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## THE ROLE OF BENTHIC FLUXES OF DISSOLVED ORGANIC CARBON IN OCEANIC AND SEDIMENTARY CARBON CYCLING

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**Abstract.** Benthic fluxes (sediment-water exchange) of dissolved organic carbon (DOC) represent a poorly quantified component of sedimentary and oceanic carbon cycling. In this paper we use pore water DOC data and direct DOC benthic flux measurements to begin to quantitatively examine this problem. These results suggest that marine sediments represent a significant source of DOC to the oceans, as a lower limit of the globally-integrated benthic DOC flux is comparable in magnitude to riverine inputs of organic carbon to the oceans. Benthic fluxes of DOC also appear to be similar in magnitude to other sedimentary processes such as organic carbon oxidation (remineralization) in surface sediments and organic carbon burial with depth.

## Introduction

In recent years, considerable effort has been devoted to the study of sediment oxygen uptake and benthic fluxes of inorganic nutrients, since these processes have been shown to play major roles in marine biogeochemical cycles [e.g., Bender et al., 1989; Martens et al., 1992]. In contrast, far less attention has been given to the examination of benthic fluxes of dissolved organic carbon (DOC), although their possible importance in sedimentary and oceanic processes has been discussed by numerous authors [Emerson and Dymond, 1984; Heggie et al., 1987; Bender et al., 1989; Mopper et al., 1991; Alperin et al., 1992; Hedges, 1992; Martens et al., 1992].

Interest in DOC fluxes from marine sediments further stems from a recognition of the need to better understand the sources and sinks of DOC in the oceans [Hedges, 1992]. This is in part driven by recent results which have reported oceanic DOC concentrations (based on high temperature catalytic oxidation (HTCO) techniques) that are 2 to 3 times higher than previously reported values determined with either UV or persulfate oxidation techniques [see Martin and Fitzwater, 1992 and references cited therein]. It has also been suggested that sediments might represent an important source of DOC to the deep ocean [Williams and Druffel, 1987; Mopper et al., 1991; Hedges, 1992], and that benthic DOC fluxes might provide an explanation for the apparent discrepancy between the "old" (~6,000 ybp) <sup>14</sup>C age of deep water DOC, the average oceanic mixing time (~1,000 yr), and other chemical properties of deep water DOC (i.e., <sup>δ</sup><sup>13</sup>C and lignin concentrations) which suggest that this material is primarily of marine origin.

In this paper, we use pore water DOC data to calculate DOC fluxes from marine sediments. The calculations use published pore water DOC data obtained by UV or wet chemical oxidation techniques, and recent pore water data obtained with HTCO techniques. Calculated DOC fluxes will also be compared with recent direct measurements of benthic DOC fluxes. All of these results will then be used to begin to quantitatively examine the role of benthic DOC fluxes in sedimentary and oceanic carbon cycling.

## Calculation of Benthic DOC Fluxes Using Pore Water Gradients

Concentration profiles of DOC have been measured in a number of marine sediments (e.g., see the references cited below and in Table 1). In general, pore water DOC concentrations are up to an order of magnitude higher than those in the overlying waters (Figure 1), suggesting the possibility of a diffusive DOC flux out of the sediments. Neglecting for now possible advective transport

across the sediment-water interface mediated by macrobenthos, this diffusive flux ( $J$ ) can be predicted by Fick's First Law, as modified for marine sediments [Berner, 1980],

$$J = -\phi_0 D_b (\partial c / \partial z)_0 \quad (1)$$

where  $D_b$  is the bulk sediment diffusion coefficient, and  $\phi_0$  and  $(\partial c / \partial z)_0$  are the porosity and concentration gradient, respectively, at the sediment-water interface.

DOC fluxes from marine sediments estimated with eqn. (1) and pore water concentration data are subject to several potential sources of error. The first involves the possibility that UV or persulfate oxidation techniques underestimate actual pore water and bottom water DOC concentrations (see discussion above). However in pore waters collected in Cape Lookout Bight, NC, the standard persulfate oxidation technique recovered  $93 \pm 2\%$  of the pore water DOC, when compared to DOC values obtained by either HTCO (at 680°C) or sealed tube combustion (at 850°C) techniques [Alperin and Martens, 1992]. Given the generally large differences between pore water and bottom water DOC concentrations, the magnitude of the DOC gradient across the sediment-water interface is predominantly determined by the pore water DOC concentration. Thus if the observations in Cape Lookout Bight pore waters are applicable to other sediments, the current controversy concerning the measurement of DOC in seawater does not appear to significantly affect the magnitude of the DOC fluxes calculated here.

A second problem in performing these calculations involves assigning a diffusion coefficient to the complex and largely uncharacterized mixture of organic compounds that makes up pore water DOC. We have chosen to adopt a range for this diffusion coefficient using the log-log relationship between  $D^*$  (the free solution diffusion coefficient) and molecular weight shown in Figure 2. This relationship is observed for organic compounds with molecular weights of 16 to ~200,000 and a wide range of molecular shapes and functional groups. The slope of this line (-0.39) is virtually identical with that predicted by the Stokes-Einstein equation ( $= -1/3$ ; Cornet et al. [1986]), providing some theoretical foundation for this observation. If pore water DOC is assumed to have an average molecular weight between 1,000 and

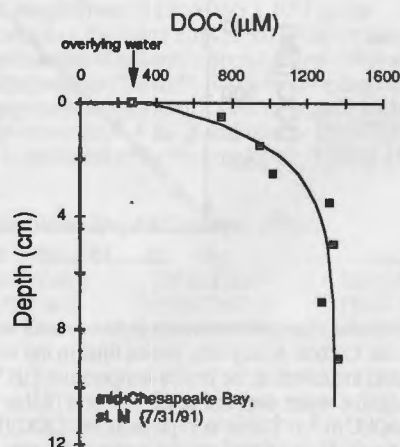


Fig. 1. Dissolved organic carbon (DOC) versus depth in a core from the mid-Chesapeake Bay (see note e in Table 1 for the location of this site). These pore waters were obtained using sediment squeezers [Burdige and Martens, 1990], although similar smooth diffusion controlled DOC profiles have also been observed in Cape Lookout Bight sediments for pore waters obtained by centrifugation [Alperin and Martens, 1992]. DOC concentrations were determined in both cases using a Shimadzu TOC-5000 Total Carbon Analyzer.

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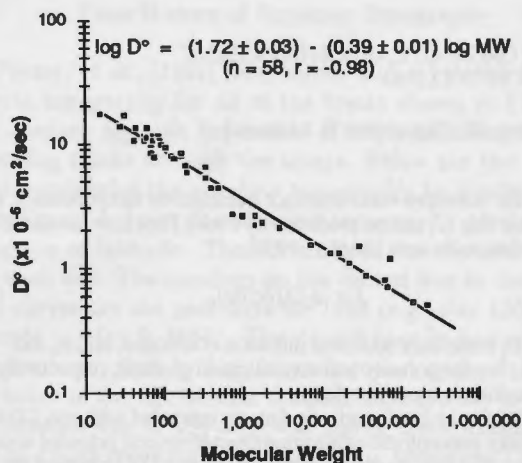


Fig. 2. Free solution diffusion coefficient ( $D^\circ$ , at 25 °C in distilled water) versus molecular weight for various organic compounds. The original data [Rossi et al., 1958; Jost, 1960; Cooper and Stepto, 1969; Sahares and Witherspoon, 1970; Lenninger, 1975; Cornel et al., 1986; The Handbook of Chemistry and Physics, 70<sup>th</sup> ed.] were corrected to 25 °C with the Stokes-Einstein equation [Jost, 1960].

10,000, this relationship predicts an average  $D^\circ$  value for pore water DOC (at 3 °C) that ranges from 0.8 to 1.9  $\times 10^{-6}$  cm<sup>2</sup>sec<sup>-1</sup>. Given the available data on the molecular weight size distribution of pore water DOC [Krom and Sholkovitz, 1981; Orem and Gaudette, 1984; Chin and Gschwend, 1991], this assumed range of molecular weights seems reasonable. It should also be noted that the approximate inverse cube root relationship between DOC diffusion coefficients and molecular weights implies that a factor of ten range in the average pore water DOC molecular weight translates into only a factor of two uncertainty in the DOC diffusion coefficient.

Several other factors may also lead to inaccuracies in benthic DOC fluxes calculated with eqn. (1). The advective transport of pore waters across the sediment-water interface by macrobenthos (i.e., "bioturbation" or "bioirrigation") could enhance the flux of DOC from marine sediments, relative to that which would occur by molecular diffusion alone [e.g., Berner, 1980]. Lastly, in making these calculations we have estimated the DOC concentration gradient at the sediment-water interface,  $(\partial c/\partial z)_0$ , by  $\Delta c/\Delta z$ , where  $\Delta c$  is the

concentration difference between the bottom waters and the pore waters in the first sediment section sampled, and  $\Delta z$  is the depth of the mid-point of this sediment section. This assumption of a linear DOC gradient near the sediment-water interface also introduces potential errors in these calculations. Enhanced DOC production or consumption near the sediment-water interface may lead to curvature in the DOC depth profile that is not observed with the 1 - 2 cm resolution of these pore water DOC data. Estimates of the DOC flux assuming a linear concentration gradient may be either over- or underestimates of their true values (e.g., see Burdige and Martens [1990] for a further discussion of this problem).

#### Comparison of Measured and Calculated Benthic DOC Fluxes

The ability of eqn. (1) to accurately estimate benthic DOC fluxes was tested by comparing directly measured and calculated DOC fluxes from two coastal sediments. In the sediments of Cape Lookout Bight, NC and the mid-Chesapeake Bay good agreement was observed between measured and calculated, benthic DOC fluxes (Figure 3). In contrast, recent benthic flux studies in California continental margin sediments did not yield a good agreement between directly measured and calculated benthic DOC fluxes (D.J. Burdige, J. Homstead and W.M. Berelson, unpub. data, 1991). In these sediments DOC fluxes calculated with eqn. (1) underestimated (by a factor of ~50 - 150) measured DOC fluxes determined with an *in situ* benthic flux lander. The causes of this discrepancy are not currently well understood, and may be related to several factors. These include: the coarse resolution (2 cm) of the pore water data used to calculate the DOC fluxes from these sediments; the suggestion that the processes controlling benthic fluxes from these sediments occur in a thin, mm scale layer of material at the sediment-water interface (W.M. Berelson, K. Johnson and K. Coale, unpub. data, 1991).

#### Results of the DOC Flux Calculations

Calculated benthic DOC fluxes and benthic carbon remineralization rates ( $\Sigma OCR$ ) from a range of sediments are compiled in Table 1. These results suggest that 1 to 25% of the remineralized organic carbon escapes these sediments as a benthic DOC flux. One of the Peru Upwelling Region sites (st. 4) may be an exception to this observation, although it is difficult to ascertain this unequivocally given the large range in the  $\Sigma OCR$  value for this site.

An attempt was also made to quantify DOC fluxes from deep-sea sediments using published pore water data from these environments [Emerson and Dymond, 1984; Heggie et al., 1987]. The DOC

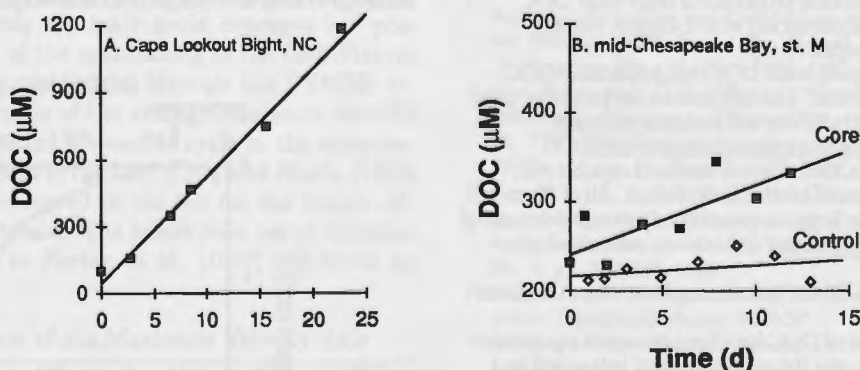


Fig. 3. DOC benthic flux measurements in two coastal sediments. Shown here are the DOC concentrations (determined using a Shimadzu TOC-5000 Total Carbon Analyzer), versus time in the waters overlying the cores. A. Cape Lookout Bight, NC. This core was collected on 10/29/90, and incubated at the *in situ* temperature (18 °C). To both stir and maintain oxic conditions during the experiment, air was bubbled through the water over the core. The best fit line through the data predicts a benthic DOC flux from these sediments of  $-0.55 \pm 0.03$  mol C m<sup>-2</sup> yr<sup>-1</sup>. For comparison, the DOC flux from these sediments calculated as described in the text using pore water DOC data was  $-0.66 \pm 0.28$  mol C m<sup>-2</sup> yr<sup>-1</sup> (Alperin et al., ms. in prep., 1992). B. Mid-Chesapeake Bay (solid squares, line labelled "Core"). This core was collected on 7/31/91, and incubated at the *in situ* temperature (26 °C). The water over the core was mixed (~8 rpm) during the experiment with a glass stirring rod attached to an electric motor. Since the bottom waters of this portion of the Bay were anoxic at this time, N<sub>2</sub> gas was blown over the water overlying the core during the experiment. Oxygen was undetectable in this water, and the faint smell of sulfide was also noted in collected water samples. Also shown here is a control experiment (open diamonds, line labelled "Control") in which bottom water collected on 7/31/91 was incubated anaerobically at the *in situ* temperature. The best fit line through the "Core" data (corrected for the slight DOC production in the control experiment) predicts a benthic DOC flux from these sediments  $-0.34 \pm 0.14$  mol C m<sup>-2</sup> yr<sup>-1</sup>. For comparison, the DOC flux from these sediments calculated as described in the text using pore water DOC data (e.g., Fig. 1) was  $-0.47 \pm 0.19$  mol C m<sup>-2</sup> yr<sup>-1</sup> (Table 1).

TABLE 1. Calculated Benthic DOC Fluxes, and Depth-Integrated Organic Carbon Remineralization Rates in Several Marine Sediments

Site	Water Depth	DOC Flux <sup>a</sup>	ΣOCR <sup>b</sup>	DOC Flux/ΣOCR
Cape Lookout Bight, NC (coastal lagoon) <sup>c</sup>	9 m	-2.2 ± 1.7 [H]	47.6 ± 5.7	0.01 - 0.08
Skan Bay, AK (intermittently anoxic marine basin) <sup>d</sup>	65 m	-3.5 ± 1.5	20.9 ± 2.4	0.10 - 0.25
site M, mid-Chesapeake Bay (estuary) <sup>e</sup>	12 m	-0.47 ± 0.19 [H]	11.1 ± 1.1	0.02 - 0.07
Buzzards Bay, MA (coastal bay) <sup>f</sup>	17 m	-0.20 ± 0.09	1.2	0.10 - 0.23
central California continental margin <sup>g</sup>				
st. 17	100 m	-0.02 ± 0.01 [H]	1.9	~0.01
st. 12	1019 m	-0.03 ± 0.02 [H]	0.5	~0.06
st. 21	2000 m	-0.04 ± 0.02 [H]	-	-
Peru Upwelling Region (continental margin) <sup>h</sup>				
st. 4	92 m	-1.14 ± 0.47	0.6 - 4.3	0.16 - >1
st. 5A	268 m	-0.18 ± 0.08	1.7	0.06 - 0.15
st. 6	506 m	-1.12 ± 0.46	-	-

DOC fluxes and ΣOCR are mol C m<sup>-2</sup> yr<sup>-1</sup>, and negative DOC fluxes are out of the sediments.

<sup>a</sup> DOC fluxes were calculated as discussed in the text using eqn. (1) and pore water data from the references cited below.  $D_s$  values were determined by the equation  $D_s = D^0/\phi F$ , where  $D^0$  values were estimated as discussed in the text.  $F$  (the formation factor) was calculated from sediment porosity data [Ullman and Aller, 1982]. The errors listed here are primarily based on the range of diffusion coefficients used in these calculations, and do not include the possible effects of the other phenomena discussed in the text. [H] indicates DOC fluxes calculated using DOC concentrations obtained with HTCO techniques (using a Shimadzu TOC-5000 Total Carbon Analyzer).

<sup>b</sup> ΣOCR = Depth integrated rate of benthic organic carbon remineralization. For Cape Lookout Bight, Skan Bay and the mid-Chesapeake Bay, ΣOCR was determined from measured ΣCO<sub>2</sub> and CH<sub>4</sub> benthic fluxes, and was independently cross-checked with estimates of this quantity based on sediment data. For the other sites, ΣOCR values were determined as indicated below.

<sup>c</sup> From Martens et al. [1992] and Alperin et al. (ms. in prep., 1992).

<sup>d</sup> From Alperin et al. [1992].

<sup>e</sup> From Burdige and Homstead (ms. in prep., 1992). This station is in the meso-haline portion of the Bay (35° 35' N, 121° 15' W).

<sup>f</sup> From Henrichs and Farrington [1987]. ΣOCR was determined by modeling sediment profiles.

<sup>g</sup> These sites are off the California coast near Pt. Piedras Blancas between 35° and 36° N, and 121° and 124° W. ΣOCR values are based on benthic lander measurements (made 6/91) of sediment oxygen and nitrate uptake (W.M. Berelson et al., unpub. data, 1991).

<sup>h</sup> DOC data from Henrichs and Farrington [1984]. The ΣOCR value for st. 5A and the lower limit for st. 4 are from Henrichs and Farrington [1984] and were determined by modeling pore water profiles. The upper limit for st. 4 is from Henrichs and Reeburgh [1987] and is based on sulfate reduction rate measurements from Rowe and Howarth [1985].

fluxes calculated here were comparable in magnitude to those reported in Table 1, suggesting that deep-sea sediments could be significant sources of DOC to the oceans. However, these calculated deep-sea benthic DOC fluxes also exceeded published ΣOCR values for the same sites, and in some cases also exceeded the rain rate of organic carbon to the sediment surface. The reasons for these inconsistencies are not currently understood. They may be related to the possibility that these measured deep-sea pore water DOC concentrations are elevated over their true *in situ* values, due to pressure artifacts associated with their collection [Emerson and Dymond, 1984; Heggie et al., 1987]. This would then lead to calculated DOC fluxes also being overestimated. As the magnitude of this potential artifact is unknown, we have chosen not to further quantitatively consider these calculated deep-sea benthic DOC fluxes.

#### Biogeochemical Implications of These Results

The data in Table 1 can be used to make a first estimate of the globally-integrated benthic flux of dissolved organic carbon to the oceans. Uncertainty in the accuracy of published pore water DOC

measurements from deep-sea sediments (see above), and the lack of any published direct DOC benthic flux measurements from such sediments, limits this calculation to sediments in water depths less than 2000 m. Since this approach does not include a major fraction of the ocean's sediments, our estimate represents a lower limit of the global benthic DOC flux. Nonetheless, the importance of this calculated lower limit will be seen below.

Marine sediments in water depths less than 2000 m were divided into four oceanic provinces [Henrichs and Reeburgh, 1987], and the data in Table 1 were used to estimate ranges of the DOC fluxes from the sediments of each region. The integrated DOC flux from these sediments was then obtained by summing the contributions from each region (Table 2). This calculation leads to an estimate of this quantity that ranges from 0.12 to 0.93 × 10<sup>14</sup> gC/yr.

A comparison of the lower limit of the global benthic DOC flux with other parameters of oceanic carbon cycling yields several interesting observations (Table 3). It can be seen that benthic DOC fluxes may represent a significant source of organic carbon to the oceans, as the lower limit of the global benthic DOC flux is comparable to estimates of the riverine inputs of either POC or DOC.

TABLE 2. Integrated Benthic DOC Flux from Marine Sediments in Water Depths Less than 2000 m

Oceanic Regime	Percent Marine Sediment Area <sup>b</sup>	Total Area <sup>b</sup> (10 <sup>12</sup> m <sup>2</sup> )	DOC Flux <sup>c</sup> (mol C/m <sup>2</sup> /yr)	Integrated Flux (10 <sup>12</sup> mol C/yr)
Shallow, highly anoxic, methanogenesis in upper 1-2 m (CLB, SB) <sup>a</sup>	0.03	0.11	-1.8 to -4.0	0.19 to 0.43
Coastal upwelling regions, 0-500 m (PU, CA [st. 17]) <sup>a</sup>	0.4	1.44	-0.01 to -1.61	0.01 to 2.32
Estuaries and bays, 0-50 m (BB, CB) <sup>a</sup>	0.6	0.66	-0.11 to -0.66	0.24 to 1.40
Shelf and slope sediments, <2000 m (CA [sts. 12, 21]) <sup>a</sup>	16.5	59.4	-0.01 to -0.06	0.59 to 3.56
			Total	1.04 to 7.74
			[= 0.12 to 0.93 × 10 <sup>14</sup> gC/yr]	

<sup>a</sup> These abbreviations indicate the sites taken to be in each of these oceanic regimes (CLB = Cape Lookout Bight; SB = Skan Bay; CB = the mid-Chesapeake Bay; BB = Buzzards Bay; CA = central California continental margin; PU = Peru Upwelling Region).

<sup>b</sup> From Henrichs and Reeburgh [1987]. The areas listed here are based on a total marine sediment area of 360 × 10<sup>12</sup> m<sup>2</sup>. Note that the sediments included in this calculation represent approx. 20% of the total marine sediment area.

<sup>c</sup> These ranges are taken from Table 1, based on the sites considered to be in each of these oceanic regimes.



TABLE 3. Organic Carbon Fluxes in the Marine Environment

Flux	Value (10 <sup>14</sup> gC/yr)
Riverine DOC input <sup>a</sup>	2
Riverine POC input <sup>a</sup>	2.3
Organic carbon oxidation in surface marine sediments <sup>a</sup>	0.2 to 5
Organic carbon burial in marine sediments <sup>a,b</sup>	0.6 to 1.4
Lower limit of the benthic DOC flux <sup>c</sup>	0.1 to 0.9
Organic carbon deposition to marine sediments <sup>d</sup>	0.9 to 7.3; 4.5

<sup>a</sup> From Emerson and Hedges [1988], Hedges [1992] and references cited therein.

<sup>b</sup> This is the organic carbon which escapes remineralization in surface sediments.

<sup>c</sup> from Table 2.

<sup>d</sup> The first range is the sum of the sediment carbon oxidation and burial fluxes, and the lower limit of the benthic DOC flux. The second value was determined with an equation for water column (sediment trap) organic carbon fluxes vs. water depth [Martin et al., 1987]. This equation was depth-integrated over 1000 m intervals and then summed over the entire ocean using data on sediment surface area vs. water depth [Open Univ., 1989].

Benthic DOC fluxes also appear to be significant when compared to estimates of organic carbon oxidation and burial in marine sediments. This suggests that these fluxes may need to be included in sediment nutrient budget calculations [e.g., Martens et al., 1992], as well as conceptual models of biogeochemical processes in marine sediments.

In summary, our results suggest that dissolved organic carbon fluxes from marine sediments may represent an important component of oceanic and sedimentary carbon cycling. However as discussed above, calculating benthic DOC fluxes in the manner described here is subject to possible inaccuracies. One way to overcome many of these problems involves the direct measurement of DOC fluxes, using either *in situ* benthic landers [e.g., Bender et al., 1989] or incubated cores. Measurements of this latter type in nearshore sediments (Figure 3) along with preliminary *in situ* measurements in continental margin sediments (K. Mopper and A.H. Devol, unpub. data, 1991; D.J. Burdige, J. Homstead and W.M. Berelson, unpub. data, 1992) have, however, yielded measured benthic DOC fluxes that are similar to the calculated fluxes in Table 1. Therefore, the results presented here represent reasonable first quantitative estimates of the role of benthic DOC fluxes in oceanic and sedimentary carbon cycling. Future direct measurements of DOC fluxes from all marine sediments will allow for a more critical examination of their role in oceanic and sedimentary processes.

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