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Nd, Sr isotopes and elemental geochemistry of surface sediments from the South China Sea: Implications for Provenance Tracing

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ABSTRACT

The mineralogy, major and trace elements, and neodymium and strontium isotopes of surface sediments in the South China Sea (SCS) are documented with the aim of investigating their applicability in provenance tracing. The results indicate that mineralogical compositions alone do not clearly identify the sources for the bulk sediments in the SCS. The Nd isotopic compositions of the SCS sediments show a clear zonal distribution. The most negative ε_{Nd} values were obtained for sediments from offshore South China (−13.0 to −10.7), while those from offshore Indochina are slightly more positive (−10.7 to −9.4). The Nd isotopic compositions of the sediments from offshore Borneo are even higher, with ε_{Nd} ranging from −8.8 to −7.0, and the sediments offshore from the southern Philippine Arc have the most positive ε_{Nd} values, from −3.7 to +5.3. This zonal distribution in ε_{Nd} is in good agreement with the Nd isotopic compositions of the sediments supplied by river systems that drain into the corresponding regions, indicating that Nd isotopic compositions are an adequate proxy for provenance tracing of SCS sediments. Sr isotopic compositions, in contrast, can only be used to identify the sediments from offshore South China and offshore from the southern Philippine Arc, as the $^{87}Sr/^{86}Sr$ ratios of sediments from other regions overlapped. Similar zonal distributions are also apparent in a La–Th–Sc discrimination diagram. Sediments from the west margin of the SCS, such as those from Beibuwan Bay, offshore from Hainan Island, offshore from Indochina, and from the Sunda Shelf plot in the same field, while those offshore from the northeastern SCS, offshore from Borneo, and offshore from the southern Philippine Arc plot in distinct fields. Thus, the La–Th–Sc discrimination diagram, coupled with Nd isotopes, can be used to trace the provenance of SCS sediments. Using this method, we re-assessed the provenance changes of sediments at Ocean Drilling Program (ODP) Site 1148 since the late Oligocene. The results indicate that sediments deposited after 23.8 Ma (above 455 mcd: meters composite depth) were supplied mainly from the eastern South China Block, with a negligible contribution from the interior of the South China Block. Sediments deposited before 26 Ma (beneath 477 mcd) were supplied mainly from the North Palawan Continental Terrane, which may retain the geochemical characteristics of the materials covered on the late Mesozoic granitoids along the coastal South China. For that the North Palawan Continental Terrane is presently located within the southern Philippine Arc but was located close to ODP Site 1148 in the late Oligocene. The weathering products of volcanic material associated with the extension of the SCS ocean crust also contributed to these sediments. The rapid change in sediment source at 26–23.8 Ma probably resulted from a sudden cessation of sediment supply from the North Palawan Continental Terrane. We suggest that the North Palawan Continental Terrane drifted southwards along with the extension of the SCS ocean crust during that time, and when the basin was large enough, the supply of sediment from the south to ODP Site 1148 at the north slope may have ceased.

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1. Introduction

The provenance of sediments and sedimentary rocks is an important component of the paleoceanography and tectonic history of a region. Geochemical methods have generally been used in tracing the provenance of sediments and sedimentary rocks, along with analyses of accessory detrital minerals, in particular the U–Pb geochronology of detrital zircon (Fedo et al., 2003 and references therein). In the absence of detrital zircon, elemental and isotopic geochemical methods are powerful tools in identifying provenance (McLennan et al., 1993), including analyses of rare earth elements (REEs) (Bhatia, 1985; McLennan, 1989) and immobile elements (Bhatia and Crook, 1986; Fralick and Kronberg, 1997). Sm–Nd isotope systematics are generally unaltered during sedimentary processes (McCulloch and Wasserburg,

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1978), meaning that Nd isotopes provide reliable evidence of provenance (Goldstein et al., 1984; Miller and O'Nions, 1984, 1985; Goldstein and Jacobsen, 1988). Previous studies have also investigated other isotopic systems, including Sr and Pb (McLennan et al., 1993; Vroon et al., 1995; Derry and France-Lanord, 1996; Graham et al., 1997; Winter et al., 1997). However, the complicated nature of sedimentary processes, and the fact that different components in sediments and sedimentary rocks may be derived from different sources, suggests that the use of a single geochemical method may not always be a reliable identifier of provenance; instead, several complementary geochemical methods should be employed (Fedo et al., 2003).

The South China Sea (SCS) is the largest marginal sea in the Western Pacific. The SCS oceanic crust emerged in the late Oligocene (~30 Ma) (Briais et al., 1993) and evolved in the context of Cenozoic tectonic activity in Southeast Asia and the Southwest Pacific (Hall, 2002). The sea–land distribution changed significantly during this period (Hall, 2002), indicating a change in the source of sediment supplied to the SCS. Therefore, an analysis of temporal trends in sediment provenance in this region may help to constrain its tectonic evolution.

One of the most significant changes in sediment provenance within the SCS occurred at around 23.8 Ma, as recorded in the sediment core recovered from ODP Site 1148 in the north basin of the SCS. At this time, the sedimentary Nd isotopes (ε_{Nd}) drifted by about 2 units (Clift et al., 2002; Li et al., 2003), coincident with significant changes in the assemblage of clay minerals (Clift et al., 2002; Tang et al., 2004), elemental compositions (Li et al., 2003), and the physical and other characteristics of the sediment (Wang et al., 2000; Li et al., 2005). A sediment core recovered from the Pearl River Mouth Basin in the northern SCS also shows a significant ε_{Nd} drift at this time (Shao et al., 2008). A similar negative ε_{Nd} drift has been reported in sediment core recovered from the Yinggehai-Song Hong and Qiongdongnan Basins in the northwest SCS, although this occurred much later, at around 13.8 Ma (Yan et al., 2007), while in the sediments from Hanoi Basin supplied by the Red River systems, there exists a significant positive ε_{Nd} drift at around 24 Ma (Clift et al., 2006a).

The interpretation of these provenance changes is a matter of debate. One explanation is the progressive headward erosion of river systems in South China, from the coastal South China region prior to 23.8 Ma, to the interior of the South China Block after 23.8 Ma (Clift et al., 2002; Shao et al., 2008). In contrast, Li et al. (2003) suggested that sediment input from the southern SCS to ODP Site 1148 ceased at 23.8 Ma, related to the progressive enlargement of the SCS. The interpretation of these changes in provenance is important because it has implications for tectonic reconstructions of the region.

This debate could be resolved by examining provenance information from geochemical proxies, in particular Nd isotopes, which are commonly used to trace the provenance of sediments and sedimentary rocks (Clift et al., 2002; Li et al., 2003; Yan et al., 2007; Shao et al., 2008, 2009). However, the applicability and limitations of Nd isotopes are not yet fully understood. Some studies have compared the Nd isotopes of sediments with those of potential source rocks (e.g., Clift et al., 2002); however, intensive chemical weathering can cause significant Nd isotope fractionation, with ε_{Nd} drifts of up to 2.5 units in secondary minerals of weathering products compared with the parent rock (Ma et al., 2010). Therefore, the mineral compositions of sediments, in particular the proportion of clay minerals (secondary minerals) and rock-forming minerals (primary minerals), should be considered when employing Nd isotopes for provenance analysis, and care should be taken to identify ε_{Nd} drifts related to chemical weathering.

The applicability and limitations of geochemical methods for tracing the provenance of SCS sediments can be assessed by analyzing the geochemical composition of surface sediments compared with that of potential source rocks. The current continent-island–sea distribution in this region was established by the late Pliocene (Hall, 2002), and the pattern of sediment supply can be roughly constrained. The Philippine island arcs form the eastern boundary of the SCS, dominated by young volcanic rocks, mainly basalt and andesite. The isotopic and elemental compositions in this region are different from those in South China and Indochina, to the north and west of the SCS, respectively (Chen et al., 1990; McDermott et al., 1993, 2005). Moreover, previous studies have reported the Nd isotopes of sediments and suspended particles in some of the main river systems around the SCS (Chen and Lee, 1990; Liu et al., 2007; Shao et al., 2009). This information is important in constraining the geochemical composition of the possible sources of SCS sediments, and in linking the sediments to potential source rocks.

Previous studies have investigated the provenance of sediments in the SCS. For example, Shi et al. (2007) examined mineral suites to identify sediment provenance around the Zhongsha Islands (Macclesfield Bank) in the northern SCS, and Liu et al. (2008) examined the assemblage of clay minerals within the fine-grained component of SCS sediments. Element discrimination diagrams has been used to identify the provenance of sediments around the Pearl River Mouth region (Yang et al., 2008), and Gui et al. (1994) mapped the distribution of strontium and oxygen isotopes in the silicate fraction of sediments from the southern SCS. In addition, the Nd isotopes of surface sediments have been used to distinguish between northern and southern provenances for sediments in the SCS (Li et al., 2003). However, few studies have integrated mineralogical, elemental, and isotopic geochemical data to investigate the provenance of sediments in the SCS. As a result, some of the key linkages relevant to provenance tracing remain poorly understood. For example, what is the relationship between the geochemical composition of SCS sediments and that of potential sources? And to what extent can these geochemical methods be used to identify the provenance of SCS sediments? It is important to answer these questions if we wish to assess the applicability of these geochemical methods in tracing the provenance of sediments in the SCS.

This study examines the mineralogy, major and trace elements, and neodymium and strontium isotopes of surface sediments throughout the SCS, with the aim of investigating their applicability in provenance tracing. The geochemical composition of sediments, which may provide provenance information, is discussed in terms of its spatial distribution, and we examine the relationship between the geochemical composition of sediments in different regions and potential sources, with a view to provenance tracing. The results contribute to our understanding of the geochemical linkage between the sediments and their sources, and to the application of these geochemical methods in provenance tracing. Based on the findings, we re-assess the significance of provenance changes recorded in sediments from ODP Site 1148 since the late Oligocene.

2. Materials and methods

Thirty-four core-top sediments in the SCS were selected for analysis. The locations of the samples are listed in Table 1. The sediments are silty clay or clay, as well as foraminifer shells.

The samples were first reacted with 2N of acetic acid (HAc) to remove biogenic carbonates. The solid material was then collected by centrifugation, oven-dried at 100 °C, and ground to powder for elemental, isotopic and mineral geochemical analysis.

The sample powders for analyses of major and trace elements, and Sr and Nd isotopes were first heated at 700 °C to destroy organic material. They were then digested by an $HNO_3 + HF$ acid mixture in high-pressure bombs for elemental analysis. Major elements were measured on a Varian Vista Pro inductively coupled plasma atomic emission spectrometer (ICP-AES) at the State Key Laboratory of Isotope Geochemistry, Guangzhou Institute of Geochemistry (GIG), Chinese Academy of Sciences (CAS). Trace elements were measured on a Perkin-Elmer Elan 6000 inductively coupled plasma mass spectrometer (ICP-MS) at the same laboratory. The analyses of major and trace

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