FINAL REPORT

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Project Title: Deposition and Fate of Modern Organic Carbon in Shelf/Upper Slope Sediments near Cape Hatteras, North Carolina

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DOE Grant #DE-FG05-95ER62073

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Summary of Accomplishments

Sulfate reduction rates (SRR) measured in North Carolina continental shelf and upper slope sediments using shipboard techniques appeared to be anomalously high relative to estimates of organic carbon input and remineralization. The SRR have also appeared to be incompatible with the relative lack of sulfate depletion seen in sediment porewaters below 30 cm depth unless relatively deep (but slow) bioirrigation is invoked. Of the possible reasons the rates could be too high, the most probable reason seemed to be the pressure shock associated with bringing deep samples to the surface, possibly releasing intracellular organic substrates usable by sulfate reducers. To test for this effect, we helped design and deployed a prototype *in situ* sediment injector to conduct the world's first *in situ* deep sea sulfate reduction rate measurements. This is a computer-controlled device, deployed by the Johnson Sea Link submarine, configured to make three 25 cm vertical line injections of radio-labeled sulfate on a single dive. Injection points were overcored immediately and the cores left in place to incubate from 24-48 hours. Parallel cores were retrieved to measure rates using standard shipboard methods. The measured rates decreased with depth from about $50 \mu\text{M} \cdot \text{d}^{-1}$ near the sediment-water interface to about $20 \mu\text{M} \cdot \text{d}^{-1}$ at 22 cm depth. The *in situ* rates agreed with and thus verified our shipboard rates. The results imply that anaerobic remineralization processes exert a much greater control on organic matter burial rates in slope environments than had been previously recognized.

Shelf and upper slope sediments comprise ~15% of the seafloor but account for ~40% of the global organic carbon burial. The large burial flux for these environments is in part due to high burial efficiency: a relatively large fraction of the deposited organic

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matter escapes remineralization. A leading theory of organic matter preservation—the Sorptive Preservation Model—states that sedimentary organic matter is protected from degradation by sorption onto mineral surfaces. An implication of this model is that organic matter must pass through a dissolved state before being preserved. Thus, factors that regulate production and consumption of porewater DOC may influence the quantity of organic matter that is ultimately buried. We developed techniques for sampling and analyzing porewater DOC and investigated the role that DOC plays in controlling burial efficiencies on the North Carolina continental slope (Alperin et al., 1999). Net DOC production rates were estimated by applying a diagenetic model to DOC concentration profiles. The model results indicated that net DOC production rates and organic carbon burial rates are highly correlated. Elevated DOC concentrations may promote organic matter preservation on the continental slope and elsewhere.

We were also directly involved in studies quantifying the rates of sedimentation and surficial sediment mixing at a series of shelf/upper slope sites. Together with benthic chamber chemical flux measurements, this work allows for the determination of organic carbon burial under a range of depths and environmental conditions as well as the fractions of new versus refractory carbon being buried.

Introduction and Purpose of Study

The atmospheric inventory of CO₂ has increased at an average rate of 3 Gt C/yr between 1981–1987 in response to the combustion of fossil fuels and tropical deforestation. The mismatch between the observed rate of increase and the well-constrained fossil fuel combustion rate (5.3 Gt/yr) has indicated the need to carefully consider the sinks of atmospheric CO₂ (Tans et al., 1990). The oceans may be the single largest sink as they appear to absorb approximately one third of the anthropogenically released CO₂, based on various computational models (Broecker et al., 1979; Toggweiler et al., 1989; and others).

Walsh et al. (1981) and Walsh (1989) proposed that a significant portion of the missing fossil fuel CO₂ is exported to the continental slope as shelf-produced organic matter. Subsequent studies have disputed the general hypothesis that the oceans could be responsible for large-scale removal of the atmospheric C via biological productivity on decadal timescales (Tans et al., 1990; Broecker, 1991; Berner, 1992). Even so, the long-term burial of organic carbon on the continental slope is expected to be an important component of the global and marine C-cycles, based on Holocene and present day sedimentary inventories of particulate organic matter (Premuzic et al., 1982; Romankevich, 1984; Walsh et al., 1985). A portion of the upper slope centered near 500–700 meters water depth has been identified as a potential depocenter for marine organic carbon (Martens, 1987).

Results of the DOE sponsored Shelf-Edge Exchange Processes (SEEP) program indicated that most of the shelf-produced organic matter was recycled on the shelf in the

Mid-Atlantic Bight (Falkowski et al., 1988; Rowe et al., 1988). However, because of uncertainties in the estimates of annual primary productivity and respiration rates, as much as 10–20% of the primary production could be exported off the shelf (Falkowski et al., 1988). The exported C was not found on the slope of the Mid-Atlantic Bight and it was argued that the POC, if it did escape the shelf, may be advected to the Cape Hatteras region (Walsh et al., 1988; Anderson et al., 1994).

The convergence of the Gulf Stream and Virginia Current over the slope offshore from Cape Hatteras may focus particulate transport into that region (Schaff et al., 1992). The presence of a physical "null" zone beneath the Gulf Stream and overlying the upper slope seabed may enhance net deposition of particles in that area (Kelchner, 1992). Following deposition, lateral sediment transport and the delivery of young, reactive organic matter to the upper slope should be facilitated by the extremely steep incline of the seabed in this region. A variety of observations indicate that particulate organic carbon is focused into this region. The upper slope of Cape Hatteras is characterized by unusually dense benthic animal communities (Blake et al., 1985; Schaff et al., 1992; Blake et al., 1993; Hecker, 1993) that must be supported by a significant flux of metabolizable organic carbon. Portions of the Hatteras slope experience sediment and organic carbon accumulation rates that appear to be 1–2 orders of magnitude greater than other sites in the Mid-Atlantic and South Atlantic Bight. Some of this material is young, and thus could be recently derived from atmospheric CO₂, as indicated by the presence of short-lived tracers, such as chlorophyll *a* ($t_{1/2}$ = 1 mo) and ²³⁴Th ($t_{1/2}$ = 22 d), in surface sediments. Some of this material is also reactive as indicated by the animal populations, benthic dissolved inorganic C (DIC) and O₂ fluxes, the pore water profiles of DIC, dissolved organic carbon (DOC), sulfate and methane, as well as measured rates of sulfate reduction (Schaff et al., 1992; Martens et al., 1987, 1994; Alperin et al., 1994).

The fate of the organic carbon deposited on the slope offshore from Cape Hatteras may differ dramatically from that deposited in other slope environments. Recent models for the Mid-Atlantic Bight (Anderson et al., 1994) and observations made in the South Atlantic Bight (Blair et al., in press) suggest that most of the deposited material is oxidized aerobically, possibly in the upper millimeters of seabed. However, biogeochemical studies of the Hatteras region noted above indicate that subsurface anaerobic processes are important. This may be because of the rapid sediment accumulation and intense bioturbation mediated by the dense infaunal communities. *In situ* tracer experiments suggest that the residence time on the surface of recently deposited organics may be as short as days as a result of deposit feeding activities (Blair et al., 1994). The rapid injection of organic matter into the seabed may be an extremely significant feature of this environment. Through controlling the balance of aerobic and anaerobic processes, the transport of the organic debris would influence the character and quantity of organic carbon ultimately buried, including that recently derived from atmospheric CO₂.

Objectives of the Proposed Research

The continental upper slope offshore of Cape Hatteras was identified as a potential depocenter for shelf and slope-derived organic carbon. The overall objectives of this study were to verify if indeed organic carbon deposition is focused into this region, and, if it is, to determine the fate of this carbon. The experimental efforts of research groups led by us and by our collaborators in the benthic processes team from other institutions including David J. DeMaster and Neal E. Blair at NC State University, and R.A. Jahnke at Skidaway, were guided by the following hypotheses:

- (1) Organic matter recently derived from atmospheric CO₂ is focused onto the Cape Hatteras upper slope.
- (2) Sedimentary organic carbon remineralization in the Cape Hatteras upper slope area is dominated by subsurface anaerobic processes.
- (3) Deposition and cycling of organic carbon in the Cape Hatteras area is controlled by mesoscale processes on the 100 km scale and bottom topography and currents on the local scale.

The primary focus of our group at the University of North Carolina at Chapel Hill was to test hypothesis number (2) above; however we also contributed substantially to obtaining data for and testing the other two hypotheses.

The sampling stations used in our research were located on several downslope transects between 300 and 1500 m along the 36°20' N and 35°23' N latitudinal lines. A second transect was run along-slope at a depth of ~700-900 m between 35°23' N and 36°20' N. The endpoints of the along-slope transect were targeted for detailed process-level work.

The depositional flux of the young organic carbon fueling anaerobic remineralization processes in slope sediments was constrained by estimating its oxidation and burial rates. The organic carbon oxidation rate, as reflected by DIC production, was determined from core incubation experiments, in situ flux measurements (by R. Jahnke) and modeled pore water DIC profiles (Table 1). Benthic O₂ flux and sulfate reduction rate measurements provided additional estimates of organic carbon oxidation rates (Table 1). Further characterization of the organic pool was made by a combination of organic geochemical and stable isotopic measurements. $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ measurements of the C_{org} and total N pools were used to constrain the relative importance of marine and terrestrial sources (Fry and Sherr, 1984).

Burial of organic matter and sediment on the Cape Hatteras slope was not expected to occur as a steady-state process. A suite of naturally occurring and bomb-produced radioactive tracers were used to evaluate these fluxes (see publications and presentations list at end of report).

Rates of Anaerobic Organic Carbon Remineralization

One of the principal objectives of the Ocean Margins Program was to determine the mechanisms by which dissolved organic carbon, particulate organic carbon and dissolved inorganic carbon, were interconverted by molecular and microbial processes in coastal sediments. Quantification of the principal respiratory modes through which organic carbon is remineralized was central to this objective. Of the alternative respiratory electron acceptors available in marine sediments, sulfate is usually dominant by virtue of its abundance in seawater. Microbial sulfate reduction can equal aerobic respiration in the overall remineralization of organic carbon in shelf/slope sediments (e.g. Jorgensen 1982) and must be considered to give an accurate view of their overall respiratory activity.

Before our study, efforts had focused on measuring benthic oxygen demand. Quantification of subsurface anaerobic respiration rate was achieved through a combination of conventional shipboard and novel, in situ rate measurements, the latter obtained using a new submarine-deployable device designed specifically for this project. The in situ device produced the first ever in situ sulfate reduction rates in the deep sea

Hypothesis 2 was a natural outgrowth of the apparent discrepancy between the conclusion of Anderson et al. (1994) and our own initial research concerning the relative importance of surface (aerobic) and subsurface (anaerobic) carbon remineralization in upper slope sediments. Anderson et al. (1994) conclude that almost all organic carbon turnover at the SEEP II sites takes place at the interface. However, at sites farther to the south near Cape Hatteras, our measurements of subsurface sulfate reduction yielded anaerobic carbon turnover rates that exceeded total carbon input estimates and oxygen uptake at the SEEP II sites.

Sulfate Reduction Rates in Upper Slope Sediment Near Cape Hatteras

Measured sulfate reduction rate profiles on the slope off Cape Hatteras were made on box core subsamples in the upper 30-40 cm of sediment, and extended through the use of Kasten core subsamples to 225-250 cm. Averaged data from box and kasten cores are illustrated in Figure 1. The results indicate that approximately half of the depth integrated sulfate reduction ($6 \text{ mmol} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$) occurs in the upper 30 cm of sediment. Rates are lower, but easily measurable to depths greater than 2 meters. This deeper sulfate reduction doubles the depth integrated rates to $12 \text{ mmol} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$.

Comparison of our rates with published values from other shelf and slope environments (Table 2) shows that they are comparable to other deep-sea sites, such as the shelf and upper slope beneath the Peru upwelling area, though somewhat higher than other shelf/slope environments.

Within the Mid-Atlantic Bight region, comparison of our sulfate reduction rates with other measures of organic carbon degradation point out the relative importance of

this process (Table 1). We do not have any direct measurements of organic carbon flux to the sediments at our sites, but at the SEEP II sites to the north, Anderson et al. (1994) measured fluxes of $3\text{--}6 \text{ mmol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ via sediment trap deployments (Table 1). Jahnke's oxygen fluxes measured via *in situ* benthic chambers at slope sites in this area range from $4\text{ to }8 \text{ mmol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ (personal communication) which would support the aerobic oxidation of the same number of moles of organic carbon, and are thus, by themselves, roughly in balance with the carbon flux measurements. Our sulfate reduction rates, however, would support the anaerobic oxidation of $16\text{--}28 \text{ mmol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ of organic carbon and are thus substantially higher than the carbon flux estimates. CO_2 flux measurements by Jahnke and Blair (personal communication) and Blair et al. (in press) are also lower than the sulfate reduction rate measurements, ranging from $2\text{--}11 \text{ mol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$.

Given that the other data in Table 1 are from different locations from those used to obtain the sulfate reduction rate data it is difficult to say if their imbalance should be regarded as problematic. It does, however, illustrate the importance of subsurface anaerobic degradation of organic carbon on the upper slope. Anderson et al. (1994) estimated the remineralization rate of organic carbon below the sediment-water interface at SEEP II sites to be only $0.4 \text{ mmol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ through use of a mixing model based on the solid phase organic carbon profile and the sedimentation rate. This result was used to argue that 90 percent of the labile organic carbon deposited at SEEP II sites was remineralized at the sediment-water interface with only 10 percent remineralized within the sediment. This conclusion followed principally from the absence of significant vertical gradient in carbon content. The model result, however, was not highly constrained due to the scatter in the organic carbon vs depth data. Based on our own model results, it is equally plausible that 80 percent of the carbon remineralization takes place below the sediment water interface.

Organic carbon deposition rates are crucial to the OMP objective of understanding the carbon cycle of shelf/slope environments. However, in most locations there will never be long-term sediment trap data from which to establish these. The best way to establish these rates may be through the summation of burial fluxes and remineralization rates at and below the sediment-water interface.

Controls on sulfate reduction rates: The role of DOC

Regardless of the magnitude of sulfate reduction rates in continental margin sediments, anaerobic decomposition plays an important role in controlling the ultimate fate of organic carbon deposited on the sea floor (Henrichs and Reeburgh, 1987). In order to understand what factors control whether sedimentary organic matter is remineralized or buried, it is necessary to elucidate the details of the anaerobic decomposition process. In marine sediments, sulfate is the dominant electron acceptor for anaerobic remineralization (Reeburgh, 1983). Therefore, understanding the controls on sulfate reduction rates will enhance our understanding of the processes that control the fate of

organic carbon deposited in margin sediments.

The key to understanding controls on sulfate reduction rates is to identify the rate limiting step in the organic matter decomposition process. It is likely that the overall rate of anaerobic decomposition is controlled by the rate at which HMW-DOC is transformed to LMW-DOC.

The focus of the research described in this section was to characterize downcore variations in the molecular weight distribution of pore water DOC in sediments from our proposed study sites. In addition to providing insight into the details of the anaerobic decomposition process, characterizing the molecular weight of pore water DOC allowed us to address several key questions pertaining to carbon cycling in margin sediments (Alperin et al., in press):

- Is a significant fraction of the pore water DOC pool of low enough molecular weight to have sufficient mobility to support an appreciable diffusive flux across the sediment-water interface?
- Is there an increase in the HMW-DOC concentration with depth that would be consistent with the formation of refractory geomacromolecules?

An important aspect of any study of sediment DOC is the sampling method. A number of studies have shown that sediment DOC concentrations can be affected by the technique used to isolate the pore water. For example, Martin and McCorkle (1993) demonstrated that whole core squeezing leads to artificially high DOC concentrations. They also showed that removing benthic fauna by sieving results in lower DOC in pore water obtained by centrifugation. Likewise, Carignan et al. (1985) report that DOC concentrations in pore water obtained by centrifugation were higher and more variable than for samples obtained by dialysis.

To avoid artifacts in DOC concentration associated with squeezing and centrifuging sediment, we collected pore water using a "sipper" device. The "sipper" is composed of a cylindrical porous (60 μm) membrane that can be inserted horizontally into the side of a core. The membrane is attached to a syringe which can be cocked to create a partial vacuum. This method uses minimal force (< 1 atm) to extract pore water from whole sediment. A direct comparison of DOC in replicate cores shows that concentrations are lower and profiles are smoother when pore water is obtained by "sipper" rather than centrifuge (Fig. 13).

Sediment depth-distributions of DOC from our down-slope transects (300-1000 m water depth) follow a trend consistent with other porewater constituents (ΣCO_2 and SO_4^{2-}) and a tracer of modern, fine-grained sediment (fallout Pu), suggesting that DOC levels are regulated by organic matter remineralization. However, remineralization rates appear to be relatively uniform across the sediment transect. A simple diagenetic model illustrates that variations in DOC profiles may be due to differences in the depth of the

active remineralization zone, which in turn is largely controlled by the intensity of bioturbation. Comparison of porewater DOC concentrations, organic carbon burial efficiency, and organic matter sorption suggest that DOC levels are not a major factor in promoting organic matter preservation or loading on grain surfaces. The DOC benthic fluxes are difficult to detect, but suggest that only ~2% of the dissolved organic carbon escapes remineralization in the sediments by transport across the sediment-water interface.

Publications and Presentations at National Meetings supported by DOE Grant #DE-FG05-95ER62073

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Figure captions

Figure 1. Average sulfate reduction rates measured at all upper continental slope stations using conventional shipboard S-35 techniques. The solid circles are data from MK-3 box core samples. The diamonds show data from deeper kasten cores.

Figure 2. A tree-way comparison of sulfate reduction rates determined using conventional shipboard S-35 techniques (open circles), deck-incubated samples using the in situ injector technology on core sediments (open squares), and in situ rate data obtained from use of the new sediment injector on the sea floor (closed circles). The latter data are the first known deep sea in situ rate measurements of sulfate reduction.

Figure 1.

THREE WAY COMPARISON OF SULFATE REDUCTION RATE RATE SAMPLE INCUBATION METHODS

SAMPLES FROM NC CONTINENTAL SLOPE
AT 35°
25°N, DEPTH = 750m

Table 1. Measures of Organic Carbon Remineralization in Slope Sediments of the Mid-Atlantic Bight

	Method	mmol m ⁻² d ⁻¹	Reference
Organic carbon flux to the sediments	Sediment traps	4 to 6	Anderson et al. (1994)
	²¹⁰ Pb flux and C _{ORG} /X _S ²¹⁰ Pb of trapped particles	3 to 6	Anderson et al. (1994)
Organic carbon burial	¹⁴ C sedimentation rate and C _{ORG}	0.5 to 2	Anderson et al. (1994)
O ₂ flux into the sediments	<i>In situ</i> benthic chamber	5 to 8	Anderson et al. (1994)
	<i>In situ</i> benthic chamber	4 to 8	Jahnke (personal comm.)
	Shipboard incubation	1 to 11	Blair (personal comm.)
ΣCO ₂ flux out of the sediments	<i>In situ</i> benthic chamber	7 to 9	Jahnke and Blair (personal comm.)
	Shipboard incubation	2 to 11	Blair et al. (in press)
	Gradient calculation	2 to 5	Blair et al. (in press)
	Shipboard incubation	11-21	Green et al. (personal comm.)
Potential integrated SO ₄ ²⁻ reduction rate	Shipboard ³⁵ S injection and incubation	8 to 14	This study
Organic carbon remineralization within the sediments	NH ₄ ⁺ flux calculated from pore water concentration gradient	0.4	Anderson et al. (1994)
	C _{ORG} gradient mixing model	< 0.4	Anderson et al. (1994)