

Carbon isotope evidence for widespread methane seeps in the ca. 635 Ma Doushantuo cap carbonate in south China

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ABSTRACT

Distinctive sedimentary structures, textures, and extremely negative $\delta^{13}\text{C}_{\text{carb}}$ values from the Ediacaran Doushantuo cap carbonate (ca. 635 Ma) in south China were taken as evidence for a methane hydrate destabilization event, but existing data for a methane-derived $\delta^{13}\text{C}_{\text{carb}}$ signature were reported from a single locality in the Yangtze Gorges area. Here we report negative $\delta^{13}\text{C}_{\text{carb}}$ values as low as -48‰ (Vienna Peedee belemnite) from two additional sections ~6 km and 55 km from the original locality. These negative $\delta^{13}\text{C}_{\text{carb}}$ values were obtained from isopachous cements that fill stromatolite-like cavities, sheet cracks, and fractures, and from partially recrystallized carbonate crusts, consistent with carbonate precipitation at cold methane seeps. The new data support a widespread methane release event following the Nantuo glaciation in south China (ca. 635 Ma), which may have contributed to the origin of unusual sedimentary and isotope features of cap carbonates.

Keywords: carbon isotopes, methane seeps, Ediacaran, Doushantuo cap carbonate, south China.

INTRODUCTION

Regionally consistent, thin carbonate rocks ubiquitously overlie late Cryogenian (ca. 750–635 Ma) glacial deposits in almost every continent. These cap carbonates are characterized by negative $\delta^{13}\text{C}$ anomalies and unique sedimentary features, which have attracted considerable attention as they may record a global-scale oceanographic and/or chemical event following the late Cryogenian glaciation (Grotzinger and Knoll, 1995; Kennedy, 1996; Hoffman et al., 1998; Jiang et al., 2003; Shields, 2005). The origin of cap carbonates has been debated; interpretations include postglacial chemical weathering of silicate and carbonate rocks (Hoffman et al., 1998; Higgins and Schrag, 2003), upwelling of alkalinity-rich deep-ocean seawater (Grotzinger and Knoll, 1995), oxidation of methane from gas hydrate destabilization (Kennedy et al., 2001; Jiang et al., 2003), and microbially mediated carbonate precipitation from deglacial meltwaters (Shields, 2005).

In south China, a 3–5-m-thick cap carbonate in the basal Doushantuo Formation overlies glaciogenic rocks of the Cryogenian Nantuo Formation. This cap carbonate, dated as 635.2 ± 0.6 Ma (Condon et al., 2005), contains a suite of distinctive sedimentary features including tepee-like structures, stromatolite-like cavi-

ties, sheet cracks, and multiple generations of cements (Jiang et al., 2003, 2006a). The presence of unusually negative $\delta^{13}\text{C}$ values as low as -41‰ in micritic crusts and isopachous cements closely associated with tepee-like structures and cavities was taken as evidence to support a methane release event. Methane release from gas hydrate destabilization was hypothesized to have contributed not only to the formation of sedimentary structures and textures in cap carbonates, but also to the negative $\delta^{13}\text{C}$ anomaly of cap carbonates.

A serious criticism of the methane hypothesis is the rarity of a methane-derived $\delta^{13}\text{C}$ signature in cap carbonates (e.g., Shields, 2005; Corsetti and Lorentz, 2006). Despite the widespread distribution of sedimentary structures and textures similar to those of modern and ancient methane seeps in the Doushantuo cap carbonate (Jiang et al., 2006a, 2006b) and in other comparable cap carbonates (Kennedy et al., 2001), highly negative $\delta^{13}\text{C}$ values have until now been obtained from only a single section in the Yangtze Gorges area (Huajipo section; location 1 in Fig. 1). Although the absence of a methane-derived carbon isotope signature in most cap carbonate outcrops may be explained in terms of the involvement of ambient seawater during carbonate precipitation and diagenetic homogenization (Jiang et al., 2003, 2006a, 2006b), its restriction to a single locality weakens the proposed connection between a local methane seep

and an inferred gas hydrate destabilization event (Shields, 2005; Corsetti and Lorentz, 2006).

Here we report highly variable $\delta^{13}\text{C}_{\text{carb}}$ values ranging from -2‰ to -48‰ from both micro-drilled and bulk samples at two additional sections of the Doushantuo cap carbonate (Jiulongwan and Wangzishi sections, locations 2 and 3 in Fig. 1) in south China. The new isotope data, along with sedimentological and petrographic observations, are consistent with carbonate precipitation at cold methane seeps and support a widespread methane release event following the Nantuo glaciation (ca. 635 Ma).

The stratigraphy, sedimentary structures and textures, and the overall carbon isotope chemostratigraphy of the Doushantuo cap carbonate have been discussed in detail elsewhere (e.g., Jiang et al., 2003, 2006a, 2006b), and are not repeated here. In this paper we focus on the new isotope data and their hosting carbonate components that may provide insights for future search of similar features in other cap carbonate sections in south China and elsewhere.

NEW ISOTOPE EVIDENCE FOR METHANE SEEPS Jiulongwan Section in the Yangtze Gorges Area

At the Jiulongwan locality (location 2 in Fig. 1), ~6 km northeast of the Huajipo section (Jiang et al., 2003), the 5-m-thick Doushantuo cap carbonate consists of predominately microcrystalline dolomite (Fig. 2A). The basal 1.2 m (C1) is strongly disrupted, with abundant fractures, bedding-parallel sheet cracks, localized breccias, and centimeter-scale stromatolite-like cavities. The disrupted basal cap carbonate grades upsection into 2-m-thick laminated microcrystalline dolomite and dolomitic limestone (C2) that show a transitional relationship with the overlying 1.9 m of silty dolomite and limestone (C3). At the C2 and C3 transition, 3–5-cm-thick silty limestone lenses are present parallel or subparallel to bedding.

Most fractures, sheet cracks, and cavities at the basal level (C1) are overprinted by silicification or recrystallization, especially near the cap

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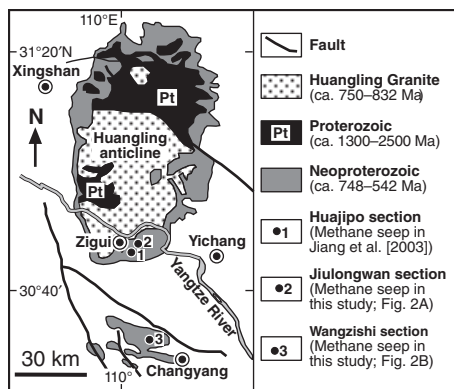


Figure 1. Simplified geological map showing locations of methane seeps in Doushantuo cap carbonate reported in Jiang et al. (2003, 2006a; location 1) and in this study (locations 2 and 3).

carbonate–diamictite contact. In the upper part of C1, locally preserved black to dark gray fractures (Fig. 3A) and cavities (Fig. 3B) are filled with fringing isopachous calcite cements. Fan-like subcrystals in the isopachous cements display undulose extinction in cross-polarized light (Fig. 3C) and encase dark spots that fluoresced bright yellow to yellow-green under ultraviolet light, possibly reflecting trapped hydrocarbons. In some cases, recrystallization partially or completely obscures the original crystal shapes, with large, inclusion-rich blocky calcite crystals characterized by intense twinning, suggesting local diagenetic modification during burial or tectonic deformation.

Isotope analyses of samples from the Jiulongwan section reveal highly variable $\delta^{13}\text{C}_{\text{carb}}$ values ranging from -2‰ to -44‰ (Fig. 2A; GSA Data Repository Table DR1¹). The most negative $\delta^{13}\text{C}_{\text{carb}}$ values (from -10‰ to -44‰) were obtained from dark gray to black isopachous cements in sheet cracks, fractures, and cavities in the lower cap carbonate (Figs. 2A, 3A, and 3B). In contrast, a microcrystalline dolomite matrix yields uniform $\delta^{13}\text{C}_{\text{carb}}$ values with an average of $\sim -3\text{‰}$ (Fig. 2A). Silty limestone lenses at the C2 and C3 transition produce moderately negative $\delta^{13}\text{C}_{\text{carb}}$ values from -5‰ to -17‰ .

Cap carbonate $\delta^{18}\text{O}$ values display two trends. Microcrystalline dolomite has highly variable $\delta^{18}\text{O}$ values from 0‰ to -12‰ that show a clear $\delta^{13}\text{C}$ – $\delta^{18}\text{O}$ covariation, while most isopachous cements have a relatively narrow $\delta^{18}\text{O}$ range from -8‰ to -10‰ (Fig. 2A). The lowest $\delta^{18}\text{O}$

¹GSA Data Repository item 2008087, Table DR1, carbon and oxygen isotopes for the Doushantuo cap carbonate at Jiulongwan and Wangzishi sections in south China, is available online at www.geosociety.org/pubs/ft2008.htm, or on request from editing@geosociety.org or Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301, USA.

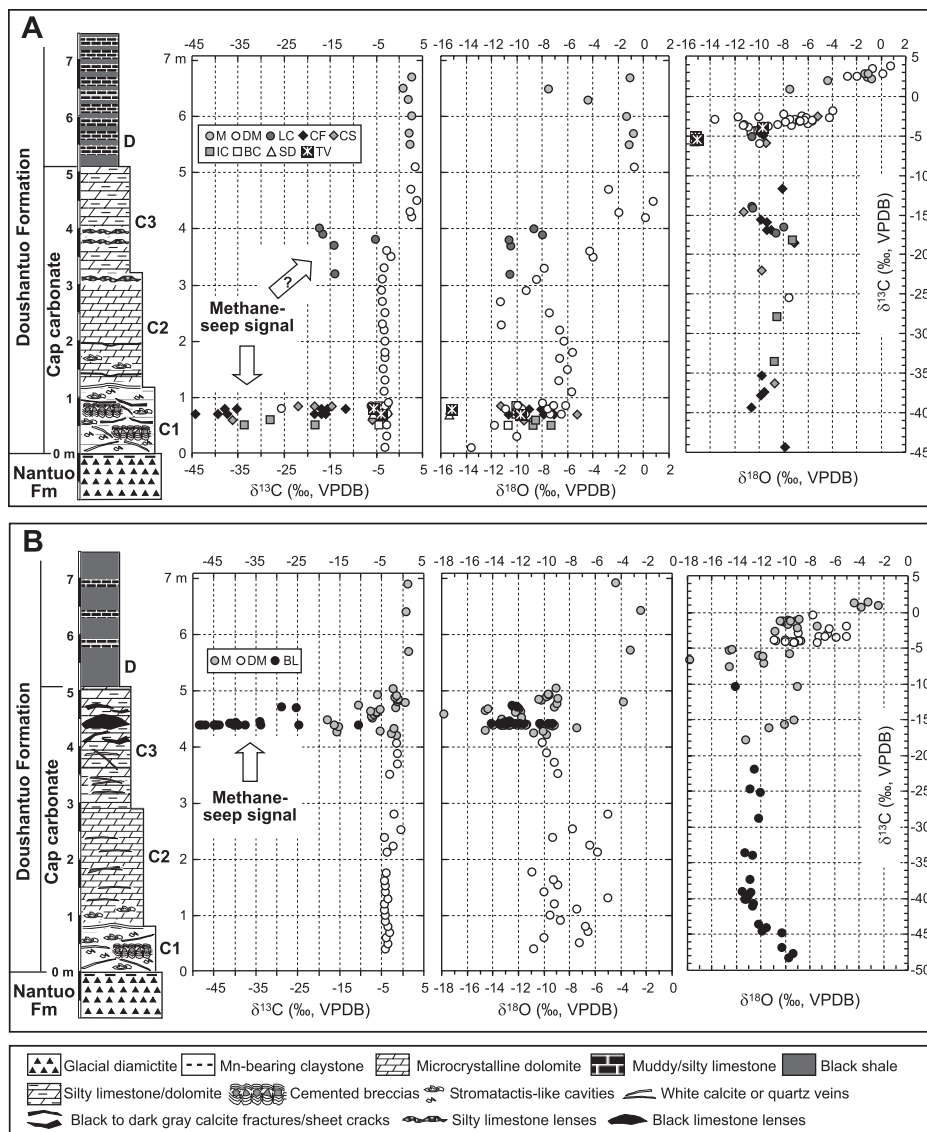


Figure 2. Carbon and oxygen isotope profiles and $\delta^{13}\text{C}$ – $\delta^{18}\text{O}$ cross-plot of Doushantuo cap carbonate at Jiulongwan (A) and Wangzishi (B) sections. Carbon isotope values ranging from -10‰ to -44‰ in lower cap carbonate at Jiulongwan and from -10‰ to -48‰ in upper cap carbonate at Wangzishi are inferred to indicate methane seeps. Moderately low $\delta^{13}\text{C}$ values (-5‰ to -17‰) from silty limestone lenses in upper cap carbonate at Jiulongwan may also have had methane influence, but evidence is not definitive. See Figure 1 for locations of sections. Carbonate components: M—microcrystalline limestone; DM—microcrystalline dolomite; LC—silty limestone lenses; CF— isopachous cements in fractures; CS— isopachous cements in sheet cracks; IC— isopachous cements in cavities; BC— blocky calcite in cavities; SD— saddle dolomite; TV— crosscutting veins; BL— black limestone lenses (matrix and cements); VPDB— Vienna Pee Dee belemnite.

values (to -16‰) are from white, crosscutting calcite veins and coarse saddle dolomite close to quartz-filled fractures. The variable $\delta^{18}\text{O}$ values suggest diagenetic modification but not complete isotope homogenization.

Carbon isotope values as low as -44‰ provide unequivocal evidence for methane influence that, when considered in context with seep-related features such as fractures, cavities, sheet cracks, and brecciation similar to those of modern and ancient methane seeps (e.g.,

Bohrmann et al., 1998; Campbell et al., 2002; Peckmann et al., 2002; Peckmann and Thiel, 2004), support carbonate precipitation in the vicinity of a methane seep. There are uncertainties concerning the moderately negative $\delta^{13}\text{C}_{\text{carb}}$ values from the silty limestone lenses at the C2 and C3 transition (Fig. 2A). They may be concretionary carbonate nodules formed under methane influence (cf. Stakes et al., 1999; Aiello et al., 2001), but further investigation is needed to evaluate their diagenetic history.

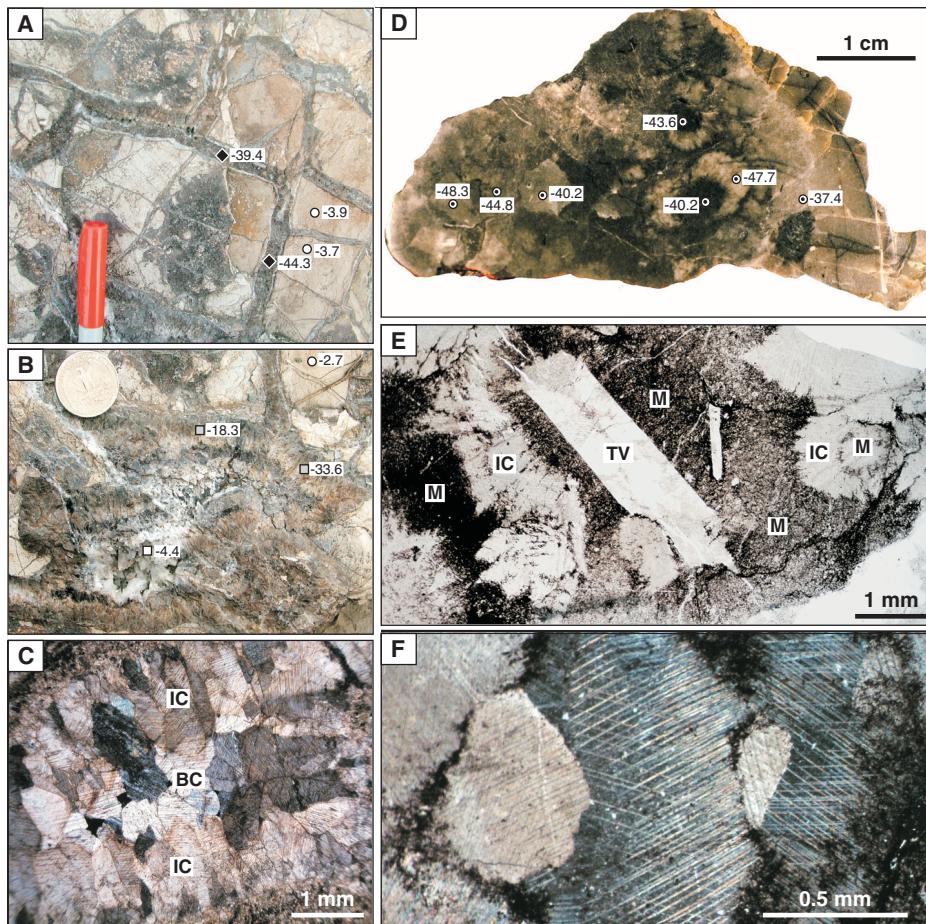


Figure 3. Cap carbonate textures and their carbon isotope compositions. **A:** Carbon isotope values from microcrystalline dolomite matrix and dark gray fracture-filling isopachous cements (0.7 m above base in Fig. 2A). Pen cap (50 mm) for scale. **B:** Carbon isotope values from microcrystalline dolomite matrix, isopachous fibrous cements, and blocky calcite within a stromatolite-like cavity (0.5 m above base in Fig. 2A). Quarter (25 mm diameter) for scale. **C:** Cross-polarized light microphotograph of isopachous cements (IC) and blocky calcite (BC) in fractures in A. Although original crystal shapes are still preserved, cements are recrystallized to large, cross-crystal calcites that show intense twinning. **D:** Polished slab of limestone lenses (carbonate crust) at Wangzishi section (4.38 m above base in Fig. 2B) showing clotted textures similar to that of carbonate crusts in modern methane seeps. Carbon isotope values of microdrilled samples are labeled. **E:** Plane-polarized light microphotograph of carbonate crusts in D. M—microcrystalline calcite and micrite; TV—late diagenetic calcite vein. **F:** Inclusion-rich, highly twinned calcite crystals overprinting micritic matrix and cements in carbonate crusts in D.

Wangzishi Section in Changyang

At the Wangzishi section in Changyang (location 3 in Fig. 1), 55 km southeast of the Yangtze Gorges area, the 5-m-thick cap carbonate consists of three lithological units (Fig. 2B): (1) a strongly deformed and silicified dolomite interval (C1) with centimeter-size cavities filled with blocky calcite and quartz (0–0.8 m), (2) a laminated microcrystalline dolomite interval (C2) with bedding-parallel and crosscutting veins filled with white, blocky calcite crystals and silica (0.8–2.9 m), and (3) a silty microcrystalline dolomite and limestone interval (C3) with black to dark gray limestone lenses that are subparallel or parallel to bedding (2.9–5.1 m). The largest

limestone lens is 15 cm thick and contains abundant white crosscutting calcite veins.

Extremely negative $\delta^{13}\text{C}_{\text{carb}}$ values from -10‰ to -48‰ were obtained from the black to dark gray limestone lenses in the upper cap carbonate (Fig. 2B; Table DR1 [see footnote 1]). On polished slabs, the limestone shows a clotted mosaic, with black clots surrounded by light gray radiating calcite spars (Fig. 3D). This clotted mosaic is similar to the globular fabrics of carbonate crusts in modern (e.g., Bohrmann et al., 1998; Greinert et al., 2002) and ancient (e.g., Campbell et al., 2002; Peckmann and Thiel, 2004) methane seeps. The limestone underwent intense recrystallization, commonly

with large (>1 mm) inclusion-rich and twinned calcite crystals (Fig. 3F) overprinting primary micrite matrix and cements, but in relatively well preserved areas, remains of micritic clots and isopachous calcite cements can still be observed (Fig. 3E).

Although some isotope variations within these limestone lenses exist, the bulk $\delta^{13}\text{C}_{\text{carb}}$ value of analyzed slabs is $<-40\text{‰}$, with a decreasing trend from the periphery to the center of the limestone lenses. In contrast, $\delta^{13}\text{C}_{\text{carb}}$ values of partially silicified microcrystalline carbonates surrounding the lenses are variable, from $+0.6\text{‰}$ to -17‰ (Fig. 2B). Oxygen isotope values from this section are 2‰ – 3‰ lower than those at the Jiulongwan section (Fig. 2A), consistent with field and petrographic observations that the cap carbonate at Wangzishi is more strongly deformed and recrystallized in general.

Extremely negative $\delta^{13}\text{C}_{\text{carb}}$ values as low as -48‰ provide strong evidence for methane-influenced carbonate precipitation at methane seeps. Although diagenetically modified, the clotted textures in better-preserved portions of the carbonate lenses (Figs. 3D and 3E) resemble carbonate crusts precipitated near the sediment-seawater interface in the vicinity of methane seeps where anaerobic methane oxidation is intensified (e.g., Bohrmann et al., 1998; Greinert et al., 2002; Peckmann and Thiel, 2004).

DISCUSSION

The discovery of extremely negative $\delta^{13}\text{C}_{\text{carb}}$ values down to -48‰ in Doushantuo cap carbonate sections at Jiulongwan and Wangzishi (Fig. 2), together with previously reported data from the Huajipo section (Jiang et al., 2003), indicates that three unequivocal methane seeps, located 60 km apart in the Yangtze platform, are now documented from the Doushantuo cap carbonate. The new isotope data strengthen the connection between methane seeps and a methane release event following the Nantuo glaciation.

Currently available data, however, are insufficient to resolve exactly when the methane release event started and how long it lasted. The syndepositional attributes of seep facies and the presence of methane-derived isotope signature in the lower, middle, and upper cap carbonate at the Jiulongwan (Fig. 2A), Huajipo (Jiang et al., 2003), and Wangzishi (Fig. 2B) sections, respectively, suggest that the duration of the methane release may be comparable to the time scale of cap carbonate deposition. However, localized but widespread seep facies are hosted in moderately ^{13}C -depleted dolomitic and microcrystalline dolomite. If the negative $\delta^{13}\text{C}$ values of the hosting dolomite at the basal level record an ocean seawater isotope signature influenced by extremely ^{13}C -depleted carbon from methane hydrate destabilization, the initial

methane release has to predate the basal cap carbonate precipitation (Jiang et al., 2006b). Considering the mixing time of methane-derived carbon in the ocean ($\sim 10^4$ yr; Dickens et al., 1997), the lag time between initial methane hydrate destabilization and cap carbonate precipitation could be on the order of 10^4 yr. In this case, seep facies in the Doushantuo cap carbonate may record the last phase of a methane release event, during which existing and syndepositional cap carbonates served as hosting sediments required for the preservation of seep facies (Jiang et al., 2006b). In addition, it is possible that the Doushantuo cap carbonate is diachronous across the basin, and the laterally discontinuous but texturally identical seep facies from the basin to platform (Jiang et al., 2006a) may record carbonate precipitation that tracked the sea level during post-glacial transgression (Hoffman et al., 2007). In this scenario, seep facies of the Doushantuo cap carbonate may record multiple methane releases associated with transgressive flooding of the basin. Additional sedimentological investigation and sampling are needed to clarify these uncertainties.

The stratigraphic occurrence of methane seeps in the Doushantuo cap carbonate provides insights into the potential relationship between methane flux and formation of carbonate crusts. The abundant cavities, breccias, and fractures near the cap carbonate–diamictite contact indicate strong disruption by gas and fluids. Yet the methane-related isotope signatures are observed above the most strongly disrupted levels in all three sections (Fig. 2; Jiang et al., 2003). While the absence of methane-derived isotope signatures may reflect intense silicification and fluid-rock interaction at the base of cap carbonates (e.g., Jiang et al., 2003, 2006a), it may also be related to high gas-fluid flow rate during early stages of methane hydrate destabilization. Recent experiments and modeling of modern methane seeps indicate that, at high gas-fluid flow rates (>90 cm/yr), methane is mostly exported to the water column and carbonate crust formation is inhibited, whereas at low gas-fluid flow rates (<90 cm/yr), methane is more likely consumed by anaerobic microbial oxidation in sediments, resulting in precipitation of thick, authigenic carbonate crusts (Aloisi et al., 2004, 2006). The strongly disrupted basal Doushantuo structures may have developed during times of high methane gas and fluid escape. Carbonate precipitation in cavities and voids may have involved more carbonate ions from seawater and thus had less extreme carbon isotope variation between carbonate components. When methane gas-fluid flow rate subsequently decreased, carbonate precipitation

involved more methane-derived biocarbonates, resulting in extreme carbon isotope variations in the Doushantuo cap carbonate.

ACKNOWLEDGMENTS

We thank J. Huang, X. Cheng, A.J. Kaufman, and M. Hailemichael for isotope analyses. We also thank P.F. Hoffman, N. Christie-Blick, J. Marshall, and H. Jenkyns for constructive reviews. This research was supported by the National Natural Science Foundation of China (40472063 and 40621002), SinoPec project (G0800-06-ZS-319), 111 Project (B08030), and National Science Foundation (grants EAR-0521196 and EAR-0545135).

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Manuscript received 22 October 2007
 Revised manuscript received 10 January 2008
 Manuscript accepted 11 January 2008

Printed in USA