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Bioavailability of Sediment-Bound Methyl and Inorganic Mercury to a Marine Bivalve

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The extent to which ingested suspended sediment can be a source of mercury for suspension feeders is largely unknown, yet this information is required to evaluate the biological significance of sediment contamination. We used radiotracer methodology to experimentally compare the bioavailability of dissolved and sediment-bound mercury for a marine bivalve. We examined the relative importance of specific sedimentary components that may exert control on the uptake of Hg(II) and $CH_3Hg(II)$ from sediments in the mussel *Mytilus edulis,* which is widely used in national monitoring programs as a bioindicator organism for detecting coastal contamination. Iron and manganese oxides, montmorillonite clay, and silica, with and without organic coatings, as well as natural sediment particles were radiolabeled with either $^{203}Hg(II)$ or $CH_3{}^{203}Hg$ using nanomolar levels of Hg. Fulvic acid coatings enhanced the sorption of both Hg(II) and $CH_3Hg(II)$ onto every type of particle, and partition coefficients were as high as 10^5. The assimilation efficiencies (AE) in mussels were very low for Hg(II) (1−9%) but were typically >30% for $CH_3Hg(II)$ and as high as 87%. The organic coatings increased AE for $CH_3Hg(II)$ bound to all types of particles, but had no impact on Hg(II). Unlike Cd, Co, and Ag, there was little (<10%) desorption of ^{203}Hg from particles at pH 5 (to simulate the behavior of food-bound metals in the acidic gut of bivalves), and it did not influence Hg assimilation in mussels. The absorption of $CH_3Hg(II)$ by mussels is rapid and efficient even from uncoated inorganic particles that were rapidly egested out of the animal's gut due to their low nutritional value. The concentration factor (10^3) of dissolved Hg(II) in mussels was half that of dissolved $CH_3Hg(II)$. These results suggest that contaminated sediments can be a significant source of Hg, especially organic mercury, to marine suspension feeders and that $CH_3Hg(II)$ is assimilable from all major sedimentary components.

Introduction

The bioavailability of sediment-bound metals is of great interest because it determines the extent to which contaminated sediments can introduce metals into aquatic food chains, but bioavailability is at present difficult to predict partly because of the presence of different types of binding sites on sediment particles (*1*). Field and laboratory studies have shown the influence of metal partitioning among sediment components on metal bioavailability to benthic invertebrates (*2*−*6*). However, the application of selective chemical extractions to understand the bioavailability of sediment-bound metals to benthic organisms is limited, and the fate of metal−sediment associations in the guts of benthic organisms has barely been explored. In principle, metals must be in solution in the gut before organisms can assimilate them (*7*), consistent with Gagnon and Fisher's (*8*) findings that the release of sediment-bound Ag, Cd, and Co from ingested particles in the acidic gut of mussels controls their assimilation. Data of this kind may be critical in evaluating existing (or establishing new) sediment quality criteria.

The bioavailability of sediment-bound mercury would be expected to be low given its strong association with sediment particles. Despite the significant role of methyl mercury in the trophic transfer of mercury (*9*−*11*), few studies have directly assessed the biological availability of sediment-bound Hg to benthic organisms. Studies of Hg binding show that methyl mercury does not bind as tightly with organic matter in sediments as does the Hg^{2+} ion (*12*), but it is still a particle-reactive contaminant (*13*). Previous investigations (*14, 15*) measured inorganic Hg incorporation by benthic fauna but did not examine methylated species, which are bioaccumulated in marine organisms to a much greater extent (*16*). Earlier laboratory studies (*17, 18*) showed that the bioavailability of $CH_3Hg(II)$ from food to bivalves was greater than inorganic mercury, but these studies did not examine sediments as a potential source of mercury.

The bioavailability of sediment-bound Hg(II) and $CH_3Hg(II)$ for benthic organisms must be evaluated with regard to key geochemical factors (e.g., organic matter, iron and manganese oxides) that vary significantly among coastal sediments. For example, inverse relationships between mercury accumulation in benthic fauna and organic carbon contents of sediments have been observed (*19*−*21*).

The principal source (particles vs dissolved phase) of mercury contamination for benthic organisms remains unknown. Despite very low release rates of $CH_3Hg(II)$ from anoxic sediments into oxic overlying water (<0.1 ng cm^{-2} yr^{-1}; *22*), uptake from solution must be considered to determine the relative importance of different uptake pathways. Earlier studies (*17, 18*) have shown that benthic organisms can accumulate Hg from the dissolved phase, although these studies used much higher $CH_3Hg(II)$ concentrations, >150 ng of Hg L^{-1}, than those typically found in marine waters, 10 pg L^{-1} (*13*); values in water columns with low oxygen content can reach 0.4 ng L^{-1} (*23*). This problem also applies to earlier studies of inorganic Hg in seawater, which used elevated Hg concentrations (e.g., 1000 ng L^{-1}) (*15*).

In this study, the assimilation of inorganic and methyl mercury from marine sediments in suspension feeding mussels was experimentally determined using radiotracer methodology. We examined the relative importance of specific sedimentary components that may control the bioavailability of inorganic and methyl mercury. Uptake of high specific activity radioactive mercury [$CH_3Hg(II)$ and Hg(II)] from water was also measured separately using lower, more realistic mercury concentrations (<1 ng L^{-1}) than in most earlier studies.

Materials and Methods

Experiments utilized the common mussel *Mytilus edulis,* a well-studied sentinel species employed in monitoring coastal contamination (*24*−*26*). The mussels were collected from the shore at Flax Pond, Long Islands and acclimated in the laboratory for 10 days in running seawater, as described elsewhere (*27*). Immediately prior to the experiments, their

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shells were cleaned of epifauna and epiflora. The general experimental protocol involved exposure of animals to radiolabeled sediment particles in suspension for a short period, followed by a 4-day exposure to unlabeled food to allow gut evacuation of ingested radiolabeled metal. ^{203}Hg ($t_{1/2}$ = 46.6 d), a γ-emitter, was used in these experiments. Since γ analysis is non-destructive, the accumulation and depuration of mercury in mussels could be followed in the same individual over time, thus reducing biological variability as a source of error in these experiments. These "pulse−chase" procedures have been employed to determine metal assimilation in deposit and suspension-feeding bivalves (*27*−*30*).

Mussels were fed with nine types of suspended particles: fulvic acid-coated and uncoated inorganic particles (iron and manganese oxides, montmorillonite clay, and silica) and natural organic-rich sediment from the salt marsh at Flax Pond. We also assessed bioaccumulation of inorganic and organic Hg from the dissolved phase. The use of radioactive Hg with high specific activity allowed work at lower concentration levels than previous studies. All seawater used in these experiments was collected from surface waters 8 km off Southampton, Long Island, NY.

Preparation of $CH_3{}^{203}Hg(II)$ of High Specific Activity. $^{203}HgCl_2$ dissolved in 1 N HCl (specific activity > 28 mCi mg^{-1}) was purchased from the Buffalo Materials Research Center and was used to synthesize radioactive methylmercury by adapting methods described previously (*31, 32*). Briefly, ^{203}Hg was methylated with methylcobalamin 0.01 N HCl (pH 1−2) for 1 h, the synthesized $CH_3{}^{203}HgCl$ was extracted with benzene, the solvent was evaporated, and the methylmercury was dissolved in a 5 mM $CaCO_3$ solution.

Preparation of Radiolabeled Suspended Particles. Iron and manganese oxides and montmorillonite clay mineral were supplied by Aldrich. Quartz silica particles were acid-washed and rinsed before radiolabeling. Particles (4−8 μm diameter) were obtained by sedimentation (*8*) and had comparable dimensions to the phytoplankton that these animals normally feed upon. A second set of the four particle types was coated with fulvic acid (FA) to simulate the organic aggregation in natural sediments. The FA, extracted from seawater (*33*), was kindly provided by G. R. Harvey. Coating the particles followed the protocol described by Decho and Luoma (*29*). Particles (150 mg L^{-1}) were mixed with a FA solution (100 mg L^{-1}) in Teflon bottles for 40 h. Natural sediment particles were also used as a food source. Surficial oxidized sediments were collected from the Flax Pond salt marsh on Long Island, and sediment particles were sieved to 100 μm prior to the size selection process.

The particles (300 mg L^{-1}) were exposed to the radioisotope in the inorganic or organic form in the dark for 48 h at pH 8.0 in 0.2-μm filtered seawater (35‰) at 20 °C. Initial concentrations of added mercury (radioisotope plus stable mercury) were as follows: Hg = 2.2 μg L^{-1} (11.0 nM); CH_3-Hg(II) = 1.0 μg of Hg L^{-1} (4.7 nM). After radiolabeling, the particles were washed with filtered seawater, centrifuged to remove unbound metal, and resuspended into unlabeled filtered seawater. Partition coefficients (K_d) were calculated as the concentration of radioisotope g^{-1} particle divided by concentration of radioisotope mL^{-1} water (dissolved). The suspension of radioactive particles was immediately used for feeding to mussels.

Feeding Experiments. Mussels were individually placed in polyethylene beakers, each containing 3 mg of suspended, labeled particles in 600 mL of filtered seawater. Four replicate mussels were examined for each treatment. Mussels were fed radioactive suspended particles for a short period (20 min) to minimize the fraction of the available metal in the dissolved state and the cycling of the radiotracer. Radioactive particles consisted of clay, hydrous ferric oxides (HFO), hydrous manganese oxides (HMO), natural sediment (NS), silica (Si), and fulvic acid-coated particles (FA-clay, FA-HFO, and FA-Si). During the radioactive feeding period, it was possible that mussels accumulated radioisotopes desorbed from radioactive particles into the dissolved phase. The extent to which this occurred was determined by exposing a fresh batch of mussels for 20 min to the feeding solution, which was filtered (0.2 μm) to remove radioactive particles. Their radioactivity was determined as above, and values were subtracted from total exposure values in all calculations presented below.

After feeding, live animals were γ-counted to determine the amount of ingested label (γ counting has no impact on the viability of the mussels), and the mussels were then allowed to depurate their accumulated mercury. They were placed in an aquarium at 12 ± 1 °C and were fed non-radioactive diatoms *(Thalassiosira pseudonana)* to purge their guts of radioactive boluses of ingested sediment particles. Fecal pellets were collected frequently as they were produced, and aquarium water was changed to minimize cycling of the released radioisotopes. Whole animals were γ-counted non-destructively throughout the depuration period (4 d). Assimilation efficiencies of ingested metals were determined after 72 h of depuration when the guts of the mussels are empty of ingested radioactive sediment particles (*27*); this was confirmed with fecal pellet analyses. After depuration, mussels were dissected, and radioactivity was determined in the digestive gland, gills, foot, mantle, and shell.

Bioconcentration from Solution. Mussels were also exposed for 14 d to dissolved $^{203}HgCl_2$ and $CH_3{}^{203}HgCl$ to estimate their bioconcentration from solution. Five individuals were maintained at 12 ± 1 °C in polyethylene containers containing 4 L of seawater. Initial concentrations were 0.4 and 0.1 ng L^{-1} of inorganic Hg and methylmercury, respectively. To assess the impact of dissolved organic matter (DOM) on uptake of Hg from the dissolved phase, some containers of seawater were enriched with 5 mg L^{-1} FA; the initial DOM concentration in the seawater, analyzed with a Shimadzu TOC 5000 analyzer, was 1.25 mg L^{-1}. Animals were periodically counted for their radioactivity throughout the exposure period (14 d). The water in the containers was changed daily with freshly contaminated seawater. During the exposure, mussels were fed marine diatoms twice a day (1 h per feeding) in a separate aquarium containing uncontaminated seawater. At the end of the experiment, mussels were dissected as described above.

Desorption from Particles. Since the gut of *M. edulis* is acidic, with a pH of 5−6 (*34*), we examined the impact of low pH on the desorption of metals associated with food particles to simulate approximately the behavior of ingested mercury. Desorption experiments were performed by resuspension of labeled particles into unlabeled seawater adjusted to pH 5 with dilute HCl, following protocols described earlier (*27, 35*). The fractionation of mercury between dissolved and particulate phases (*36*) was determined over 24 h.

Radioactive Counting. The γ emissions of ^{203}Hg (279 KeV) in whole organisms were determined using a large well NaI-(Tl) γ detector interfaced to a Canberra 35+ multichannel analyzer. Activities in all other samples (water and filtered particles) were determined with an LKB Pharmacia Wallac 1282 Compugamma counter, also equipped with a NaI(Tl) well detector. The two instruments were intercalibrated daily with appropriate radioactive standards, analyses were decay-corrected, and samples were counted for sufficient time to yield propagated errors ≤6%.

Results

Mercury Adsorption onto Particles. The extent of metal adsorption to the nine particle types is presented as partition coefficients (K_d values), calculated at 48 h as dpm ^{203}Hg g^{-1} particle divided by dpm mL^{-1} dissolved (Figure 1). The adsorption of Hg(II) and $CH_3Hg(II)$ to particles was influenced

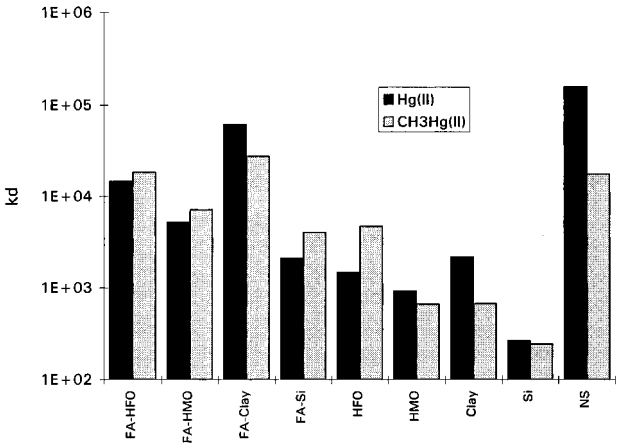

FIGURE 1. Partition coefficients (K_d) for Hg(II) and $CH_3Hg(II)$ for suspended particles. HFO, hydrous ferric oxide; HMO, hydrous manganese oxide; Si, silica beads; NS, natural sediment particles. FA prefix: particles coated with marine fulvic acids.

TABLE 1. Assimilation Efficiencies (%) of Inorganic and Methylmercury from Suspended Particles in Mussels[a]

particle	Hg	CH_3Hg
HFO	9 ± 6	27 ± 8
FA-HFO	7 ± 6	87 ± 1
HMO	8 ± 7	32 ± 6
FA-HMO	3 ± 2	36 ± 19
clay	5 ± 2	5 ± 4
FA-clay	1 ± 2	49 ± 12
Si		37 ± 6
FA-Si	9 ± 6	81 ± 5
NS	2 ± 2	55 ± 5

[a] Values are means of four individuals (±1 SD). HFO, hydrous ferric oxide; HMO, hydrous manganese oxide; Si, silica beads; NS, natural sediment; FA, fulvic acid coating.

by the type of particle and the particle coating. Adsorption onto silica particles was the lowest (K_d values $< 10^3$) and greatest on natural sediment particles (as high as 10^5). Generally, Hg and CH_3Hg displayed comparable K_d values. Organic coatings enhanced the adsorption of both Hg and CH_3Hg onto every type of particles ($P < 0.05$).

Assimilation Efficiencies. Figure 2 shows the retention over 4 d of the ingested mercury during the depuration period. The egestion of inorganic Hg was generally faster than egestion of the organic Hg. The assimilation efficiencies (AE), defined here as the proportion of an element remaining in the mussels after 72 h of depuration (*27*), are shown in Table 1 for each food type. In some cases, a small fraction of assimilated Hg was released from the mussels prior to 72 h (hence our AE values may be slight underestimates), but these amounts were insignificant relative to the variability in the data. AE values were very low for Hg(II) (1−9%), while they were typically >30% for $CH_3Hg(II)$ and as high as 87%. The organic coatings increased the AE values for $CH_3Hg(II)$ bound to all types of particles but had no significant effect on AE values for Hg(II).

No significant correlations were evident between desorption of Hg(II) from particles at pH 5 over a 1-h period and the assimilation efficiency of ingested Hg(II) in the mussels; however, $CH_3Hg(II)$ bound to uncoated particles showed a significant correlation ($r = 0.87$) between desorption and AE (Figure 3). Mercury desorption from organic-coated particles was generally low (<10%), and no significant correlation was noted for desorption from these particles and AE values.

Bioaccumulation from Solution. Figure 4 presents concentration factors (CF) of Hg(II) and $CH_3Hg(II)$ accumulated from solution in mussels over 14 d. CF (equal to dpm $^{203}Hg\ g^{-1}$ wet wt mussel divided by dpm $^{203}Hg\ mL^{-1}$ in solution) for the organic Hg (2×10^3) were twice as high as for inorganic Hg after 2 wk exposure. Bioaccumulation of dissolved Hg(II) appeared to slow down after 10 d, whereas the $CH_3Hg(II)$ CF continued to increase steadily at a rate of 160 d^{-1}. The addition of FA to the seawater decreased the CF of Hg(II) by 16% and that of $CH_3Hg(II)$ by 13%. The difference in bioaccumulation observed between these two Hg species in solution was much lower than differences observed for AE values from food (Table 1).

Tissue Distribution of Mercury in Mussels. The distribution of Hg in mussel tissues following exposure by different pathways is given in Table 2. For all mussels, regardless of the source, most of the Hg(II) and $CH_3Hg(II)$ were in the digestive gland and mantle. Large differences were not apparent in tissue distributions of Hg(II) or $CH_3Hg(II)$ between source terms (dissolved vs ingested) or between fulvic acid-coated particles and uncoated particles.

Discussion

The greater assimilation efficiencies of $CH_3Hg(II)$ than of Hg(II) from ingested abiotic particles in mussels are consistent with earlier studies involving marine bivalves feeding on phytoplankton (*17, 18, 37, 38*) as well other invertebrates (39) and fish (*40, 41*). In mussels inhabiting different sites with varying degrees of mercury contamination, the percentage of total mercury that is methylated can vary from 17% to near 100%, with the highest percentages of methylmercury in relatively unpolluted areas (*38, 42, 43*). The inorganic:methyl mercury ratios in mussels can in part be explained by the greater depuration rates of inorganic mercury than methylmercury (this study; *38*). Mercury concentrations in *M. edulis* also vary seasonally, reflecting the composition of suspended particulate matter (*44*) and in particular mercury complexation by particulate organic matter (*21*). However, the form of bioavailable mercury bound to sediment has not been directly investigated.

The low AE values observed here for inorganic Hg (ranging from 1% to 9%) are comparable to the findings (about 5%) of an earlier report for mussels feeding on phytoplankton (*38*). Given the experimental errors, it was not possible to distinguish differences in the low AE values for Hg(II) between the various types of particles. AE values in mussels for Cd and Ag bound to the same sediment particles are as high as 20%, and these metals desorbed from particles at pH 5 to a greater extent than Hg (Cd: 50−90%, Ag: 20−60%) (*8*). Mercury may be more strongly bound to organic matter, and this may explain the lower desorption of Hg from organic coatings. The release of metal from ingested particles in the acidic gut during digestion may be a primary control on the assimilation of metals. K_d values do not provide specific information on the behavior of particle-bound metals in an acidic gut, and thus the relationships between AE and K_d values are not expected to be linear (our observations gave r values < 0.7). The very low Hg desorption (<10%) under acidic conditions is evidence for the its binding strength to the ingested particles. Generally, it is thought that metals must be in solution within the digestive tract of an organism before they can be assimilated (*7*), and in fact, strong correlations between assimilation efficiency and metal desorption from organic-coated particles at a pH similar to that of a bivalve's gut were noted for Cd, Ag, and Co (*8*). As with these other metals, there is a correlation ($r = 0.87$) between AE and desorption for $CH_3Hg(II)$ bound to uncoated particles, despite their short residence times in the gut, indicating that this Hg species is more rapidly absorbed than inorganic Hg and other metals (Cd, Co, Ag). Little is known about the behavior of methylmercury in the digestive system of bivalves, but the digestive lining in fish has been reported to be permeable to methylmercury (*41*).

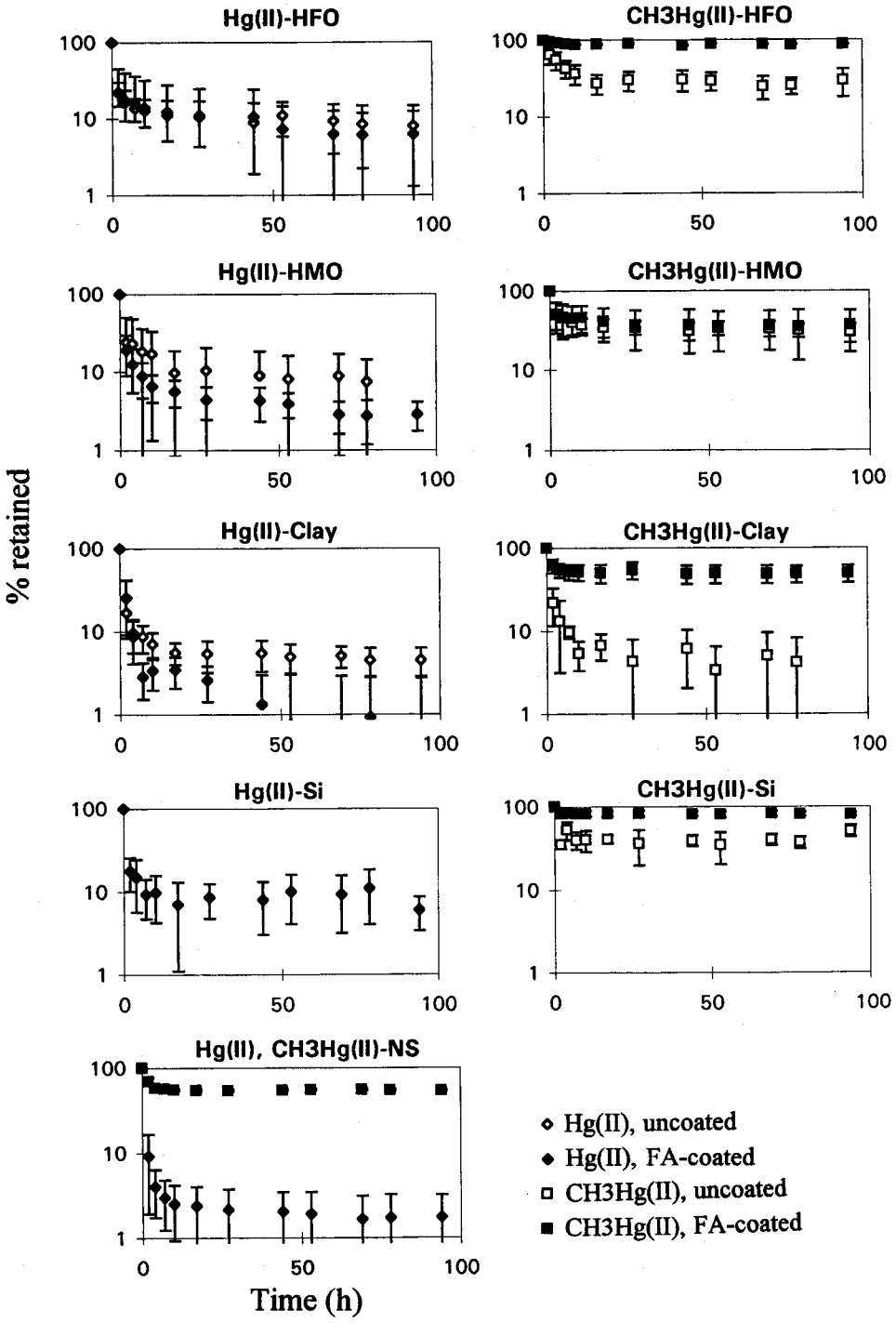

FIGURE 2. Retention of Hg(II) and $CH_3Hg(II)$ in mussels after ingestion of radiolabeled particles. HFO, hydrous ferric oxide; HMO, hydrous manganese oxide; Si, silica beads; NS, natural sediment particles; FA, fulvic acids. Data points are means of four individual mussels (±1 SD error bars).

Desorption of metals bound to particles is a time-dependent process. The residence time of an ingested particle in an animal's gut is a key factor that may control the assimilation of particle-bound metals (*8, 27, 29*). Wang and Fisher (*30*) found excellent correlations with several metals (Cd, Co, Ag, and Zn) between AE and gut passage time. Organic-coated particles may undergo intracellular digestion (relatively slow and intensive) whereas inorganic particles may only be processed by extracellular digestion (relatively rapid). This biphasic digestion is evident in the assimilation of some ingested metals in clams and mussels (*27, 45*). Low correlations between metal desorption and AE of Cd, Co, and Ag bound to inorganic particles were explained by short gut transit times (*8*). Consistent with these findings, no correlation between AE and desorption was observed for Hg(II) bound to inorganic particles.

Different AEs were observed for $CH_3Hg(II)$ among various types of particles having a coating of fulvic acid, despite the fact that $CH_3Hg(II)$ desorption at pH 5 was generally comparable for all coated particles. The amount of fulvic acid adsorbed to mineral particles depends on their adsorptive capacity, which probably varied among the particles types examined (but was not tested). If the extent of organic coating influences gut residence time in the mussel, as shown previously (*8, 29*), then the variations observed in AE for CH_3-Hg(II) from different particles may be related to variability in the gut residence times of the different particles.

TABLE 2. Mercury Distribution (%) in Tissues of Mussels Contaminated by Food and from Dissolved Phase[a]

	Hg(II)					$CH_3Hg(II)$				
treatment	gills	mantle	foot	gland	shell	gills	mantle	foot	gland	shell
HFO	2	30	1	63	4	17	34	10	32	7
HMO	15	35	9	16	25	7	38	6	40	8
clay	11	48	4	26	11	12	49	7	31	1
Si	nd	nd	nd	nd	nd	13	35	11	25	16
FA-HFO	12	36	2	51	0	5	39	6	46	4
FA-HMO	11	25	10	22	32	nd	nd	nd	nd	nd
FA-Clay	8	44	3	46	0	9	38	6	42	5
FA-Si	11	36	2	51	0	7	39	7	42	6
NS	5	42	3	38	13	8	40	9	38	5
dissolved[b]	4	48	4	38	5	14	55	11	16	4
dissolved + FA[c]	7	54	5	26	8	14	48	13	20	5

[a] FA, fulvic acid; nd, not determined. [b] Coastal water; 1.25 mg L^{-1} DOC. [c] Coastal water + 5 mg L^{-1} FA added.

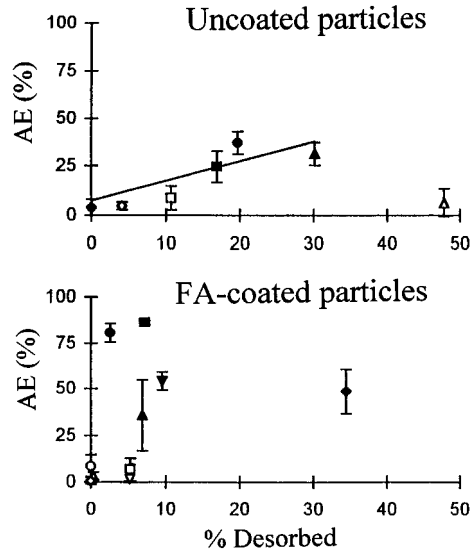

FIGURE 3. Relationship between the assimilation efficiency (AE), as percent, of ingested Hg(II) (open symbols) and $CH_3Hg(II)$ (solid symbols) in mussels and the fraction of Hg desorbed from radiolabeled particles into seawater at pH 5 after 1 h. Particles consisted of clay (◇), hydrous ferric oxides (□), hydrous manganese oxides (△), silica beads (○), and natural sediment (▽). The regression line drawn for uncoated particles is for $CH_3Hg(II)$; $y = 1.078x + 7.443$, $r = 0.87$. All other correlations were not significant. FA, fulvic acid.

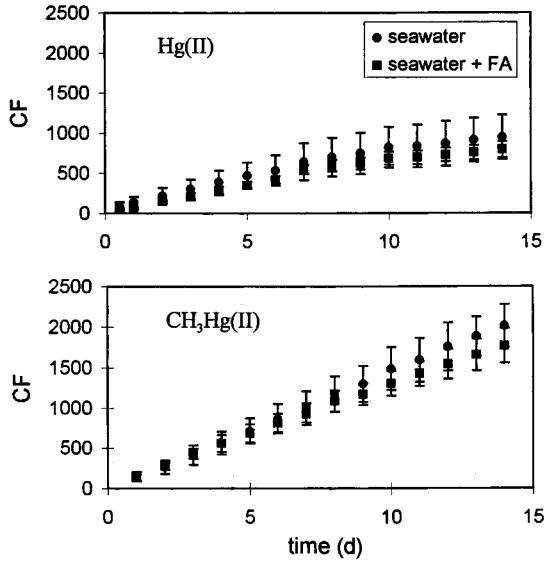

FIGURE 4. Concentration factors (CF) in mussels of Hg(II) and CH_3-Hg(II) accumulated from the dissolved phase. Initial concentrations were 0.4 and 0.1 ng L^{-1} for Hg(II) and $CH_3Hg(II)$, respectively. Data points are means of five individual mussels (±1 SD error bars). FA, fulvic acid.

Unlike Ag, Cd, and Co, correlations were weak between AE and metal desorption from organic-coated particles for both $CH_3Hg(II)$ and Hg(II). The desorption of Hg(II) and $CH_3Hg(II)$ bound to organic-coated particles was not appreciably affected at pH 5 (<10%). The stability of Hg binding to organic coatings under acidic pH conditions may explain the poor correlation between metal desorption and AE. Complexation of released Hg(II) in solution by dissolved FA may influence its assimilation, as indicated in Figure 4. Observations from a study on thiol complexation (*46*) suggest that Hg(II) would be more organically complexed than methylmercury in seawater and may also help explain the low AE of Hg(II) from coated particles. High assimilation of $CH_3Hg(II)$ despite low desorption suggests that metal desorption from organic coatings under acidic conditions does not control the assimilation of $CH_3Hg(II)$ in mussels. Solubilization under other digestive conditions, including surfactants in the digestive fluids, may explain the assimilation of $CH_3Hg(II)$. Bivalve crystalline styles have a powerful emulsifying activity (*47*), and experiments with crystalline style extract from *Mytilus edulis* showed that this material can detach microorganisms from sediment particles (*48*). These surfactants might also increase lipophilic contaminant exposure to these animals, as suggested for some deposit feeders exposed to contaminated sediments (*49*).

Although ingestion of sediment-bound $CH_3Hg(II)$ appears to be an important route of exposure, both dissolved and particulate sources must be considered when evaluating the bioavailability of a contaminant to aquatic organisms. Bivalves can potentially accumulate metals from solution as well as from ingested particles (*50*). Our results show that uptake of both dissolved and particulate Hg species can occur and that the dissolved phase may be a significant source for mussels. This finding, based on experiments using low, environmentally realistic Hg concentrations, is similar to results from some (*18, 19*) but not all (*37*) earlier studies which used much higher Hg concentrations, suggesting that over a broad range of Hg concentrations the dissolved phase is likely to be an imporant source of Hg for mussels. Dissolved organic matter such as fulvic acids can complex Hg (*51, 52*) and therefore decrease its bioavailability from the dissolved phase (Figure 4; *16, 53*).

Whereas differences in assimilation efficiencies were substantial between the two particulate Hg species, the concentration factor (10^3) of Hg(II) in mussels exposed to the dissolved species at these low concentrations was only half that of dissolved $CH_3Hg(II)$. This observation confirms that mechanisms of absorption differ between the uptake pathways. Redistribution of Hg and $CH_3Hg(II)$ within the mussel

after assimiliation may make it impractical to use mussel tissue distributions to distinguish the sources of Hg uptake for mussels in the field.

In conclusion, sediment-bound methylmercury can be an important source of Hg to suspension feeding mussels and appears to be assimilable from all major sedimentary components. Organic coatings favor the assimilation of CH_3-Hg(II) from ingested particles; inorganic Hg(II) bound to sediments is assimilated at a much lower efficiency. Dissolved mercury—inorganic and organic—is concentrated very appreciably by mussels and dissolved Hg(II), relatively abundant as compared to $CH_3Hg(II)$, and should be considered a potential source for mussels. However, in turbid coastal waters, dissolved mercury commonly comprises only a small proportion of the total mercury in the water. Although humic materials can help maintain mercury in the dissolved phase, a major fraction of the most accumulable species, $CH_3Hg(II)$, in marine and estuarine waters can be primarily associated with suspended particles. Particulate sinking is the ultimate removal mechanism of dissolved $CH_3Hg(II)$ from waters (*13*, *23*). The high AE of particulate $CH_3Hg(II)$ and the physical speciation of $CH_3Hg(II)$ in waters suggest that particulate CH_3-Hg(II) can be a major source of Hg to mussels.

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