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SULFATE REDUCTION IN DEEP-SEA SEDIMENTS

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ABSTRACT. Sulfate reduction rates calculated from about 200 DSDP pore water sulfate profiles have been contoured and plotted on a map covering most areas of the world ocean. Rates show a remarkable spatial consistency, with high rates observed near the continental margins, becoming progressively lower toward the central ocean basins. Relatively elevated rates are also found in the eastern equatorial Pacific, a site of upwelling and correspondingly high rates of primary organic production. Overall, the distribution of sulfate reduction in pelagic sediments looks very similar to the distribution of primary organic carbon production. When rates are directly compared, however, the correlation between sulfate reduction and primary production is only moderately strong. Perhaps the most important influence on sulfate reduction is sediment deposition rate and the control this has over the fraction of the sedimentary organic carbon flux that becomes available for sulfate reduction. The slower the rate of sediment deposition the more time for oxic respiration and the less organic carbon that escapes to the zone of sulfate reduction. To predict most accurately sulfate reduction rates, however, the variables of primary production, water depth, and sediment deposition rate must all be integrated.

INTRODUCTION

Numerous processes are responsible for the oxidation of organic carbon in marine sediments including oxic respiration, denitrification, metal oxide reduction, sulfate reduction, and methanogenesis (for example, Emery and Rittenberg, 1952; Stumm and Morgan, 1970; Froelich and others, 1979; Berner, 1980; Henrichs and Reeburgh, 1987). Worldwide, oxic respiration is quantitatively the most important, with sulfate reduction next (Jørgensen, 1983; Henrichs and Reeburgh, 1987). The relative importance of the these two processes changes, however, such that in near-shore sediments equal amounts of organic matter are oxidized by sulfate reduction and oxic respiration (Jørgensen, 1982, Canfield, 1989), whereas in deep-sea sediments oxic respiration dominates (Bender and Heggie, 1984; Canfield, 1989).

In recent years the mechanisms controlling the rates of oxic respiration in sediments, including the variables of primary production, water depth, and bottom water oxygen level, have been explored (Smith and Hinga, 1983; Jørgensen, 1983; Emerson, 1985). A similarly comprehensive look at sulfate reduction in deep-sea sediments has not been taken. The present paper takes advantage of a large set of sulfate reduction rate data obtained from about 200 DSDP (Deep-Sea Drilling Project) sulfate profiles. These data allow a look at the global distribution of sulfate reduction rates and promote a refined understanding of the processes controlling sulfate reduction in deep-sea sediments.

METHODS

DSDP sulfate profiles were used to obtain sulfate reduction rates in deep-sea sediments by assuming that sulfate reduction within the sediment column is at steady state; that is, that the profiles are not presently changing. At steady state, the rate of sulfate reduction will be balanced by sulfate inputs into the sediment, which include molecular diffusion and advection into the zone of sulfate reduction. To use molecular diffusion coefficients to calculate transport rates, it is further assumed that pore water disturbance by sediment mixing does not occur within the zone of sulfate reduction. The latter assumption is likely valid for most pelagic sediments, because sulfate reduction occurs over depths ranging from 10's to 100's of meters, far too deep to be influenced by bioturbating organisms. The steady state assumption, however, is only approximate, but small deviations from true steady state will not affect the interpretations offered here.

Mathematically, the following formulation was used to obtain sulfate reduction rates from sulfate profiles:

$$\text{rate} = \phi D_s dC/dx + \phi_o \omega C_o - \phi_b \omega C_b \quad (1)$$

where ϕ is the average sediment porosity in the zone of sulfate reduction, C is the concentration of sulfate, and C_o and C_b are, respectively, the concentrations of sulfate at the sediment water interface and at the base of the sulfate reduction zone (where sulfate no longer changes with depth). Sediment porosity at the sediment surface is given by ϕ_o and at the base of the sulfate reduction zone, by ϕ_b, x is sediment depth, D_s is the diffusion coefficient for sulfate at 4°C (Li and Gregory, 1974), corrected for tortuosity (Berner, 1980), and ω (cm yr^{-1}) is the average rate of sediment deposition in the zone of sulfate reduction (see also Canfield, 1989).

In some instances only minimum sulfate reduction rates could be obtained, either because few data exist near the sediment surface where maximum sulfate gradients are observed, or because some profiles exhibit considerable scatter in their data. Where appropriate, these data have been identified. The accuracy of sulfate reduction rate calculations is difficult to assess, but, in cases other than those just mentioned, gradients could be resolved to better than a factor of two. A complete list of data is available upon request.

RESULTS AND DISCUSSION

Sulfate reduction rates have been plotted and contoured on a world map, which is shown in figure 1. There is an impressive spatial coherency in the regional patterns of sulfate reduction, and this coherency is not compromised by mixing older and newer (likely of higher quality) DSDP sulfate data. When older and newer sites are in the same region, rates are usually similar. Overall, relatively elevated rates are observed nearest to the continental margins, including marginal seas (Bering Sea, Gulf of Mexico, Mediterranean Sea, Greenland Sea), with low rates in

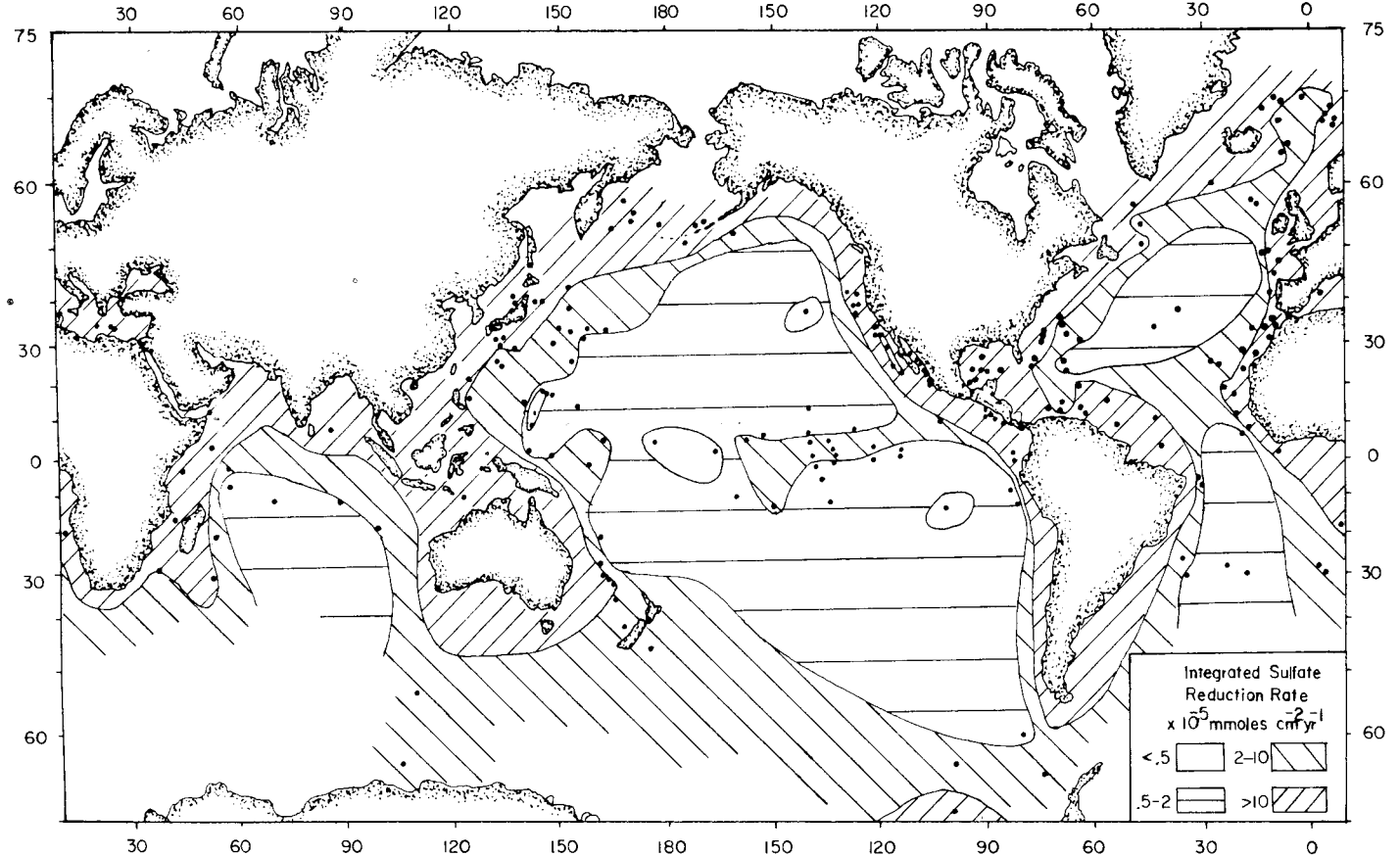


Fig. 1. World map of sulfate reduction rates in deep-sea sediments. Data from about 200 DSDP stations were used, with station locations given by black dots.

the ocean interiors, and the lowest rates in red clay regions. Due to a paucity of pore water sulfate data in red clay sediments, the extent of very low sulfate reduction rates may be underestimated.

Sulfate reduction rates from figure 1 are comparable to rates predicted from the distribution of organic carbon in deep-sea sediments as reported by Waples and Sloan (1980). For example, at DSDP site 444 organic carbon decreases almost linearly from about 0.3 to 0.07 wt percent over a depth interval of 55 m, corresponding to about 4 my of burial. By 55 m depth no further decrease in organic carbon concentration is observed. Assuming that changes in organic carbon concentration are due to organic matter oxidation and not variable input concentrations, with a sediment water content of 50 percent (by weight; deVries Klein, Kobayashi, and others, 1980), organic matter is oxidized at a rate of 23.7×10^{-5} mmoles cm^{-2} yr^{-1}. If sulfate reduction is the dominant oxidation pathway, an average sulfate reduction rate of half this value, or 11.9×10^{-5} mmoles cm^{-2} yr^{-1}, is calculated. At DSDP sites 442, 443, and 445 (Waples and Sloan, 1980), the distribution of organic matter would suggest sulfate reduction rates of 28×10^{-5} mmoles cm^{-2} yr^{-1}, 28×10^{-5} mmoles cm^{-2} yr^{-1}, and 14×10^{-5} mmoles cm^{-2} yr^{-1}, respectively. Sulfate profiles have not been reported for these stations, but, from figure 1, a similar rate of about 10×10^{-5} mmoles cm^{-2} yr^{-1} is observed in the same region (approx 25°–30°N, 132°–137°E).

From the above depth-integrated rates of sulfate reduction, average per volume rates range between about $1–5 \times 10^{-5}$ mmoles l^{-1} (whole sediment) yr^{-1}. These sulfate reduction rates are extremely slow, about 100 to 1000 times slower than the slowest rates reported from radiotracer studies (Volkov and Rozanov, 1983, p. 386–387). Even at such slow rates, sulfate reduction still probably results from the biogenic dissimilatory oxidation of organic matter, as opposed to a slow abiotic process. Two arguments can be offered to support the biogenic nature of sulfate reduction in deep-sea sediments. First, the available literature, though not extensive, shows that finely dispersed reduced sulfur in deep-sea sediments (usually in very low concentration) is isotopically light, leaving isotopically heavy pore water sulfate (Volkov and Rozanov, 1983; Claypool and Kaplan, 1974; Goldhaber and Kaplan, 1974), indicative of biologically mediated sulfate reduction. Second, as discussed by Waples and Sloan (1980) and Claypool and Kaplan (1974), in deep-sea sediments at several hundred meters depth, below the zone of sulfate depletion (when this occurs), isotopically light methane may be found. Isotopically light methane forms from the biogenic breakdown of organic matter (Claypool and Kaplan, 1974) and would suggest that organic matter oxidation processes which precede methane production, such as sulfate reduction, may also have been mediated by bacteria.

Returning to figure 1, the global distribution of sulfate reduction is somewhat similar to that observed for water-column primary production (Koblentz-Mishke, Volkovinsky, and Kabanora, 1970; Berger and others, 1987; Berger, 1989) and the distribution of phytoplankton

pigment concentrations as determined by satellite imaging (National Geographic, 1988). This is especially true in the eastern equatorial Pacific Ocean where a tongue of elevated sulfate reduction rates underlies a similar distribution of high rates of primary production (Koblentz-Mishke, Volkovinsky, and Kabanora, 1970; Berger and others, 1987; Berger, 1989), which are fueled by the upwelling of nutrient-rich water. (A similar region of high equatorial rates of sulfate reduction in the Atlantic is not well defined and should be viewed only as suggestive.) The apparent correlation between sulfate reduction and primary production implies that the production and resulting flux of marine organic carbon to deep-sea sediments may be a dominant factor controlling rates of sulfate reduction.

Such a conclusion would be consistent with other measures of benthic metabolism which have also been found to correlate with water column primary production. For example, Lyle (1983) observed that in the eastern equatorial Pacific, the depth of the brown/green color transition, which is a rough measure of surface and near surface organic matter decomposition rates, correlated with overlying water-column primary production rates. Also, Smith and Hinga (1983) reported that rates of benthic oxic respiration correlated well with rates of primary production. However, when rates of sulfate reduction are directly compared to rates of primary production at each DSDP site (primary production rates obtained from the maps of Berger and others, 1987; and Berger 1989), only a weak correlation results (fig. 2). Such a weak correlation may reflect inaccuracies in measuring rates of primary production, or interpreting sulfate profiles, or both. Also potentially important is that rates of sulfate reduction in deep-sea sediments integrate up to a few million years of organic carbon input (see Toth and Lerman, 1977; Waples and Sloan, 1980), whereas primary production rates are an instantaneous rate measurement, not necessarily reflective of organic matter production in the past.

Indeed, there is some evidence that primary production rates have varied from present values. For example, Pederson (1983) and Pederson and others (1988) showed that organic carbon accumulation rates were higher than present in the equatorial Pacific during the last glacial maximum, about 18,000 yrs ago, and suggested that rates of primary production were elevated at this time. Also, during the same glacial maximum, rates of primary production in the Atlantic Ocean may have been, on average, about 20 percent higher than at present (Mix, 1989). Beyond these observations it appears that relatively high rates of primary production have persisted in the eastern equatorial Pacific through the Pleistocene and probably from the mid-Miocene (Lyle, Murray, and Finney, 1980; Thayer and others, 1985). However, it is not yet known how these long-term elevated rates of primary production compare to modern rates, or how they changed over time.

The potential time-variable nature of ocean primary productivity may help to explain why sulfate reduction rates in deep-sea sediments

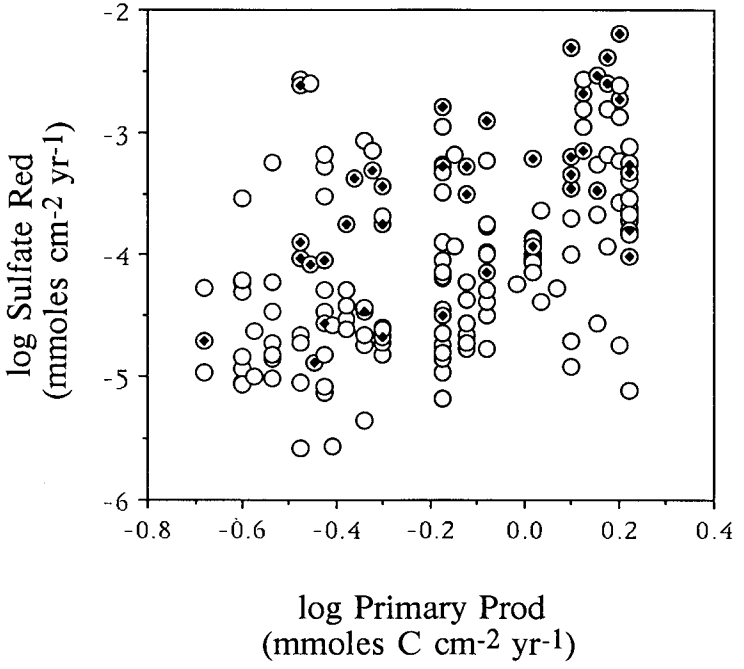


Fig. 2. Rates of sulfate reduction in deep-sea sediments are compared to rates of overlying water-column primary production. Some sulfate reduction rate calculations yielded minimum values, with rates potentially higher than shown. These data have been designated with small diamonds (see text for details).

correlate only poorly with modern rates of primary production. However, rates of water-column organic production may not be the most important factor influencing rates of sulfate reduction. From figure 2, primary production rates vary by about a factor of 10 in non-coastal ocean waters, whereas sulfate reduction rates in deep-sea sediments vary by a factor of nearly 10,000. Such a large difference in the magnitudes of variability of the two processes would suggest that additional factors are important in controlling rates of sulfate reduction.

Several authors have stressed that rates of sediment deposition are strongly correlated with sulfate reduction rates (Toth and Lerman, 1977; Berner, 1978; Canfield, 1989), which means that the rate of organic carbon delivery to sulfate reducers must also correlate with sediment deposition rate (Berner, 1978). Of interest, then, are the factors that control the delivery rate of organic carbon to sulfate reducers. Primary production, as discussed above, must have some control over the flux of organic carbon to the sediment surface. However, only some of this carbon will become available for sulfate reduc-

tion, and as shown by Canfield (1989; see also Jørgensen, 1982), the proportion of the total sedimentary carbon flux used in sulfate reduction is a strong function of sediment deposition rate. In general, as sedimentation rate slows, there is more time for processes preceding sulfate reduction (of which oxic respiration is the most important, Henrichs and Reeburgh, 1987) to oxidize organic matter, leaving less for sulfate reducers.

Two variables, then, likely control sulfate reduction rates in deep-sea sediments. The first is the sedimentary flux of carbon, and the second is sedimentation rate as it affects the proportion of the total carbon flux made available for sulfate reduction. Both these variables may be quantified and used to predict sulfate reduction rates with just a few easily-obtained parameters, which are available for all the DSDP sites discussed here. Calculated rates may be compared to rates obtained from DSDP sulfate profiles to assess whether the two variables are sufficient to account for the flux of carbon to sulfate reducers and the resulting rates of sulfate reduction. Also, how the two variables combine may be explored to determine the importance of each in regulating sulfate reduction rates.

The proportion of the total carbon flux made available for sulfate reduction is quantified using the data in Canfield (1989). From Canfield (1989, fig. 4) a good relationship is observed between the total amount of organic matter decomposed in marine sediments, which is equivalent to the sedimentary flux of metabolizable organic matter, and sediment deposition rate. The relationship is expressed mathematically in eq 2:

$$\log (C) = 0.68 \log (\text{sed rate}) + 0.6 \quad (2)$$

where C is the carbon oxidation rate (mmoles C cm^{-2} yr^{-1}) and sed rate is the sediment deposition rate (g cm^{-2} yr^{-1}). A good trend was also observed between sulfate reduction rate and sedimentation rate (eq 3):

$$\log (SR) = 1.52 (\text{sed rate}) + 0.6 \quad (3)$$

where SR is the rate of sulfate reduction converted to carbon oxidation rate assuming a 2/1 stoichiometry between carbon oxidized to sulfate reduced (mmoles C cm^{-2} yr^{-1}). The proportion of the total carbon flux used in sulfate reduction is given by eq 4, which is the difference between eqs 2 and 3:

$$SR/C = (\text{sed rate})^{0.84} \quad (4)$$

Two different scenarios will be considered for the flux of sedimentary organic carbon. In the first case, the flux is calculated by augmenting the rate of primary production overlying each DSDP site, as presented earlier, with an estimate of organic matter decomposition during settling to the sediment surface (for example, Suess, 1980; Betzer and others, 1984). As previously discussed, such an estimate must be viewed

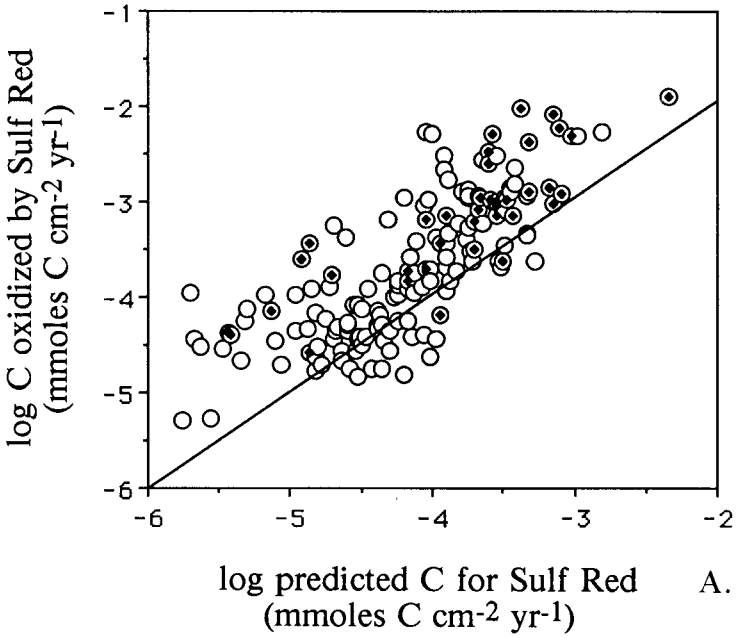
with caution because modern rates of primary production may not compare with rates in the past over the time scales by which sulfate reduction occurs. Also, organic matter may not settle directly to the sea bottom but may be transported and redistributed within the ocean by currents (for example, see Hollister, Ewing, and others, 1972, p. 141). To calculate sedimentary organic carbon flux, the following water depth/organic carbon flux relationship was used (Berger, 1987). This was considered by Berger (1987) to give the best overall prediction of organic matter oxidation during settling to the sea floor:

$$J_{sed} = 9\ PP/Z + 0.7\ PP/Z^{0.5} \quad (5)$$

where J_{sed} is the organic carbon flux to the sediment surface (mmoles C $cm^{-2}\ yr^{-1}$), PP is the rate of primary production (mmoles C $cm^{-2}\ yr^{-1}$), and Z is water depth (meters). This relationship allows two types of organic matter to result from primary production (see Westrich and Berner, 1984): a rapidly decomposing fraction whose extent of decomposition depends on $1/Z$ (see also Suess, 1980), and a fraction that decomposes more slowly, at a rate proportional to $1/Z^{0.5}$.

The parameters of water depth, primary productivity, and sedimentation rate are known for the different DSDP sites, and rates of organic carbon oxidation by sulfate reduction may thus be computed by multiplying the results from eqs 4 and 5. The accuracy of this estimate is not much compromised by ignoring the refractory sedimentary carbon flux that is not metabolized, because refractory carbon in deep-sea sediments makes up only between about 1 to 10 percent of the total flux (Henrichs and Reeburgh, 1987). The estimated flux of organic carbon for sulfate reduction is compared to organic carbon oxidation rates computed from sulfate profiles in figure 3A. Despite the caveats outlined above, the predicted rates of organic carbon oxidation by sulfate reduction are fairly close (on avg about 50 percent higher) to rates obtained from DSDP pore water data. It would appear that the magnitudes of the organic matter oxidation processes described by eqs 2 to 5 are roughly correct.

A second case considered is where the extreme assumption is made of a constant worldwide sedimentary organic carbon flux. With this scenario, only sedimentation rate and its control over the flux of organic carbon delivered to sulfate reducers influences predicted sulfate reduction rates. Using 0.01 mmoles C $cm^{-2}\ yr^{-1}$ as a typical value for the sedimentary flux of carbon (results from eq 5), organic carbon oxidation rates by sulfate reduction are computed and compared to DSDP-obtained values in figure 3B. The slope of the trend is much different from in figure 3A and shows that at higher rates of sulfate reduction (y axis) the predicted rates (x axis) become progressively underestimated. This demonstrates that higher rates of sulfate reduction must be driven, in part, by higher sedimentary fluxes of organic carbon, as predicted by the relatively weak, but positive trend between sulfate reduction rate and primary production in figure 2.



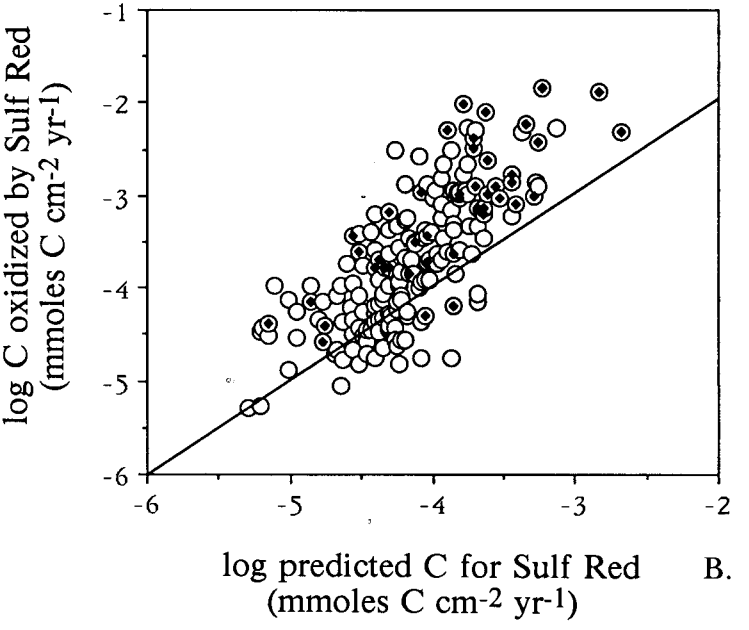


Fig. 3(A) The flux of organic carbon to the zone of sulfate reduction is calculated by combining the sedimentary organic carbon flux from eq 5 with eq 4 (see text for details). These results are plotted againt the amount of carbon oxidized by sulfate reduction, which is obtained by using the DSDP sulfate reduction rate data and assuming that two moles of carbon are oxidized for each mole of sulfate reduced. The line designates a 1/1 relationship. (B) The same as (A) except that eq 4 was combined with an assumed constant sedimentary organic carbon flux of 0.01 mmoles C cm^{-2} yr^{-1}. As in figure 2, results yielding minimum rates of sulfate reduction have been designated with small diamonds.

The patterns of sulfate reduction rate in figure 1, then, likely result from an interplay between sedimentation rate and its control over the proportion of the total sedimentary organic flux made available to sulfate reducers and primary productivity as it controls the sedimentary organic carbon flux. In fact, the trends in figure 1 are as much reminiscent of expected patterns in sediment deposition rates (highest nearer to the continental margins) as they are of patterns in primary production. This is even true in the eastern equatorial Pacific, where, from Thayer and others (1981), elevated rates of sediment deposition are found. Since high sedimentation rates in this area result from a high rain rate of biogenic debris from the productive surface waters, the two factors of sedimentation rate and organic carbon flux probably act in concert.

CONCLUSIONS

Sulfate reduction in deep-sea sediments is usually the terminal phase of organic matter decomposition, which begins in the surface ocean and continues as organic matter falls to the sediment surface. Further decomposition occurs during sediment burial by oxic respiration and other metabolic pathways. Algorithms may be written that reasonably quantify the amount of organic carbon lost during each phase of the transit. It is certain that these algorithms are only approximations, and that additional factors must also contribute to the ultimate fate of organic matter in sediments. However, the three variables of primary production rate, water depth, and sediment deposition rate, and their control over organic matter decomposition, combine to give a reasonably good approximation of sulfate reduction rates in deep-sea sediments. This implies that although not perfect, our knowledge of the processes controlling organic matter oxidation is fairly good (organic matter preservation in sediments is another matter).

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