



## Dissolved organic carbon dynamics in anaerobic sediments of the Santa Monica Basin

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Received 23 May 2012; accepted in revised form 11 February 2013; available online 27 February 2013

### Abstract

Cycling of dissolved organic carbon (DOC) was investigated in anoxic sediments of the Santa Monica Basin, California Borderland, by analyzing the concentration and isotopic signatures ( $\Delta^{14}\text{C}$  and  $\delta^{13}\text{C}$ ) of pore-water DOC and dissolved inorganic carbon (DIC), and organic compound classes extracted from the bulk sediments. DOC and DIC increased across the sediment–water interface, indicating net efflux of these solutes out of the sediments. Throughout the depth interval examined (0–30 cm), the  $\Delta^{14}\text{C}$  value of DOC ( $\Delta^{14}\text{C}_{\text{DOC}}$ ) was similar to, or higher than, that of bulk sedimentary particulate organic carbon (POC), indicating degradation of relatively  $^{14}\text{C}$ -rich components of POC. There were prominent peaks in both  $\Delta^{14}\text{C}_{\text{DOC}}$  and  $\Delta^{14}\text{C}_{\text{DIC}}$  in the uppermost 2 cm of the sediment column, indicating degradation and remineralization of  $^{14}\text{C}$ -rich, labile organic matter in the near-surface sediments. However, below these sub-surface maxima,  $\Delta^{14}\text{C}_{\text{DOC}}$  and  $\Delta^{14}\text{C}_{\text{DIC}}$  decreased with depth by  $\sim 200\%$  and  $\sim 50\%$ , respectively. Given the diffusive time scales, these decreases were too large to be explained by  $^{14}\text{C}$  loss due to radioactive decay. To help explain these observations, we constructed and implemented a selective degradation model that considers bulk pore-water DOC to be the sum of three kinetically- and isotopically-distinct sub-components. Based on this model, the most reactive DOC fraction, which supported  $\sim 60\%$  of the DIC production, had a  $\Delta^{14}\text{C}$  value indicating the presence of bomb- $^{14}\text{C}$ . The intermediate fraction had a  $\Delta^{14}\text{C}$  value of  $\sim -60\%$  and accounted for most of the pore-water DOC standing stock. The least reactive fraction was virtually non-reactive in these sediments, and had a  $\Delta^{14}\text{C}$  value of  $\sim -500\%$ . The benthic DOC flux of this  $^{14}\text{C}$ -depleted, poorly-reactive DOC fraction may represent a source of pre-aged, refractory DOC to the oceans.

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### 1. INTRODUCTION

Continental margin sediments play a key role in the marine and global cycles of carbon through active organic

matter degradation and burial (Hedges and Keil, 1995; Burdige, 2007). While particulate organic carbon (POC) undergoing degradation is mostly remineralized to dissolved inorganic carbon (DIC) and inorganic nutrients, it has been found that  $< \sim 10\%$  of POC undergoing degradation accumulates as dissolved organic carbon (DOC) in the pore waters (Alperin et al., 1999; Burdige, 2002; Fig. 1). This net DOC production in turn supports an efflux of DOC out of the sediments to the water column. Globally, marine sediments are estimated to supply DOC to the water column at a rate comparable to global riverine DOC fluxes ( $\sim 0.18 \text{ Gt yr}^{-1}$ ; Burdige et al., 1999). Despite this quantitative importance, the significance of net DOC production in the marine carbon cycle remains unclear, because of a lack

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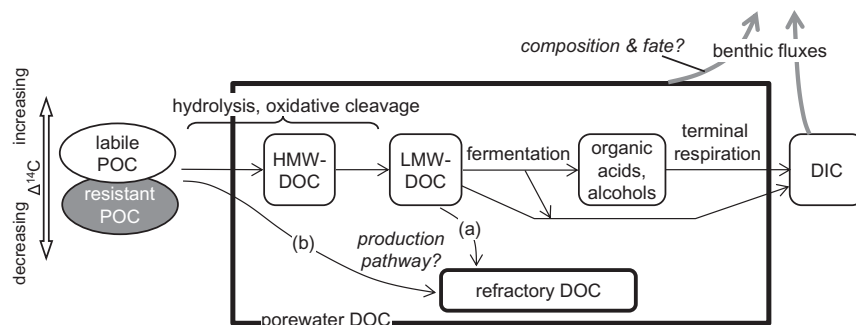


Fig. 1. Organic carbon degradation in sulfate-reducing sediments (see e.g., Fenchel et al. (1998) and Arnosti (2010) for further details). POC is first converted into high-molecular-weight DOC (HMW-DOC) via hydrolysis and/or oxidative cleavage. HMW-DOC is further transformed into low-molecular-weight (LMW) DOC, such as amino acids and monosaccharides. Fermentation converts LMW-DOC into short-chain organic acids and alcohols (along with  $\text{CO}_2$  and  $\text{H}_2$ ), which are respired to DIC. Sulfate reducers can also directly oxidize some LMW-DOC to DIC. Two possible pathways for the formation of refractory DOC are shown: (a) internal conversion of originally-labile HMW- or LMW-DOC (Burdige and Gardner, 1998), (b) direct production from POC solubilization (Robador et al., 2010; Komada et al., 2012). Benthic DOC fluxes are supported by a mixture of labile and refractory porewater DOC.  $\Delta^{14}\text{C}$  signatures were used in this study as proxy for OC reactivity (far left of the figure; see text for details).

of understanding of the mechanism for DOC production, and the composition and reactivity of DOC that accumulates in the pore waters.

Pore-water DOC composition has been investigated through quantification of specific compounds (e.g., short-chain organic acids, amino acids; Sansone and Martens, 1982; Burdige and Martens, 1990) and major classes of biomolecules (Lomstein et al., 1998; Burdige et al., 2000). These studies show that such identifiable components can be relatively abundant near the sediment–water interface, but overall are a minor fraction of the pore-water DOC pool due to their high reactivity. The majority of pore-water DOC instead consists of moieties that are not readily characterized by conventional analytical techniques (Burdige, 2001, 2002). This apparent structural complexity of pore-water DOC is consistent with findings from ultra-high-resolution mass spectrometry (Koch et al., 2005; Tremblay et al., 2007; Schmidt et al., 2011). Investigation into the molecular size distribution of pore-water DOC further shows that with the exception of near-surface sediments where high-molecular-weight (>1 kDa) DOC is relatively abundant, pore-water DOC is dominated by components of lower molecular weight (Burdige and Gardner, 1998). This has led to the hypothesis that analogous to DOC in the water column (Amon and Benner, 1996), most pore-water DOC consists of highly degraded, low-molecular-weight material of limited reactivity (Burdige and Gardner, 1998). The presence of poorly-reactive DOC in sediment pore waters has been supported by laboratory incubation experiments (Hee et al., 2001; Komada et al., 2012), and through modeling studies (Burdige, 2002; Komada et al., 2004).

The occurrence of DOC that resists degradation in the pore waters leads to two major questions (Fig. 1). First, how is refractory DOC produced in sediments, and how is its production related to the overall degradation and remineralization of organic carbon (OC) in the sediments? There are two standing hypotheses for the origin of recalcitrant DOC in sediments: production within the pore waters

themselves through alteration of originally labile monomers (pore water size reactivity model; Burdige and Gardner, 1998), and production of refractory DOC directly from POC hydrolysis (Robador et al., 2010; Komada et al., 2012). Second, what is the composition of DOC that is exported to the water column, and what is the fate of this material in the water column? Modeling studies show that the benthic flux is dominated by labile, high-molecular-weight DOC whose production rate is high near the sediment–water interface (Burdige, 2001, 2002). This suggestion is consistent with high  $\Delta^{14}\text{C}$  values reported for pore-water DOC collected from the surface sediments (0–10 cm depth) of the Santa Monica Basin, California Borderland, and Station N, located on the continental rise of the eastern North Pacific Ocean (Bauer et al., 1995). However, benthic DOC fluxes must also be supported at least in part by refractory DOC (Fig. 1). The significance of benthic DOC fluxes in the marine carbon cycle would depend strongly on the magnitude of this refractory DOC flux, and the reactivity of this material in the oxic water column (Alperin et al., 1999).

The goal of this study was to further investigate the mechanism of pore-water DOC accumulation, and to better understand the role of pore-water DOC in the overall organic matter degradation process. To achieve these goals, we determined  $^{14}\text{C}$  and  $^{13}\text{C}$  signatures of pore-water DOC and DIC in the suboxic to anoxic sediments of the Santa Monica Basin (SMB), California Borderland, and analyzed them using a selective degradation model. The  $^{14}\text{C}$  signatures were used as proxies for OC reactivity, where high  $^{14}\text{C}$  abundance (high  $\Delta^{14}\text{C}$ , young radiocarbon age) was interpreted to indicate the presence of young, reactive material, and low  $^{14}\text{C}$  abundance (low  $\Delta^{14}\text{C}$ , old radiocarbon age) was interpreted to indicate the presence of aged material of limited reactivity (e.g., Trumbore, 2000; Mayorga et al., 2005; Repeta and Aluwihare, 2006; Blair and Aller, 2012; Fig. 1). As shown below, results obtained applying our selective degradation model to the pore water data support these assumptions. Finally, in addition to pore water constituents, we applied  $^{14}\text{C}$  and  $^{13}\text{C}$  measurements to

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