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Behavior of mercury in the Patuxent River estuary

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Abstract. An overview of a comprehensive study of the behavior and fate of mercury in the estuarine Patuxent River is presented. Total Hg (Hg_T) and methylmercury (MeHg) exhibited weakly non-conservative behavior in the estuary. Total Hg concentrations ranged from 6 ng L^{-1} in the upper reaches of the sub-urbanized tidal freshwater river to $<$0.5 ng L^{-1} in the mesohaline lower estuary. Filterable (0.2 μm) Hg_T ranged from 0.2 to 1.5 ng L^{-1}. On average, MeHg accounted for $<$5% of unfiltered Hg_T and $<$2% of filterable Hg_T. Dissolved gaseous Hg (DGHg) concentrations were highest (up to 150 pg L^{-1}) in the summer in the mesohaline section, but were not well correlated with primary production or chlorophyll a, demonstrating the complex nature of Hg^0 formation and cycling in an estuarine environment. Organic matter content appeared to control the Hg_T content of sediments, while MeHg in sediments was positively correlated with Hg_T and organic matter, and negatively correlated with sulfide. MeHg in sediments was low (0.1 to 0.5% of Hg_T). Preliminary findings suggest that net MeHg production within sediments exceeds net accumulation. Although Hg_T in pore waters increased with increasing sulfide, bulk MeHg concentrations decreased. The concentration of MeHg in sediments was not related to the concentration of Hg_T in pore waters. These observations support the hypothesis that sulfide affects the speciation and therefore bioavailability of dissolved and/or solid-phase Hg for methylation. Comparison with other ecosystems, and the negative correlation between pore water sulfide and sediment MeHg, suggest that sulfide limits production and accumulation of MeHg in this system.

Introduction

Mercury contamination of estuaries is a concern because estuaries provide unique habitats for a number of species, and because they are often heavily used for commercial and recreational fishing. These coastal ecosystems often lie in close proximity to major population centers, so they may be strongly impacted by human activities. In addition, estuaries provide a link between the marine and terrestrial environment, therefore, understanding the behavior and fate of Hg in estuaries has implication for larger scale Hg biogeochemical cycles.

With the advent of non-contaminating techniques in the late 1980's, it has been possible to measure Hg concentrations in coastal waters, and a number of recent studies have examined concentration patterns in estuaries (Cossa et al. 1988; Mason et al. 1993; Leemakers, et al. 1995; Vandal & Fitzgerald 1995; Stordal et al. 1996; Gagnon et al. 1996). In this study, we examine the behavior of Hg species along an estuarine gradient, focusing on processes that control their distribution in the water column and sediments, including the biogeochemical control of Hg and MeHg patterns in sediment; efflux from and deposition to sediments; Hg^0 volatilization; and Hg methylation.

Study site

The Patuxent River is a two layer coastal plain estuary, and a subestuary of Chesapeake Bay, USA. Located in southern Maryland, much of its upper watershed lies within the suburbs of Washington and Baltimore (Figure 1). The area of the drainage basin is 2.39×10^9 m^2, and 38% of this area is developed (USEPA 1994). The surface area of the estuary, defined here as the span from the limit of tidal influence to the mouth, is 1.36×10^8 m^2. The estuary volume is 6.52×10^8 m^3 (Cronin & Pritchard 1975), the average freshwater inflow rate for 1977 through 1994 was 3.3×10^8 m^3 y^{-1} and the freshwater replacement time ranges seasonally from 30 to 100 d (Hagy 1996). However, the inflow of higher salinity bottom water from Chesapeake Bay represents the largest portion of the water budget for the Patuxent, with flow across the mouth of the river about ten times higher than freshwater flow (Hagy 1996). The river is seasonally stratified as far north as XED 4892, where a sill results in upward mixing of saline bottom waters, and a turbidity maximum. The river is generally saline to PXT 0402 and tidal to PXT 0494; this area of the river is surrounded by tidal marshes.

Methods

Non-contaminating techniques (e.g. Gill & Fitzgerald 1987; Flegal et al. 1991) were employed during all stages of sample collection, handling and analysis for Hg. Water column samples were collected either by boat or from shore by peristaltic pumping through acid-leached Teflon® and C-Flex® tubing. Filtration was performed in the field using a precleaned in-line 0.45 μm Calyx® capsule filter. Sediment samples were collected with a non-metallic Soutar box corer, and subcores were taken in acid-cleaned acrylic or PVC core tubes. Cores were extruded and sectioned in an O_2-free glovebox, and

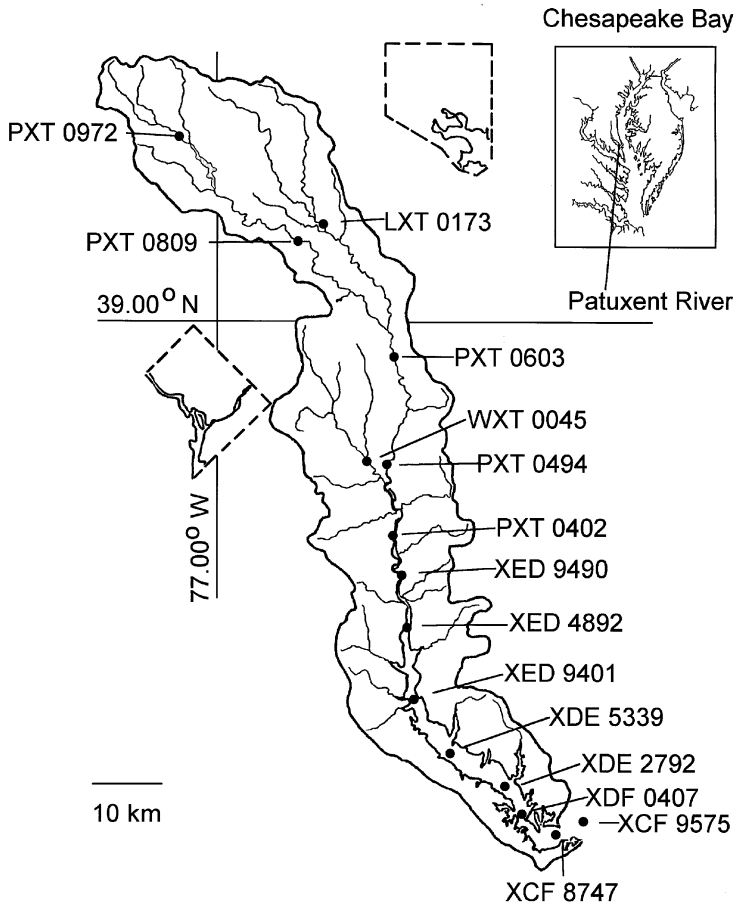

Figure 1. Map of the Patuxent River and its watershed showing the sampling stations used for this study.

porewaters were separated via vacuum filtration in precleaned 0.2 μm Nalge polycarbonate filter units with cellulose nitrate filters.

Water samples were analyzed for Hg_T following the method of Gill & Fitzgerald (1987) and Bloom & Fitzgerald (1988). Sediment samples were digested with a 5:2 mixture of HNO_3:H_2SO_4 prior to analysis. Dissolved gaseous Hg (DGHg) was determined within six hours of sample collection (Mason et al. 1993). MeHg analysis in both waters and sediments was carried out by distillation (Horvat et al. 1993a,b), then aqueous phase derivitization (Bloom 1989).

The detection limit for MeHg in aqueous samples, based on $3\times$ the standard error of sample blanks and 150 ml sample size, averaged 20 pg L^{-1}.

The average recovery of MeHg spikes (at 100 pg spike per 150 ml sample) was 80±26% (n = 21). Duplicate analysis of 17 samples yielded an average relative percent deviation (RPD) of 28. The detection for Hg_T in aqueous Patuxent River samples of about 100 ml averaged 0.14 ng L^{-1} in 1994 and 0.07 in 1995–1997 in unfiltered samples, or 0.14 and 0.16 for filtered samples in the same years. Spike recoveries for inorganic Hg (100 pg per 100 ml sample) averaged 103±30% (n = 21) in 1995 and 87±12% (n = 7) 1996. Analysis of 18 samples in duplicate yielded an average RPD of 6.3±7.4%. Details on SRMs, intercalibrations, and quality assurance for analysis of total and methyl Hg sediments can be found in Gilmour et al. (this volume).

Sulfide subsamples from porewaters were preserved in sulfide anti-oxidant buffer and measured using an ion specific electrode. Sulfate was measured by ion chromatography. The organic matter content of sediments was estimated from loss on ignition (LOI). Sediment/water fluxes were measured in large (0.5 m^3) continuous flow mesocosms, as described in Sanders & Riedel (1993). This design provides a measure of diffusive plus non-diffusive flux, as benthic infauna were present and water was moving over the sediment surface. The large chamber volume also minimizes sorption of Hg to surfaces. Mesocosms contained 0.22 m^2 sediment trays filled with multiple intact sediment box cores. Water in the mesocosms was replaced at 10% per day with 1 μm filtered Patuxent River water and was bubbled with air. Four mesocosms were set up for each sediment site, plus four water only controls. Net flux was calculated from steady-state dissolved concentrations relative to no-sediment control chambers.

Results and discussion

Distribution of total and methylmercury in the water column

The distribution of filtered and unfiltered Hg_T and MeHg in surface water in July 1993 is plotted against station in Figure 2. There was a decline in both filtered and unfiltered Hg_T concentration moving downriver, however, the unfiltered concentration declined more rapidly, so that the filterable fraction tended to increase. In general, a greater proportion of the MeHg was filterable compared to Hg_T. The particle-water partition coefficient (K_D, L kg^{-1}) ranged from 6×10^4 to 5×10^5 for Hg_T and from 7×10^3 to 1×10^4 for MeHg, falling within the fairly wide range seen in other estuaries (Leermakers et al. 1995; Stordal et al. 1996).

MeHg concentrations also decreased downriver, but on this date increased dramatically at station XEF9575, which is in the main bay. MeHg constituted $<$5% of the total filterable Hg_T except at the mouth of the river, where it

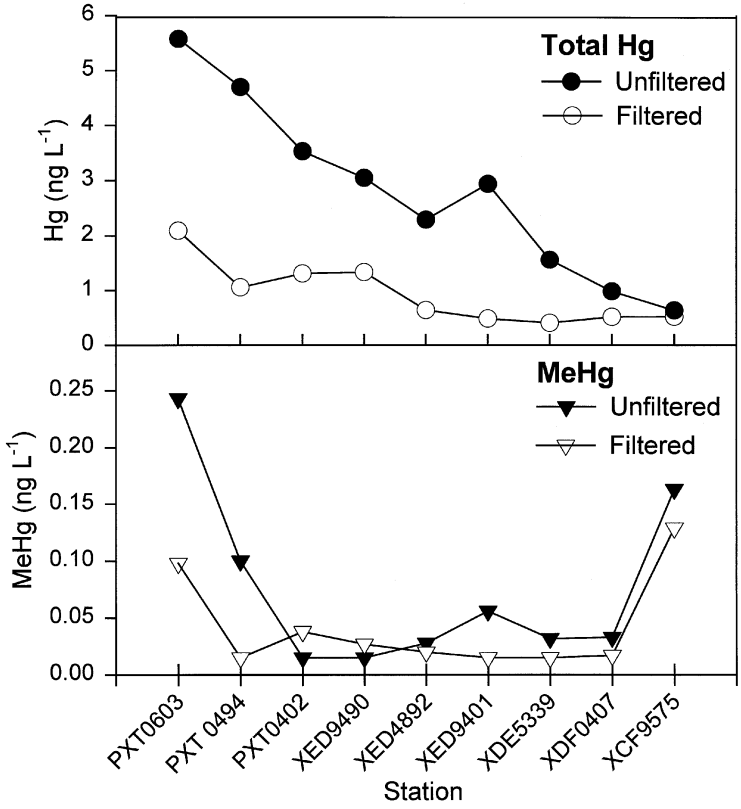

Figure 2. Surface water Hg_T and MeHg concentrations, 7/2/93, along the main river plotted by station from freshwater at the left to the estuarine Chesapeake Bay at the right. The average errors between duplicates for this data set were 150 pg Hg and 36 pg MeHg L^{-1}.

increased to 25%. High levels of MeHg have also been found at the mouth of the Scheldt Estuary (Leermakers et al. 1995). We speculate that the high MeHg concentration at this station may result from upward mixing or tilting of anoxic bottom water, which is present in the mainstem of the Chesapeake Bay during fully stratified summer conditions. An increase in dissolved MeHg in anoxic bottom waters in stratified lakes has been commonly observed (Bloom et al. 1991; Watras et al. 1995; Henry et al. 1995). However, movement of anoxic Chesapeake bottom waters into the Patuxent may occur only during wind-driven tilting events. The degree of Hg and MeHg enrichment in Chesapeake Bay bottom waters is poorly characterized.

The somewhat non-conservative behaviors of both Hg_T and MeHg are illustrated in Figure 3. The Hg_T concentration and distribution pattern along the estuary is similar to that seen in other moderately contaminated estuaries such as the Scheldt (Leermakers et al. 1995); Galveston Bay (Stordal et al.

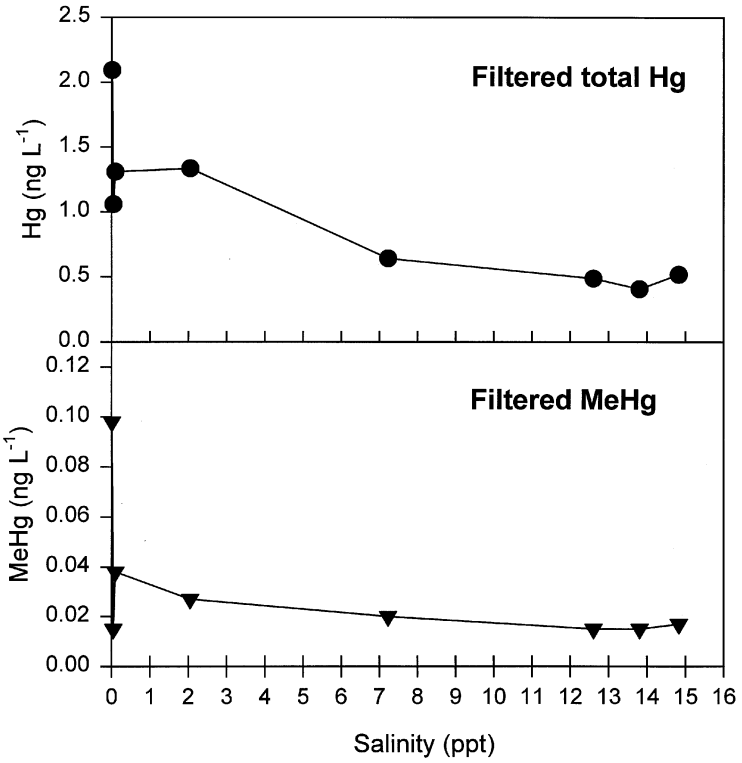

Figure 3. Filterable Hg_T and MeHg concentration plotted against salinity for 7/2/93. The points at zero salinity represent three stations above the upper limit of salinity.

1996) and the St. Lawrence (Cossa et al. 1988). Maximum Hg removal occurs in these estuaries at <7–10 ppt salinity. However, estuaries with different mixing patterns exhibit different Hg distributions (Coquery et al. 1995; Stordal et al. 1996). In addition, given the relatively long residence times of water in the Patuxent, the saline end members may not be reflective of current freshwater inputs.

Total Hg concentrations through time (1995–1996) are shown for three stations in Figure 4. These distributions reinforce the spatial trend measured in July 1993. Particulate Hg was removed rapidly from the oligohaline into the mesohaline section. Filterable Hg_T accounted for 35% of the unfiltered Hg_T on average at the mesohaline station, but only about 15% at the tidal freshwater station. During the sampling period, Hg_T concentrations were highest during high water flow in the winter and spring. A period of exceptionally high flow occurred in winter 1996 (USGS Surface Water Information Dept., Towson, MD). The average concentration of filterable MeHg at these three stations in the estuary did not exceed the detection limit of 20 pg L^{-1} (data not shown).

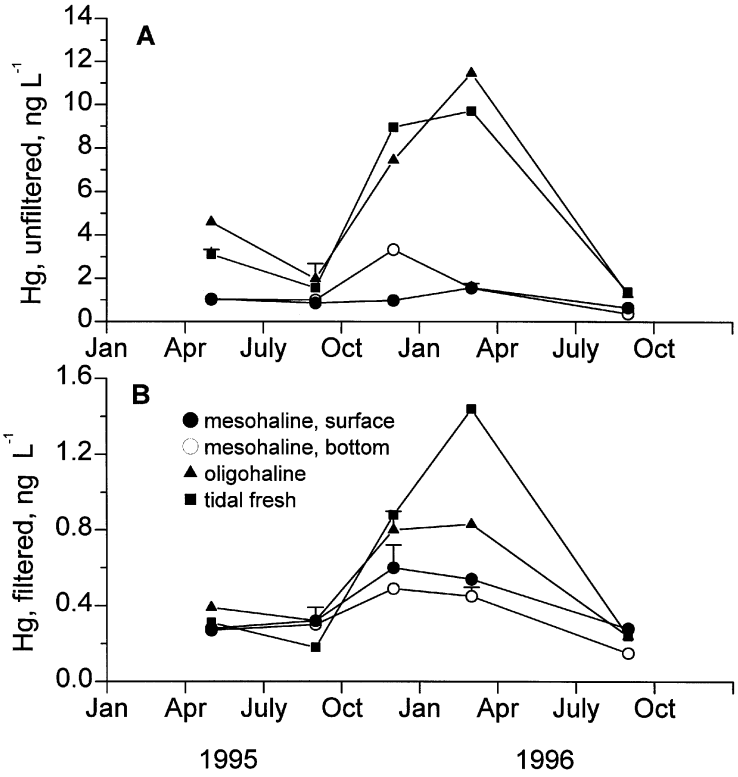

Figure 4. Unfiltered (A) and filterable (B) Hg_T concentrations at three Patuxent River stations during five sampling periods in 1995 and 1996. Error bars represent errors between duplicates; points without error bars represent single measurements, or errors less than the size of the symbol. The average errors between duplicates for this data set were 60 and 76 pg Hg L^{-1} for filtered and unfiltered samples, respectively. All are surface water concentrations except that a 6 m sample was also taken at the mesohaline station, which undergoes salinity stratification and periodic anoxia in the summer.

Unfiltered MeHg was only slightly higher, averaging 25 pg L^{-1}, and only exceeding 50 pg L^{-1} in one instance. The percent MeHg in filtered samples averaged $<5\%$, in unfiltered samples $<2\%$.

Dissolved gaseous Hg (DGHg) concentrations, shown for three stations in Figure 5, varied both spatially and temporally in the estuary. During the winter months, DGHg was near the detection limit at all of the sites. Similarly, our Hg^0 measurements throughout the Chesapeake Bay in February 1997 were all less than 40 pg L^{-1}. These results suggest low Hg^0 production during the winter months. Intermediate concentrations were observed in the spring and fall, but high concentrations were found at XED 5339 in the summer of 1995. In contrast, our measurements in summer 1996 (August; 13 pg L^{-1} at

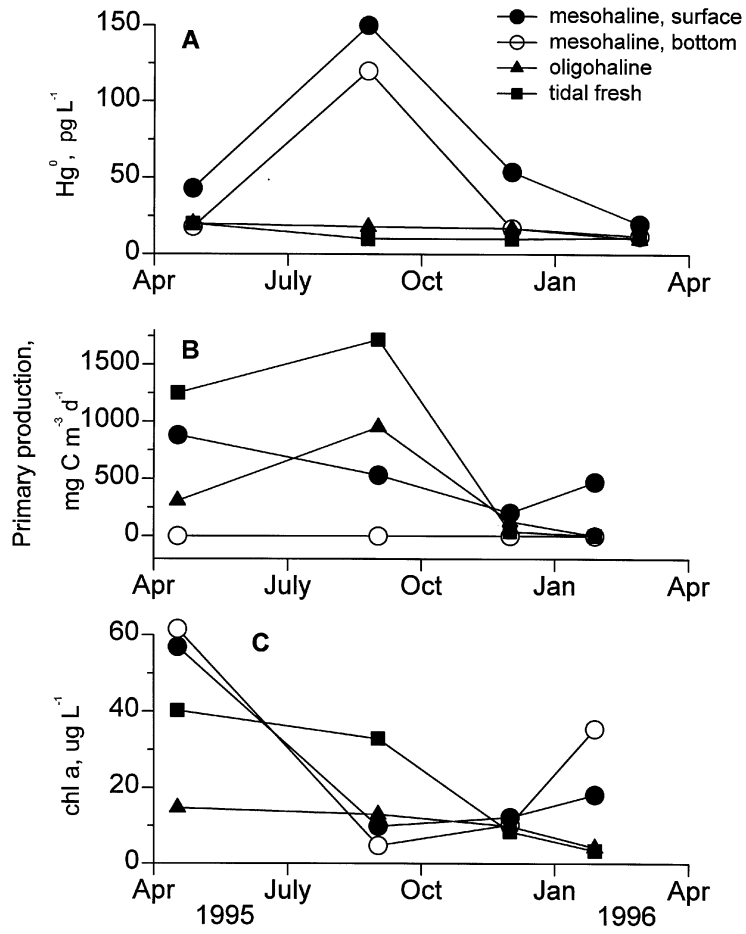

Figure 5. Dissolved gaseous Hg concentrations (A) primary production (B) and chlorophyll a (C) for three estuarine stations on four dates in 1995 and 1996. Stations as in Figure 4. Primary production and chlorophyll from Lacouture et al. 1996.

XDE 5339 and 26 pg L^{-1} at XED 4892) were much lower. Comparison of DGHg with primary production and chlorophyll a (Figure 5) suggests that DGHg production is not directly linked to primary production. However, DGHg production might be linked to the activity of specific types of algae, or to bacterial activity, as has been observed elsewhere, and in accord with experimental work on phytoplankton reducing capacity (Mason et al. 1995). Results from the Patuxent demonstrate the complex nature of Hg^0 formation and cycling in an estuarine environment compared to the open ocean where biotic processes play a more dominant role in Hg^0 production (Mason et al. 1995).

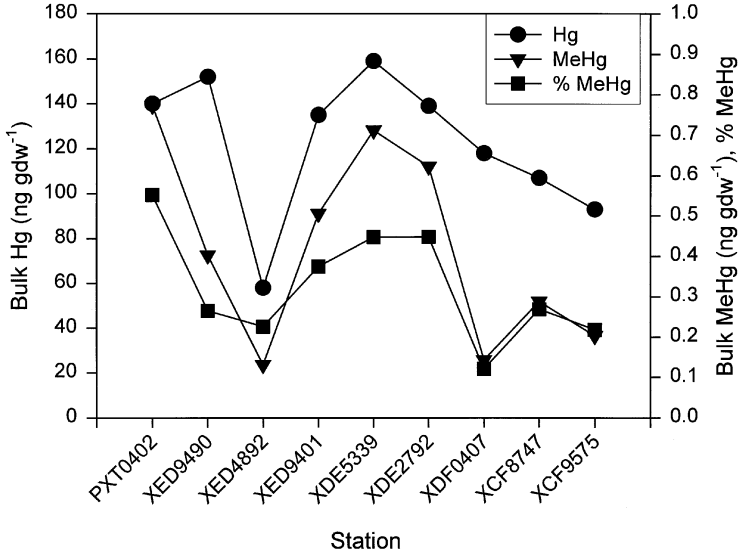

Figure 6. Bulk Hg_T and MeHg concentrations in estuarine sediments. The average concentrations in the top 4 cm of sediment taken in Sept. 1995 are shown on a dry weight basis, along with MeHg as a percentage of Hg_T (%MeHg).

Distribution of total and methylmercury in sediments and sediment pore waters

The distributions of Hg_T, MeHg, and MeHg as a percent of Hg_T (%MeHg) in the solid phase surficial sediment are shown in Figure 6 for the estuarine stations. Moving down estuary, there was a general decreasing concentration trend in all, punctuated by a dip at XED 4892, which is an area of sandy sediments. The bulk Hg_T concentration distribution followed trends in organic matter (OM) content, and the two parameters were moderately correlated (r^2 = 0.54). When normalized to organic matter, Hg_T increased slightly moving toward the bay (Figure 7). This distribution is consistent with removal of dissolved Hg_T from the water column via scavenging onto organic particles and deposition, as has been described (e.g. Cossa et al. 1988). The lower Hg:OM ratio in lower salinity sediments might reflect salinity-driven differences in Hg scavenging, changes in the Hg:C ratio during remineralization of sedimented OM, or the dilution in the upper river from relatively large OM inputs. The OM normalized MeHg concentration shows no distinct trend and exhibits a more irregular shape. However, MeHg was significantly correlated with Hg_T, as discussed below.

The porewater distribution of Hg_T, MeHg and sulfide along the salinity gradient of the estuary exhibited some suggestive trends (Figure 8). A small

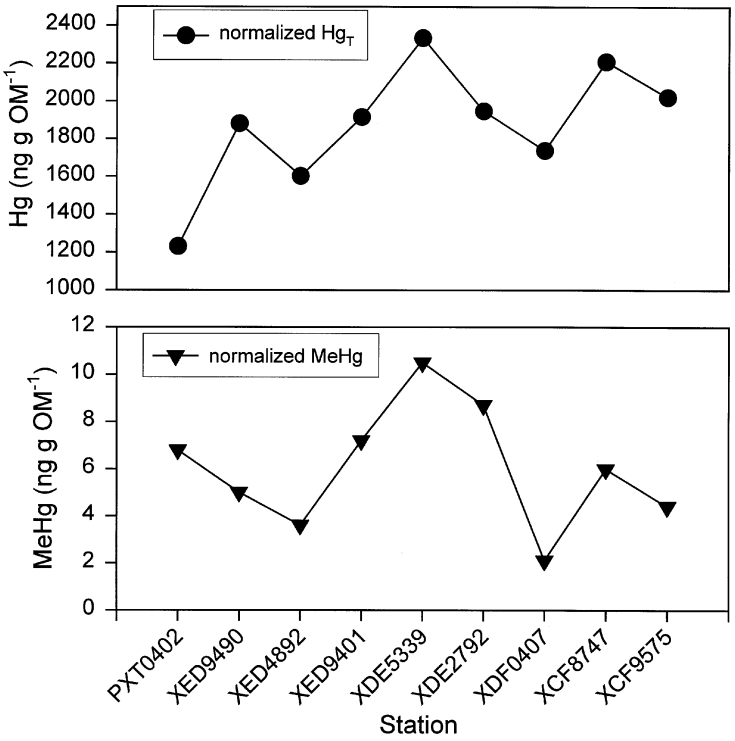

Figure 7. Bulk Hg_T and MeHg concentrations normalized to organic matter content. The concentrations were converted using the %LOI measured in sediments at each site. It was assumed that all of the loss on ignition was attributable to destruction of organic matter.

peak in porewater Hg_T and MeHg occurred at the three up-estuary stations which had low organic matter content (XED 9490, XED 4892, and XED 9401), but porewater MeHg was undetectable (D.L. = 0.03 ng/L) at all other sites. In contrast, the highest porewater Hg concentrations were observed at the stations with elevated sulfide concentrations, and across sites pore water Hg was significantly correlated with dissolved sulfide (r^2 = 0.69). This result suggests that the presence of excess sulfide increases the solubility of Hg(II), which is consistent with previous models of cinnabar dissolution (e.g. Schwarzenbach & Widmer 1963; Dyrssen & Wedborg 1991). Other mechanisms may increase the solubility of Hg(II) under anoxic conditions, such as release of Hg coprecipitated with Fe oxyhydroxides (Gobeil & Cossa 1993), but sulfide control of Hg dissolution appears to be an important mechanism in the Patuxent.

Relative to many lakes and wetlands (Gilmour et al. 1992; St. Louis et al. 1994; Verta & Matilainen 1995), %MeHg in estuarine sediments is generally

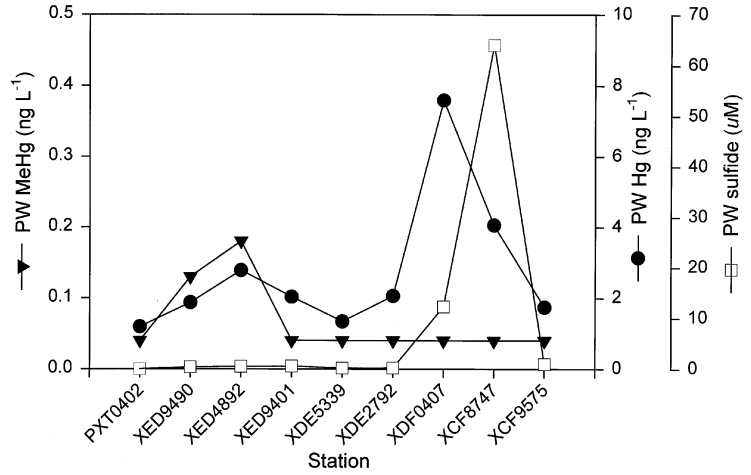

Figure 8. Porewater Hg_T, MeHg, and sulfide concentration in estuarine sediments. The porewater concentration (passing through a 0.2 μm filter) in the top for 4 cm of sediment is shown. Undetectable MeHg concentrations were plotted at the detection limit (0.03 ng L^{-1}).

low, <0.5% (e.g. Andren & Harris 1973; Bartlett & Craig 1981; Gobeil & Cossa 1994; Gagnon et al. 1996). The Patuxent follows this trend with 0.3% MeHg on average for the estuarine sediments. Because sulfate-reducing bacteria (SRB) are important mediators of methylation (Compeau & Bartha 1985; Gilmour et al. 1992), a general relationship between %MeHg and sulfate reduction rate has been postulated for aquatic sediments (Gilmour & Henry 1991). At relatively low sulfate concentrations, sulfate stimulates both sulfate-reduction and Hg methylation. However, at high sulfate concentrations, methylation rates and %MeHg are often low, probably due to inhibition of methylation by sulfide (Compeau & Bartha 1985; Choi & Bartha 1994). Figure 9 shows %MeHg as a function of sulfate concentration for a number of aquatic systems we have studied, including the Patuxent.

The factors most related to %MeHg in Patuxent sediments were Hg_T, sulfide, and organic carbon. In this estuary there was no significant relationship between porewater sulfate and %MeHg. The significant positive correlation (r = 0.78, α = 0.05) between Hg_T and MeHg concentrations (Figure 10) suggests that they are co-deposited or that MeHg production *in situ* is a function of bulk Hg_T concentration. Organic matter was correlated with both Hg_T and MeHg, which may reflect the affinity of both for depositing organic matter. In addition, organic material in sediments may stimulate bacterial activity and hence methylation. MeHg in both solid phase and porewater was highest when dissolved sulfide was lowest (compare Figures 6 and 8).

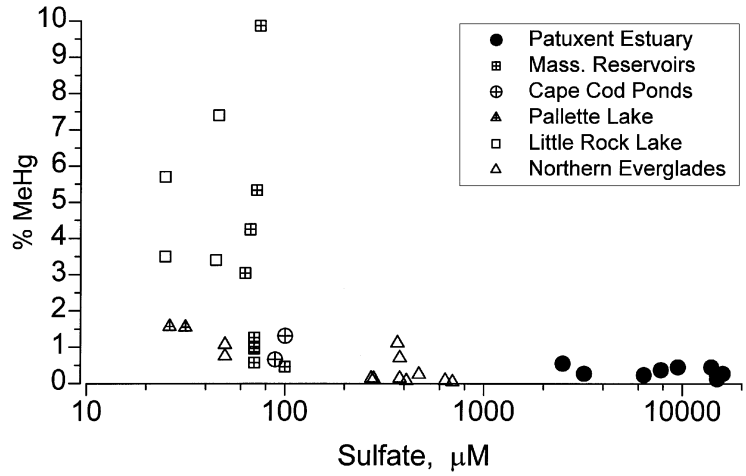

Figure 9. The relationship between sulfate concentration in surficial (0–4 cm) sediment porewaters and %MeHg in the bulk phase of surficial (0–4 cm) sediments from a number of aquatic environments, including the estuarine Patuxent River. Data from Gilmour et al. 1992, 1997; Gilmour & Riedel 1995; and unpublished data.

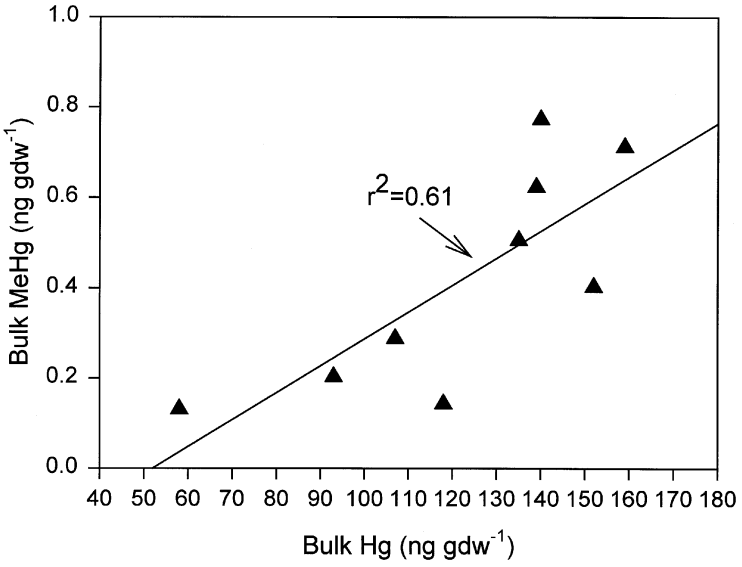

Figure 10. Bulk Hg_T versus bulk MeHg in estuarine sediments of the Patuxent river. Data were derived from Figure 6.

Reduction in the solubility of Hg(II) via precipitation of $HgS_{(s)}$ has been suggested as the mechanism for sulfide inhibition of methylation (Compeau & Bartha 1985; Choi & Bartha 1994). Since the dissolved concentration of Hg_T

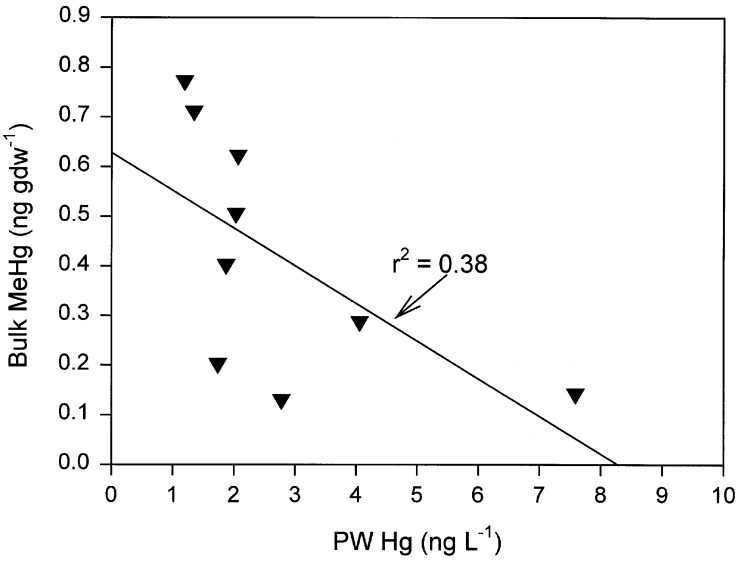

Figure 11. The relationship between porewater Hg_T concentration and %MeHg in the bulk phase of estuarine sediments of the Patuxent River. Data were derived from Figures 6 and 8.

in these sediments *increases* with increasing dissolved sulfide, we speculate that other mechanisms are more important. According to previous models of Hg-sulfide complexation (e.g. Swarzenbach & Widmer 1963; Dyrssen & Wedborg 1991), the speciation of Hg at higher sulfide concentrations shifts away from the HgS^0 complex towards disulfide complexes (mainly $HgHS_2^-$ at pH 7.0). We hypothesize that HgS^0 may be more available for microbial methylation because it more readily diffuses across cell membranes compared to disulfide complexes. Formation of charged Hg-disulfide complexes may therefore inhibit Hg methylation under sulfidic conditions.

If total dissolved Hg is the substrate for methylation, a positive relationship between porewater Hg_T and bulk MeHg might result, but this was not the case in Patuxent sediments (Figure 11). In addition to shifts in dissolved Hg speciation with sulfide concentration, another possible explanation for this observation is availability of solid-phase Hg for methylation, i.e Hg uptake is not mediated by dissolution. The significant positive relationship between bulk Hg_T and MeHg concentrations supports this idea. Alternatively, the lack of relationship between porewater Hg_T and MeHg could also result if the uptake of dissolved Hg by bacteria in close proximity to particles is rapid relative to its rate of release from the solid phase. In this case, uptake rate will depend on dissolved Hg, but the ambient porewater concentrations will not reflect the concentration at the cell surface.

Net methylation

A limited number of measurements of net MeHg production within Patuxent surficial sediments suggest that methylation is sufficient to support ambient MeHg concentrations. Net MeHg production within sediments was estimated by incubating unamended, intact sediment cores over 15 days, as described in Gilmour & Riedel (1995). The concentration of MeHg in sediments after incubation, relative to the initial concentration, is a measure of MeHg production plus MeHg degradation (demethylation). Net methylation in the top 4 cm of Baltimore Harbor sediments sampled in April 1992 was 4.9±0.7 pg (g wet weight)$^{-1}$ d^{-1} (n = 3); from XED 4892 and PXT 0402 sediments sampled in May 1993, 2.0±.2 and 1.6±0.4 pg (g wet weight)$^{-1}$ d^{-1} (n = 5). Replicates represent individual sediment cores.

Annual MeHg production in river sediments was estimated by applying the net methylation rate from PXT 0402 and XED 4892 to the depositional area of the upper and lower sections of the Patuxent, respectively, for 182 d y^{-1}. Using sediment deposition rates of 4.9 and 2.5 kg m^2 y^{-1} (Boynton et al. 1995) for the upper and lower river, and an average dry weight for surficial sediments of 25%, MeHg production was estimated at 450 g y^{-1}. Annual net MeHg burial was estimated by multiplying measured summer MeHg concentrations, of 0.44 and 0.30 ng gdw^{-1} for the upper and lower Patuxent, by annual deposition rates, yielding 110 g y^{-1}. By this estimate, MeHg production within sediments exceeded burial by about a factor of four. If the estimate is correct, a removal mechanism other than demethylation or burial for MeHg from sediments must be present. However, the estimates of net MeHg sedimentation and removal presented here are very preliminary.

Sediment efflux

Sediments provide a potential source of Hg_T and MeHg to the water column through the diffusive flux of dissolved species as well as bioaccumulation through the benthic food web or porewater advection. In the summer of 1996, the sediment-water flux of Hg_T and MeHg was measured in mesocosms containing sediments collected near station XDE 2792 in the Patuxent and from Baltimore Harbor. Hg_T concentrations in water-only control tanks were about 100 pg L^{-1} throughout the study while MeHg was undetectable at the D.L. for this experiment, which was 20 pg L^{-1}, based on three times the standard deviation of the filtration blanks. Flux of Hg_T from Patuxent River sediments was 59±7 ng and 89±5 ng m^{-2} d^{-1} after 5 and 21 days, respectively, while flux from Baltimore Harbor sediments was greater, 130±11 and 169±20 ng m^{-2} d^{-1}. These experiments suggest that direct flux of dissolved

Hg may be a small source to the river, but that flux of dissolved MeHg from sediments is not an important source.

Conclusions

The distribution of Hg in the Patuxent estuary reflects both external inputs and internal transformation. The DGHg distribution demonstrates the complex nature of Hg^0 formation and cycling in the estuary. Scavenging by organic matter appears to control the removal of Hg(II) from the water column and explains the weakly non-conservative behavior of Hg_T and MeHg in the estuary. As such, bulk Hg_T was strongly correlated with the OM content of sediments. MeHg in sediments was low (0.1 to 0.5% of Hg_T), positively correlated with both OM and Hg_T, and negatively correlated with sulfide. A preliminary comparison of net methylation rates with burial rates suggests that new production exceeds sedimentation, and that there is significant removal of MeHg from sediments. Since net MeHg production within sediments appeared to exceed net accumulation, co-distribution of Hg_T and MeHg suggests that *in situ* production is a function of total substrate (Hg_T) concentration. However, Hg speciation also appears to play a role in MeHg production and distribution. Although dissolved Hg_T increased with increasing sulfide in sediment porewaters, bulk MeHg concentrations decreased. This observation supports the hypothesis that sulfide affects the speciation and therefore bioavailability of dissolved and/or solid-phase Hg(II) for methylation.

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