chlorite influx (Liu et al. 2007a). Further south, the Mekong submarine delta, which is the primary depocentre for the modern river, is dominated by illite with lesser amounts of smectite, kaolinite and chlorite (Szczyciński et al. 2013). Further illite is supplied from the rivers of northern Borneo (Liu et al. 2007c).

Differences in clay mineralogy of Holocene and recent sediment in the deep basin, as well as those found in older deposits, have been used to separate the different contributions from potential sources (Boulay et al. 2005; Liu et al. 2010b). Figure 4 shows a triangular diagram and the type of mixing arrays that have been proposed in the past to separate the influence of these different source terrains. In this particular example, we see that sediment from Ocean Drilling Program (ODP) Site 1144 lies close to the field of Taiwan, allowing the provenance of this deposit to be constrained to this island (Hu et al. 2012). Similar approaches have been applied to modern sediment in the northern South China Sea (Z. Liu et al. 2003; J. Liu et al. 2014) and used to infer sediment transport directions, driven by bottom currents.

Unfortunately, this method is prone to problems because it is based on the assumption that river clay mineral assemblages have been the same in the past as they are at the present. There seems little doubt that environmental changes have caused changes in clay assemblages over long periods of geological time (Clift et al. 2002; Clift et al. 2014; Wan et al. 2007), largely as a result of changes in the monsoon, so that the end members cannot be considered stable over long periods of time as the climate in SE Asia has evolved. Furthermore, the clay mineral evidence for the origin of the Holocene sediment at ODP Site 1144 appears to show that the sediment is not entirely derived from Taiwan, despite the fact that other proxies indicate that it would seem to be most likely (Hu et al. 2012). Why then do these data not plot directly in the modern Taiwan field? The difference was attributed by Hu et al. (2012) to weathering of the sediment on the exposed continental shelf during the Last Glacial Maximum and then reworking during the Holocene before final sedimentation. Sediment buffering between source and sink is often associated with chemical weathering (Lupker et al. 2012), the net result of which is a transformation of the clay mineral assemblage relative to the modern river composition. The method is also open to error if the sequences are affected by burial diagenesis, as may be the case in deep boreholes where burial heating can be significant.

An additional complexity was recognized by Steinke et al. (2008), who showed that the clay mineralogy at any one site on the Sunda Shelf was largely controlled by sea level, which acted to disrupt the large drainage systems that existing during the Last Glacial Maximum. In particular, sediment rich in kaolinite derived from the south, as well as from the exposed Sunda Shelf itself, is strongly reduced during the Holocene as flux from the Mekong increased relative to the southern sources, as their river mouths retreated from proximity to the shelf edge. Moreover, these workers recognized that, as sea level rose, little of this material was reaching the deep basin, but, rather, was sequestered in terrestrial flood plains and submarine delta clinoforms. Furthermore, studies of the clays around the Mekong delta shows that these differ from east to west in the modern system, partly reflecting the preferential settling of some clays close to the river
mouth (Xue et al. 2014). Sediments from the east, on the South China Sea side of the delta, look like the Mekong River itself, while those to the west have much less illite and chlorite, and imply reduced fine particle inputs from the Mekong River towards the Gulf of Thailand.

Most seriously of all is the recognition that the composition of the modern rivers is not in equilibrium and may bear little resemblance to the natural state of the river before a few thousand years ago. It has increasingly been demonstrated that human settlement has had a major impact on landscape evolution and, in particular, has encouraged the rapid erosion of soils as agriculture became prevalent over the last several thousand years (Syvitski et al. 2005; Syvitski & Kettner 2011). Data from the delta of the Pearl River suggests that human settlement in southern China had become highly disruptive to the river by around 2500 years ago (Hu et al. 2013) and similar patterns might be anticipated for Indochina. More recently, the development of more sophisticated industrial cultures has resulted in the damming of major river systems so that the rate of sediment delivery to the ocean has been reduced in recent times.

A number of studies have now shown that the alteration state of sediment reaching the delta since the onset of agriculture has increased (Bayon et al. 2012; Hu et al. 2013), consistent with the increased influence from anthropogenic contaminants at that time (Zong et al. 2010). For example, the Pearl River shows a large dominance of kaolinite in the modern system, but cores taken in the river mouth show that the situation only arose at around 2500 ka and that before that time smectite was much more important, as shown by the difference between modern Pearl River sediments and those from the Holocene delta (Fig. 4). Thus, to assume that much of the smectite in the South China Sea was derived from Luzon or from central Vietnam prior to 2500 ka would be incorrect, and before that time similar size variations can be linked to climate change during the early Holocene when the monsoon was stronger (Wang et al. 2001; Dykoski et al. 2005). Unless the impact of modern human disruption to river systems and past climate change can be accounted for, therefore, it seems that clay mineralogy by itself is a rather unreliable provenance proxy unless simply applied to the present day. Although in deep-water settings, where coarser sediment is not found, it may be one of the few methods that can even be attempted.

**Bulk geochemistry**

An alternative method to using weathering regime as a tool for sediment provenance is to look at the

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**Fig. 4.** Clay mineralogy of the Holocene sediments analysed from ODP Site 1144 (Hu et al. 2012) compared to modern river and offshore sediment compositions in the possible source regions (Liu et al. 2010a). Holocene Pearl River data are from Hu et al. (2013). The arrow shows how the Pearl River composition changes after 2.5 ka.
bulk geochemistry. This approach is potentially useful in sediment of many different grain sizes, including the muds that are not easily analysed using the thermochronology methods discussed below. Care must, however, be exercised in comparing geochemical data between sediments of approximately similar size as hydrodynamic sorting of minerals and preferential alteration are methods by which different grain sizes can end up with quite different chemistries, despite coming from the same original source.

The premise of this particular method is that elements that are particularly soluble in aqueous solution are depleted in rocks and sediment, which has experienced more chemical alteration, and are mobilized and removed compared to immobile elements such as aluminum or silicon (Nesbitt & Young 1982; Galy & France-Lanord 1999). This means that rivers in warmer, wetter environments tend to have more altered sediment in them than those in drier, colder places. Nonetheless, it has to be recognized that topography is also important because fast-flowing, high-energy rivers from steep mountains, such as Taiwan, tend to transport sediment quickly, leading to less chemical weathering. Liu et al. (2007b) surveyed the rivers of the South China Sea and showed that the Pearl River tended to include sediment that was more weathered than that in the Mekong, which in turn is more weathered than that in the Red River. This study was able to do this using the geochemical proxy ‘chemical index of alteration’ (CIA: Fig. 5), which was developed for use in soils (Nesbitt & Young 1982). Despite showing some overlap, Liu et al. (2007b) did highlight differences between the rivers when a limited grain size was considered. The CIA proxy was not developed for marine sediments and should probably not be applied in this situation, but has, nevertheless, a long history of being used to look at the alteration state of deep-water marine sediments.

Application of proxies such as the CIA require that only limited grain-size fractions should be compared with one another because of the effects of the hydrodynamic sorting of different mineral species that has a dominant control on bulk chemistry. Furthermore, marine sediments are often contaminated by biogenic calcium, which needs to be corrected for if the proxy is to have any meaning whatsoever (Singh et al. 2005). Likewise, sodium may be increase in marine sediments as a result of seawater in pores, which has to be flushed from

![Fig. 5. Chemical index of alteration (CIA) plotted against silica content for sand and silt samples from the modern Red River (black dots) and older borehole samples from the Hanoi Basin labelled with depositional age (open circles) (Clift et al. 2008b). Fields for modern trunk Red, Mekong and Pearl rivers are from Liu et al. (2007b), and include only fine-grained sediments.](http://sp.lyellcollection.org/)
the sample if the analysis is to have any significant meaning. However, with appropriate sample preparation, these issues can be overcome.

Figure 5 shows how the data presented by Liu et al. (2007b), which was generated after decarbonation, compares with a series of more sandy sediments from the Red River alone (Clift et al. 2008b). What is apparent is that more sandy material tends to have lower CIA values, but that the scatter of data is very significant. The distribution of values within a single river is so great that it is probably unrealistic to hope to find a characteristic value for any given river system. This also suggests that using CIA or other chemical weathering proxies as a way to track provenance is probably useless unless this is restricted to the present day and/or to a very limited grain-size range. Geochemical studies of the Yellow Sea region do, however, show that with appropriate filtering for grain size and with correction for biogenic carbonates that some degree of provenance can be successfully achieved using bulk sediment major-element chemistry (Kim et al. 1999; Lim et al. 2013).

Other types of bulk sediment geochemical data have been used in the past to track provenance, most notably rare earth elements (REEs). These are more uniformly immobile to aqueous chemical weathering and might be expected to better preserve the average composition of the source bedrocks that are generating the sediment. However, much of the upper continental crust tends to have a gentle enrichment in light REEs (LREEs) and to be often similar between different basins (Rudnick & Gao 2003). Total REE content is not useful because the REEs are strongly concentrated in a number of heavy minerals so that the concentrations are largely a reflection of the absolute abundance of these minerals in the sample selected (Garc¸on et al. 2014). This in turn is related to hydrodynamic sorting during sedimentation, so that samples taken from different parts of a single bedform might show a quite different REE content. Studies of the REEs in the Red River showed a wide scatter of REE concentrations and ratios, but no systematic difference in REE character between different tributaries, suggesting that large tracts of the upper continental crust have quite similar degrees of enrichment and that, therefore, large rivers with diverse catchment geologies will often have a similar chemical character (Clift et al. 2008b).

In contrast, some studies claim that key REE ratios such as La/Yb or Gd/Yb, as well as the relative europium anomaly, can be distinctive of different source regions, although this will be less true in larger river basins where more averaging can occur (Vital & Stattegger 2000; Jung et al. 2012). As with other forms of chemistry, REE chemistry may be affected by variations in grain size and by fractionation of heavy minerals, and so are best used on a only a limited size fraction.

In general, REEs do not seem like promising provenance proxies, with the exception of potentially finding sediment derived from the Luzon Arc, which, being a more primitive, mantle-derived piece of crust, would be associated with more LREE-depleted compositions (i.e., low La/Yb values).

### Bulk isotope character

Combined Nd and Sr isotopes have an established track record in terms of resolving provenance in many basins worldwide and specifically in the South China Sea (Clift et al. 2002; Li et al. 2003). Nd, in particular, is generally recognized as being immobile to chemical weathering, and is not fractionated by erosion, weathering and transport (Goldstein & Jacobsen 1988). Sr, however, is more mobile, and is fractionated so that more weathered sediments tend to have higher $^{87}\text{Sr}/^{86}\text{Sr}$ values that represent both source composition and weathering intensity (Derry & France-Lanord 1996). Nd is a powerful provenance proxy, although, again, grain size may have an influence because Nd is largely dominated by monazite content (Garçon et al. 2013). The method has most successfully been applied to fine-grained sediments, where it gives an estimate of the relative age of the continental crust from which the sediment is derived (Allegre & Ben Othman 1980), although it could be applied to coarser materials provided that these data were compared with other coarse sediment measurements.

Figure 6 shows the range of measured Sr and Nd isotope ratios for a series of modern rivers draining into the basin, together with selected analyses from marine cores, largely in the northern part of the sea. It is clear that the modern Pearl River and some parts of the modern Red River are characterized by very high $^{87}\text{Sr}/^{86}\text{Sr}$ values and might be resolvable in this respect. However, this ignores the fact that these rivers are anthropogenically disrupted and are presently carrying much more altered sediment than has been typical for the Holocene.

Sediment in rivers draining Luzon is truly unique in Sr and Nd isotopes, in having both very low $^{87}\text{Sr}/^{86}\text{Sr}$ values and very high $\varepsilon_{\text{Nd}}$ values. Because Nd is resistant to change during weathering, the influence of Luzon in providing sediment should be easily resolved using this approach. However, many possible sources cluster around $^{87}\text{Sr}/^{86}\text{Sr}$ values of 0.72 and $\varepsilon_{\text{Nd}}$ values of $-10$, including the modern Mekong and Red rivers, Taiwan, and the Holocene of the Pearl River. Not surprisingly, many of the cored sediments analysed for these isotopes also plot in this region, suggesting that this
approach may not be the best at resolving sources in SE Asia. It is noteworthy that ODP Site 1144 shows overlap with Taiwan and has the lowest $^{87}\text{Sr}/^{86}\text{Sr}$ values, which do not seem unique to that source compared to sources in southern China or Indochina. The relative lack of variation in $\varepsilon_{\text{Nd}}$ values reflects the fact that many of the blocks in SE Asia have similar crustal residence ages, but it should be noted that when rivers have connected to sources in the much older Yangtze Craton in the past then much lower $\varepsilon_{\text{Nd}}$ values were recognized, most notably in the Red River delta (Song Hong-Yinggehai Basin) (Clift et al. 2006a).

Apatite fission track

Comparison of fission-track ages between sediments and bedrock sources has been a powerful method for sediment provenance for some time (Hurford & Carter 1991; Carter 2007), and is one with some of the greatest potential in the South China Sea. The apatite fission-track method records cooling of rocks through approximately 60–125°C over timescales of 1–10 Ma (Green et al. 1989) and, provided that the sediment has not been buried and reheated again since deposition, allows single grains to be tied to sources with unique exhumation histories. This favours sedimentary systems in SE Asia because different parts of the margins have experienced different deformation and erosion histories. Fission-track methods are best applied to sediment that is fine sand or coarse in grain size, reflecting the need to measure track densities and the 16 µm length of new tracks. However, coarse silts can be used in extreme circumstances.

Luzon is less easily recognized in this approach because basalts that dominate the arc are not rich in apatite but should be Oligocene or younger if present, given the known range of magmatism.

Fig. 6. Sr and Nd isotopic plot showing the variability in Holocene and modern sediments from ODP Site 1144 (Hu et al. 2012) and in Lower Miocene–Recent sediments at ODP Site 1148 compared to modern potential sources around the South China Sea. The diagram shows the general similarity of the sediment with modern Taiwanese rivers and bedrock samples (Chen & Lee 1990; Lan et al. 2002), and the differences with sediments in the modern Pearl River (Liu et al. 2007b) and with potential volcanic sources in the Philippine island of Luzon (Zhou et al. 2002). The Neogene sediments at ODP Site 1148 show an overlap with both Taiwanese rivers and the Holocene sediments of the Pearl River Estuary (Hu et al. 2013). Data from the Red and Mekong rivers are from Liu et al. (2007b) and Clift et al. (2008b).
(Defant et al. 1989). Figure 7 shows a range of kernel density estimate (KDE) plots designed to demonstrate the most likely cooling ages for each source (Vermeesch 2012). Taiwan is unique in this system, with very young ages that post-date the start of collision at c. 5 Ma, making grains derived from this island easy to resolve within mixtures. Fission-track cooling ages from Cathaysia in southern China, and from the Khorat Plateau, the Vietnamese Central Highlands and the Kontum Massif in Indochina have a similar range of central ages, especially around 30–60 Ma. This age reflects cooling driven by the break-up of the South China Sea and the subsequent erosional degradation of the rifted margins. However, because this process is common, sediment derived from these different sources cannot be resolved with this technique, with the possible exception that the Central Highlands has slightly younger ages than other parts of the western margin because of rejuvenation of that region during emplacement of volcanic sequences at approximately 8 Ma (Carter et al. 2000).

An intermediate grouping of fission-track ages are those grains with cooling ages of c. 20 Ma. These are typical of bedrocks in the Red River Fault Zone, but are found throughout the SE flank of the Tibetan Plateau, especially in the gorges of SW China (Clark et al. 2005) and are not specific to the fault zone. This cooling is related to the deformation and rock uplift associated with the southeastward propagation of the Tibetan Plateau during the Miocene (Schoenbohm et al. 2006). This explains why grains of this age dominate the modern Red River (Fig. 7) which derives its sediment from this region (Clift et al. 2006b). In contrast, the Mekong River has a subtly different age spectrum, with a younger peak at approximately 14 Ma and a long tail of older ages. The younger aged grains in the Mekong are believed to be derived from younger sources in its upper reaches in Tibet and the older ages from the lowlands of Indochina, including the Khorat Plateau (Clift et al. 2006b). For whatever reason, the fission-track ages of these two rivers contrast with each
other and the presumed spectrum for the Pearl, which must be dominated by the older ages from Cathaysia.

Apatite fission-track thermochronology thus holds great promise in being able to distinguish sediment from the three major rivers, as well as Taiwan, even if there is more ambiguity concerning grains dated at 30–60 Ma, which may be eroded from either the basin’s western or northern margins.

**Muscovite Ar/Ar dating**

There is much less existing $^{40}$Ar/$^{39}$Ar data from potential bedrock sources around the South China Sea compared to fission-track data, yet this method is also useful at pinpointing sources and can be complementary to the lower temperature method. The $^{40}$Ar/$^{39}$Ar method has been applied in several Asian provenance studies (Najman et al. 1997; White et al. 2002; Szulc et al. 2006; Hoang et al. 2010) and allows the age when each single mica grain cooled through an isotherm of around 300°C to be determined (Hodges 2003). Triassic cooling ages are common in SE Asia and are related to the Indosinian Orogeny (Huang et al. 2003; Lepvrier et al. 2004). However, there are important discrepancies to this general picture, and resolvable differences between sources around the northern and western edges of the basin that make this a good provenance tool. Single-grain mica dating requires substantial crystals and even multiple grain methods mean that the method is really only applicable to sands, generally limiting its application to the proximal parts of a given sedimentary system.

Analysis of the modern Red and Mekong rivers shows that the Mekong has a generally younger set of Indosinian micas than seen in the Red River; 150–210 Ma compared to 210–250 Ma (Clift et al. 2006b) (Fig. 8). This difference must reflect contrasting cooling ages of Indosinian metamorphic rocks in the two drainage basins and when transferred to the marine realm would allow sediment from each river to be resolved within offshore depocentres. Both rivers contain muscovite micas dating to c. 20–30 Ma, similar to the rocks of the

![Fig. 8. Probability density diagrams for Ar/Ar ages of detrital muscovites within the Mekong and Red rivers compared to known ranges from possible source regions. Central Vietnam data are from Lepvrier et al. (1997) and Nagy et al. (2000). Ailao Shan/Red River Fault Zone data are from Leloup et al. (1993, 2001), Harrison et al. (1996), P. L. Wang et al. (1998), Jolivet et al. (1999) and Maluski et al. (2001), and data from Songpan Garze are from Reid et al. (2005). Qiangtang Block data are from Kapp et al. (2000).](http://sp.lyellcollection.org/)

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Red River Fault Zone and equivalent structures across SE Tibet and Indochina, such as the Mae Ping and Three Pagoda faults (P. L. Wang et al. 1998; Morley 2002). As a result, these ages are only partially source diagnostic, and are only related to sources from SE Tibet. The timing of motion on these faults is commonly Oligo-Miocene, making it impossible to resolve between the different sources. Only the Xianshuie–Xiaojiang Fault of SW China has continued rapid motion until recent times (E. Wang et al. 1998). Nonetheless, the presence of 20–30 Ma micas does at least rule out erosion from southern China. \(^{40}\text{Ar}/^{39}\text{Ar}\) data from southern China–Cathaysia is sparse, but that which does exist tends to be of Indosinian age, 207–215 Ma (Wang et al. 2005), but critically slightly younger than dated basement rocks in central Vietnam, which tend to cluster at about 240 Ma (Lepvrier et al. 1997). This latter tectonic block is, however, marked by a minor population of micas dating at 110–130 Ma, which is not found further north.

Luzon is not a significant source of muscovite because of its dominant basaltic oceanic arc character and any mica derived from the Central Ranges of Taiwan must have very young ages of <5 Ma if they have been reset by the orogeny on that island. If muscovite grains from Taiwan have not been reset — for example, in the lower structural levels of the thrust wedge — then they would carry the same array of ages as seen in SE China and would not be resolvable using this method. Although the \(^{40}\text{Ar}/^{39}\text{Ar}\) method is generally slower and more expensive than that of the fission track method, it can form an important tool for quantifying erosion budgets in the South China Sea because of the array of unique sources that characterize some of the major rivers, especially along the western margins of the basin.

Zircon U–Pb dating

U–Pb dating of zircon has become one of the most popular forms of provenance analysis, following the improvement in dating technology that now allows large numbers of grains to be quickly and cheaply dated using laser ablation inductively coupled plasma mass spectrometry (ICP-MS) instead of the more traditional mass spectrometry. This method can only be applied to grains that are large enough to be dated, which usually means >50 \(\mu\text{m}\) (coarse silt), as smaller grains are too small to be targeted by a laser ablation ICP-MS that dominates the approach. This means that the method is less useful in the most distal, deep-water deposits in the basin centre.

U–Pb ages in zircon are considered to reflect the time of zircon crystallization at around 750°C (Hodges 2003), so that they may be interpreted to record the last growth phase in a rock’s history. However, zircons are known to be resistant to physical abrasion during erosion and transport, as well as to chemical weathering, making them susceptible to multiple phases of reworking (DeCelles et al. 2004; Campbell et al. 2005). Although it is impossible to say precisely where a single grain with a single age is derived from, it is possible to suggest the most likely source rock unit for many grains, making U–Pb dating a powerful provenance tool in this area. Because the different possible source terrains have unique age spectra, it is usually possible to at least exclude possible dominant sources based on the U–Pb age of the sediment.

As with the \(^{40}\text{Ar}/^{39}\text{Ar}\) ages, Triassic Indosinian ages are very common in many of the source regions and tend not to be diagnostic by themselves. Figure 9h shows that the Qiangtang Block of Tibet and its equivalent terrane, Sibumasu, in SE Asia (Fig. 2) are particularly dominated by this age range and have relatively few older grains, resulting in a potentially unique signature. Likewise, Red River Fault Zone rocks yield Cenozoic ages that are synchronous with the motion on that fault (Fig. 9g), and which are effectively unknown outside this zone and presumably equivalent fault zone rocks in the other major strike-slip zones of SE Tibet–SW China. In any case, observations from the Mekong and Red rivers indicate that the fault zone rocks are not major contributors to the net flux to the ocean (Fig. 9a, b). A high proportion of grains dated between 700 and 1000 Ma is typical of bedrock from the Yangtze Block. These grains are also common in Cathaysia-derived sediment, but these have a slightly older peak at about 900 Ma rather than about 800 Ma, and also contain significant populations clustered around 1800 and 2400 Ma, which are present but are not so abundant in the Yangtze Block. The Songpan Garze terrane shows intermediate character, having relatively few 700–1000 Ma grains, but common approximately 1800 and 2400 Ma grains, as well as a strong 400–500 Ma population (Fig. 9d), which is unknown in the Yangtze Block, rare in Cathaysia but is also known in Indochina in the Khorat Plateau, the Kontum Massif and the Central Highland areas that fringe the western edge of the South China Sea.

Consideration of the spectra of zircon ages in modern Red and Mekong river sediments shows that while they share many of the same populations there are differences between these systems that reflect the contrasting ages in the bedrock sources. The Red River has a much stronger peak at around 800 Ma compared to the Mekong River, which may be interpreted to indicate more erosion from
Table 1. Summary of the different provenance methods reviewed in this paper and simplified results for the different major source terrains that are supplying sediment to the modern South China Sea

<table>
<thead>
<tr>
<th>Method</th>
<th>Taiwan</th>
<th>South China</th>
<th>Indochina</th>
<th>Luzon</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bulk sediment chemistry</td>
<td>Variable</td>
<td>Variable</td>
<td>Variable</td>
<td>Volcanic</td>
</tr>
<tr>
<td>Clay minerals</td>
<td>Chlorite and illite</td>
<td>Kaolinite, smectite and illite</td>
<td>Kaolinite, smectite and illite</td>
<td>Smectite</td>
</tr>
<tr>
<td>Nd isotopes</td>
<td>−9 to −11</td>
<td>−8 to −11</td>
<td>−9 to −13, except the Central Highlands</td>
<td>+4 to +8</td>
</tr>
<tr>
<td>Apatite fission track</td>
<td>&lt;3 Ma</td>
<td>30–60 Ma</td>
<td>17–23 Ma (S. Chay Massif), 27–50 Ma (Kontum)</td>
<td>&lt;45 Ma</td>
</tr>
<tr>
<td>Zircon fission track</td>
<td>&lt;5 Ma</td>
<td>90–120 Ma</td>
<td>90–550 Ma</td>
<td>N/A</td>
</tr>
<tr>
<td>Ar–Ar muscovite</td>
<td>&lt;6 Ma</td>
<td>200–220 Ma</td>
<td>160–250 Ma</td>
<td>N/A</td>
</tr>
<tr>
<td>U–Pb zircons</td>
<td>&lt;500 and c. 1800 Ma</td>
<td>800–1500 Ma</td>
<td>240–450 and c. 1800 Ma</td>
<td>&lt;45 Ma</td>
</tr>
</tbody>
</table>

Fig. 9. Probability density diagrams showing the detrital U–Pb ages of zircon grains known from the major tectonic blocks in SE Asia. Data for Ailao Shan and Red River Fault Zone rocks are from Schärer et al. (1990, 1994), Zhang & Schärer (1999), Nagy et al. (2000) and Carter et al. (2001). Data for Cathaysia are from Li et al. (1989, 2001, 2005). Data for Indochina sources (including the Khorat Plateau, the Kontum Massif and the Central Highlands of Vietnam) are from Carter & Moss (1999), Carter et al. (2001), Nagy et al. (2001) and Carter & Bristow (2003). Data for the Songpan Garze Terrane are from Hu et al. (2005), Bruguier et al. (1997) and Weislogel et al. (2006, 2010). Data for the western Yangtze Craton are from Sun et al. (2009), Xu et al. (2008) and Yang et al. (2005). Data for the Qiangtang Block are from Roger et al. (2000, 2003).
the Yangtze Craton, consistent with the known extent of the modern drainage. Furthermore, Clift et al. (2006b) demonstrated that the Indosinian peak in the Mekong River is slightly younger than in the Red River, a difference that could be exploited to resolve and quantify relative input from the two drainages.

Although rivers from Luzon have not yet been analysed for zircon U–Pb ages, those sources are not considered to be important to the zircon budget of the basin because of the relative lack of zircons in basaltic and basaltic andesite lavas in that arc. When they are present then they must be younger than the Oligocene when that arc became active and thus could only be potentially confused with sediment from the Red River Fault Zones. Metamorphic rocks in Taiwan were not heated sufficiently to reset the U–Pb system in zircon during that arc–continent collision and are anticipated to yield typical Cathaysian ages based on the sources to that part of the passive margin (Lan et al. 2014). Fortunately, Taiwan has a unique signature in terms of lower temperature thermochronology systems so that its contribution is not dependent on U–Pb zircon dating. It appears that U–Pb dating can be a useful component to a provenance study in the South China Sea, but often yields mixed or ambiguous signals because many of the major rivers share similar aged bedrocks.

Discussion and conclusions

In this review of the major provenance methods now applied in the South China Sea, it is clear that a single method is generally insufficient to quantify the relative sediment flux contributions from multiple terrains into a given sediment. Table 1 shows a summary of the character of the sources on three sides of the basin. There is relatively little information about the character of sources in Borneo or peninsular Malaysia, both of which may have been more important in the past when sea level was lower and when the Molengraaff River may be fed material to the Sunda Shelf and into deep-water basin, at least at its SW end (Voris 2000; Steinke et al. 2008; Hanebuth et al. 2011). When sea level was higher, sediment flux into the South China Sea from these sources was generally minimal. More work needs to be carried out to look at additional methods that should be effective in this basin, such as heavy mineral studies, but at the time of writing insufficient is known about the typical mineralogy of the modern drainages to assess whether this would actually work on ancient sequences or not. Only the continental shelf of Vietnam has been investigated for heavy minerals (Jagodziński 2005).

What is clear is that proxies that are strongly affected by chemical weathering make unreliable provenance proxies for sediments predating the last few hundred years. The modern rivers are known to be significantly disrupted in terms of bulk sediment chemistry and clay mineralogy, so that these compositions cannot be used to fingerprint ancient sediments. The same is also true of Sr isotopes. Even if the pre-modern composition can be fixed by looking at Holocene sediments under the most recent cover, it must be realized that the sediment composition is influenced by the changing climate, especially the intensity of the East Asian Monsoon.

Nd isotopes are only of limited use, except to resolve input from the young and primitive rocks of Luzon and in the case where rivers are draining sediment out of central southern China where the ancient crust of the Yangtze Craton results in very negative $\varepsilon_{Nd}$ values. Nd compositions in Indochina and southern China are very similar and largely overlap, resulting in serious ambiguity in terms of resolving the effects of different sources. However, coherent changes may be detected and could be useful in discerning some changes depending on the size and heterogeneity of the basement rocks involved, since not all parts of Cathaysia or Indochina are identical. Erosion of the Miocene volcanic rocks from the Vietnamese Central Highlands would be anticipated to yield more positive $\varepsilon_{Nd}$ values than is typical, for example, for the rest of the Vietnamese coast.

The most useful provenance proxies are those related to detrital thermochronology, but even here it is generally most effective to use more than one method to achieve a robust result. Apatite fission track is probably the best single method because of the contrasting timing of moderate amounts of exhumation around the basin, which allow most sources to be distinguished, although there is some overlap between parts of Indochina and southern China, not only in apatite but also in muscovite Ar/Ar. Fortunately, these sources can be separated on the basis of U–Pb zircon dating. Provenance in the South China Sea is complex, but it is not an impossible task and one that is essential to solve if we are to reconstruct the palaeogeography, drainage evolution and chemical weathering regimes in SE Asia during the Cenozoic. Because this is a classic area for collision tectonics, associated drainage reorganization and the development of the monsoon, the sedimentary archives in the South China Sea are invaluable to efforts to address these issues and require provenance control if the records are to be understood.

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