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Dissolution of particle-reactive radionuclides in deposit-feeder digestive fluids

Abstract—Naturally occurring radionuclides such as ^{234}Th , ^7Be , and ^{210}Pb are important tracers for quantifying sediment-mixing and sediment-accumulation rates. Profiles of these radionuclides in marine sediments are strongly influenced by particle displacement due to deposit feeding. Observations of rapid dissolution and high concentrations of dissolved metals in deposit-feeder digestive fluids suggest that particle-bound radionuclides could also undergo dissolution during deposit-feeder gut passage. We investigated this possibility in laboratory experiments examining radionuclide dissolution into the digestive fluids of the lugworm, *Arenicola marina*. Experiments with artificially labeled particles indicated that significant fractions of ^{234}Th , ^7Be , and ^{210}Pb dissolved from labeled algal detritus and clay particles at low particle concentrations.

^{137}Cs was also dissolved from clays. However, if unlabeled sediment particles were added to reach sediment:fluid ratios similar to those in *A. marina* midguts, little net dissolution occurred, which implies resorption of dissolved radionuclides by the added solid phases. Partition coefficients of these radionuclides in mixtures of digestive fluid and the various solid phases imply that relatively more ^{234}Th resorbs to the residual organic phase following digestion, compared to ^{210}Pb and ^7Be , which partition more strongly to the inorganic sediment phases. Despite little net dissolution, the phase change from algal detritus to either mineral surfaces (for ^{210}Pb) or undigested organic matter (for ^{234}Th) implies that ^{234}Th would serve as a better tracer for organic-matter mixing in sediments compared to ^{210}Pb , which would better trace bulk sediment mixing.

Deposit feeders are often the numerically dominant trophic group in muddy sediments. Scaling arguments suggest that the reworking activities of deposit feeders may control rates of sediment mixing due to their rapid ingestion rates and large size relative to the mixed-layer depth (Wheatcroft et al. 1990). Sediment-mixing rates are frequently determined from vertical profiles of the particle-reactive radionuclides ^{234}Th , ^7Be , and ^{210}Pb . Profiles of excess activities of these tracers in marine sediments may result primarily from particle reworking by deposit feeders (Rice 1986; Shull 2001).

In addition to physically displacing sediments, deposit-feeder gut passage can influence sediment chemistry. Benthic deposit feeders secrete digestive agents such as hydrolytic enzymes and surfactants to hydrolyze and solubilize absorbable food items from sediments (Mayer et al. 1997). Digestive fluids of deposit feeders contain high concentrations of various dissolved organic compounds that can serve as ligands for binding and solubilizing metals (Mayer et al. 1996; Chen and Mayer 1999; Chen et al. 2000).

Dissolved metals are highly concentrated in luminal fluids of deposit feeders. Metal concentrations within the digestive tract appear to be regulated by simple metal-ligand interactions (Chen et al. 2000). For example, Cu in digestive fluids is complexed primarily by amino acids such as histidine (Chen and Mayer 1998, 1999). Solubilization kinetics can be complex, involving rapid dissolution followed by slower dissolution or resorption onto the solid phase (Chen and Mayer 1999).

Deposit-feeder digestive tracts may therefore be active environments for solubilizing particle-reactive, metallic radionuclides such as ^{234}Th , ^7Be , ^{210}Pb , and ^{137}Cs . Dissolution during gut passage would challenge the assumption of postdepositional immobility required for using these radionuclides to track particle transport processes such as bioturbation and sedimentation. However, the extent of dissolution of naturally occurring radionuclides in deposit-feeder digestive fluids is unknown. Field and laboratory studies indicate that ^{234}Th and ^{210}Pb experience dissolution and perhaps diffusive redistribution in the sediment (Santschi et al. 1983). Cochran et al. (1986) attributed dissolution of ^{234}Th in sediments to the reduction and dissolution of adsorptive Fe and Mn oxides that host these radionuclides. ^{210}Pb is also associated with reducible solid phases and can be subject to dissolution and mobilization in lacustrine sediments (Benoit and Hemond 1991). Dissolution during deposit-feeder gut passage could contribute to diffusive mobilization of these tracers and thereby affect the measurement of sediment-mixing and sediment-accumulation rates. Dissolution followed by resorption could result in a change in the solid phase binding these tracers without diffusive mobilization.

The objective of this research is to determine the extent of dissolution of these radionuclides in the digestive fluids of the deposit-feeding polychaete *Arenicola marina*. We focus on this species because the digestive system of *A. marina* is perhaps the best studied among benthic deposit feeders and is somewhat representative of polychaetous deposit feeders in general (Mayer et al. 1997). We address this issue with a series of laboratory experiments using natural and artificially labeled sediment particles. Experiments with la-

beled particles were designed to simulate the scavenging of these radionuclides by particles in the water column followed by deposition and ingestion by deposit feeders.

Methods—In our experiments, we used digestive fluids from the lugworm, *Arenicola marina*, collected from a sand-flat near Lubec, Maine. Individuals were maintained in seawater for 3 to 5 h to allow the worms to purge sediment from their digestive tracts. Afterward, lugworm midguts were exposed by dissection and luminal fluids were collected by perforating the gut wall with a pipette and withdrawing the fluid. Midgut fluids were used because dissolved metal concentrations are typically highest in this region, along with high enzyme activities and high concentrations of amino acids and surfactants (Mayer et al. 1997; Chen et al. 2000). Additional digestive fluids were collected by perforating and draining the digestive glands, located anterior to the midgut, which have characteristics similar to midgut fluids. Sediment particles were removed from the fluids by centrifugation at $8,000 \times g$ at 4°C for 30 min. Clarified fluids were then stored at -80°C until they were used for incubation experiments. We have observed little loss of enzyme activity in *A. marina* digestive fluids stored at this temperature.

To determine the extent of dissolution into digestive fluids of naturally occurring radionuclides, *in vitro* digestion experiments were conducted with surficial sediment collected from Lowes Cove, Maine, adjacent to the Damariscotta estuary, and sediment from Bailey Cove, adjacent to Montsweag Bay, Maine. Particles were also collected by use of a sediment trap (PVC tube, 140-cm length, 10-cm inner diameter) deployed at a depth of approximately 16 m in the Damariscotta River near the Darling Marine Center ($43^\circ56'10''\text{N}$, $69^\circ35'\text{W}$). Three grams of wet sediment (1.5 g dry sediment) were added to 3 ml *A. marina* digestive fluids (or to $0.45\text{-}\mu\text{m}$ filtered seawater as a control, collected from the Damariscotta River estuary), vortexed, and incubated for 3 h on a shaker. Digestive fluids were shaken to reduce the potential for dissolution rates to be limited by rates of diffusion of digestive enzymes and dissolved ligands. After incubation, particles were separated from the fluid phase by centrifugation at $20,000 \times g$ for 30 min at 4°C . The fluid phase was then passed through a $0.45\text{-}\mu\text{m}$ filter to safeguard against contamination by particulate material and transferred to a 5-ml scintillation vial for counting. Radionuclide activities in the digestive fluid and particulate phases were measured by use of an intrinsic germanium gamma spectrometer with a well detector (Canberra GCW 3523/S) equipped with ultra-low background lead shielding (Canberra Model 777). Activities in digestive fluid prior to the *in vitro* digestion experiments were also measured. Detector efficiency at each energy peak was calculated by use of an energy-efficiency curve determined by counting multinuclide standards (Isotope Products). Particulate-phase and fluid-phase standards were created with the same geometry and self-absorption characteristics as the samples.

Labeled particles were also used for *in vitro* digestion experiments. We radiolabeled algae (*Tetraselmis* 2K instant algae, Reed Mariculture) and clay particles ($<2\text{-}\mu\text{m}$ fraction of sediment, collected from Miller's Island, Maine) by incubating algae or clay with particle-reactive radioisotopes in

solution. We obtained ^7Be for the incubations from Brookhaven National Laboratories (in 0.5M HCl). We used ^{210}Pb and ^{137}Cs from a multinuclide standard (Isotope Products 7500). We separated ^{234}Th from a solution of its parent ^{238}U (SPEX CertiPrep) by ion chromatography using Dowex 1×8 100–200 mesh resin (Anderson and Fleer 1982). These radioisotopes, in acidic aqueous solution, were added to seawater, and the pH was adjusted to eight by addition of NaOH. This solution was then added to a suspension of seawater and algae (or clay particles). These suspensions were incubated on a shaker for 4 d. The resultant activities of the algae were $5,000 \text{ Bq g}^{-1}$ (^{210}Pb), 800 Bq g^{-1} (^{234}Th), and $2,800 \text{ Bq g}^{-1}$ (^7Be). We did not employ sterile conditions, so the algae underwent decomposition by bacteria present in the seawater and the resultant particulates resemble an algal-detritus complex as it might occur after settling through a marine water column. Radiolabeled particles were then separated from solution by centrifugation at $8,000 \times g$ for 10 min.

To examine dissolution kinetics, 7–8 mg algal detritus (2 mg dry weight) and 1.0 g wet sediment from the lugworm collection site (0.8 g dry weight) were added to six centrifuge tubes containing 2.1 ml digestive fluid. The centrifuge tubes were vortexed for 1 min and then incubated on a shaker for 10, 20, 40, 80, 160, and 320 min. After incubation, the fluid and particulate phases were separated by centrifugation and filtration and both were counted with the gamma detector. Counts of the particulate phase before and after digestive-fluid incubation were compared to determine activity balance.

To test for the influence of varying amounts of sediment on net dissolution, 3–4 mg algal detritus were added to 3 ml digestive fluid (or to 0.45- μm -filtered seawater as a control, collected from the Damariscotta River estuary) to approximate the ratio of algal detritus to digestive fluid we expected to find in lugworms in the field. A range of masses of wet sediment from the lugworm collection site was added (0.1 to 1.8 g dry weight) to five centrifuge tubes. The highest sediment mass approximates the ratio of sediment to digestive fluid often found in lugworm midguts. The five treatments were incubated for 3 h, and the fluid and particulate phases were separated and counted as in the kinetics experiment. To determine net dissolution under the simpler conditions of varying amounts of algal detritus or varying amounts of sediment without algal detritus, this experiment was repeated using (a) 50 mg (wet weight) radiolabeled clay (25 mg dry weight) incubated in digestive fluid containing 0.1 to 1.8 g dry weight of unlabeled sediment and (b) radiolabeled algal detritus incubated in digestive fluid containing a range of masses of unlabeled algal detritus (1 to 90 mg dry weight). This experimental design enabled us to infer differences in radionuclide sorption affinities for algal detritus and sediment mineral phases by comparing partition coefficients by use of analysis of covariance with particle concentration as the covariate.

Results—Experiments with natural sediments (surface sediment and sediment-trap material) showed little dissolution of radionuclides into *A. marina* digestive fluids. How-

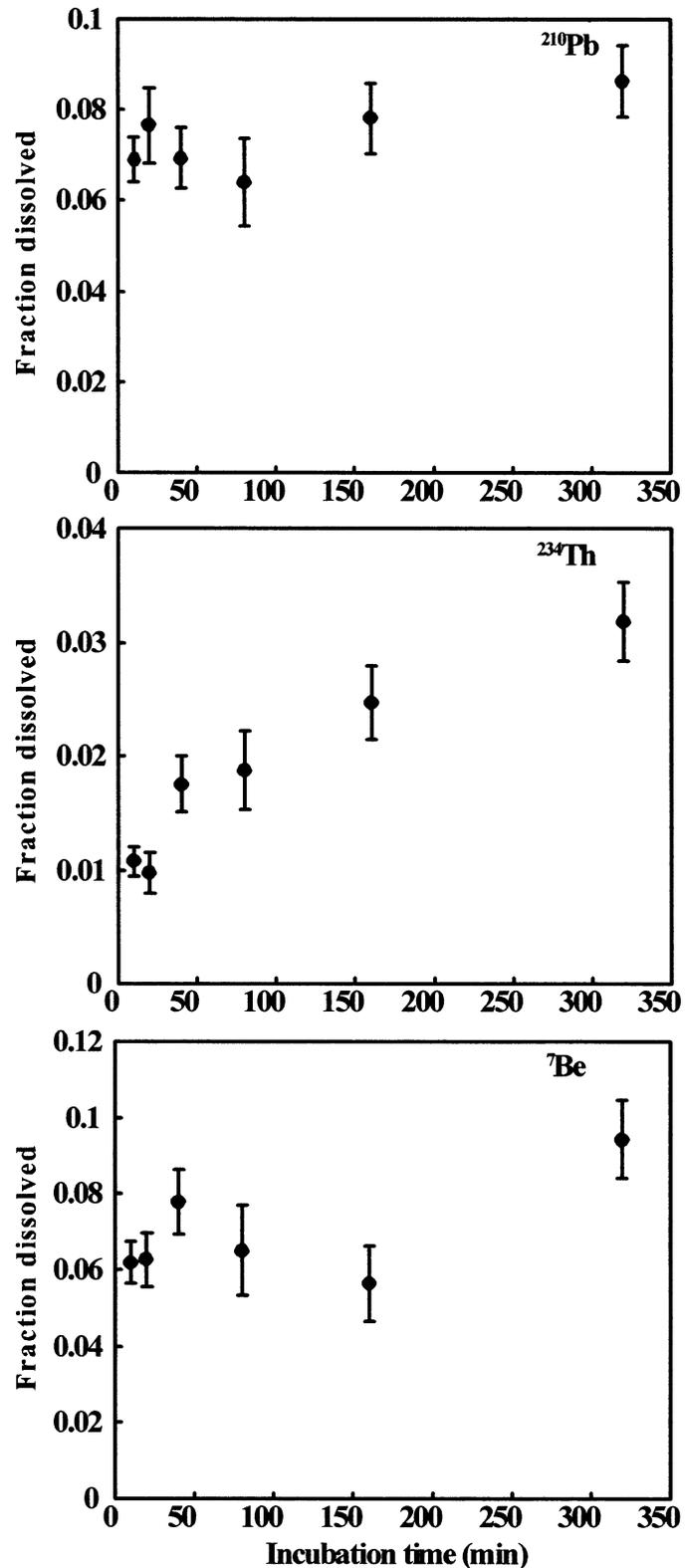


Fig. 1. Kinetics of ^{210}Pb , ^7Be , and ^{234}Th release from labeled algal detritus into *A. marina* digestive fluid in the presence of sediment (0.8 g dry weight). Fraction dissolved is the ratio of fluid-phase activity to total activity (dissolved activity/[dissolved + particulate activity]). Error bars represent analytical error (1 SD).

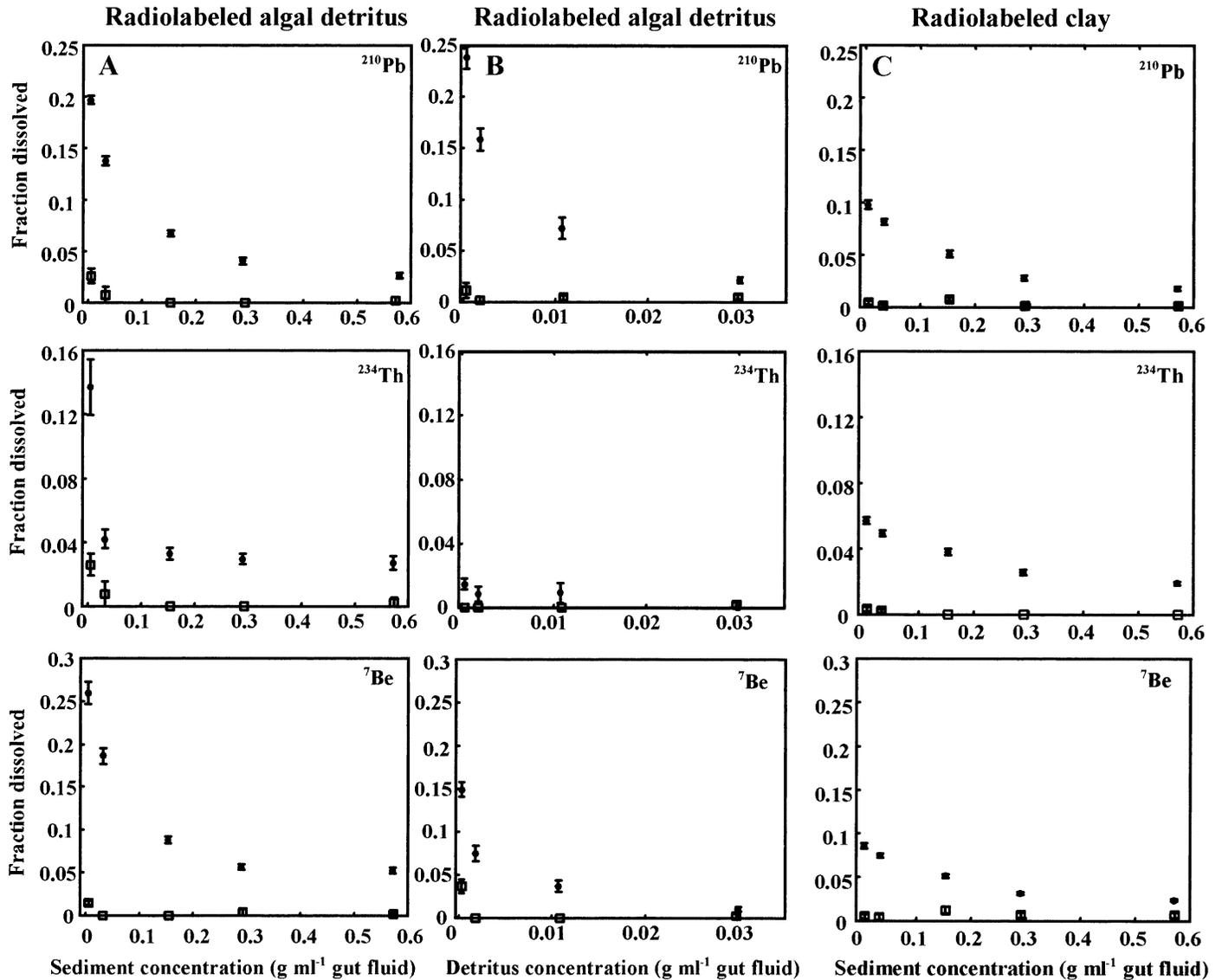


Fig. 2. Fraction of ^{210}Pb , ^{7}Be , and ^{234}Th released from labeled algal detritus into *A. marina* digestive fluid over a range of added particle concentrations. (A) Dissolution of radionuclides initially bound to algal detritus over a range of unlabeled sediment particle concentrations. (B) Dissolution of radionuclides initially bound to algal detritus over a range of unlabeled algal-detritus concentrations. (C) Dissolution of radionuclides initially bound to clay over a range of unlabeled sediment particle concentrations. Solid circles represent fraction radionuclides dissolved into digestive fluids. Open squares represent fraction dissolved into seawater. Error bars represent analytical error (1 SD).

ever, experiments with radiolabeled algal detritus and clay particles showed measurable dissolution. Small fractions of radionuclides, initially bound to labeled algal detritus, dissolved over the 5-h period in the presence of digestive fluids and sediment (Fig. 1). We define dissolved as not subject to centrifugation at $20,000 \times g$ and passing a $0.45\text{-}\mu\text{m}$ filter. This fraction may also contain colloiddally bound radionuclides. Most dissolution for ^{210}Pb and ^{7}Be occurred during the first 10 min of incubation followed by steady state or equilibrium. For ^{7}Be , there was a slight rise in the fraction dissolved after 3 h of incubation. ^{234}Th activity in the dissolved phase continued to rise during the 5-h experiment. Approximately 6% to 9% of ^{210}Pb and ^{7}Be initially bound to algae were found in the dissolved phase. Only 3% of ^{234}Th was found in the dissolved phase after 5 h. Subsequent ex-

periments were conducted with a 3-h incubation time. Although ^{234}Th would not reach steady state or equilibrium within this incubation period, a longer incubation time would be unrealistic relative to gut passage times of deposit feeders.

Adding unlabeled algal detritus or sediment particles to the labeled ones decreased the net dissolution of radionuclides into digestive fluids (Fig. 2). Between 14% and 26% of the three radionuclides was dissolved from algal detritus at the lowest particle concentration (4 mg unlabeled sediment ml^{-1} digestive fluid, Fig. 2A). Less than 5% was dissolved in treatments with sediment concentrations approximating that found in the midgut of *A. marina* (0.7 g ml^{-1} unlabelled sediment). In seawater controls, less than 3% of ^{234}Th , ^{7}Be , and ^{210}Pb was found in the dissolved phase, even at the lowest algal particle or sediment particle concentra-

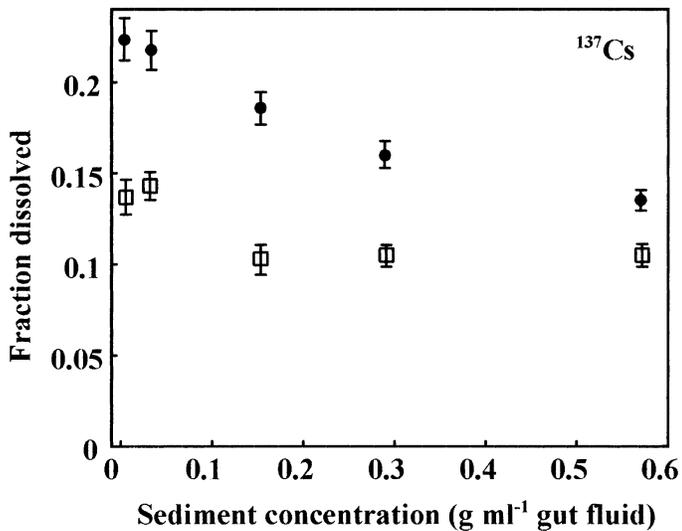


Fig. 3. Fraction ^{137}Cs released from labeled clay into *A. marina* digestive fluid over a range of unlabeled sediment particle concentrations. Solid circles represent fraction radionuclides dissolved into digestive fluids. Open squares represent fraction dissolved into seawater. Error bars represent analytical error (1 SD).

tion. Little or no activity was observed in the dissolved phase for treatments with higher particle concentrations.

The addition of unlabeled algal particles to the digestive-fluid incubations also decreased the fraction dissolved (Fig. 2B). Algal detritus was more sorptive than sediment, with little ^{234}Th dissolution in treatments with low concentrations of unlabeled algal particles. Dissolution of ^{210}Pb and ^7Be also dropped with increasing concentrations of unlabeled algal particles. Again, much less dissolution occurred in the seawater controls (Fig. 2B).

For ^{210}Pb and ^7Be , a smaller fraction of radionuclide was dissolved from labeled clay particles than from labeled algal detritus (Fig. 2C). For ^{137}Cs , 14% to 22% was dissolved by digestive fluid (Fig. 3). At the lowest sediment particle concentration, seawater dissolved nearly 15% of the ^{137}Cs initially sorbed to clay.

The relative adsorptivity of added algal detritus versus added sediment becomes apparent by recasting these data as partition coefficients, K_D (Fig. 4). The partition coefficient is the ratio of the solid-phase activity per mass of solids to the dissolved-phase activity per volume of fluid (units: ml digestive fluid g^{-1} particle). This recasting linearizes the relationships and facilitates statistical comparisons. In both the labeled algal detritus and labeled clay experiments, K_D decreased significantly and in a linear fashion, as particle concentration increased ($F > 40$, $p < 0.001$). The slopes of the $\log K_D$ versus \log (algal-detritus concentration) regression lines were not significantly different from the $\log K_D$ versus \log (sediment concentration) regression lines for ^{210}Pb or ^{234}Th ($p > 0.05$) but were different for ^7Be ($p = 0.022$). Analysis of covariance allows removal of the effect of solids concentration in comparing K_D between solids types. For ^{234}Th , the $\log K_D$ quantifying the partitioning between digestive fluid and algal detritus was higher than the $\log K_D$ quantifying partitioning between digestive fluid and sedi-

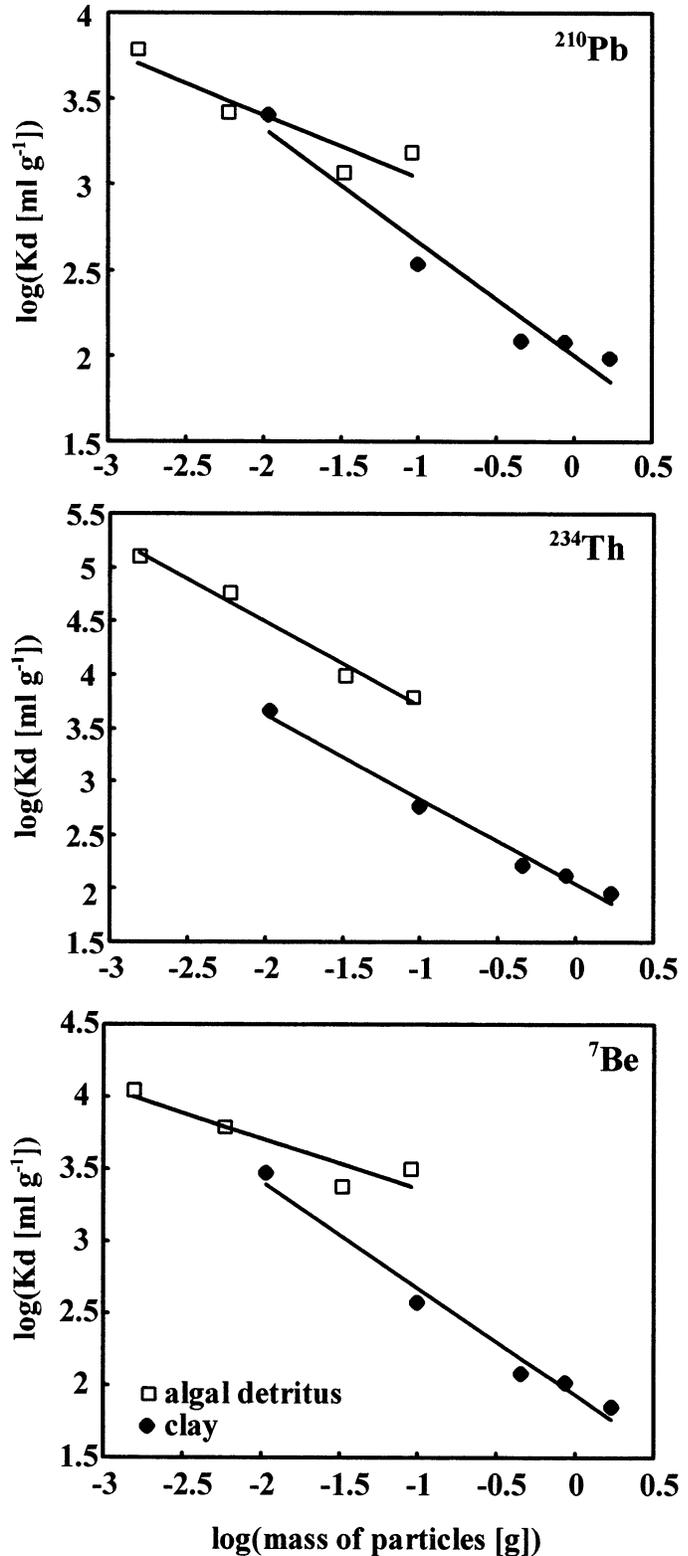


Fig. 4. Partition coefficients for ^{210}Pb , ^7Be , and ^{234}Th (ml fluid g^{-1} particle) as a function of particle concentrations. Open squares indicate incubations with unlabeled algae. Closed circles indicate incubations with unlabeled sediment particles. Partition coefficients were calculated from the dissolution data displayed in Fig. 2B (algal particles) and 2C (sediment particles). Solid lines represent least-squares fits.

ment particles (ANCOVA, $F_{1,6} = 121$, $p < 0.0001$). For ^{210}Pb , the $\log K_d$ for algal detritus was not significantly higher than the $\log K_d$ for sediment particles (ANCOVA, $F_{1,6} = 2.3$, $p = 0.18$).

Discussion—Dissolution kinetics of Cu, Cd, and stable Pb in *Arenicola marina* digestive fluids have been examined previously (Chen and Mayer 1999). Dissolution of these metals involves rapid initial solubilization followed by stabilization of dissolved concentrations or resorption onto solid phases. In our time-series experiment, we found rapid dissolution of ^{210}Pb and ^7Be , followed by steady state or equilibrium. In contrast, ^{234}Th showed slower dissolution kinetics. This finding is surprising in light of observations that ^{234}Th sorption/dissolution kinetics with natural organic matter is rapid (on the order of minutes) and irreversible (Quigley et al. 2001). There are several possible explanations for relatively slow dissolution of ^{234}Th in digestive fluid. It is possible that the digestion of algal detritus may have generated colloidal material over the course of the incubation. Thorium is strongly sorbed to colloids (Honeyman and Santschi 1989; Clegg and Whitfield 1993) and any colloidal ^{234}Th would be included in the dissolved phase by our separation techniques. Alternatively, as thorium possessed a higher affinity for algal detritus than the other radionuclides tested, dissolution of this detritus complex during digestion might control the rate of thorium dissolution, along with complexation-induced displacement from solid phases.

The greater dissolution of these radionuclides in gut fluid relative to the seawater control implies that gut fluid contains ligands with high affinity for these radionuclides. Despite the increased partitioning toward the fluid phase that results from these ligands, the relative partitioning between fluid and solid phases is similar to that found in other systems. The rank order of partition coefficients for radionuclides between sediment particles and lugworm digestive fluids (Th > Pb > Be > Cs) is the same as the rank order of partition coefficients between clay particles and seawater (Li 1981). Higher affinity of thorium for algal detritus compared to lead has been observed in phytoplankton and biogenic debris in the water column (Fisher et al. 1987, 1988), consistent with our finding of stronger partitioning of Th toward the algal-detritus phase than for the other radionuclides. This attraction could also result from very high affinity sorption sites for thorium associated with bacterial cells likely associated with the algal detritus (Hirose and Tanoue 2001). The lesser enhancement of ^{137}Cs dissolution in gut fluid relative to the seawater control is consistent with the lower affinity of Cs for organic ligands such as those present in gut fluid.

A significant fraction of radionuclides initially bound to algal detritus was dissolved in the presence of digestive fluids at algal-detritus concentrations similar to those in the field. However, in the presence of nonalgal particles at concentrations mimicking those in *A. marina* guts, net dissolution was strongly reduced. This reduction may explain why our digestive-fluid incubation experiments with natural sediments indicated undetectable levels of dissolution. The maintenance of low dissolved radionuclide concentrations differs from results in a water column situation, in which protistan grazing on ^{234}Th associated with bacteria was found

Particle-reactive radionuclide phase changes

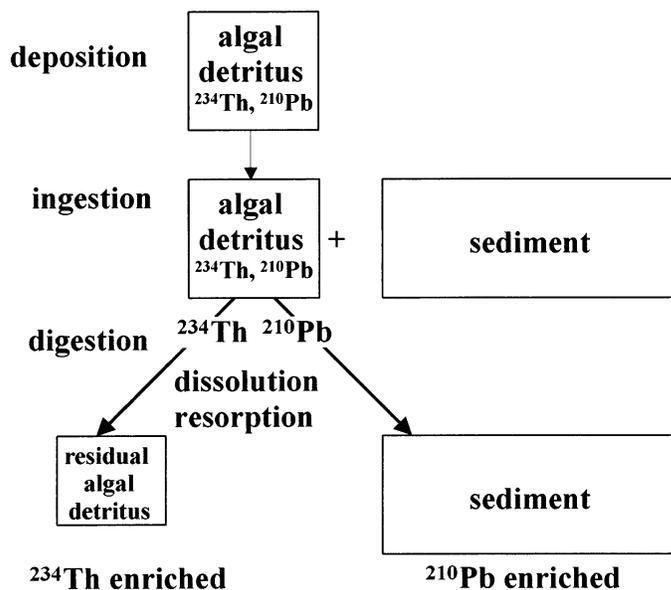


Fig. 5. Illustration of the transfer of ^{234}Th and ^{210}Pb from algal detritus to different particle phases due to dissolution and resorption during deposit-feeder gut passage.

to partition the thorium to the dissolved phase (Barbeau et al. 2001). In this latter case, particulate concentrations in the water column are too low to allow significant resorption.

Our results imply a shift in the phase association of ^{210}Pb from the food items (algal detritus) to the matrix within which they are mixed during deposit feeding. This is due to much higher concentrations of non-algal-detritus particles in sediments. ^{234}Th will undergo a similar process but will remain more closely associated with the nondigested residual algal-detritus fraction (Fig. 5). Partition coefficients indicate ^7Be displays behavior intermediate between ^{234}Th and ^{210}Pb over the particle concentrations examined. Initial dissolution followed by resorption has also been observed for Cu, Cd, and stable Pb during time-course incubations with deposit-feeder digestive fluids (Chen and Mayer 1999). Resorption results in little net dissolution of ^{210}Pb , ^{234}Th , and ^7Be from algal detritus and clay during gut passage in the presence of high particle concentrations. Nevertheless, the apparent transfer of these radionuclides from algal detritus to other sediment surface ligands has implications for the use of these tracers in determining rates of particle mixing.

Generally, ^{234}Th is mixed more rapidly than ^{210}Pb in surface sediments. This observation has been ascribed to age-dependent mixing, in which recently deposited particles of high food value, enriched in short-lived radionuclides, are mixed more rapidly than particles associated with longer lived radionuclides due to particle selection by deposit feeders or other processes (Smith et al. 1993; Fornes et al. 2001). Our findings support and modify this hypothesis by providing a mechanism for transferring ^{234}Th and ^{210}Pb onto different types of particles that would be subject to differential selection by deposit feeders. For example, if 20% of excess ^{210}Pb were dissolved from organic detritus (Fig. 2A) and transferred to inorganic sediment particles during each gut

passage, roughly 99% would be transferred after 23 gut passages. Owing to differences in sorption affinities, a higher fraction of dissolved ^{234}Th would resorb onto the residual organic phase. Selective feeding on organic-rich particles by deposit feeders would then result in more rapid mixing of ^{234}Th relative to ^{210}Pb . Although mixing mechanisms that do not involve gut passage might play an important role in determining differences in bioturbation rates for these tracers (Levin et al. 1997; Fornes et al. 2001), dissolution followed by sorption onto different kinds of particles would provide an explanation for differences in mixing rates among these tracers due to selective deposit feeding. Under this scenario, ^{210}Pb would be a better tracer for bulk sediment, whereas ^{234}Th would be a better tracer for particles of high food value.

Despite appreciable dissolution of radionuclides from algal detritus, resorption by other solid-phase ligands within the gut limits the potential for remobilization and diffusive redistribution of the dissolved phase within the sediment. The commonly used tracers ^{234}Th , ^{210}Pb , and ^7Be appear to be essentially conservative with respect to gut passage. The small fraction of dissolved radionuclides that persists within digestive fluids at high sediment concentrations could contribute, however, to the rates of dissolution and mobilization of these radionuclides observed in natural sediments (Santschi et al. 1983; Cochran et al. 1986; Benoit and Hemond 1991).

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