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Early diagenesis in the sediments of the Congo deep-sea fan dominated by massive terrigenous deposits: Part II – Iron–sulfur coupling



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ABSTRACT

Deep-sea fans are well known depot centers for organic carbon that should promote sulfate reduction. At the same time, the high rates of deposition of unconsolidated metal oxides from terrigenous origin may also promote metal-reducing microbial activity. To investigate the eventual coupling between the iron and sulfur cycles in these environments, shallow sediment cores (< 50 cm) across various channels and levees in the Congo River deep-sea fan (~5000 m) were profiled using a combination of geochemical methods. Interestingly, metal reduction dominated suboxic carbon remineralization processes in most of these sediments, while dissolved sulfide was absent. In some 'hotspot' patches, however, sulfate reduction produced large sulfide concentrations which supported chemosynthetic-based benthic megafauna. These environments were characterized by sharp geochemical boundaries compared to the iron-rich background environment, suggesting that FeS precipitation efficiently titrated iron and sulfide from the pore waters. A companion study demonstrated that methanogenesis was active in the deep sediment layers of these patchy ecosystems, suggesting that sulfate reduction was promoted by alternative anaerobic processes. These highly reduced habitats could be fueled by discrete, excess inputs of highly labile natural organic matter from Congo River turbidites or by exhumation of buried sulfide during channel flank erosion and slumping. Sulfidic conditions may be maintained by the mineralization of decomposition products from local benthic macrofauna or bacterial symbionts or by the production of more crystalline Fe(III) oxide phases that are less thermodynamically favorable than sulfate reduction in these bioturbated sediments. Overall, the iron and sulfur biogeochemical cycling in this environment is unique and much more similar to a coastal ecosystem than a deep-sea environment.

1. Introduction

Understanding the biogeochemical processes involved in the respiration of natural organic matter (NOM) in marine sediments is critical, as sediments represent the ultimate sink of carbon on Earth and the release of dissolved inorganic carbon (DIC) to the overlying waters, as by-product of respiration processes, may affect CO₂ inputs from the atmosphere to the oceans and, therefore, ocean acidification. Although aerobic respiration is the most efficient NOM degradation process (Hedges et al., 1995; Burdige, 2007), anaerobic processes are generally recognized to be significant in sediments exposed to high sedimentation

rates. In these sediments, the high concentrations of NOM settled to the seafloor promotes consumption of the majority of the oxygen available by diffusion across the sediment-water interface (Burdige, 2006). These environments include estuaries (Martens et al., 1978; Aller and Yingst, 1980; Meiggs and Taillefert, 2011), coastal bays (Thamdrup et al., 1994), deltas (Aller et al., 1986; Aller, 1998; Pastor et al., 2011), and some continental margins dominated by large inputs of fine grained sediments from large rivers (Aller et al., 1986; McKee et al., 2004; Rabouille et al., 2009; Beckler et al., 2016). In these sediments, sulfate reduction is often the main anaerobic terminal electron accepting process (e.g. Jørgensen, 1982; Howarth, 1984; Hoehler et al., 2001)

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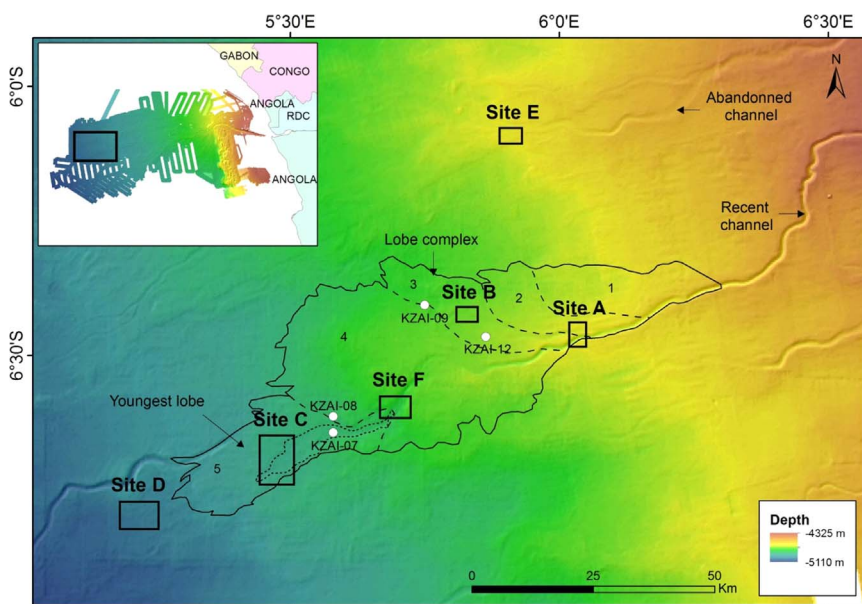


Fig. 1. Maps of the stations in the Congo River Fan, including the high resolution bathymetry obtained at the site during the same cruise (courtesy of Laurence Droz, ©Ifremer, Congolobe cruise).

given the large concentrations of sulfate in seawater and the fact that one mole of sulfate is able to oxidize two moles of organic carbon all the way to DIC. Evidence, however, indicates that the reduction of Mn(IV/III) and Fe(III) oxides by metal-reducing bacteria may also contribute to a significant fraction of carbon remineralization processes in environments with significant inputs of minerals (Canfield et al., 1993; Lowe et al., 2000; Thamdrup and Canfield, 1996; Kostka et al., 2002; Meigs and Taillefert, 2011; Beckler et al., 2016), despite the fact that two moles of Mn(IV) and four moles of Mn(III) or Fe(III) are required to oxidize one mole of organic carbon. In these environments, the importance of metal oxides on carbon remineralization processes partly relies on their recycling upon exposure to oxygen (van der Zee et al., 2002; Wehrmann et al., 2014). As the abiotic oxidation of Fe^{2+} by dissolved oxygen is extremely fast at circumneutral pH (Sung and Morgan, 1980; Davison and Seed, 1983; Millero et al., 1987), a significant fraction of the oxygen consumption rate in sediments may be attributed to the recycling of Fe(III) oxides near the sediment-water interface. On the other hand, the abiotic oxidation of Mn^{2+} by dissolved oxygen is slow at circumneutral pH (Diem and Stumm, 1984; Davies and Morgan, 1989) and Mn^{2+} -oxidizing bacteria are required to facilitate the recycling of Mn(IV) oxides (Tebo et al., 2004).

Unfortunately, microbial reduction of Mn(IV/III) and Fe(III) oxides is difficult to demonstrate directly in the field, as unique molecular or genetic markers for these processes have yet to be identified (DiChristina et al., 2005). In addition, dissolved sulfide produced by sulfate reduction is a great reductant of both Mn(IV/III) (Burdige et al., 1985; Herszage and dos Santos Afonso, 2003) and Fe(III) oxides (dos Santos Afonso and Stumm, 1992; Peiffer et al., 1992). As these reactions simultaneously remove dissolved sulfide and generate Mn^{2+} and Fe^{2+} in pore waters, it is difficult to differentiate sulfide-driven reduction of these metal oxides from microbial metal reduction, especially in iron-rich sediments where a cryptic sulfur cycle may be responsible for the production of Fe^{2+} in the pore waters (Aller et al., 2010). Similarly, the precipitation of FeS in the presence of excess dissolved sulfide (Pyzik and Sommer, 1981; Rickard, 1995) and the eventual generation of pyrite (Rickard and Luther, 1997) represent two additional processes that scavenge reduced iron and dissolved sulfides from the pore waters. The occurrence of these processes can be inferred from the solid phase, however, as FeS is a dense black precipitate. Chemical tracers of microbial iron reduction and precipitation of FeS were recently adopted to identify the zones of sulfate reduction in situ without the addition of radiotracers (Beckler et al., 2016). In this approach, organic-Fe(III) complexes detected by voltammetric mercury-gold amalgam (Hg/Au)

microelectrodes below the oxygen penetration depth were used as proxy for microbial iron reduction, as these complexes are produced as intermediates during the reduction of Fe(III) oxides by several strains of the *Shewanella* genus (Taillefert et al., 2007; Jones et al., 2010). In addition, the existence of organic-Fe(III) complexes in oxygen-depleted pore waters indicates that sulfate reduction is not ongoing, as the reduction of these Fe(III) complexes by dissolved sulfide is extremely fast (Taillefert et al., 2000a). Simultaneously, $\text{FeS}_{(\text{aq})}$ clusters detected by voltammetric Hg/Au microelectrodes may be used as proxy for the zone of sulfate reduction, as these clusters are intermediates in the precipitation of FeS (Theberge et al., 1997; Rickard et al., 1999; Taillefert et al., 2000a; Rickard, 2006).

In this paper, the coupling between iron and sulfur biogeochemistry in the sediments from the active lobe complex of the Congo River deep-sea fan is examined using both field measurements and theoretical model considerations. Sediments from the levees along the main channel of this deep-sea fan are compared to those off axis, including an old extinct channel. In addition, the sediment geochemistry is examined in discrete habitats colonized by micro- and megafauna along the main channel to determine the biogeochemical conditions that lead to the existence of these unusual ecosystems.

2. Sampling and methods

Sediment cores from the active lobe complex of the Congo River deep-sea fan were obtained using either a multicorer or by the ROV Victor 6000 (Ifremer, Brest, France) from 5 stations approaching 5000 m depth and up to 800 km offshore (Fig. 1) during a cruise on the N/O *Pourquoi Pas?* (Ifremer, Brest, France) from December 2011 to January 2012 (Rabouille, 2011). Stations were located along two perpendicular transects: one along-axis extending from the entrance of the main channel to the main present deposition zone (Site A, 4755–4769 m; Site F, 4861–4868 m; Site C, 4944–4967 m); and one off-axis from the entrance of the canyon (Site A), to a station ~10 km north from the active channel (Site B, 4816–4823 m), and to an extinct channel no longer receiving turbidite flows (Site E, 4750 m). The multicorer was used to sample sediments from the levees along the active channel, whereas the push cores were deployed from the ROV Victor 6000 around discrete habitats that were either deeply reduced, populated by microbial mats, or colonized by chemosynthetic megafauna, including species of *Vesicomysidae* bivalves (Table 1). More details on the stations and the sampling locations along the Congo River deep-sea fan are provided in the first paper of this special edition

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