

# Monsoon forcing, hydrodynamics of the Kuroshio Current, and tectonic effects on sedimentary carbon and sulfur cycling in the Okinawa Trough since 90 ka

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Received 6 November 2005; revised 16 January 2006; accepted 30 January 2006; published 9 March 2006.

[1] Monsoon forcing and hydrodynamic effects controlled carbon and sulfur biogeochemistry over the last 90 ka in a 43-m core from the Okinawa Trough (OT). Total organic carbon (TOC) contents co-vary with summer insolation at 30°N suggesting monsoon forcing of primary productivity and organic carbon burial. Before the last glacial maximum (LGM), total sulfur (TS) contents varied concomitantly with TOC. However, sea level rise and intensified Kuroshio inflow during the Holocene enhanced deepwater ventilation, which resulted in TS-depleted sediments with low degree of pyritization (DOP). By contrast, DOP values were high during the previous highstand (at ~80 ka BP), when sea level was similar to that in the Holocene, as well as during the LGM when deepwater circulation was relatively weak. A topographic barrier is proposed to have sufficiently blocked the Kuroshio Current out of the OT during the previous sea level highstand, which weakened deepwater ventilation and led to reducing diagenetic conditions. Tectonic rifting in the southern OT at 60–30 ka BP enabled the major change in hydrodynamics and sediment biogeochemistry. **Citation:** Kao, S. J., A. P. Roberts, S. C. Hsu, Y. P. Chang, W. B. Lyons, and M. T. Chen (2006), Monsoon forcing, hydrodynamics of the Kuroshio Current, and tectonic effects on sedimentary carbon and sulfur cycling in the Okinawa Trough since 90 ka, *Geophys. Res. Lett.*, 33, L05610, doi:10.1029/2005GL025154.

## 1. Introduction

[2] The evolution of the Kuroshio Current (KC) flow path and variations in the volume of water that it transports are important because, as the Western Boundary Current of the western Pacific, it carries large amounts of heat from equatorial to mid-latitudes, thereby strongly influencing climate over the northwestern Pacific region. The present KC enters the Okinawa Trough (OT) through the Yonaguni Depression and flows along the outer edge of the East China Sea Shelf to the northeast and bifurcates across the Ryukyu Arc through Tokara Strait and into the Yellow Sea and

Japan Sea, respectively (Figure 1). However, the KC might have migrated to the east of the Ryukyu Islands (Figure 1) rather than flowing into the OT during the last glacial maximum (LGM) due to lowered sea levels and/or the hypothetical emergence of a Ryukyu-Taiwan land bridge [Ujiié *et al.*, 1991, 2003]. Intensified KC inflow at the beginning of the Holocene introduced abundant warm water fauna into the OT [Jian *et al.*, 2000; Ujiié *et al.*, 2003; Ijiri *et al.*, 2005], and Japan Sea [Oba *et al.*, 1990], which affected the carbon and sulfur cycling in both basins synchronously [Kao *et al.*, 2005]. In the OT, most previous studies of KC history rely upon short marine cores and little attention has been paid to sedimentary diagenesis and biogeochemistry. In this paper, we present biogeochemical data for a giant piston core that penetrated down to 93 ka BP. Based on these biogeochemical analyses, we demonstrate that organic carbon and sulfur burial were primarily controlled by monsoon forcing, with superimposed sea level fluctuations and tectonic effects contributing to hydrodynamics and deep ventilation within the OT.

## 2. Materials and Methods

[3] The studied giant piston core, IMAGES-MD012404 (total length 43 m), was recovered at 125.81°E, 26.65°N (Figure 1) at a water depth of 1397 m. The sediment core was sliced into 1-cm-thick segments during the cruise and preserved. TS and TOC analyses are detailed by Kao *et al.* [2005]. For TOC/TN,  $\delta^{13}\text{C}_{\text{TOC}}$  and  $\text{Fe}_A$  analyses, 1 N HCl was applied onto the samples for 16 hours to remove carbonate; the sediments were then freeze-dried and centrifuged. The supernatants were measured for Fe concentration in triplicate using an ICP-OES system (Optima 3200DV, Perkin-Elmer™ Instruments, USA).  $\text{Fe}_A$  is an operationally defined fraction that represents the maximum value of reactive iron in the sediments, which is the sum of the highly and the poorly reactive iron [Raiswell and Canfield, 1998]. The acidified sediments were determined for TOC/TN and  $\delta^{13}\text{C}_{\text{TOC}}$  using a Carlo-Erba EA 2100 elemental analyzer and a Thermo Finnigan Delta<sup>plus</sup> Advantage IRMS with detection limits of 5  $\mu\text{gC}$  and 0.1  $\mu\text{gN}$ . Carbon isotopic compositions are presented in the standard  $\delta$  notation with respect to PDB carbon. The age model for core MD012404 was reported by Chang *et al.* [2005].

## 3. Results and Discussion

[4] Numerous studies in a variety of oceanic settings have demonstrated a positive relationship between surface primary production, organic carbon export out of the euphotic zone and organic carbon burial [e.g., Calvert *et*

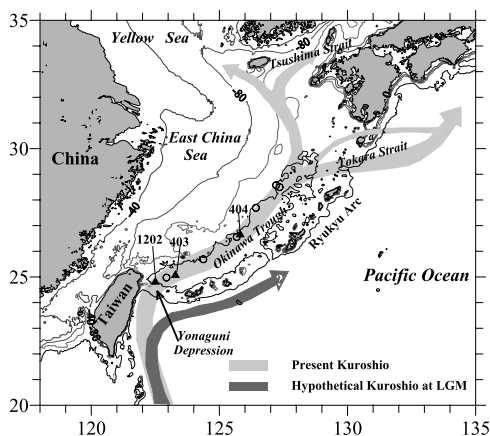
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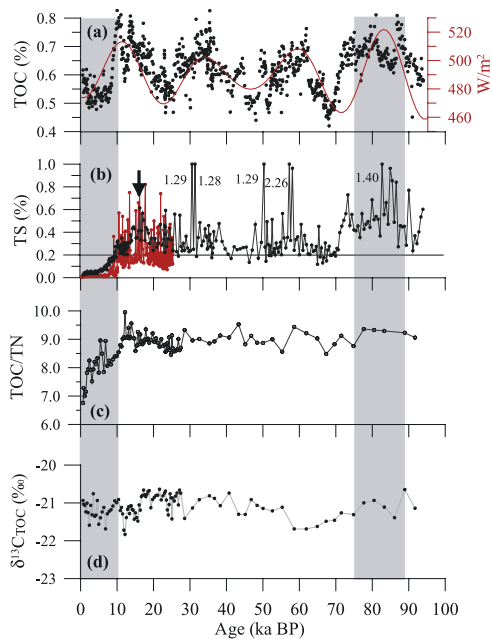
**Figure 1.** Location map for IMAGES cores MD012404 and MD012403, and ODP Site 1202 and the core sites (○) reported by Ujiie *et al.* [2001]. The  $-40$ ,  $-80$ ,  $-120$  and  $-1000$  m isobaths are shown.

*al.*, 1995]. TOC contents are therefore used as a paleo-productivity proxy. TOC values vary between 0.4% and 0.8% (Figure 2a), with a periodicity of  $\sim 23$  kyr, which is close to that of orbital precession cycles and of variations in Northern Hemisphere summer insolation (June at  $30^\circ\text{N}$ ; Figure 2a) [Berger and Loutre, 1991]. Surface ocean productivity, which is reflected in organic carbon burial in sediments, therefore appears to have responded to local insolation changes.

[5] Apart from surface primary production, changes in terrestrial organic inputs, water column conditions, oxygen exposure time, and sedimentation rate might also have affected burial of organics over time. Sedimentation rate (SR) apparently has an insignificant effect on fluctuating TOC contents in the OT since the range of TOC values in core MD012404 (SR,  $45 \pm 15$  cm/kyr) is consistent with those in core MD012403 (see location in Figure 1) despite its much higher and more variable SR ( $153 + 128$  cm/ky) [Kao *et al.*, 2005]. TOC/TN, which is indicative of organic source [Meyers, 1997], has an upward decreasing trend (Figure 2c), which resembles those documented in core MD012403 [Kao *et al.*, 2005] and in six other sediment cores from the OT [Ujiie *et al.*, 2001]. From this pattern, we suggest that the contribution of marine organics increased gradually during the post-glacial period [Ujiie *et al.*, 2001]; yet the mean  $\delta^{13}\text{C}_{\text{TOC}}$  has a typical end-member value of  $-20$  to  $-22\text{‰}$  [Meyers, 1997] revealing a major contribution from marine sources without changes in the upper part of the core as suggested by TOC/TN. The cause of the observed inconsistency between the two proxies is not known. However, other surface ocean conditions could have affected the isotopic composition of the marine end-member, including the rate of primary production, temperature, pH and  $\text{CO}_2(\text{aq})$  [Kienast *et al.*, 2001]. Since neither of the source-related proxies correlates with TOC fluctuation, we suggest that the observed fluctuation of TOC is primarily a response to changes in surface primary production driven by solar insolation rather than changes in fractional contribution from marine and terrestrial organic sources.

[6] During burial of sedimentary organic matter, oxygen is rapidly consumed to low levels. Instead, porewater sulfate and sedimentary iron oxides (with varying reactivity)

become more important oxidants of organic carbon. The relative abundances of labile organic carbon, porewater sulfate and available iron for sulfidation closely link together to affect total sulfur burial [Thamdrup and Canfield, 1996]. Thus, the iron-sulfur-organic carbon relationship may reveal oxidation conditions in sediments. In this study,  $\text{Fe}_A$  has insignificant temporal trends (not shown) and a mean value of  $1.0 + 0.1\%$  ( $n = 104$ ), which is slightly lower but comparable to values observed in core MD012403 ( $1.3 + 0.1\%$ ;  $n = 80$ ) from the southern OT [Kao *et al.*, 2005]. Down-core TS contents vary by more than an order of magnitude and have significantly different patterns before and after the LGM (marked by the arrow in Figure 2b). Before the LGM, spiky but persistently higher TS contents are observed. During this period, the TS pattern mimics TOC variations with higher TS values, including the TS spikes, occurring at times of high TOC contents (pyritized fossils, which were identified frequently in core MD012403 [Kao *et al.*, 2005], account for the TS spikes). Measured TS can be converted to the portion of metabolized organic carbon decomposed via sulfate reduction if we assume that all reduced sulfur produced through sulfate reduction was fixed as iron sulfide without re-oxidation [Morse and Berner, 1995]. This assumption is supported by the relatively higher reactive iron compared to the TS contents, implying an excess of reactive iron available for sulfidation throughout the last 90 ka. Thus, co-varying patterns in TS and TOC (Figures 2a and 2b) indicate that both the buried



**Figure 2.** Down-core profiles for core MD012404: (a) total organic carbon content and solar insolation at  $30^\circ\text{N}$  (red curve), (b) total sulfur content, (c) TOC/TN ratio, and (d) isotope composition of total organic carbon ( $\delta^{13}\text{C}_{\text{TOC}}$ ). Gray columns mark periods of sea level height of  $> -40$  m (see Figure 3a). The arrow indicates the Last Glacial Maximum. A reference line of 0.2% is shown in Figure 2b. Out range values in Figure 2b are numbered aside peaks. In Figure 2b, TS values (in red) for MD012403 are shown for comparison.

(or preserved) portion and the mineralized portion (via sulfate reduction) of the organic matter delivered to the coring site were controlled by monsoon forcing.

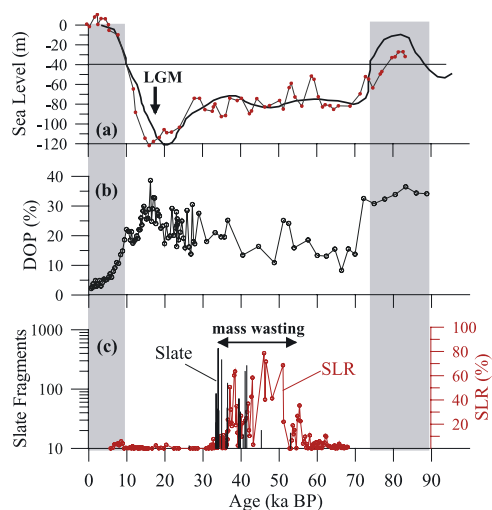
[7] The sedimentary sulfur biogeochemistry of core MD012404 has been altered significantly by changes in hydrological conditions. TS contents decreased continuously to low levels (Figure 2b) in association with increasing sea levels since the LGM (Figure 3a) regardless of fluctuations in TOC contents. The upward decreasing trend in TS contents resembles the TS pattern (in terms of age, Figure 2b) found in core MD012403 from the southern OT [Kao *et al.*, 2005]. Further down-core sulfate reduction might have contributed to the TS profile to some degree; however, the pattern in the upper part of the core is attributed to circulation changes. Synchronous decreases of TS in two cores from the central and southern trough (despite a  $\sim 4$  times higher sedimentation rate in the southern core) indicate that bottom water circulation change is a basin-wide phenomenon. Such a basin-wide hydrological change, rather than primary production (organic delivery), provides the primary control on TS burial in the Holocene sediments. Overall, monsoon forcing regulated the organic matter flux to the sediments and thus created a positive correlation between sediment-bound TS and TOC contents in the OT. However, additional input of oxygen due to enhanced deepwater ventilation, which was driven by post-glacial sea level rise and/or by the Holocene KC intensification, switched the sedimentary organic matter (mainly the labile fraction) oxidation pathway from sulfate reduction to aerobic respiration, and, consequently, resulted in TS-depleted sediments.

[8] Since the TS content of sediments is sensitive to hydrological conditions (regardless of whether hydrological conditions were mainly driven by sea level rise and/or KC intensification), an obvious question relates to why TS-depletion is not observed for the previous sea level highstand at around 80 ka BP. The degree of pyritization (DOP), which quantifies the extent to which reactive iron is fixed as pyrite [Bernier, 1970], has been successfully used to indicate euxinic conditions [Raiswell *et al.*, 1988] and to distinguish oxygenation conditions in the Japan Sea [Tada, 1994].

$$\text{DOP}(\%) = (\text{pyritic iron})/(\text{total reactive iron}) \times 100\%, \quad (1)$$

where the pyritic iron can be converted through measured TS stoichiometrically by assuming that all sulfur is in the form of pyrite ( $\text{FeS}_2$ ). The total reactive iron is the sum of pyritic iron and  $\text{Fe}_A$ . DOP values in core MD012404 are similarly high for the LGM and during the period around 80 ka BP (Figure 3b), when sea levels differed by over 100 m (Figure 3a). DOP values vary as a mirror image of the sea level curve since 70 ka BP. The fact that DOP values decrease with increasing sea level suggests that sea level change can generate deepwater ventilation in the OT. However, different deepwater circulation patterns appear to have been necessary to generate the reverse DOP pattern at the two sea level highstands.

[9] A recent sedimentological study [Huang *et al.*, 2005] on the 400-m ODP core 1202 (water depth 1274 m; Figure 1) from the southwestern corner of the OT revealed a massive turbidite layer (at a depth of 150 m). The reported silt-sand layer ratio (SLR: total thickness of silt and sand layers/1.5-m core section) increases significantly from 55 ka BP to 35 ka



**Figure 3.** (a) Sea level curves. The red dot-line and the continuous curve represent data digitized, respectively, from Saito *et al.* [1998] and Cutler *et al.* [2003]. (b) Down-core pattern of degree of pyritization (DOP) for core MD012404. (c) Occurrence of slate fragments ( $>250 \mu\text{m}$ ; column) and silt-sand layer ratio (SLR: total thickness of silt and sand layers/1.5 m of core; red line with circles for data points) in core MD012404. The arrow indicates the age interval with massive turbidite layers (mass wasting) at ODP Site 1202 (data taken from Huang *et al.* [2005] and age model based on Wei *et al.* [2005]).

BP (Figure 3c). Volcanic detritus was found to be one of the major components in the coarse sediment fraction ( $>250 \mu\text{m}$ ) in this turbidite-rich interval suggesting extensive submarine volcanic activity. Abundant shallow-marine fossils, including echinoids, bryozoans and mollusks, are also present in the turbidite layer. This layer was related to slope failures associated with frequent earthquakes induced by active opening of the OT back-arc basin offshore of NE Taiwan. Following opening of the OT near Taiwan, they found a dramatic increase in the abundance of slate fragments (Figure 3c), which were delivered primarily from the Miocene slate belt of the Central Range in northern Taiwan.

[10] ODP site 1202 is located at the upstream area of the KC entrance (near the Yonaguni Depression). The major mass wasting sequence recorded between  $\sim 55$  and 35 ka BP suggests the opening of a passage for the KC to enter the OT and thus explains the hydrodynamic differences at two similar sea level highstands. During the  $\sim 80$  ka BP sea level highstand this topographic barrier had blocked the KC resulting in weakened deepwater circulation in the OT and high DOP values. Blocking of the KC during the previous sea level highstand might have also prevented formation of the warm Tsushima Current, which has been reported to not have flowed into the Japan Sea at  $\sim 80$  ka [Oba *et al.*, 1991]. The hydrodynamic change associated with inferred tectonic opening of the Yonaguni depression enabled deepwater ventilation in the Holocene highstand that supported low DOP values.

#### 4. Conclusions

[11] Sedimentary biogeochemistry (C-S-Fe relationships) in the OT indicates that fluctuations in burial of organic

carbon reflect variations in primary productivity that were driven by solar insolation/monsoon forcing. Markedly different C-S relationships are observed for sea level highstands at ~80 ka BP and in the Holocene. Extensive pyritization of sediments in the earlier highstand indicates poor ventilation in the OT, whereas low DOP values in the Holocene suggest good ventilation resulting from inflow of the KC. The difference in deepwater hydrodynamics for the two highstands is attributable to documented tectonic changes in the Yonaguni depression at 60–30 ka BP that allowed incursion of the KC into the OT during the Holocene highstand. Heat transport of the Kuroshio Current and associated climate change in the northwestern Pacific will have therefore undergone substantial changes in the past as a result of evolving hydrodynamics that resulted from global sea level change and local tectonic changes in the OT.

[12] **Acknowledgments.** This study was supported by grants (NSC94-2611-M-001-004) from the National Science Council of Taiwan. We acknowledge the crew and scientists on board the RV *Marion Dufrenoy* for retrieving IMAGES core MD012404.

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