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The remineralization of sedimentary organic carbon in different sedimentary regimes of the Yellow and East China Seas

Bin Zhao^{a,c,d}, Peng Yao^{a,b,*}, Thomas S. Bianchi^d, Ana R. Arellano^{d,e}, Xuchen Wang^{a,c}, Jianbin Yang^{a,c}, Rongguo Su^{a,c}, Jinpeng Wang^{a,c}, Yahong Xu^{a,c}, Xinying Huang^{a,c}, Lin Chen^{a,c}, Jun Ye^{a,c}, Zhigang Yu^{a,b}

^a Key Laboratory of Marine Chemistry Theory and Technology, Ministry of Education, Ocean University of China, Qingdao 266100, China

^b Laboratory for Marine Ecology and Environmental Science, Qingdao National Laboratory for Marine Science and Technology, Qingdao 266071, China

^c College of Chemistry and Chemical Engineering, Ocean University of China, Qingdao 266100, China

^d Department of Geological Sciences, University of Florida, Gainesville, FL 32611, USA

^e College of Marine Science, University of South Florida, Saint Petersburg, FL 33701, USA

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ABSTRACT

We investigated the remineralization of sedimentary organic carbon (SOC) at 12 sites in East China Sea mobile-muds (ECSMMs) and South Yellow Sea central mud deposits (SYSMDs) - using a time-sequence sediment incubation experiment. We examined pore-water dissolved inorganic carbon (DIC), dissolved organic carbon (DOC), fluorescent dissolved organic matter (FDOM), dissolved inorganic nitrogen (DIN) nutrients (NH_4^+ , NO_3^- , and NO_2^-), redox sensitive elements (Fe^{2+} and Mn^{2+}), and major anions (SO_4^{2-} and Cl^-) in incubated sediments, to better constrain controlling mechanisms of SOC remineralization under different sedimentary regimes. Lower DIC production rates in SYSMDs ($2.36\text{--}3.13\text{ mmol m}^{-2}\text{ d}^{-1}$) than those in ECSMMs ($2.94\text{--}13.5\text{ mmol m}^{-2}\text{ d}^{-1}$), were mainly attributed to cold bottom water masses and a relatively stable sedimentary environment in SYS. Higher DIC production rates were observed mostly at offshore sites of ECSMMs that had relatively enriched ^{13}C of SOC - which indicated preferential degradation of labile SOC of marine origin. When compared with tropical mobile-muds, higher bottom-water temperatures, thicker mobile-muds, and large inputs of reactive terrestrial OC resulted in more intense remineralization of SOC in Amazon mobile-muds than in ECSMMs. Lower ratios of DOC/DIC production rates in ECSMMs (0.11–0.72) were likely indicative of efficient transformation of OC, and largely due to sulfate reduction. A rapid increase in marine protein-like FDOM components during the incubation indicated that less stable marine SOC was preferentially converted to DOC - and then to DIC. Our SOC budget indicates that 16.8% of SOC was decomposed in sediments of ECSMMs, but only about 5.4% of SOC was decomposed in SYSMDs, suggesting lower SOC preservation efficiency in mobile-muds than distal muds.

1. Introduction

Marginal seas are major sinks for > 90% of organic carbon (OC), both terrestrial and marine sources, buried in the shallow marine environment (Bianchi and Allison, 2009; Hedges and Keil, 1995; Liu et al., 2010a, 2010b). Annually, marginal seas can receive a large quantity of land-derived freshwater, sediments, and associated OC from adjacent rivers (McKee et al., 2004). Large amounts of nutrients and terrestrial materials carried by rivers into marginal seas, especially in large-river delta-front estuaries (LDEs) (McKee et al., 2004; Bianchi and Allison,

2009), results in higher rates of primary productivity and deposition compared to other shelf regions and the open ocean (Liu et al., 2006, 2007). Despite large OC inputs, recent studies demonstrated that mobile-muds in LDEs play an important role in remineralization of sedimentary OC (SOC) (Aller et al., 1996, 2008; Aller, 1998). Low preservation efficiency of both terrestrial and marine SOC in mobile-muds is largely attributed to an abundant and diverse microbial community, enhanced oxygen exposure, and efficient metabolite exchange - caused by physical reworking and benthic community activities (Aller, 2004; Blair and Aller, 2012).

* Corresponding author at: Key Laboratory of Marine Chemistry Theory and Technology, Ministry of Education, Ocean University of China, Qingdao 266100, China.

E-mail address: yaopeng@ouc.edu.cn (P. Yao).

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There is increasing evidence that SOC firstly solubilizes into high-molecular-weight DOC (HMW-DOC) through extracellular enzymatic hydrolysis and then further transfers to low-molecular-weight DOC (LMW-DOC) through fermentation and/or hydrolysis, which is then oxidized to DIC via terminal respiration process such as sulfate and iron reduction (Alperin et al., 1994; Burdige et al., 2016; Komada et al., 2012). Numerous studies have shown that sedimentary dynamic processes (e.g., physical reworking activities, seasonal erosion and re-deposition) are key factors controlling degradation of SOC especially in mobile-muds (Aller et al., 1996; Alongi et al., 2012; Yao et al., 2014; Zhao et al., 2017). In particular, LDEs and their adjacent marginal seas, with high SOC inputs from riverine sources and primary production, provide an ideal location for examining factors controlling remineralization of SOC in a diversity of sedimentary regimes.

The East China Sea (ECS) and Yellow Sea (YS), located in the western Pacific Ocean, receive large inputs of freshwater, sediments, OC, and nutrients from the Changjiang and Yellow River (Hu et al., 2013; Milliman and Meade, 1983; Milliman and Syvitski, 1992). These riverine-derived terrestrial materials are subject to long-term hydrodynamic sorting and reworking processes that result in the formation of several large mud deposits in the eastern China marginal seas (Li et al., 2014a; Liu et al., 2006, 2007; Qin et al., 1996). Similar to mobile-muds in the Amazon delta, the ECS mobile-muds (ECSMMs) have high sedimentation rates, but the preservation efficiency of terrestrial SOC is relatively low (about 30%) (Blair and Aller, 2012; Yao et al., 2014). Compared with ECSMMs, the central South Yellow Sea mud deposits (SYSMDs) also has large mud pools but they are relatively stable and less influenced by terrestrial inputs (Dong et al., 2011; Zhou et al., 2015). While past studies on diagenetic processes and remineralization of SOC (e.g., Aller et al., 1985; Song et al., 2016; Yao et al., 2014; Zhao et al., 2017) and SOC budgets (Deng et al., 2006; Song et al., 2016; Tao et al., 2016) in the YS and ECS have provided valuable insights of SOC cycling, a comparison of remineralization rates in these different sedimentary regimes has yet to be explored.

Here, we build on previous work to further study remineralization of SOC in the mud deposits of ECS and YS, using time-sequence sediment incubation method. We provide an SOC budget that includes remineralization rates for ECS and YS, which allows for a better understanding of the role of mobile-muds in SOC remineralization. Finally, we also make comparisons with mobile-muds in tropical estuarine regions (e.g., Amazon mobile-muds and Gulf of Papua) to better constrain early diagenetic cycling and controlling mechanisms of remineralization under different sedimentary regimes.

2. Materials and methods

2.1. Site description

The ECS and YS are typical passive marginal seas characterized by their shallow and broad epicontinental shelf (Blair and Aller, 2012; Liu et al., 2003; Milliman et al., 1989; Zhu et al., 2013). The ECS is a subtropical marginal sea covering an area of approximately $0.77 \times 10^6 \text{ km}^2$, with typical water depths of $< 200 \text{ m}$, and large inputs of freshwater ($9.0 \times 10^{11} \text{ m}^3 \text{ yr}^{-1}$) and sediment ($4.70 \times 10^8 \text{ t yr}^{-1}$) from the Changjiang River (Milliman and Farnsworth, 2011; Yang et al., 2011). Once discharged to the coast, the fate of these terrestrial materials delivered by the Changjiang Diluted Water (CJDW), is mainly controlled by seasonal variations in the Taiwan Warm Current (TWWC), Zhe-Min Coastal Current (ZMCC) and the Yellow Sea Coastal Current (YSCC) (Liu et al., 2006, 2010a, 2010b). In fall and winter, the CJDW is constrained by an intensified ZMCC southward along the inner shelf that extends to the northeast in summer - due to an intensified TWWC (Liu et al., 2006). These complex hydrographic dynamics preclude sediments from escaping the Changjiang Estuary and ECS inner shelf, and result in the formation of a mobile-mud belt (within the 75 m isobath) that extends from the Changjiang Estuary to the Taiwan Strait

(Fig. 1a) (Qin et al., 1996; Liu et al., 2003, 2006, 2007). Mud deposits in the estuarine and inner ECS shelf regions occur in a diverse range of sedimentary regimes - largely due to the complex sedimentary dispersal dynamics and textural/instability differences of the these deposits (Li et al., 2011; Wang et al., 2015; Yao et al., 2015; Zhao et al., 2017). Recent studies have shown that frequent physical reworking and summer bottom-water hypoxia in mobile-muds of ECS inner shelf play an important role in remineralization of SOC and other diagenetic processes (Xu et al., 2015; Yao et al., 2014; Zhao et al., 2017; Zhu et al., 2016).

The YS is a temperate semi-enclosed marginal sea that receives large amounts of freshwater and sediments from several large rivers, including the Yellow River, Changjiang, Xinyi River, Sheyang River, Han River and Geum River (Fig. 1b) (Zhou et al., 2015). The circulation pattern of the YS in winter is characterized by a counterclockwise gyre, with a northward flowing Yellow Sea Warm Current (YSWC) and southward flowing Yellow Sea Coastal Current (YSCC) (Hu, 1984; Hu et al., 2013). In summer, the YSWC turns eastward into the Cheju Strait followed by a flow to the north. The YSWC is a branch of the Tsushima Current that carries saline and warm water into the YS and plays a critical role to the formation of the Yellow Sea Cold Water Mass (YSCWM), which has great influence on primary production and hydrographic features of the SYS (Hu, 1984; Shi et al., 2003; Zhang et al., 2008). Relatively weak hydrodynamic conditions, combined with the YSWC and cold eddies in the SYS, contribute to the formation of the SYSMDs (Dong et al., 2011). Recent studies showed that the main sources of sediments in the SYSMDs are the Yellow River and the Old Yellow River Delta (Hu et al., 2013; Zhou et al., 2015).

2.2. Sample collection

Four cruises were conducted onboard the R/V Haijian 47 in July 2012, R/V Dongfanghong 2 in July 2013, R/V Kexue 3 in August 2013, and R/V Kexue 1 in June 2014, respectively. Samples were obtained from 12 sites in ECSMMs and SYSMDs (Fig. 1). Water depth, bottom-water temperature, and salinity at each site were measured using a CTD-rosette system (Seabird 911 Plus, USA). Surface sediments (ca. 3 cm in depth) were sampled at each site using a 0.25 m^2 stainless-steel box-corer and stored in pre-combusted small alumina boxes at -20°C prior to laboratory analyses. Six short cores were sub-sampled in the same box for sediment incubation experiments (see details below).

2.3. Sediment incubation and pore water collection

Net remineralization flux of SOC was determined using anaerobic whole sediment core incubations, sampled at intervals of 20 cm over 30 days, according to Aller et al. (1996), as modified by Yao et al. (2014) (see Text S1 in the supporting information for details of sediment incubation). At incubation times of 0 d, 10 d and 30 d, pore-water samples were collected at 1 cm intervals. This incubation method provided a relatively labor-free method for determining net remineralization rates of SOC and solutes released during SOC remineralization (Aller and Mackin, 1989; Aller et al., 1996). However, the reaction rates estimated by this incubation method are likely to represent a conservative estimate of SOC remineralization rate, due to the absence of physical and biological disturbances commonly found in these dynamic sedimentary regimes (Aller et al., 1996; Yao et al., 2014).

Pore-water samples were extracted immediately after section using Rhizon samplers (Netherlands) inserted into sediments and attached to vacuum test tubes by 23G needles in N_2 glovebox, and first 1 mL pore-water was discarded (Seeberg-Elverfeldt et al., 2005; Yao et al., 2014). Approximately 7 mL of pore-water were collected from each sediment section within 2 h of collection. After collection, pore-water samples were further split into acidified or non-acidified sub-samples and stored at 4°C for subsequent analysis. Non-acidified samples were analyzed for DIC, dissolved inorganic nitrogen (DIN) nutrients (NH_4^+ , NO_3^- , and

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