



CONTAMINANT FLUXES FROM SEDIMENT DUE TO TUBIFICID OLIGOCHAETE BIOTURBATION

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Abstract—The release of the hydrophobic organic compounds pyrene, dibenzofuran and phenanthrene from bioturbated freshwater sediments was studied in laboratory microcosms. Initial Tubificid oligochaete densities of 0, 6700 and 2.67×10^4 individuals $\cdot m^{-2}$ were employed. Under oxygen saturated conditions, the difference between the contaminant fluxes from the high-density bioturbated microcosms and controls remained essentially constant at 37 and 70 $ng \cdot cm^{-2} \cdot d^{-1}$ for pyrene and phenanthrene, respectively, corresponding to effective mass transfer coefficients of 0.16 and 0.37 cm/y . Under hypoxic conditions, worm defecation on the sediment surface increased and led to significantly increased fluxes to a maximum of 380, 490 and 940 $ng \cdot cm^{-2} \cdot d^{-1}$, for pyrene, phenanthrene and dibenzofuran, respectively. Average bioturbation fluxes in the high-density microcosms of 246, 258 and 310 $ng \cdot cm^{-2} \cdot d^{-1}$ for the respective compounds corresponded to effective mass transfer coefficients of 1.7, 3.2, and 7.5 cm/yr . Initial release rates from medium-density microcosms (25% of high density) were typically half the release rate of the high-density microcosms, indicating greater organism activity per individual at the lower density. The increased flux with the more soluble compounds likely reflects more rapid release at the sediment surface and the increased importance of porewater pumping over sediment particle reworking for migration of these compounds.

Key words—sediment, bioturbation, tubificid oligochaetes, PAH, contaminant flux

INTRODUCTION

Hydrophobic organic and metal contaminants are strongly associated with the particulate fraction of the sediment and partition only weakly into the porewaters and overlying waters. This often leads to the retention of hydrophobic organic contaminants in sediments long after the original introduction of the contaminant to the environment. Bed sediments are often assumed to act as a sink for such contaminants although several mechanisms may disperse contaminants.

Singh *et al.* (1988) identified the possible transport pathways for hydrophobic contaminants from bed sediments to overlying water, including particle resuspension processes, diffusion, advection and processes mediated by organic colloidal material and benthic organisms. Medine and McCutcheon (1989) and Reible *et al.* (1991) recently reviewed the mathematical modeling of these processes, and suggest that activities that result in particle and sorbed contaminant transport, are generally responsible for the largest contaminant fluxes to the overlying water column. In low energy environments in which sediment resuspension by erosive forces is

negligible, however, contaminant migration is likely to be dominated by the indirect action of the normal life cycle activities of benthic organisms, or bioturbation (Reible *et al.*, 1991).

Bioturbation is defined as the sediment processing by animals during burrowing, sediment ingestion/defecation, tube building and biodeposition. The net result of bioturbation is the vertical and horizontal movement of sediment particles and pore water, mixing the upper sediment surface (Robbins, 1982; Robbins *et al.*, 1979). Some larger benthos are particularly effective bioturbators and even surface dwellers that do not directly process sediment can still cause significant particle and contaminant movement by disturbing the surface (McCall and Fisher, 1980). Those organisms that process sediment with their head down into the sediment and defecate on the sediment surface (conveyor-belt species) have perhaps the greatest impact on sediment conditions (Cochran and Aller, 1979; Robbins, 1982; Robbins *et al.*, 1979).

Several recent reviews describe the importance of bioturbation as a transport mechanism for inorganic and organic contaminants from sediment (Lee and Schwartz, 1980; Aller, 1982; Gschwend *et al.*, 1987). Although bioturbation is generally recognized as an effective means of transport of hydrophobic organic

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compounds and elemental species, it is difficult to quantify. Typically effective diffusion models, are in a crude state of development. Matisoff (1982), for example, presents more than 80 different measurements of effective bioturbation diffusion coefficients that vary over 4 orders of magnitude. In addition, the use of an effective diffusion coefficient is likely to be a poor representation of the process of bioturbation by conveyor-belt feeders. Insufficient data exist, however, to differentiate between the mechanisms and rates of bioturbation-related contaminant transport by various organisms.

The current work is directed toward quantifying the increased flux of hydrophobic organics from contaminated sediment due to bioturbation by freshwater oligochaetes belonging to the family Tubificidae. Members of this family feed using the conveyor-belt method, are small (2–5 cm in length) but are abundant infauna in many lakes, slow-moving streams and brackish-water estuaries (Brinkhurst, 1975; Brinkhurst and Gelder, 1991). They live with a head-down posture within the sediment, bringing sediments from depth to the surface in the form of long, cylindrical fecal pellets. Tubificid oligochaetes exhibit a high tolerance for hydrophobic organic and other contaminants (Chapman and Brinkhurst, 1993) and are able to adapt to contamination (Klerks and Levinton, 1989). Tubificids often dominate benthic macrofaunal populations in contaminated sediments (Pennak, 1989). Karickhoff and Morris (1985) demonstrated that *Tubifex tubifex* can be an effective bioturbator and measured contaminant fluxes from the sediment as a result of their activities in laboratory microcosms.

The purpose of this work was to measure the magnitude of bioturbation by tubificid worms in the laboratory to further modelling of contaminant flux and, ultimately, understand its ecological implications. Our experimental system was composed of small sediment microcosms, with or without bioturbation, contaminated with hydrophobic compounds (polynuclear aromatic hydrocarbons) that served as tracers of flux from sediments. This system allowed us the opportunity to examine the influence of various factors, including oxygen tension, worm density and contaminant loading on bioturbation effects. In addition, the tracers varied in their solubility allowing us to examine the influence of chemical properties on flux from sediments.

EXPERIMENTAL PROCEDURE

Apparatus

The laboratory experiments were conducted in small chambers 15 cm long, 5 cm wide and 4.5 cm deep and constructed from 0.64 cm thick plexiglas. The depth of the sediment layer was 3.5 cm. A total of 12 microcosms were employed in each of the experiments, with four replicates at the 0, 2700 and 26,700 individuals $\cdot m^{-2}$ animal densities.

All experiments were conducted at room temperature. Two small reservoirs were located at each end of the rectangular test section. All microcosms had a removable lid. During an experiment, the bottom of the chamber was filled with the model sediment. Water was introduced to the entrance reservoir, where it passed over the sediment-filled region and exited the chamber at the other end. Continuous water flow across the sediment bed was provided by a peristaltic pump with flow rate of about 500 ml $\cdot h^{-1}$. The flowrate was selected to reduce the influence of water-side mass transfer resistances without further complicating the chemical analysis by dilution of the collected water. This flowrate corresponds to a linear velocity of about 3 cm/min in the cells. The chamber shape and dimensions are shown in Fig. 1.

Artificial pond water (0.5 mM NaCl, 0.2 mM NaHCO₃, 0.05 mM KCl, 0.4 mM CaCl₂) was formulated and kept in a reservoir tank to supply incurrent water. The antibiotic Manacyn® at 30 mg/l was added to the tank and, in the first experiment, the tank was initially aerated to remove chlorine. This resulted in saturation with respect to dissolved oxygen at levels of 9 mg/l. At the conclusion of the first experiment aeration was stopped, reducing the dissolved oxygen level of the incurrent water supply to 2 mg/l. The second experiment was conducted without aeration. Oxygen levels in the microcosms was not measured, but would be expected to be lower than the incurrent supply. Tubificid worms are very tolerant of low dissolved oxygen, typically living in waters with high oxygen demand under hypoxic conditions (Brinkhurst, 1975; Bonacina *et al.*, 1987).

Sediment

Sediment was obtained from Bayou Manchac, Baton Rouge, La. In order to achieve measurable contaminant fluxes, the sediment was inoculated with pyrene, phenanthrene and dibenzofuran by tumbling a sediment-filled vessel on which the compounds had previously been plated (Means *et al.*, 1980). These compounds were selected for two important reasons: (i) they are significant pollutants in contaminated sediments in several regions of the United States, and (ii) they have widely varying aqueous solubilities; pyrene the least soluble exists mostly in association with the sediment, whereas, the most soluble, dibenzofuran exists in porewaters. Soxhlet extraction (EPA Method 3540) was employed to determine the initial sediment loading. The physical properties of the compounds and the sediment characteristics are summarized in Table 1. Analytical difficulties precluded monitoring of dibenzofuran during the first experiment but all compounds were monitored during the second experiment.

During both experiments, the effluent water was analyzed for tracer concentration to infer contaminant fluxes as a function of time from the sediment bed. During the first experiment described herein, 0.8 l water samples were collected weekly from the effluent of the experimental cells. The sample was added to 0.5 l of hexane and extracted over 8 h in a continuous extractor. The hexane was then concentrated to 2 ml, and then exchanged with acetonitrile during evaporation with nitrogen. Analysis was then conducted via u.v. detection high performance liquid chromatography (Hewlett-Packard Model 1090 HPLC) with a 70/30 acetonitrile/water carrier and 150 \times 4.6 mm column packed with Envirosep-PP (a reversed phase C-18 column). Average recovery of pyrene and phenanthrene by this process was 93.3 and 92.7%, respectively. Dibenzofuran results during the first experiment were unreliable.

During the second experiment, the continuous liquid-liquid extraction was replaced with a three stage batch extraction procedure so that samples could be conveniently taken and analyzed twice per week. Eighty ml of hexane was

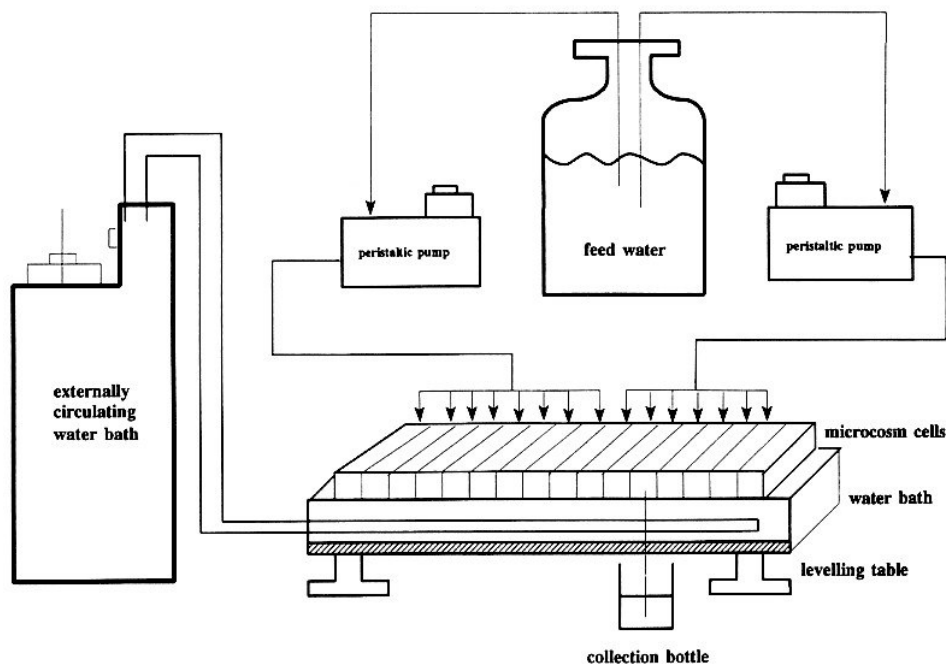


Fig. 1. Depiction of experimental system.

added to 700 ml of sample and shaken for 45 min in the first stage. 60 ml of hexane was then added twice with 15 min of shaking each time. Hot water immersion of the sealed sample was used to break any emulsions formed during shaking. The combined hexane extract was concentrated by evaporation to 2 ml. The solvent was then exchanged for acetonitrile and analyzed as in the previous experiment. Recovery from matrix samples averaged $77 \pm 4.3\%$ for pyrene, $93.70 \pm 1.1\%$ for phenanthrene and $83.9 \pm 6.5\%$ for dibenzofuran. Although average recoveries were less by this method, it was felt that increased sample frequency was desirable and the method was employed throughout the experiment.

Bioturbators

Tubificid worms are widespread ecologically and are easy to culture and grow to about 2–5 cm in length. The first experiment utilized *Tubifex tubifex* obtained from a commercial supply house. This supply was interrupted just

before the start of the second experiment due to late-season cold front that depressed culture densities available to suppliers throughout North America. We therefore collected and cultured a local species (*Limnodrilus hoffmeisteri*). *L. hoffmeisteri* and *T. tubifex* are closely related taxonomically and are similar in size and in several important ecological characteristics (Chapman and Brinkhurst, 1993). Individuals of both species selected for use in these experiments were of very similar body mass, averaging 0.57 mg dry weight, and ingestion/defecation rates were very similar (see below). In both experiments, initial densities of 0 (control), 6700 and 2.67×10^4 organisms $\cdot m^{-2}$, were obtained by adding the appropriate number of tubificids to the sediment microcosms, with four replicates at each density. Tubificids adapt quickly to the new surroundings and burrowed into sediments within a few hours and began to feed almost immediately.

Figure 2 shows the observed tube building behavior of the worms in the experimental cells. Large volumes of processed sediment is also brought to the surface as a result of deposit

Table 1. Sediment and chemical characteristics

	Pyrene	Phenanthrene	Dibenzofuran
Water solubility, S (mg/l)	0.13	1	10
Organic-carbon partition coefficient $\log(K_{oc})^a$	4.8	4.2	3.7
Henry's law constant, H (atm \cdot m ³ /mol)	1×10^{-5}	6×10^{-5}	7.9×10^{-5}
Water diffusivity, D_w , (cm ² /s)	5.5×10^{-6}	5.8×10^{-6}	6.0×10^{-6}
Retardation factor, R_f	1325	333	105
Loading, Experiment 1 (W_λ , mg/kg)	90	60	—
Loading, Experiment 2 (W_λ , mg/kg)	52 ± 4	30 ± 3	15 ± 2

Bayou Manchac sediment—physical properties:

organic carbon content, $f_{oc} = 0.021$;
 bulk density, ρ_b (g/cm³ = 1.0);
 porosity, $\epsilon = 0.59$;
 sand, % = 29;
 silt, % = 37;
 clay, % = 34.

^aLog (K_{oc}) estimated from water solubility. All other quantities are measured in our laboratory.

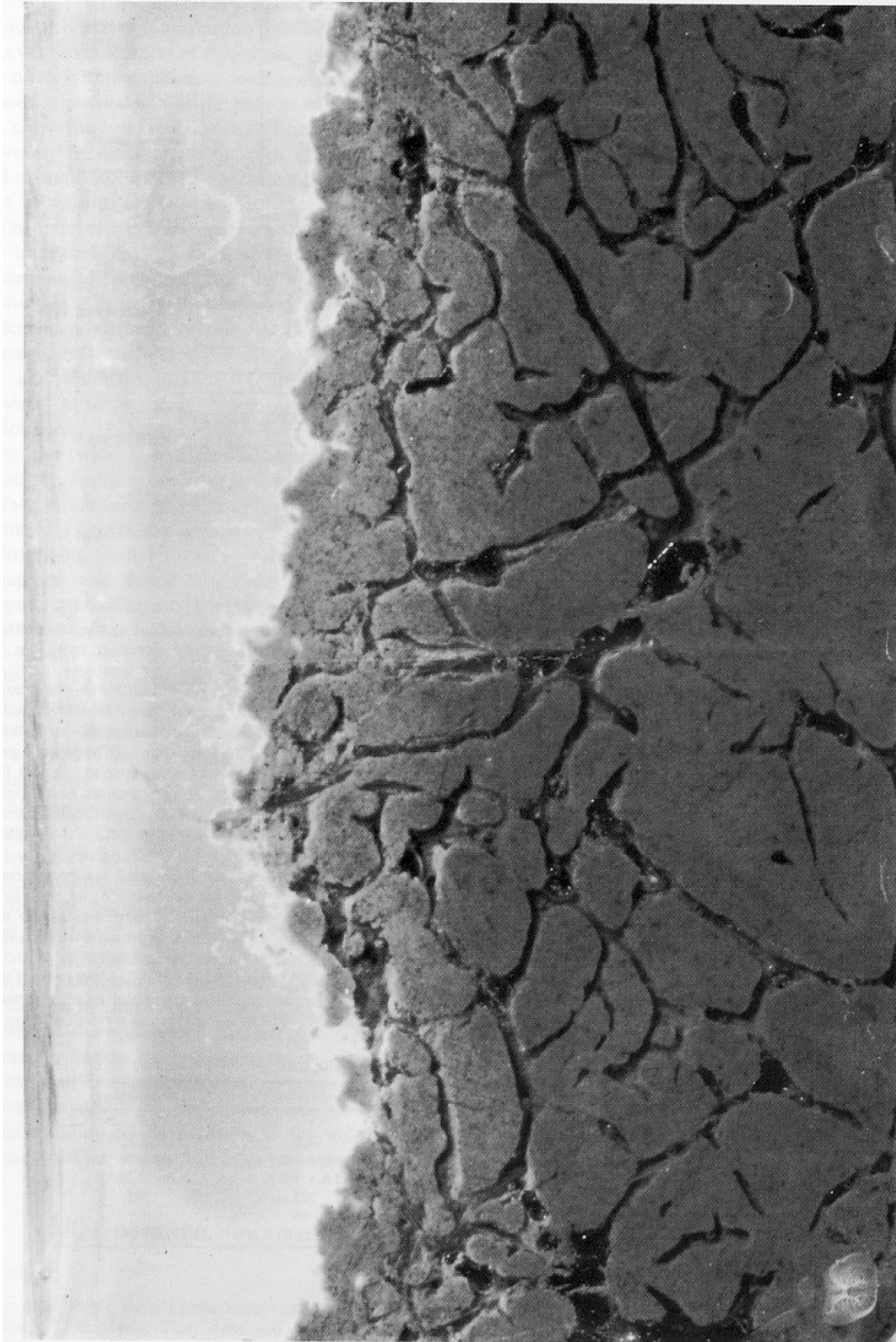


Fig. 2. Photograph of the side of an experimental microcosm showing *Tubificex* tube-building activities.

feeding. *Tubifex tubifex* has been shown to ingest and defecate several times its body weight daily (Kaster *et al.*, 1984). As shown in the photograph, a worm is partially exposed to the overlying water. Defecation while residing in this posture can release any contaminants in the feces directly to the water column. Significant secondary porosity is also developed within the sediment as a result of these activities.

Ingestion/defecation rate is sometimes used as a measure of animal activity because tubificids are bulk feeders, obtaining energy from the organic matter of sediments. Defecation rates were estimated coincident with each flux experiment, following the procedures of Kaster *et al.* (1984). Two sets of four 9.3×2.7 cm vials were filled three quarters full with Bayou Manchac sediment and 15 tubificid worms were introduced into each vial. One set of vials contained contaminated sediment and the other contained control sediment. Tubificid density was essentially identical to that in the high-density experimental cells. The surface of the sediment was covered with a layer of polyester aquarium floss and a disc of cheese cloth was placed on top. Both floss and cheese cloth were held in place by a teflon split-ring compressed inside the vial. Then the vials were filled with water and each set was introduced into separate aquaria. Most worms protruded their posterior ends through the floss and the cheese cloth into the water and began to defecate within 2 h. Feces were deposited on the top of the cheese cloth and could be easily removed by carefully inverting the vial and rinsing the fecal matter using a squeeze bottle. For a period of twenty d (after the first 48 h), samples of fecal material were collected at 24 h intervals, filtered through a $8.0 \mu\text{m}$ filter, dried overnight at 60°C and weighed. After each collection, the teflon ring was pushed gently downward to eliminate any water space between the floss and the sediment caused by worm activity.

The *L. Hoffmeisteri* body burden of pyrene was determined at the conclusion of the second experiment by Soxhlet extraction and HPLC. Worms were placed in clean water for 24 h after collection to allow purging of sediment from their bodies. The worms were then sacrificed and the contaminants extracted in a manner identical to that used for the analysis of sediment samples.

RESULTS

Experiment 1

The measured fluxes from both the control and high organism density cells during the first experiment are shown in Figs 3 and 4 for pyrene and phenanthrene respectively. The solid symbols represent the chemical flux in the bioturbated cells while the open symbols represent the flux from the control cells. The solid lines represent the difference between the two fluxes and thus represent the estimated flux attributable to bioturbation of the sediment. The error bars indicate the standard deviation in the measured fluxes over the four cells at each organism density ($26,700 \text{ m}^{-2}$ and 0).

The fluxes from the control channels decreased rapidly during the course of the experiment. Separate experiments to monitor the dynamics of colloidal material in the laboratory cells suggest that the colloidal flux, as measured by dissolved organic carbon (DOC), also decreases rapidly (Valsaraj *et al.*, 1993). Since pyrene would be expected to sorb strongly to the dissolved organic carbon, this could account for the rapid decrease in measured pyrene

flux. Due to the high water flow rate in the experiments and consequent dilution of the dissolved organic carbon concentration, it was not possible to verify the DOC concentrations or fluxes.

Although the flux in the control cells decreased rapidly, the difference in flux between the populated cells and the control cells, i.e., the flux due solely to bioturbation, remained relatively constant under oxygen-saturated conditions for both compounds. The measured pyrene bioturbation-related flux of about $37 \text{ ng} \cdot \text{cm}^{-2} \cdot \text{d}^{-1}$ corresponds to about 5–15% of the pyrene in the $2 \text{ mg} \cdot \text{d}^{-1}$ of sediment (90 mg/kg pyrene) processed by each organism in the contaminated sediment. In the case of the more soluble phenanthrene 14 to 44% of that was processed was released to the water column with an average flux of $70 \text{ ng} \cdot \text{cm}^{-2} \cdot \text{d}^{-1}$.

During most of the period included in Figs 3 and 4, the oxygen level in the incurrent water was essentially saturated at about $9 \text{ mg} \cdot \text{l}^{-1}$. At these high dissolved oxygen levels, *T. tubifex* tended to remain completely within the sediment and defecated below the sediment–water interface, without noticeable mounding at the surface. Viewing from the side of the transparent cells suggested that worm passages penetrated to near the bottom of the cell, 3–3.5 cm below the sediment surface, in a manner similar to that depicted in Fig. 2. This also likely led to a reduction in the influence of bioturbation on the flux. After 25 d of experimentation, aeration of the feed water was stopped resulting in movement of the organisms to the surface to improve oxygen uptake. This led to an increase in the contaminant flux from the bioturbated channels by a factor of 4–6 to an average of about $157 \text{ ng} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$ for pyrene and about $480 \text{ ng} \cdot \text{cm}^{-2} \cdot \text{d}^{-1}$ for phenanthrene. Assuming that the organisms were still processing of the order of 2 mg dry sediment $\cdot \text{worm}^{-1} \cdot \text{d}^{-1}$ under hypoxic conditions, this implies that the greater surface interaction of the organisms increased the pyrene released to the overlying water to about 50% of that processed. The increase in pyrene and phenanthrene fluxes is also shown in Figs 3 and 4 in which the portion of the flux attributed to bioturbation, that is, the difference between the fluxes in the control and the high organism density microcosms, is plotted versus time.

An effective bioturbation mass transfer coefficient can be defined as the ratio of the flux due to bioturbation and the contaminant concentration in the sediment.

$$k_{\text{bio}} = \frac{\text{Observed Flux} - \text{Control Flux}}{W_A \rho_b}$$

Here the bioturbation flux is estimated by subtracting the flux measured in the control chambers from that measured in the bioturbated chambers. Because of the high fluxes from the control chambers during much of the experiment, the bioturbation flux

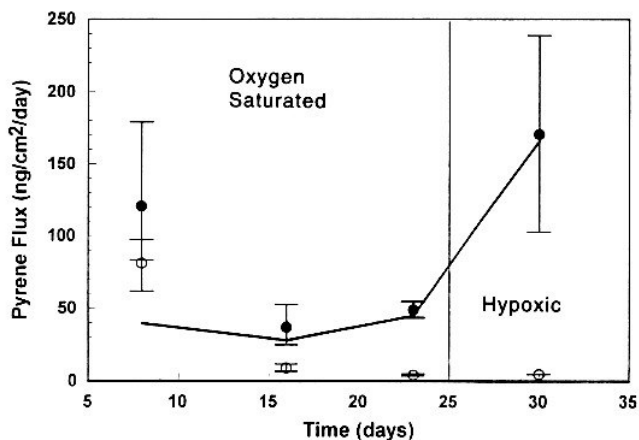


Fig. 3. Pyrene flux from the experimental cells during the first experiment. (●) Flux from high density bioturbated cells. (○) Flux from control cells. (—) Estimated flux due to bioturbation (by difference). Ranges are standard deviations.

from sediments with a low tubificid density was not significantly different from controls. The bioturbation flux from the high-density microcosms, however, was more reliably estimated and these fluxes and the estimated mass transfer coefficient are included in Table 2. After the change to hypoxic conditions, the bioturbation flux increased by a factor of 4–6.

Experiment 2

In order to more fully explore the change in tubificid behavior and the increase in contaminant fluxes under hypoxic conditions, a second experiment was initiated to focus on these conditions. Hypoxic conditions are quite common in the contaminated sediments and waterway conditions of interest. In addition, it was decided to initiate water flow in the microcosms two weeks before the addition of animals to allow the bulk of the colloidal material responsible for the initial transient to leave the system.

The average and standard deviation in the flux of pyrene, phenanthrene and dibenzofuran for all three tubificid densities are shown versus time in Figs 5, 6 and 7. Bioturbation resulted in significantly increased contaminant fluxes over controls and over the fluxes estimated under oxygen-saturated conditions. The fluxes from the sediments in the controls without bioturbation were essentially constant throughout the experiment. A rapid decrease in flux with time would normally be expected as a result of depletion of the near surface sediment by retarded diffusion into the overlying water. It is postulated that the low, nearly constant flux observed during this experiment was the result of diagenetic processes leading to the production of colloidal organic carbon to which the contaminant could sorb and whose subsequent release served to sustain the observed fluxes (Valsavaj *et al.*, 1993). The effect of microbial activity may not have been observable during the first

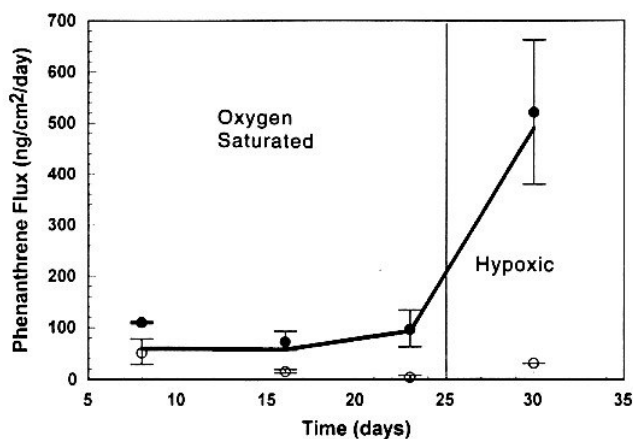


Fig. 4. Phenanthrene flux from the experimental cells during the first experiment. (●) Flux from high density bioturbated cells. (○) Flux from control cells. (—) Estimated flux due to bioturbation (by difference). Ranges are standard deviations.

Table 2. Bioturbation release rates high organism density cells—oxic conditions (fluxes in $\text{ng} \cdot \text{cm}^{-2} \cdot \text{d}^{-1}$)

Cell	Phenanthrene	Pyrene
Sediment loading $\pm \sigma$ (mg/kg)	60	90
Net bioturbation flux (observed-control) $\pm \sigma$	70 ± 16	37 ± 7
Effective bioturbation mass transfer coefficient (cm/yr)	0.42	0.15
Contaminant release fraction (%) ^a	22% (14–44%)	8% (5–15%)

^aBioturbation flux + maximum flux with average defecation rate of $2.1 \text{ mg} \cdot \text{d}^{-1} \cdot \text{worm}^{-1}$. Range given in parentheses assumes standard deviation of defecation rate is $1.1 \text{ mg} \cdot \text{d}^{-1} \cdot \text{worm}^{-1}$.

experiment as a result of the steep transient associated with the release of colloidal organic carbon and contaminants during the experiment. The relatively short duration of the first test may also have led to minimal diagenetic activity.

The release rates during the hypoxic condition experiments are summarized in Table 3 in which the difference between the observed flux and the control flux is used to estimate an average bioturbation flux and effective mass transfer coefficient as defined previously. The effective mass transfer coefficients are 4–6 times those observed under oxygen saturated conditions, apparently as a result of the increased surface defecation. It is important to note that the magnitude of fluxes with worms during experiments 1 and 2 were similar indicating that although two different types of tubified species were used their behavior with respect to bioturbation are identical.

Relationships of contaminant release rates to defecation rates

The defecation rates measured in separate experiments varied from 2 to $4 \text{ mg} \cdot \text{worm}^{-1} \cdot \text{d}^{-1}$ for both species. The measured defecation rates indicated that the presence of the relatively high level of contaminants in the first flux experiment reduced the organism activity by a factor of 2–3. Upon introduction to uncontaminated sediment, a defecation rate of about $3 \text{ mg} \cdot \text{worm}^{-1} \cdot \text{d}^{-1}$ was

measured, while by 17–22 d after introduction, the rate had increased to $9\text{--}11 \text{ mg} \cdot \text{worm}^{-1} \cdot \text{d}^{-1}$ due to growth and acclimation of the animals. The defecation rate in contaminated sediment remained about 2 mg/worm/d throughout the defecation rate experiment, averaging $2.1 \pm 1.1 \cdot \text{worm}^{-1} \cdot \text{d}^{-1}$ or about $3.7 \pm 1.9 \text{ mg dry sediment per d per mg of worm dry weight}$.

During the second experiment, a lower chemical loading resulted in minimal difference between the defecation rates in separate experiments in contaminated and uncontaminated soil. In both cases the defecation rate was initially about $2 \text{ mg} \cdot \text{worm}^{-1} \cdot \text{d}^{-1}$ and increased over 20 d to $3\text{--}5 \text{ mg} \cdot \text{worm}^{-1} \cdot \text{d}^{-1}$. The average defecation rate in the contaminated sediments, however, changed little from the first experiment at $2.3 \pm 1.2 \text{ mg per worm per d}$. Higher defecation rates were measured near the end of these experiments as a result of the acclimation of the animals to the sediment and apparent population growth. In addition, the animal density at the end of the experiment in randomly selected cells was about 50% higher than at the outset of the experiment. At least part of this is expected to have resulted from reproduction and animal growth. The effect of animal reproduction, growth and acclimation is reflected in the increasing flux of pyrene over the course of the experiment as shown in Fig. 5. Assuming an average sediment processing rate

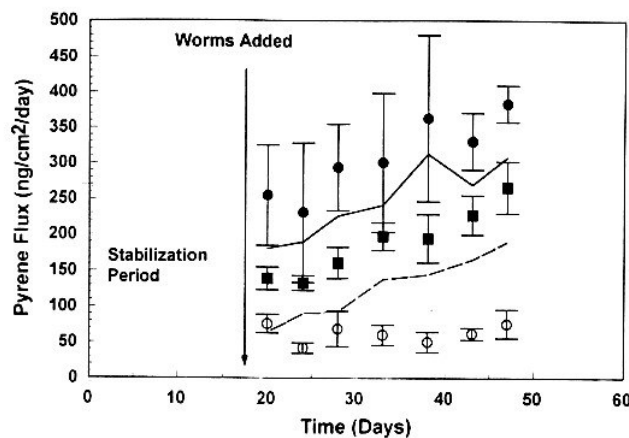


Fig. 5. Pyrene flux from the experimental cells under hypoxic conditions during the second experiment. (●) Flux from high density bioturbated cells. (■) Flux from the medium density bioturbated cells. (○) Flux from control cells. (—) Estimated flux in high density cells due to bioturbation (by difference). (---) Estimated flux from medium density cells due to bioturbation (by difference). Ranges are standard deviations.

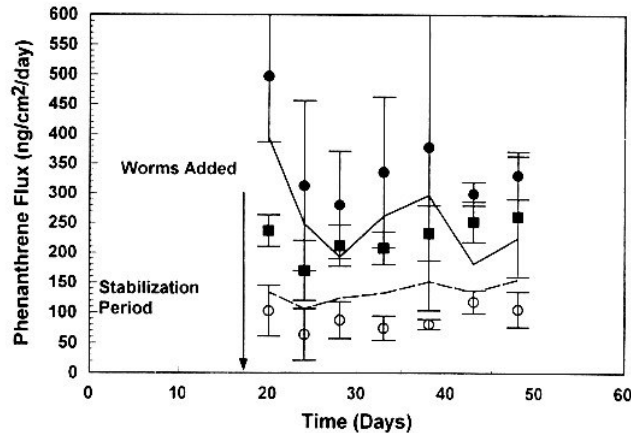


Fig. 6. Phenanthrene flux from the experimental cells under hypoxic conditions during the second experiment. (●) Flux from high density bioturbated cells. (■) Flux from the medium density bioturbated cells. (○) Flux from control cells. (—) Estimated flux in high density cells due to bioturbation (by difference). (---) Estimated flux from medium density cells due to bioturbation (by difference). Ranges are standard deviations.

of $2.3 \text{ mg} \cdot \text{worm}^{-1} \cdot \text{d}^{-1}$, the average observed bioturbation flux for pyrene of $246 \text{ ng} \cdot \text{cm}^{-2} \cdot \text{d}^{-1}$ is consistent with 77% of the pyrene in the processed sediment being released at the sediment-water interface. Because of the variability in the defecation rates, this release fraction is not significantly different from 100% of the pyrene released by the sediment processing component of bioturbation.

A different behavior was observed with phenanthrene and dibenzofuran. The observed average phenanthrene bioturbation flux was 50% larger than that expected from the sediment processing rate of the organisms and for dibenzofuran, more than 3 times larger. Tube dwellers such as tubificids process water in addition to sediment and their movement and burrowing activities can also lead to the exchange of pore water with the overlying water, especially in

the more permeable bed resulting from the burrows (see Fig. 2). Thus transport in the dissolved state via pore water is likely to be much more important for the more water soluble compounds dibenzofuran and phenanthrene (see Table 1) and thus the contaminant migration rate can be greater than that which would be expected on the basis of the particle reworking alone.

DISCUSSION

The impact on flux of contaminants by bioturbation was great, especially under hypoxic conditions. Bioturbation rates (as measured by ingestion/defecation) for *Tubifex tubifex* and *Limnodrilus hoffmeisteri* were very similar, enhancing the flux by a factor of 4 to 6 under high-density conditions over non-

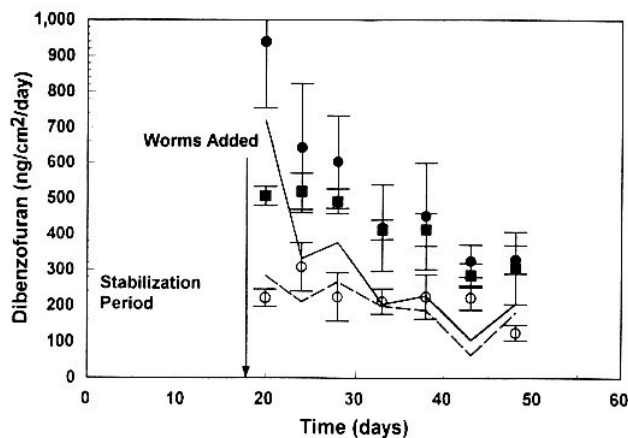


Fig. 7. Dibenzofuran flux from the experimental cells under hypoxic conditions during the second experiment. (●) Flux from high density bioturbated cells. (■) Flux from the medium density bioturbated cells. (○) Flux from control cells. (—) Estimated flux in high density cells due to bioturbation (by difference). (---) Estimated flux from medium density cells due to bioturbation (by difference).

Table 3. Bioturbation release rates—hypoxic conditions (fluxes in $\text{ng} \cdot \text{cm}^{-2} \cdot \text{d}^{-1}$)

Cell	Dibenzofuran	Phenanthrene	Pyrene
Flux $\pm \sigma$ (control)	220 \pm 54	90 \pm 19	62 \pm 13
Sediment load $\pm \sigma$ ($\mu\text{g}/\text{cm}^3$)	15.1 \pm 2.1	29.7 \pm 2.7	52.1 \pm 4.4
Medium density cells—6700 individuals/ m^2			
Observed flux $\pm \sigma$	419 \pm 95	225 \pm 31	188 \pm 49
Net flux bioturbation	199	135	126
Effective bioturbation mass transfer coefficient (cm/yr)	4.8	1.7	0.9
High density cells— 2.67×10^4 individuals/ m^2			
Observed flux $\pm \sigma$	530 \pm 219	348 \pm 73	308 \pm 56
Net flux bioturbation	310	258	246
Effective bioturbation mass transfer coefficient (cm/yr)	7.5	3.2	1.7
Contaminant release fraction, (%) ^a	340% (220–700%)	140% (92–290%)	77% (50–160%)

^aNet bioturbation flux + maximum flux based on a defecation rate of $2.3 \text{ mg sediment} \cdot \text{day}^{-1} \cdot \text{worm}^{-1}$. Numbers greater than 100% likely indicate uncertainty in determination of actual defecation rates in the experiments and the increased importance of pore water pumping processes for more soluble compounds. Ranges based on standard deviation in measured defecation rate given in parentheses.

bioturbated sediments. Tubificids not only directly bring contaminated sediments to the sediment–water interface in large quantities (upto several times their body weight per d of sediment is defecated at the surface, Kaster *et al.*, 1984), but their actions alter sediment porosity as well (McCall and Fisher, 1980). Advective processes driven from below or by currents in the overlying water would likely be considerably enhanced by the presence of this secondary porosity. In addition, the organisms also process water, moving contaminants in the dissolved state or associated with colloidal organic matter in the pore water.

The ratio of contaminant fluxes between the medium and high-density microcosms differed by only about a factor of two although the animal densities differed by a factor of four. The reduced activity per organism in the high-density microcosms could have been the result of increased intraspecies competition, perhaps for food at the higher animal densities. Consistent with this is a model of organism activity, and thus bioturbation, dependent upon the square root of density. Forbes and Forbes (1994)

analyzed 10 datasets given by Matisoff (1982) in which the bioturbator, biomass density and the resulting effective bioturbation diffusion coefficient were measured. They concluded that the effective diffusivity was proportional to biomass to a power of 0.52 ± 0.15 . It thus appears that our observation of proportionality to the square root of biomass may be more generally applicable. The effective bioturbation diffusion coefficients and biomass reported by Matisoff are shown in Fig. 8. Also included in Fig. 8 is the best fit geometric curve to Matisoff's reported data ($0.5 \times 10^{-8} [\text{Biomass density}]^{0.52}$) as well as the individual measurements of effective bioturbation diffusion coefficient. The bioturbation diffusion coefficient data reported by Matisoff includes measurements from marine organisms (*Yoldia limatula* and *Pectinaria gouldi*) and freshwater macroinfauna (*Limnodrilus*, *Ponoporeia hoyi* and *Tubifex tubifex*) in both laboratory and field settings. Given the variety of data sources, the effective bioturbation diffusion coefficients correlate with biomass remarkably well.

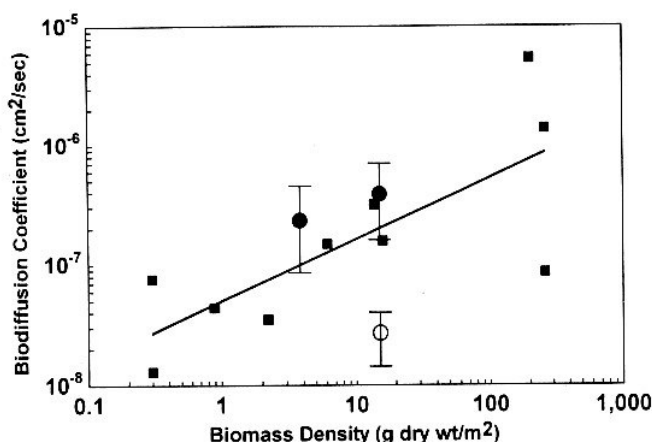


Fig. 8. Effective bioturbation diffusion coefficient compared with biomass density (dry weight basis). (■) Measurements reported by Matisoff (1982). (—) $D_{\text{bio}} = 0.5 \times 10^{-8} \text{ cm}^2/\text{s} (\text{biomass density})^{0.52}$. (●) Estimated bioturbation diffusion coefficient from this work—hypoxic conditions. (○) Estimated bioturbation diffusion coefficient from this work—oxygen saturated conditions.

The observed effective bioturbation mass transfer coefficients were also converted to effective bioturbation diffusion coefficients and plotted in Fig. 8. Reference to film theory analogies of mass transfer were made to calculate the bioturbation diffusion coefficient from

$$D_{\text{bio}} \approx k_{\text{bio}} H \quad (H = 3 \text{ cm})$$

Here H is the depth of the bioturbation layer, limited to 3 cm in the experimental cells. The effective bioturbation diffusion coefficients measured under hypoxic conditions (solid symbol) agree well with the correlation based on the previous data. The range shown in the figure indicates the range of average release rates observed with the three different compounds. The effective bioturbation diffusion coefficient under oxygen-saturated conditions (open symbol) falls considerably below the correlation.

It should be noted that the measured mass transfer coefficients are also comparable to other field measurements. Matisoff (1982) presents more than 80 measured values of effective diffusion coefficients for bioturbation, of which more than 2/3 fall in the range of 0.3–30 cm²/yr. If the region influenced by bioturbation is of the order of 3 cm, this range is equivalent to an effective mass transfer coefficient range of 0.1–10 cm/yr. All of the laboratory measurements presented here fall within that range.

The importance of bioturbation from the perspective of this work is the resulting increased contaminant flux to the overlying water from bioturbated cells relative to control cells. The magnitude of the increase can also be illustrated by comparing the measured effective bioturbation mass transfer coefficients with the effective mass transfer coefficient assuming that only molecular diffusion of the dissolved compound is causing release to the water column. As shown by Reible *et al.* (1991) the flux from an initially uniformly contaminated sediment via molecular diffusion only can be written

$$\text{Flux}_{\text{diff}} = \left[\frac{D_w \epsilon^{4/3}}{R_f \pi t} \right]^{1/2} \rho_b W_A \quad (3)$$

where D_w is the molecular diffusivity in water, $\epsilon^{4/3}$ corrects the diffusivity for the porosity (ϵ) and tortuosity of the sediment (Millington and Quirk, 1961), and $R_f = \epsilon + K_d \rho_b$ is the retardation factor associated with the sorption of the contaminant onto the (immobile) solid phase, K_d is the sediment–water partition constant for the contaminant and ρ_b is the bulk sediment density. For pyrene in the Bayou Manchac sediment this value is approx. 1325. The ratio of the bioturbation to the diffusion flux is then given by

$$\frac{\text{Flux}_{\text{bio}}}{\text{Flux}_{\text{diff}}} = k_{\text{bio}} \left[\frac{R_f \pi t}{D_w \epsilon^{4/3}} \right]^{1/2} \quad (4)$$

Initially, diffusion is dominant because contaminated sediment would be present at the sediment–water interface. Over time, however, this region would become depleted of contaminant and the rate of diffusion would slow. Bioturbation, however, would extend at least several cm into the sediment and the rate would be maintained for long periods of time. For pyrene in the Bayou Manchac sediment, the flux from bioturbated sediment after 10 yr would be about 370 times greater than the flux from a non-bioturbated sediment exhibiting only diffusional release to the overlying water column.

The ecological importance of this level of flux into the sediments is less well known. The contaminant that enters the water column will likely be quickly sorbed to organic matter, transported and re-deposited in the sediments dispersing contaminants. Nevertheless, such levels may bring about sublethal effects (Kennish, 1992). Bioaccumulation is also likely since tubificids are important dietary components in fishes and other aquatic species.

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