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An experimental approach to quantify biologically mediated dissolved silicate transport at the sediment-water interface

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ABSTRACT: Fluxes of dissolved silicate (diss. Si) across the sediment-water interface were measured by incubating cores from the Bay of Fundy, Canada, and the Dutch Wadden Sea. The influence of infauna on diss. Si transport was estimated by inactivation of infauna with formalin poisoning and 'asphyxiation' Release of diss. Si in the Bay of Fundy ranged from 2.2 to 6.9 mmol Si m^{-2} d^{-1} (temp. 18 to 22 °C) before, and from 0.7 to 2.5 mmol m^{-2} d^{-1} after fauna-inactivation. In the Dutch Wadden Sea these data were −1.2 to 21.3 and −0.05 to 9.7 mmol diss. Si m^{-2} d^{-1} (temp. 13.5 to 20 °C), respectively. Effect of formalin-poisoning and 'asphyxiation' compared well. With only one exception fluxes of diss. Si after inactivation of sediment fauna approached, within a factor of 2, fluxes calculated from pore-water gradients by assuming that molecular diffusion is the only transport mechanism for diss. Si exchange.

INTRODUCTION

As a consequence of organic matter decomposition, dissolved substances are released and exchanged across the sediment-water interface, affecting overlying water composition. Especially in estuarine and coastal marine environments nutrient regeneration in the sediment can be an important factor in determining primary production in the overlying water (Nixon et al. 1976, Billen 1978).

The exchange between sediment and overlying water can be enhanced considerably by turbulence in overlying water (Vanderborght et al. 1977) and by the presence and activity of benthic organisms, which rework the sediment and irrigate the pore water (Aller 1980, Aller & Benninger, 1981, Gust & Harrison 1981, Henriksen et al. 1982, Boudreau 1984, Emerson et al. 1984, Aller & Yingst 1985, Aller et al. 1985).

In this paper we compare dissolved silicate (diss. Si) fluxes from coastal sediments incubated in the field and in the laboratory with diss. Si fluxes in 'poisoned' cores, to assess the influence of infauna on the diss. Si fluxes. Formalin was chosen as the 'poison' because it is reported to be effective for discrimination of 'biological' and 'chemical' oxygen uptake by sediments (Pamatmat 1971a, b, Hargrave 1972, Smith 1974, Granéli 1977, Hargrave & Phillips 1981, Andersen & Hargrave 1984).

Inactivation of infauna is also achieved when the oxygen level in overlying water is allowed to decrease naturally in incubated cores. This 'asphyxiation technique' was used by Rutgers van der Loeff et al. (1984) to distinguish between diss. Si exchange governed by molecular diffusion and the exchange accelerated by bio-irrigating infauna. We therefore compared the effect of formalin poisoning with the asphyxiation technique.

The reasons for using diss. Si as a natural tracer of sediment-water exchange in our study were both practical and theoretical. First, in coastal sediments there is usually a sharp concentration gradient of diss. Si across the sediment-water interface, enabling diss. Si fluxes to be detected relatively easily. Second, the analysis of diss. Si is simple and formalin does not interfere with it. Third, and most important, modelling of silicate pore-water profiles has revealed that silicate is not significantly involved in redox-reactions. Steady-state pore-water profiles are mainly determined by transport across the sediment-water interface and by a temperature-dependent first-order dissolution reaction of

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biogenic silicate. The rate of this reaction is proportional to the difference between saturation concentration of dissolved Si and actual Si pore-water concentration (Fanning & Pilson 1971, Hurd 1973, Berner 1974, Schink et al. 1975).

By applying a diagenetic model that accounts for the presence of burrows in sediment, Aller (1980) was able to quantify the influence of irrigated burrows on vertical distributions of pore-water solutes, and illustrated that profiles of diss. Si, subject to first-order dissolution kinetics, are less sensitive to bio-irrigation than profiles of solutes with zero-order reaction kinetics such as NH_4^+. Fluxes of diss. Si, in contrast to those of NH_4^+, are very sensitive.

Here we report results from experiments carried out, in 1982, through *in situ* incubation of cores at 2 locations in the intertidal area of the Bay of Fundy (Canada). We continued our work in 1985, with cores from 4 localities around the Island of Texel, The Netherlands. During the latter period, cores were incubated in the laboratory and, in addition to formalin-poisoning, an asphyxiation experiment was done for comparison of the 2 techniques.

MATERIAL AND METHODS

Bay of Fundy. Two intertidal locations, Anthony Park on the south side of Cobequid Bay, described by Hargrave (1978), and Peck's Cove on the north-west side of Cumberland basin, described by Gordon et al. (1980), were visited twice and once, respectively, during August-September 1982. Sediments at both locations consisted of well-sorted fine silt (mean grain size 30 µm). Surface sediments at both locations had an organic carbon content of 0.6 to 0.7 % C and contained 0.06 to 0.1 % organic nitrogen (Hargrave 1978, Gordon et al. 1980).

Predominant benthic macrofauna at both sites are the deposit-feeding mollusc *Macoma balthica* and the amphipod *Corophium volutator*. At Anthony Park densities of 1500 and 150 m^{-2} respectively were recorded (Hargrave 1978) and at Pecks Cove these were 150 and 1200 m^{-2} (Gordon et al. 1980).

Plexiglass core-tubes (diameter 5.7 cm; 11 cm length) were carefully inserted into the sediment at low tide. Overlying, filtered (Whatman GF/C) water (60 to 110 ml) was carefully siphoned into the core tubes above the sediment surface giving an overlying water column of 2.5 to 4.5 cm. Usually 2 cores were poisoned with 1 % buffered (pH 7.2) formalin solution. Incubation time of the cores was ≤ 200 min and chosen in such a way that overlying water O_2 concentrations did not decrease below 50 % saturation value. O_2 concentration was measured with an YSI Model-57 O_2-probe. Samples for diss. Si analyses were taken by syringe equipped with a filter-holder (Millipore 0.2 µm, cellulose acetate).

Island of Texel. Sediment cores were taken at 4 locations around the island: Hoornderslag, a sandy beach on the North Sea coast, and Mokbaai 1, Mokbaai 2, and Oost on the Wadden Sea. The latter had sandy-silty sediments with Mokbaai 1 as the coarsest and Oost as the finest (Andersen & Helder 1987). Organic C content ranged from 0.02 % (Hoornderslag) to 0.4 % at Oost. Macrofauna at the Texel locations consisted of bivalves (*Macoma balthica, Cardium* sp. and *Mya* sp.) and polychaetes (*Heteromastus filiformis, Nereis virens, Arenicola marina, Scoloplos* sp., *Lanice* sp. and *Phyllodoce* sp.). At Mokbaai 1 densities of bivalves and polychaetes were 125 and 340 m^{-2}, at Mokbaai 2 1100 and 350 m^{-2} and at Oost 880 and 260 m^{-2}, respectively.

Undisturbed cores were taken by hand about 3 h after high water slack in 0.5 m water depth. The Plexiglass core tubes were 30 cm long, 5.2 cm diameter and usually contained 6 to 7 cm overlying water upon recovery. Cores were handled carefully and brought to the lab within 0.5 h where they were placed in a thermostated room at the *in situ* temperature. During the incubation the cores were closed with a lid containing a magnetic stirrer and sampling ports. Samples for silicate analyses were taken at regular intervals. Poisoning with formalin was carried out as before and the asphyxiation experiment was carried out with cores from 'Oost'. During this last experiment the O_2 concentration in overlying water was followed with an O_2 electrode (Radiometer Blood Gas Analyser) according to Andersen & Helder (1987).

Pore-water profiles of diss. Si were obtained from all experimental sites by squeezing 0.5, 1.0 or 2.0 cm segments (depending on sediment depth) of the cores in Teflon Reeburgh-type squeezers (Reeburgh 1967). Silicate analyses were done on an auto-analyser according to the procedure given in Strickland & Parsons (1972). Calibration curves made with diss. Si additions to filtered seawater and with diss. Si additions to seawater containing 1 % formalin did not show significant differences.

Porosity of sediments was measured by weight loss of 5 cm^3 samples of wet sediment after heating at 75 °C to constant weight.

At the Texel sites resistivity profiles were made to assess the influence of tortuosity on molecular diffusion coefficients (Berner 1980). The resistivity measurements were carried out with a modified (Pt wires instead of stainless steel) 4-terminal resistivity probe (Andrews & Bennett 1981).

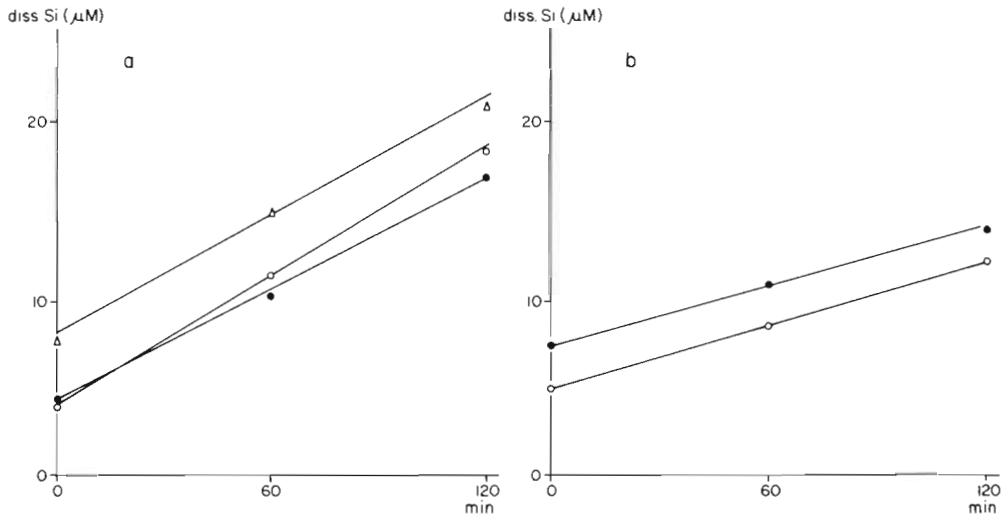

Fig. 1. Evolution of dissolved Si in overlying water of cores at Anthony Park, Bay of Fundy. (a) Without formalin; (b) after addition of formalin. 12 Aug 1982; 22 °C

RESULTS

Bay of Fundy

Fig. 1 shows the increase of diss. Si concentrations in overlying water during *in situ* incubation of cores at Anthony Park. The rate of increase of the diss. Si concentration is constant, but was significantly slower in the formalin-poisoned cores. The observed rates, the overlying water volume in the cores, and the sediment surface area (25.5 cm^2) were used to calculate diss. Si fluxes. We refer to these as experimental fluxes without (−formalin) or with formalin addition (+ formalin).

When molecular diffusion is assumed to be the only transport mechanism for sediment-water exchange, then the diss. Si fluxes can be calculated by application of Fick's first law:

$$J = -\phi D_s \left(\frac{dc}{dx}\right)_{x=o} \quad (1)$$

where ϕ = porosity; D_S = whole sediment molecular diffusion coefficient; and $(dc/dx)_{x=o}$ is the interfacial concentration gradient. Porosity (ϕ) was measured in our experiments and profiles of porosity are shown in Fig 2. Given the similarity of the porosity profiles from Anthony Park and Pecks Cove we applied for both sites the relation between formation factor (F) and porosity (ϕ) as given by Andrews & Bennett (1981) for Pecks Cove: $F^{-1} = 1.0240^{2.077}$ to modify the molecular diffusion coefficient of diss. Si in free solution (Wollast & Garrels 1971) to a whole sediment diffusion coefficient (Berner 1980).

The interfacial diss. Si gradients were calculated from diss. Si pore-water profiles of which examples are shown in Fig. 3. Fig. 3 indicates that at both sites diss. Si reaches distinct maxima of about 550 µm. Calculated Si fluxes are given in Table 1, in which they can be compared with the experimental fluxes obtained from core-incubation with and without formalin addition.

Texel

Cores from the 4 locations at Texel were, in contrast to the experiments carried out in the Bay of Fundy, incubated in the laboratory at *in situ* temperature and at constant stirring rate. After following the development of diss. Si concentrations in overlying water of the

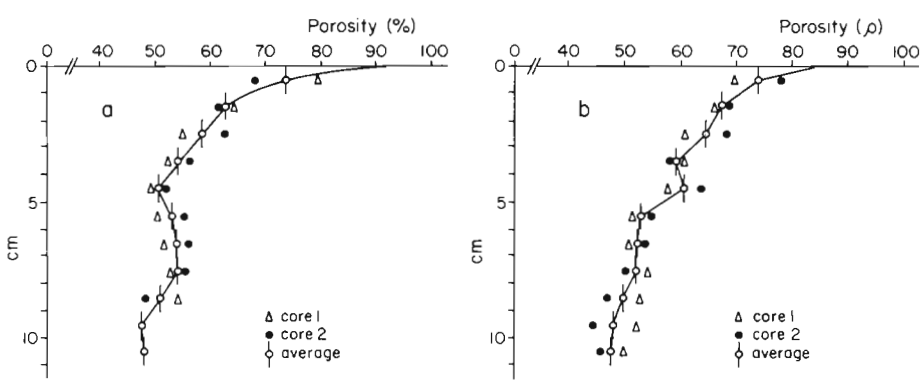

Fig 2. Porosity profiles in Bay of Fundy, August 1982. (a) Anthony Park; (b) Peck's Cove

incubated cores for a certain time, formalin was added and the development of diss. Si concentrations was followed again. Fig. 4a gives an example of such an experiment.

Results of the asphyxiation experiment are shown in Fig. 4b. When the oxygen concentration in the incubated cores decreased to about 20% saturation (50 μMO_2) a significant change in the diss. Si flux

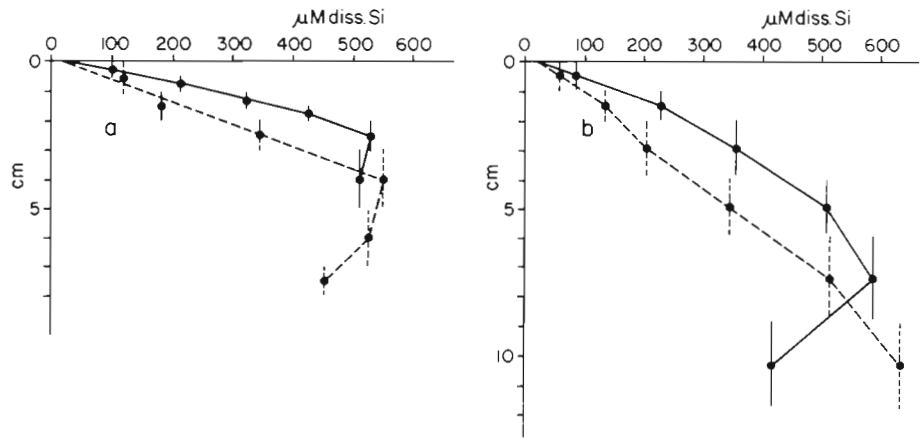

Fig. 3. Pore-water profiles of dissolved Si in 2 cores from (a) Anthony Park (12 Aug 1982; 22 °C); and (b) Peck's cove (13 Sep 1982; 21 °C)

Table 1. Comparison of observed and calculated dissolved Si fluxes (mmol m^{-2} d^{-1}) at Bay of Fundy locations. Numbers in parentheses indicate SD and number of measurements

Parameter	Anthony Park		Peck's Cove
Date	12 Aug 82	8 Sep 82	13 Sep 82
Temperature (°C)	22	18	21
Experimental flux			
− formalin	6.9 (1.2, n=3)	2.8 (1.3, n=5)	2.2 (0.8, n=5)
+ formalin	2.5 (0.2, n=2)	0.7 (0.1, n=2)	0.8 (n=1)
Calculated flux assuming molecular diffusion	1.0 (0.5, n=3)	0.4 (0.1, n=2)	0.5 (0.1, n=2)
Diss. Si concentration gradient at interface (µmol cm^{-4})	0.274 (0.127, n=3)	0.096 (0.02, n=2)	0.122 (0.03, n=2)
Ratio of − formalin/+ formalin flux	2.8	4.0	2.7

Table 2. Comparison of observed and calculated fluxes of dissolved Si (mmol m^{-2} d^{-1}) at Texel locations. Numbers in parentheses indicate SD and number of measurements

Parameter	Hoornderslag		Mokbaai 1	Mokbaai 2	Oost	
Date	25 Jun 85	26 Jun 85	2 Jul 85	19 Jul 85	29 Jul 85	5 Aug 85
Temperature (°C)	13.5	13.5	13.5	17.7	19.0	20.0
Experimental flux:						
− formalin	−1.2 (1.1; n=4)	−0.5 (0.1; n=4)	−4.3 (0.3; n=4)	21.2 (7.7; n=4)	21.3 (4.8; n=8)	18.8 (13.0; n=8)
+ formalin	−0.5 (0.6; n=4)	−0.02 (0.4; n=4)	−0.7 (0.4; n=2)	9.7 (2.1; n=4)	–	4.8 (1.0; n=8)
+ asphyxiation	–	–	–	–	4.0 (0.4; n=8)	–
Diss Si gradient at interface (µmol cm^{-4})	Not measurable		0.073 (n=2)	0.181 (n=2)	1.408 (n=2)	1.219 (n=4)
Calculated flux assuming molecular diffusion	~0	~0	−0.35	1.0	7.3	6.5
Ratio of − formalin/ + formalin flux	–	–	6.1	2.2	–	3.9
Ratio − formalin/+ asphyxiation flux	–	–	–	–	5.3	–

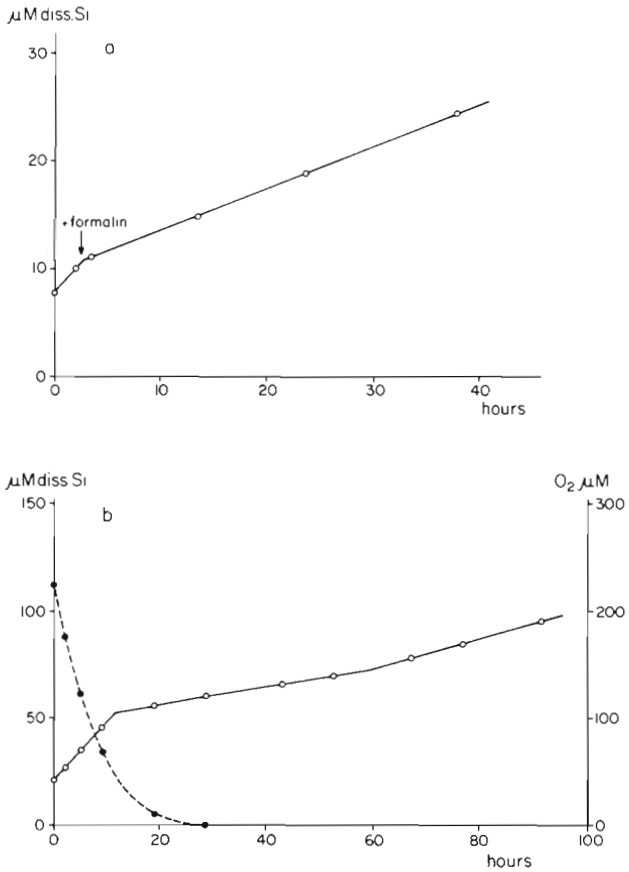

Fig. 4. Evolution of dissolved Si in overlying water of representative cores (n=8) from 'Oost' location at Texel. (a) Formalin poisoning: time of formalin addition indicated by arrow. (b) Asphyxiation experiment. (---) Oxygen concentration in overlying water during the experiment

occurred. Dead macrofaunal species were sometimes observed at the sediment surface after asphyxiation as well as after formalin addition. After about 50 to 60 h of incubation a small increase in the diss. Si flux occurred in all asphyxiated cores (e.g. Fig. 4b).

Experimental fluxes, obtained from the diss. Si versus time plots, are given in Table 2, where they can be compared with the fluxes calculated from application of Fick's first law assuming molecular diffusion.

Profiles of diss. Si in pore water are shown in Fig. 5. Whole sediment diffusion coefficients (D_s) were calculated from $D_s = D/\phi F$ and the formation factor (F) was calculated from profiles of electrical resistivity in sediment (R) and overlying water (R_o), with $F = R/R_o$ (Berner 1980), according to the procedure of Andrews & Bennett (1981). All results from the experiments at Texel locations are summarized in Table 2.

DISCUSSION

Experimentally determined diss. Si fluxes in the Bay of Fundy and around Texel ranged from 2 to 7 mmol m^{-2} d^{-1} and 0 to 21 mmol m^{-2} d^{-1}, respectively. The experiments were carried out in Summer when highest temperatures occur in both areas. The observed flux rates must be considered as maxima, bearing in mind the considerable temperature dependence of Si dissolution rates (Hurd 1973) as well as of macrofauna-enhanced diffusion coefficients (Aller & Benninger 1981, Rutgers van der Loeff at al. 1984). The maximum diss. Si flux rates at our Texel locations are comparable to maximum rates reported for other coastal areas, e.g. 19 mmol m^{-2} d^{-1} in Potomac River estuary by Callender & Hammond (1982), 20 mmol m^{-2} d^{-1} in Long Island Sound by Aller & Benninger (1981) and 13.2 mmol m^{-2} d^{-1} in the mouth of the Changjiang estuary (China) by Aller et al. (1985). The negative diss. Si fluxes that we found at the Texel sites Hoornderslag and Mokbaai 1 are in agreement with the direction of the interfacial diss. Si gradient at these locations.

The addition of formalin resulted at all locations in a decrease of diss. Si flux rates. At the Bay of Fundy locations fluxes decreased by a factor of 2.8 to 4, and at the Texel locations by a factor of 2 to 6 (Tables 1 & 2), compared to experimentally determined flux rates in the absence of formalin. 'Asphyxiation' of sediment macrofauna also resulted in a decrease of the diss. Si flux rates (Fig. 4b; Table 2) and the effects of formalin poisoning and asphyxiation agree quantitatively. The small increase of diss. Si concentration in overlying water of asphyxiated cores after 50 to 60 h of incubation is possibly associated with increased dissolution rates of siliceous material in the sediment at lowered redox potential. Also the presence of distinct maxima in diss. Si pore water profiles (Fig. 3 & 5) suggests that a source for enhanced diss. Si production is present at a particular depth horizon

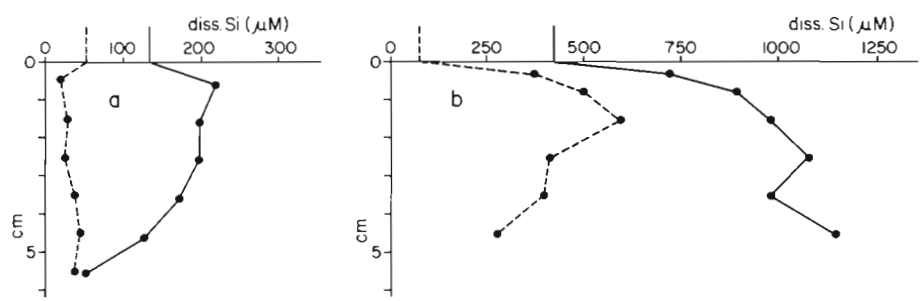

Fig. 5. Dissolved Si pore-water profiles at Texel. (a) Mokbaai 1 location (---) and Mokbaai 2 location (—). (b) 'Oost' location before (---) and after incubation of cores (—). Data points represent averaged values of 2 to 4 pooled cores

in the sediment. A mechanism that can qualitatively explain both the increase of diss. Si evolution in overlying water of asphyxiated cores and the presence of maxima of diss. Si in pore water profiles has been given by Aller & Yingst (1985). These authors proposed that reduction of Fe^{3+} to Fe^{2+} in poorly crystallized Fe silicates or removal of Fe oxide coatings from siliceous surfaces during Fe reduction are responsible for enhanced dissolution of Fe containing siliceous material at low redox potential. The small increase in diss. Si evolution after prolonged incubation (~60 h), which we found in asphyxiated cores, cannot be detected in the formalin-poisoned cores (Fig. 4a, b). This might indicate the effectiveness of formalin as an inhibitor of microbial processes that tend to lower the redox potential in the sediment.

Flux-rates after formalin addition and after asphyxiation are within a factor of 2 of calculated fluxes, based on interfacial diss. Si gradients assuming molecular diffusion as the only transport mechanism.

We conclude from these observations that formalin poisoning and asphyxiation of sediment fauna are both effective in reducing dissolved Si transport across the sediment-water interface by decrease of an effective diffusion coefficient (D_e) in the presence of bio-irrigating macrofauna to a molecular whole sediment diffusion coefficient (D_s) in the absence of active macrofauna. The ratios of D_e/D_s that we found in the Bay of Fundy area (2.7 to 4.0) and at the Texel locations (2.2 to 6.1) compare well with earlier reports: Callender & Hammond 1982 (2.6 to 6.1); Lyons et al. 1982 (3 to 8); Rutgers van der Loeff at al. 1984 (2 to 10); Aller & Yingst 1985 (2 to 5). Estimation of the effect of macrofauna on flux rates is carried out in many cases by comparing experimentally determined fluxes with those calculated from pore-water gradients assuming molecular diffusion. Our method of formalin addition as applied during 1985 at Texel has, like the asphyxiation technique described by Rutgers van der Loeff at al. (1984), the advantage that both the bioturbation-enhanced flux and the diffusion flux after inactivation of macrofauna are determined in one and the same core and that the diffusion flux is measured instead of calculated. Thus we obtain a ratio of D_e to D_s that does not depend on spatial homogenity of diss. Si pore-water profiles, or on accurate determination of diss. Si interfacial gradients which is not achieved when diss. S concentrations are determined in 0.5 or 1.0 cm sediment segments.

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